From: Gildersleeve, Melissa (ECY)
Sent: Friday, May 04, 2012 4:42 PM
To: Niemi, Cheryl (ECY)
Subject: FW: Invitation to Fish Consumption Rate Meeting

fyi

From: Laurie, Tom (ECY)
Sent: Friday, May 04, 2012 4:38 PM
To: Sturdevant, Ted (ECY)
Cc: Pendowski, Jim (ECY); Susewind, Kelly (ECY); Gildersleeve, Melissa (ECY); Hankins, Martha (ECY)
Subject: Re: Invitation to Fish Consumption Rate Meeting

Good set-up

Tom Laurie
(360)407-7017 desk
(360)790-4110 cell

From: Sturdevant, Ted (ECY)
Sent: Friday, May 04, 2012 04:30 PM
To: Gary Chandler <GaryC@AWB.ORG>; Courtney Barnes <courtneyb@AWB.ORG>; chris@nwpulpandpaper.org <chris@nwpulpandpaper.org>; fholmes@wspa.org <fholmes@wspa.org>; Ken Johnson <ken.johnson@weyerhaeuser.com>; Bill Taylor <BillT@taylorshellfish.com>; Dewey, Bill
Cc: Laurie, Tom (ECY)
Subject: RE: Invitation to Fish Consumption Rate Meeting

I've heard from a few of you with the suggestion that I invite municipalities to this meeting as well. I encountered more than slight resistance to that idea when vetting with the tribal leaders, fearing it could dilute the meeting given the time available, though they expressed interest in such a meeting in the future. I won't jam them by forcing the issue right before the meeting, nor would I have done so to you had the situation be reversed.

I understand your interest, and agree with at least part of it – this is not, and should be framed as, a "business vs. Tribes" issue. The reality is that this issue is far bigger than these two (or three) interest groups anyway. However, it is fair to say that business and Tribes have been the most vocal to date. I also strongly believe that a frank discussion with Ecology, business and the Tribes is in our mutual interest. At best, I believe it can help us understand and appreciate each other's real interests and make possible a more collaborative path. More specifically, I hope that the Tribes gain a better understanding of the value of the implementation tools rulemaking, and hope that you hear from them that they're interested in finding a feasible path. At the least, it can provide a frank assessment of what the real lay of the land is.

My intention is to keep this meeting at a high level, to elicit from each party their fundamental goals and concerns, and see if there is common ground to be built upon. I am optimistic that such ground exists. The draft agenda below is a simple one designed to meet that end. I am open to suggested improvements.

Also, you should be receiving today from my staff the response to the questions posed by NWPPA in earlier conversations.

Ted

Business/Ecology/Tribe Meeting
Ecology HQ, Lacey
1:00 – 2:30

Draft Agenda

1. Welcome / Introductions
2. Purpose of Meeting (Ted)
   Key issues and Ecology's approach to date
3. Key Concerns / Issues– Tribal perspective
4. Key Concerns / Issues– Business perspective
5. Discussion
6. If time, more discussion of specific policy choices ahead (Ted)
7. Timing and steps ahead

Ted Sturdevant, Director
Department of Ecology
(360) 407-7001
tstu461@ecy.wa.gov

From: Gary Chandler [mailto:GaryC@AWB.ORG]
Sent: Wednesday, May 02, 2012 8:13 PM
To: Sturdevant, Ted (ECY)
Cc: prigdon@yakama.com; rhepfer@elwha.nsn.us; jpeters@nwifc.org; awhitener@squaxin.nsn.us; dherrera@skokomish.org; syanity@stillaguamish.com; Brian Cladoosby; Courtney Barnes; chris@nwpulpandpaper.org; fholmes@wspa.org; Ken Johnson; Bill Taylor; Fran Wilshusen; Ann seiter; Pendowski, Jim (ECY); Susewind, Kelly (ECY); Laurie, Tom (ECY); North, Teri (ECY)
Subject: Re: Invitation to Fish Consumption Rate Meeting

Thanks Ted

I will be there

Sent from my iPad

On May 2, 2012, at 3:53 PM, "Sturdevant, Ted (ECY)" <TSTU461@ECY.WA.GOV> wrote:

As you may know, I am convening a small table of tribal and business leaders and my lead staff to discuss Ecology's work on updating fish consumption rates and water quality compliance tools.

The purpose of the meeting will be to exchange perspectives on the rule amendments and to discuss concerns that have arisen. Changing and implementing new standards across the clean-up and water quality programs is an important process, and direct dialogue will help us fully understand our respective interests and get to the best possible outcomes.

I want to invite each of you to join me on **May 9 from 1:00 to 2:30 p.m.** at Ecology for this purpose. I understand that some of you may not be available but if your schedule changes, I sincerely hope that you will attend if possible.

Please let me know if you are able to attend this important meeting.

Ted

Ted Sturdevant, Director
Department of Ecology
(360) 407-7001
tstu461@ecy.wa.gov

From: Paul Lumley <plumley@critfc.org>
Sent: Wednesday, October 10, 2012 10:38 AM
To: Dianne Barton; Laurie, Tom (ECY)
Cc: Gildersleeve, Melissa (ECY)
Subject: Re: FW: October 29th--First Water Quality Standards Water QualityPolicy Forum meeting

Tom and Melissa:

Thank you for the invitation. I appreciate the clarification that this meeting is not a government-to-government meeting. The CRITFC tribes met on Monday and directed me to participate on October 29. Dainne Barton will accompany me. Can you please send me more specifics on the time and location of the meeting? I do not see this information in the emails below.

Regards,

Paul Lumley
Columbia River Inter-Tribal Fish Commission
503-238-0667

>>>
From: "Laurie, Tom (ECY)" <tlau461@ECY.WA.GOV>
To: "Paul Lumley (plumley@critfc.org)" <plumley@critfc.org>, "(bard@critfc.org)" <bard@critfc.org>
CC: "Gildersleeve, Melissa (ECY)" <MGIL461@ECY.WA.GOV>
Date: 10/04/12 8:16 AM
Subject: FW: October 29th--First Water Quality Standards Water QualityPolicy Forum meeting

Paul & Dianne, please see the note below from Melissa Gildersleeve who is managing our water quality standards revisions.

We are looking to get more tribal participation in the Policy Forum that we are setting up to work with Ecology as we work to establish Human Health Criteria in our water quality standards which will include a fish consumption rate. This Policy Forum will be open to the public but will revolve around a "Delegates Table" representing key constituencies.

We recognize that this is not a substitute for government to government consultation but hope that tribes will also participate in Delegates Table to provide their expertise and perspective on the important policy choices that lie before us over the next year.

We would welcome direct participation from CRITFC and any help you can offer in helping to identify a few more tribal representatives to this forum.

The letters that went to tribal governments inviting them to this forum are on our website at:
http://www.ecy.wa.gov/programs/wq/swqs/humanhealthcritpolltr.html

Information about the Policy Forum is at:
http://www.ecy.wa.gov/programs/wq/swqs/hhcpolicyforum.html

- Tom

C. Thomas Laurie | Executive Advisor for Tribal & Environmental Affairs | Washington State Dept. of Ecology PO Box 47600, Olympia, WA 98504-7600 http://www.ecy.wa.gov/
email: tom.laurie@ecy.wa.gov phone: 360/407-7017 cell: 360/790-4110

From: Gildersleeve, Melissa (ECY)
Sent: Wednesday, October 03, 2012 3:57 PM
To: Fran Wilshusen (fwilshus@nwifc.org)
Cc: Laurie, Tom (ECY); Ann seiter
Subject: FW: October 29th--First Water Quality Standards Water Quality Policy Forum meeting

Fran---FYI-This just went out today for the first Water Quality Standards Policy Forum. I would sure like to have some staff from NWIF and CRITFC serve as delegates to the table. I think it would help the conversation as well as help you all as you advise others in the tribal community. This is going to be a pretty technical conversation and think your staff will have the capacity and maybe time...to digest all the topics that will be a part of developing human health criteria. That being said I know that you are still determining how to engage. Would you be available to talk with Tom and I and Dianne Barton at CRITFC to see if we can talk you all into participating?--Melissa

From: Gildersleeve, Melissa (ECY)
Sent: Wednesday, October 03, 2012 3:52 PM
To: 'carls@awcnet.org'; 'hkibbey@ci.everett.wa.us'; 'sandra.kilroy@kingcounty.gov'; 'BRawls@spokanecounty.org'; 'nbell@advocates-nwea.org'; 'JStuhlmiller@wsfb.com'; 'JHellman@washingtonports.org'; 'don.hurst@colvilletribes.com'; 'troutt.david@nisqually-nsn.gov'; 'paulm@pgst.nsn.us'; 'rorourke@pgst.nsn.us'; 'slape.james@nisqually-nsn.gov'; 'margaretbarrette@pcsga.org'; 'jstuhlmiller@wsfb.com'; 'jhellman@WASHINGTONPORTS.ORG'; 'dave.steele@centurylink.net'
Cc: Merriman, Scott; 'Williams, Dave'; 'garyc@awb.org'; 'dmparnel@aol.com'; 'jbrimmer@earthjustuce.org'; 'bv@columbiariverkeeper.org'; 'Johnson, Eric'; Conklin, Becca (ECY); Niemi, Cheryl (ECY); Susewind, Kelly (ECY)
Subject: October 29th--First Water Quality Standards Water Quality Policy Forum meeting

Dear Water Quality Standards Policy Forum Delegates-

Welcome. We have picked October 29th as the first Water Policy Forum/Delegates Table meeting. This first Delegate's Table meeting will be held here, in Olympia, at the Ecology Building. We will do the same thing we did with the August 28th workshop and have video participation in each of our regional offices. Right now our plan is to spend the morning with the Delegates Table and then open the afternoon up to the Policy Forum engagement.
Draft Agenda
Our Draft agenda for the morning looks like this-we might rearrange the order but these are the subjects we want to cover:

- Orientation to all that is in motion, and how we want to run the Forums
- Update on Implementation Tool Rulemaking
- Update on Contract for Facilitator
- Introduction to the Delegates Table: each delegate introduces themselves, who they are representing and their interests/concerns related to this process
- Agency Introductions (staff and staff expertise)

* Specific Topics:

-Walk through generic scenario that we will build upon as we discuss implementation and Human Health Criteria and discuss questions we want the scenario to address -Talk about the cost analysis that will be done as part of the rule development

In the afternoon we will open up the discussion to the broader Policy Forum and get ideas and feedback on the agenda items from the morning.

Logistics

We are still working on the best way to set up this forum, so please give us feedback. Our plan for this first meeting is to have the Delegates meet here in Olympia. The morning will be just the Delegates working through the agenda, discussing items, and asking questions. In the afternoon we will open the meeting up to the broader Policy Forum. Several of you have identified alternates and technical support staff . We are going to ask that the alternates and support staff sit in the front row behind you so we can have room for all the designated delegates.

Makeup of the Delegates Table

Below is a list of groups we invited to participate as Delegates in the Policy Forum. We are still working with some organizations to get this list finalized:

* Tribes (Ecology recognizes that this does not replace formal Government to Government consultation.)

o Don Hurst (Delegate)	Colville Indian Reservation
o Paul McCollum (Delegate)	Port Gamble S'Klallam Tribe
o Rory O'Rourke (Alternate)	Port Gamble S'Klallam Tribe
o James Slape, Jr (Delegate)	Nisqually Indian Reservation
o David Troutt (Alternate)	Nisqually Indian Reservation

* AWB - 4 delegates TBD

* Association of Cities - 2 delegates

o Carl Schroeder (Delegate)	Association of Washington Cities
o Heather Kibbey (Delegate)	City of Everett

* Association of Counties - 2 delegates

o Sandra Kilroy (Delegate)	King County
o Bruce Rawls (Delegate)	Spokane County

* Environmental Groups - 2 delegates

o Nina Bell (Delegate)	NW Environmental Advocates

o TBD Earth Justice and Riverkeepers

* Recreational fishery - 1 delegate TBD

* Commercial Fishery (Association of Shellfish Growers) - 1 delegate

o David Steele (Delegate) Pacific Coast Shellfish Growers

o Margaret Barrette (Alternate) Pacific Coast Shellfish Growers

* Washington Farm Bureau - 1 delegate

o John Stuhmiller

* Washington Ports Association - 1 delegate

o Johan Hellman
As you look at this and have questions or are unclear please give Becca (360)407-6413 or me a call or shoot us an email. The YouTube Videos from the August 28th workshop are up on the website (http://www.ecy.wa.gov/toxics/fish.html bottom of page). I encourage those of you that did not see Cheryl Niemi's presentation on Human Health Criteria 101 to watch that video. She does an excellent job of walking through the development of Human Health Criteria which is a preview for all our discussions.

See you on the 29th and call us if you have questions or need anything. - Melissa

Melissa Gildersleeve, Section Manager, Water Quality, Department of Ecology 360-407-6461 P Please consider the environment before printing this e-mail.

From: Bolster, Todd <tbolster@nwifc.org>
Sent: Monday, October 28, 2013 1:56 PM
To: (Group) Coordinated Tribal Water Quality Program; Ann Seiter; Lauren Rich; Lisa Hainey
Subject: Fwd: Revised agenda and material for FCRWQS call Tues Oct 29 at 1:30
Attachments: EPA WQ stds Comparison-of-Existing-Rule-Language-and-Proposed-Rule-Language.pdf; CTWQP-workshop-OVERVIEW_Oct-16-17_2013_re-FCR-WQS.docx; CTWQP-workshop-notes-TRIBES-ONLY_Oct16-17_2013-2.docx; CTWQP-workshop-notes-WITH-ECOLOGY_Oct17-2013-2.docx; FCR-WQ Update for Tribal Leaders Oct-28-2013_without-attachments.docx

Hi All,
Please find below information regarding tomorrow's coordination conference call on FCR.

---------- Forwarded message ----------
From: **Seiter, Ann** <aseiter@nwifc.org>
Date: Mon, Oct 28, 2013 at 1:44 PM
Subject: Revised agenda and material for FCRWQS call Tues Oct 29 at 1:30
To: "(Group) Tribal Fish Consumption Workgroup" <fishconsumption@nwifc.org>
Cc: Zach Welcker <zwelcker@kalispeltribe.com>

Tribal Fish Consumption Work Group and CTWQP,

This is a reminder that there will be a conference call for the folks interested in the FCR/Water Quality Standards issue on Tuesday October 29th from 1:30 to 3 pm. The call in number is 206-553-1454. There is no pass code. .

Revised Agenda:

- Updates on leadership meetings

- Preparation for November 6th Dept. of Ecology public meeting on rule-making for WQ standards. Please see site at: http://www.ecy.wa.gov/programs/wq/swqs/publicmtgs.html in order to reserve a seat or register for the webinar.

- October 16-17 Workshop follow-up and development of recommendations on implementation options

- Other FCR issues and responses

NEW: EPA Water Quality Standards -- analysis of proposed changes and potential call with EPA

New material attached:

- Notes from the Oct 16-17 Workshop: There are 3 parts, a short overview, the Tribes-only session, and the session with Ecology
- Comparison of existing and proposed EPA water quality standards (thanks, Kalispel)
- An update that is going out to tribal leaders this week. (I did not include their attachments since it is redundant to other material sent to you previously.)

Please call me at 360-683-5725 if you have any questions.

---Ann

This information is confidential and is intended for tribal policy representatives and technical/legal staff only. Please do not forward this message or attachments outside of tribal organizations. Thank you.

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Ann Seiter
aseiter@nwifc.org
Coordinator: Fish Consumption Rate Project
PO Box 2201; Sequim, WA 98382
Home Office: 360-683-5725

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Todd Bolster
Habitat Policy Analyst
Northwest Indian Fisheries Commission
tbolster@nwifc.org
(360) 528-4347

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Comments to Idaho Department of Environmental Quality
Risk, Human Health, and Water Quality Standards

Please accept these comments, which respond to the Idaho Department of Environmental Quality (IDEQ) *Discussion Paper #7: Risk Management and Human Health* (Dec. 2014)[1] [hereinafter IDEQ Risk Discussion Paper] and follow up on discussion during the public meeting held by IDEQ on December 2, 2014. These comments reflect the views of the author. Although they raise concerns about the impacts of Idaho's water quality standards on tribes, they do not purport to represent the perspective of any tribe; those perspectives must be obtained directly from each tribe.

I. Background

Idaho's deliberations involve risk in the context of its water quality standard-setting efforts. This context is significant, because it constrains the debate in important ways. Among other things, the discussion here must be framed and bounded by the relevant legal provisions, including treaties and other instruments securing tribes' fishing rights, and including statutory directives under the Clean Water Act (CWA). Thus, discussions of risk in other regulatory contexts, or discussions of risk generally, may have more or less relevance for Idaho's deliberations, depending on how far afield they are from the context at hand.

Under the CWA, water quality standards are health-based standards. The touchstone for agencies' efforts is human health. Fish are the primary route of human exposure to PCBs, mercury, dioxins, and a host of toxic chemicals that are harmful to human health. Health-based water quality standards are set to ensure that humans can safely consume fish, without also being exposed to contaminants in harmful amounts. Pursuant to Environmental Protection Agency (EPA) guidance, agencies enlist quantitative risk assessment methods to set standards for both threshold and non-threshold contaminants. For threshold contaminants, standards are set so that contaminants don't exceed levels that are safe for humans. For non-threshold contaminants, including carcinogens, exposure to any non-zero amount has the potential to cause cancer; standards are set so that contaminants don't exceed a risk level

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[1] Idaho Department of Environmental Quality, Idaho Fish Consumption Rate and Human Health Water Quality Criteria – Discussion Paper #7: Risk Management and Protection of Human Health (Dec. 2014)[hereinafter IDEQ, Risk Discussion Paper] *available at* http://www.deq.idaho.gov/media/1118404/58-0102-1201-discussion-paper7.pdf.

901 12th Avenue, Sullivan Hall P.O. Box 222000 Seattle, WA 98122-1090 www.law.seattleu.edu

determined to be "acceptable." In either case, agencies then work with the risk assessment equation to "solve" for the concentration of each chemical that will be permitted in the waters that support fish.

Idaho's water quality standards affect the rights, resources, and well-being of numerous tribes in the region. In fact, when the waters that support fish are allowed to be contaminated, tribes' interests are profoundly affected and tribal people disproportionately among the most exposed. It is therefore troubling that the IDEQ Risk Discussion Paper contemplates tolerating a greater level of cancer risk than Idaho has in the past – particularly as studies have made clear that tribal people would be the ones who disproportionately would have to bear this risk.

These comments begin in Part II by discussing some historical background specific to EPA's approach to health-based standards under the CWA's water quality standards provisions. Part III sketches the various considerations relevant to a risk's "acceptability," and distinguishes between assessments of risk in the water quality standard-setting context and assessments of risk in other contexts. Part IV responds to the IDEQ Risk Discussion Paper's discussion of "voluntary" risks. Parts V and VI raise two issues not discussed in the IDEQ Risk Discussion Paper: risks disproportionately borne by tribal people and risks that impair tribes' legally protected fishing rights. Part VII considers the direction provided by the relevant EPA guidance for states' water quality standard-setting efforts.

II. For Carcinogens, the Recommended Concentration to Protect Human Health is Zero

EPA, in a prominent 1984 criteria document for dioxin, made clear that it understood that human health could only be ensured for this contaminant's non-threshold effects if risk were set at zero.

> For the maximum protection of human health from the potential carcinogenic effects due to 2,3,7,8 - TCDD exposure through Ingestion of contaminated water and contaminated aquatic organisms, the ambient water concentration should be zero. This criterion is based on the non-threshold assumption for 2,3,7,8 - TCDD.[2]
>
> Under the Consent Decree in NRDC vs. Train, criteria are to state "recommended maximum permissible concentrations (including where appropriate, zero) consistent with the protection aquatic organisms, human health, and recreational activities." 2,3,7,8 -TCDD is suspected of being a human carcinogen. Because there is no recognized safe concentration for a human carcinogen, the recommended concentration of 2,3,7,8 -TCDD in water for maximum protection of human health is zero.[3]

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[2] U.S. Environmental Protection Agency, Ambient Water Quality Criteria for 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin x (Feb. 1984), *available at* http://water.epa.gov/scitech/swguidance/standards/upload/2008_09_10_criteria_dioxincriteria.pdf.

[3] *Id.* at C-180.

While EPA went on to offer calculations based on three risk levels, 10^{-7}, 10^{-6}, and 10^{-5}, it took pains to point out that these were all in the realm of the second best, i.e., that they would not result in a truly health-based standard, because only a standard permitting zero risk could do so.[4]

EPA's statements in this criteria document are notable for three reasons. The first is simply that, for non-threshold contaminants in our waters such as dioxins, PCBs, and other carcinogens, any non-zero concentration is inadequate to protect human health; any non-zero amount will result in quantifiable levels of risk. EPA's statements recognize and preserve the difference between "zero" and "some," between protecting human health and permitting an amount of risk to remain. A risk quantified at 10^{-7}, 10^{-6}, or 10^{-5} can't be converted into zero risk by simply eliding this difference, nor by quoting terms (e.g., "*de minimis*" or "essentially zero") applied in other contexts (a point discussed further below).

The second notable aspect of EPA's statements is that the cancer risk levels EPA deemed relevant to water quality standard-setting at the time ranged from 10^{-7} to 10^{-5}. EPA's 1980 guidance on water quality standard-setting similarly embraced risk levels that range from 10^{-7} to 10^{-5}.[5] And EPA's current 2000 guidance, its *Ambient Water Quality Criteria Methodology* (EPA AWQC Guidance) continues to state that the range of acceptable risk levels runs from 10^{-7} to 10^{-5} – with the caveat that risk levels at the less protective end of this range will be scrutinized for their impact on highly exposed subgroups and may be rendered unacceptable if they result in risks greater than 10^{-4} to members of such subgroups:

> With AWQC derived for carcinogens based on a linear low-dose extrapolation, the Agency will publish recommended criteria values at a 10^{-6} risk level. States and authorized Tribes can always choose a more stringent risk level, such as 10^{-7}. EPA also believes that criteria based on a 10^{-5} risk level are acceptable for the general population as long as States and authorized Tribes ensure that the risk to more highly exposed subgroups (sportfishers or subsistence fishers) does not exceed the 10^{-4} level.[6]

The IDEQ Risk Discussion Paper omits this background and this current EPA language, selectively quoting material from the EPA AWQC Guidance that refers to a range that, at its most protective, reaches only 10^{-6}.[7] As such, it may portray 10^{-6} as an upper limit, whereas EPA offers 10^{-6} as somewhere in the middle.

The third notable feature of EPA's statement in its dioxin criteria document that the "recommended concentration …[for the] protection of human health is zero" is that EPA thus expressed what Douglas

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[4] *Id.* at xi, C-180 to C-181 (explaining that because attaining zero concentration level might not be achievable in some cases and because the criteria document was intended to assist states and the EPA in calculations of water quality standards, EPA was providing concentrations corresponding to a range of risk levels).

[5] U.S. Environmental Protection Agency, Methodology for Deriving Ambient Water Quality Criteria for the Protection of Human Health 1-8 (2000)[hereinafter EPA, AWQC Guidance], *available at* http://water.epa.gov/scitech/swguidance/standards/upload/2005_05_06_criteria_humanhealth_method_complete.pdf.

[6] *Id.* at 1-12.

[7] IDEQ, Risk Discussion Paper, *supra* note 1, at 5-6.

Kysar has termed a "moral remainder."[8] That is, it registers the "sense of regret" when there remains "a shortfall between statutory command and societal achievement."[9] The lives lost and harms permitted as a result of such shortfalls, however, "are viewed as tragic, lamentable consequences of human fallibility and finitude –a moral remainder that provides enduring motivation for surviving members of society to seek ways of doing better in the future."[10] The Columbia River Inter-Tribal Fish Commission (CRITFC), in comments to then-administrator Browner similarly called attention to this moral remainder.[11] CRITFC reminds us that zero risk is the only level that will actually protect human health, and so, importantly should remain our ultimate goal in enacting health-based standards.[12]

III. "Acceptable" Risk is a Judgment of Value that is Context-Specific

As soon as we move away from zero, there is potential for harm. How much risk (and of what sort, borne by whom) we are willing to tolerate requires a judgment of value. It is a judgment that involves nothing less than deciding, to paraphrase Annette Baier, which harms to notice and on whom we will with good conscience impose "death [or] risk of death."[13]

A. Context is Crucial to the Kind and Amount of Risk Collectively Thought Tolerable

At the individual and collective levels, this judgment of value is context specific. As a general matter, a risk's acceptability can turn on a host of factors respecting the nature of the risk (including, e.g., its familiarity, controllability, etc.); whether the risk is sought out or undertaken voluntarily (please see discussion below); what is at stake/the seriousness of the harm (including, e.g., death, irreversible neurological impairment, cancer); whether the risk is equitably distributed (including, e.g., whether those who bear the risk also benefit from the risk-producing activity); whether subpopulations of particular concern will bear the risk (including, e.g., children); and whether the risk attends the exercise of practices that are important or to which people have rights.[14]

Thus, risks are not fungible (except in the actuarial sense). Judgments of "acceptability" made in one context (e.g., the occupational context) can't simply be transferred to another context (e.g., the environmental context).

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[8] DOUGLAS A. KYSAR, REGULATING FROM NOWHERE: ENVIRONMENTAL LAW AND THE SEARCH FOR OBJECTIVITY 19-20 (2010).
[9] *Id.* at 20.
[10] *Id.*
[11] Columbia River Inter-Tribal Fish Commission, Comments to Administrator Browner on the Draft Revisions to the Methodology for Deriving Ambient Water Quality Criteria for the Protection of Human Health (1999).
[12] *Id.* at 3 (arguing that only "zero" risk will actually protect the health of tribal members).
[13] Annette Baier, *Poisoning the Wells,* in VALUES AT RISK 49 (Douglas MacLean, ed., 1986)("When is a public policy that entails death for some and risk of death for more a policy that offends our moral standards? … It is not merely a question of whose lives we should save by what measures with whose money, but whom, among those whose cooperation and whose taxes we use, we will with good conscience kill, cause to die, or let die, and by what measures or neglect.")
[14] See, e.g., Molly J. Walker Wilson, *Cultural Understandings of Risk and the Tyranny of the Experts*, 90 OREGON L. REV. 113 (2011); see generally, VALUES AT RISK (Douglas MacLean, ed., 1986).

Thus, comments at the December 2nd public meeting to the effect that society finds risks as high as 1 in 100 or 1 in 1,000 to be acceptable in the occupational context (with the implication that we should therefore be undaunted by a similar risk level in the environmental context) miss the mark without more. Specifically, their import depends on an inquiry into the similarities in and differences between these two contexts (e.g., risks on the job are undertaken by adults as part of an consensual contractual arrangement for compensation; whereas risks from environmental sources are imposed on all humans whose ordinary practices – e.g., breathing, eating, drinking – leave them exposed to contaminants) and, importantly, why these similarities/differences ought to matter. Consent, in particular, is understood to be among the relevant considerations to evaluations of a risk's acceptability.[15]

Similarly, it is unhelpful to point to figures about the lifetime risk of cancer that we all currently face (i.e., 1 in 3 for women; 1 in 2 for men), as the IDEQ Risk Discussion Paper does.[16] While presumably offered to situate the present risk debate in context, such figures do the opposite: they lump together all cancers, from all causes – devoid of context. This approach doesn't permit inquiry into any of the relevant considerations noted above. Importantly, it doesn't ask *whether* we, as a society, think this is okay. In fact, one of the pioneers of the field of risk perception studies, Paul Slovic, has found that most people believe current risk levels to be too high:

> Another consistent result from psychometric studies of expressed preferences is that people tend to view current risk levels as unacceptably high for most activities. The gap between perceived and desired risk levels suggests that *people are not satisfied with the way that market and other regulatory mechanisms have balanced risks and benefits.*"[17]

In short, the fact *that* we currently face a certain level of risk doesn't tell us whether that level is desirable or, crucially, whether it is ethically defensible.

B. Statutory Context and Constraints

Moreover, as a society, we *have* collectively determined that some risks – such as those from environmental contaminants – should be reduced. And, through democratic processes, we have enacted an array of environmental, health, and safety laws that direct agencies to require risk reduction. These statutes establish various mechanisms for regulating the entities and processes that produce contamination, i.e., for seeking risk reduction from risk-producers. These statutes enlist different approaches and permit different considerations, depending on context (e.g., consumer protection, worker safety, children's health). They reflect our collective judgments regarding the degree of risk reduction to be achieved and the appropriateness of considering relevant tradeoffs for each particular

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[15] See, e.g., Douglas MacLean, *Risk and Consent: Philosophical Issues for Centralized Decisions*, in VALUES AT RISK 17 (Douglas MacLean, ed., 1986)(discussing why the concept of consent must play a crucial role in justifications for governments' decisions to impose risk). As will be discussed, risk in an occupational context is also governed by different statutory commands, namely, the Occupational Health and Safety Act.

[16] IDEQ, Risk Discussion Paper, *supra* note 1, at 2.

[17] Paul Slovic, *Perception of Risk*, 236 SCIENCE 280, 283 (1987) (emphasis added).

statutory program. Notably, these statutory programs sometimes strike different balances regarding risk.

So, the fact that a certain level of risk has been found permissible by a sister agency (e.g., the Food and Drug Administration; the Nuclear Regulatory Commission)[18] or sister program within EPA (e.g., the Superfund program),[19] does not answer the question whether it ought to be viewed as acceptable in the current context, i.e., under the CWA's water quality standards program for Idaho. Yet the IDEQ Risk Discussion Paper presents the results of these agencies' deliberations as if they had equal and obvious precedential force. Again, we would need more information about the context in order to assess whether the reasons offered in support of accepting a particular risk level in a different regulatory context ought to hold sway in the context at hand. In particular, we would need to have more information about the governing statutory instructions, in order to ensure that we were comparing apples to apples. Some environmental statutory provisions permit cost-benefit balancing; some preclude agencies from considering costs. Some direct agencies to set standards based on what is healthful; some direct agencies to set standards that are technologically feasible or achievable. It is not appropriate (and may not be legal) to import results reached under one set of statutory directives (e.g., "as low as reasonably achievable")[20] into a decision making process under another statutory directive.[21]

It is also problematic, as Michael Livermore and Richard Revesz have recently discussed, for agencies to make decisions on the basis of an "unacknowledged factor," such as cost.[22] As noted above, the CWA's water quality standards provisions are health-based; they take human health, not technological feasibility, as their touchstone. Yet the IDEQ Risk Discussion Paper presents a "partial list of potential criteria for considering risk to be acceptable" that includes several entries explicitly or implicitly calling for the weighing of costs. While it adds a note that "[n]ot all of the above are based solely on health risk; some clearly involve cost-benefit analysis," it doesn't make the meaning of this note clear. Members of the public might be misled into believing that weighing the costs of risk reduction is permissible and/or what IDEQ intends to do.

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[18] IDEQ, Risk Discussion Paper, *supra* note 1, at 3-4, 8.

[19] *Id.* at 4-5.

[20] *Id.* at 8.

[21] Thus, the IDEQ Risk Discussion Paper's account of the FDA's attempt to arrive at a defensible risk level under the Food, Drug, and Cosmetics Act's "Delaney Clause" appears to have been given emphasis out of proportion with its relevance to water quality standards under the CWA. *Id*. at 3-4. The FDA was laboring under a particular statutory directive with a unique history; its efforts to determine a "safe" level reflect the language and constraints of the statutory provisions for food additives. The IDEQ Risk Discussion Paper appears to make the leap to a claim that the 10^{-6} risk level ultimately arrived at in that context can be equated with "essentially zero" risk in every other context. Such a claim would require more support. See generally, William Boyd, *Genealogies of Risk: Searching for Safety, 1930s-1970s*, 39 ECOLOGY L. Q. 895 (2012).

[22] Michael A. Livermore & Richard L. Revesz, *Rethinking Health-Based Standards*, 89 NYU L. REV. 1184, 1233 (2014).

IV. “Voluntary” Risks

The IDEQ Risk Discussion Paper delves into one factor understood to be relevant to a risk’s acceptability: whether the risk can be said to have been “voluntarily” undertaken. The assumptions and conclusions embedded in this discussion are troubling and warrant extensive comment. This discussion raises issues discussed in the risk literature as voluntariness, responsibility, and self-relevance.

A. Voluntariness

The IDEQ Risk Discussion Paper accurately states that a risk’s acceptability turns in important part on whether it is considered to be voluntary or involuntary. Researchers from various disciplines have studied this intuition, and it has proven to be stable when tested using a variety of disciplinary methods.[23]

Whether any particular risk is properly viewed as voluntary, however, is not self-evident. In fact, the determination of voluntariness is value-laden and often complex; there may be considerable disagreement in particular cases over whether a particular activity or practice – and the risk it entails – is voluntary. Yet agencies, and other expert or individual evaluators sometimes simply label certain activities as “voluntary” or “involuntary” – without offering justifications for doing so. The act of valuation becomes invisible, as a judgment of voluntariness is presented as a natural, immutable “fact” about the world. The IDEQ Risk Discussion Paper, for example, simply states as fact that: “Examples of voluntary risks are those associated with driving, skiing, and tobacco use. Involuntary risks include exposure to pollutants in air or drinking water.”[24]

Judgments of voluntariness implicate views about whether a particular activity, practice, or lifeway is laudable, important, or essential to living a human life. They rest on particularized understandings of what a practice involves and what, therefore, is at stake. These judgments also implicate perspectives on whether the risks that are entailed when a particular practice brings humans in contact with contamination can be avoided readily or cheaply – or whether risk avoidance would be impossible or would burden fundamental rights or would mean profound loss. The determinations of importance, necessity, ease, and possibility are judgments of value that are recognized to be culturally influenced.[25]

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[23] See, e.g., Walker Wilson, *supra* note 14, at 149-50, 165, 168-69; Cass R. Sunstein, *A Note on “Voluntary” Versus “Involuntary” Risks*, 8 DUKE ENVTL L. & POLICY FORUM 173 (1997). The IDEQ Risk Discussion Paper cites Chauncey Starr; Starr’s early estimates are now viewed as likely overstating the magnitude of this effect, but a significant effect has still been evidenced in more recent studies. *See* Walker Wilson, *supra* note 14, at 168-69.

[24] IDEQ, Risk Discussion Paper, *supra* note 1, at 7.

[25] See, generally, Walker Wilson, *supra* note 14 ; Dan M. Kahan, *Two Conceptions of Emotion in Risk Regulation*, 156 U. PA. L. REV. 721 (2008); Catherine A. O’Neill, *Risk Avoidance, Cultural Discrimination, and Environmental Justice for Indigenous Peoples*, 30 ECOLOGY L. Q. 1 (2003)[hereinafter O’Neill, *Risk Avoidance and Cultural Discrimination*]; Catherine A. O’Neill, *No Mud Pies: Risk Avoidance as Risk Regulation*, 31 VERMONT L. REV. 273 (2007)[hereinafter O’Neill, *No Mud Pies*].

The assignment of a label of voluntariness is thus of particular concern where an agency or other evaluator espouses the perspective of the dominant society, but the risk-bearers are Native people or members of other non-dominant groups. As I have observed elsewhere: "[t]he dominant society's understandings of the value of the practices in question and the ease or anguish with which avoidance would be undertaken will often be different, perhaps profoundly so, from the understandings of the indigenous peoples on whom the burden of risk avoidance will fall."[26]

Yet, a risk may be proclaimed to be voluntary as if everybody, including Native Peoples, agreed it were so, when only (some) members of the dominant society share this perspective. The IDEQ Risk Discussion Paper goes nearly this far. It opines that "given the availability of other healthy food choices, consuming large amounts of fish must be considered a voluntary risk."[27] It allows that, "in some cases, the voluntary nature of fish ingestion risk is tempered by financial need or cultural factors."[28] Its bottom line, however, is firm: "Still, fish consumption is a voluntary behavior."[29] Whereas "we do not have a choice about breathing air and drinking water," fish consumption is deemed a matter of choice.[30]

In an article published in the *Ecology Law Quarterly*, I considered the different understandings typical of dominant society evaluators, on the one hand, and Native Peoples of the Pacific Northwest, on the other, respecting the value of the practices at stake when fish have become contaminated and respecting the possibility of risk avoidance:

> *Value, Necessity of the Pursuit*
>
> For dominant society evaluators, fishing is likely to be viewed primarily as a recreational pursuit and secondarily as an economic activity. Fishing is therefore likely to be understood as a pursuit that is not necessary for most practitioners, but important for recreational or economic reasons for some. Fish are likely to be recognized by those in the dominant society as a palatable, efficient, and relatively inexpensive source of protein and other nutrients for humans, although not the only such source. Fish consumption is therefore likely to be valued, but unlikely to be thought indispensable.
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> For Native peoples of the Pacific Northwest, by contrast, the various aspects of fishing are constitutive of their identity as peoples. Fish, fishing, and fish consumption are understood to be vital for the physical, social, economic, political, spiritual, and cultural health of these peoples and their members. Proper practice includes protecting and tending to fish and shellfish habitat, fishing for or gathering fish and shellfish, preparing, consuming and using fish and shellfish, all attended by appropriate methods, prayers, and ceremonies. Fish, fishing, and fish consumption

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[26] O'Neill, *Risk Avoidance and Cultural Discrimination*, *supra* note 25, at 28.
[27] IDEQ, Risk Discussion Paper, *supra* note 1, at 7.
[28] *Id.*
[29] *Id.*
[30] *Id.*

are understood to be necessary, an indispensable part of what it means to be Nez Perce or Nisqually. Fishing and eating fish are important occasions for the inter-generational transfers of knowledge, including the ecological, historical, social, and spiritual knowledge that is a central part of the inheritance of succeeding generations. Fishing is also important for economic reasons, as fishers can feed their families or sell their catch or harvest for income. The inestimable value that the various Native peoples of the Pacific Northwest attach to fish, fishing and fish consumption is marked in stories and ceremonies, language, treaties negotiated with the invading peoples, past and present fisheries management practices, contemporary leadership in restoration efforts, and the ongoing political and legal struggle for the survival of the salmon, fish, and shellfish and the flourishing of their fishing cultures. Del White, Nez Perce, explains: “People need to understand that the salmon is part of who the Nez Perce people are. …

Possibility and Costs of Avoiding the Attendant Risk

Dominant society evaluators are likely to believe that there are a host of alternatives to fishing and substitutes for eating fish, each of which might involve some costs, but all of which would be reasonable means of avoiding the risks that fishing and fish consumption have come to entail. To the extent that the dominant society views fishing as a recreational pursuit, fishing in different places, practicing “catch and release” fishing, or taking up alternative pastimes might suit nearly as well. Because the dominant society is less likely to attach any significance to the consumption of particular species or parts of fish and shellfish, risk avoidance measures that advised against consumption of certain species or certain parts would be unproblematic, apart from small compromises in terms of money (perhaps the prohibited species is less expensive to purchase or catch) and predilection (perhaps the prohibited part is a delicacy). Similarly, because the dominant society is less likely to consume fish and shellfish at particular times and frequencies in accordance with seasonal availability or ceremonial requirements, risk avoidance measures that entail consuming at reduced rates or measured frequencies (e.g., “eat no more than one fish meal per week”) would visit little or no hardship on its members, although it might entail some inconvenience (perhaps it is difficult to identify dietary substitutes that provide the nutritional benefits of fish). And, because the dominant society is less likely to employ the particular preparation methods that advisories recommend against, these risk avoidance measures are unlikely to implicate practices that are thought to be culturally important.

From the perspectives of the various Native peoples of the Pacific Northwest, such risk avoidance measures would occasion profound loss. Given that fish, fishing, and fish consumption is part of who these peoples are, it is simply not fathomable for them to avoid the attendant risks by ceasing to fish and eat fish. Indeed, it would be unthinkable….

It would also not be appropriate or possible in most cases to fish “elsewhere.” As the Columbia River Inter-Tribal Fish Commission explains: “Salmon and the rivers they use are part of our sense of place. The Creator put us here where the salmon return. We are obliged to remain and

> to protect this place." Moreover, various tribes' aboriginal and treaty-based claims to the fish and other resources are tied to specific places; the legal protections that flow from these claims cannot simply be re-established somewhere else. In addition, the particularized skills and ecological knowledge that indigenous peoples have developed over centuries are also place-specific and, therefore, are not transferable to other locations.
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> Similarly, it would be unimaginable from the perspective of these peoples to undertake risk avoidance that required consuming fish and shellfish at reduced rates or frequencies, given that ceremonial observance necessitates consumption of large quantities during certain events timed in accordance with seasonal, traditional or cultural dictates. … In short, the loss occasioned by the potential risk avoidance measures would be profound and felt along cultural, spiritual, social, ecological, economic, and political dimensions.
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> In sum, as these examples help to illustrate, it will often be the case that the practices that have come to entail risk because of environmental contamination are valued differently by the dominant society on the one hand and indigenous peoples on the other. Where this is so, avoidance measures that ask risk-bearers to abandon or alter these practices are unlikely to be understood as particularly burdensome by dominant society evaluators –although they may be understood as impossibly burdensome by indigenous risk-bearers. Because environmental policy is likely nonetheless to reflect the dominant society's understandings of what is at stake, the risk avoidance measures that are adopted will likely be the very ones that encroach most profoundly on the expression of indigenous cultures and the exercise of indigenous rights.[31]

While the discussion excerpted above considers the issue in general, the tribes whose practices and rights are affected are the only ones who can speak properly to the question whether, from their perspectives, the relevant risks ought to be considered "voluntary." Although the IDEQ Risk Discussion Paper appropriately acknowledges that other perspectives exist ("For subsistence fishers, [catching and eating fish] is a way to obtain a high quality protein source inexpensively. Native American cultural identity with fish harvest and consumption also casts the voluntary nature of the risk in a somewhat different light"), it effectively dismisses them in the next breath, delivering its bald conclusion that "fish consumption is a voluntary behavior."[32] Yet the affected tribes have spoken repeatedly to this question as part of public processes and have provided numerous written statements to the rulemaking document.[33] These statements *by the affected tribes* indicate a quite different perspective than that asserted by IDEQ.

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[31] O'Neill, *Risk Avoidance and Cultural Discrimination*, *supra* note 25, at 35-40 (citations omitted; please consult original for supporting authorities).

[32] IDEQ, Risk Discussion Paper, *supra* note 1, at 7.

[33] Idaho Department of Environmental Quality, Water Quality: Docket No. 58-0102-1201-Negotiated Rulemaking (please see tribes' comments regarding their fishing rights, and the importance of fishing and fish to the tribes and their members); see, generally, Affiliated Tribes of Northwest Indians, Resolution #13-44 (2013).

B. Responsibility

An issue related to labeling a risk as "voluntary," as recognized by the IDEQ Risk Discussion Paper, is that it involves judgments about matters of "responsibility."[34] The IDEQ Risk Discussion Paper's discussion here is slippery. After having deemed fish harvest and consumption "voluntary" in the previous paragraph, it states:

> If a risk is voluntary, the question of individual responsibility arises. When voluntary behaviors lead to risk, to what extent is it the responsibility of the government to reduce that risk? When regulatory efforts have reduced the risk associated with fish consumption to the extent possible, individual responsibility still plays a role in managing risk associated with fish consumption.[35]

It purports to raise a question, but buries within it a number of unstated assumptions. First, it portrays the risk as the consequence of the practices themselves ("When voluntary behaviors lead to risk ...."; and, to start off the previous paragraph, "The amount of contaminants in fish to which we are exposed is a function of the amount of fish we consume."). *But fish, if they aren't permitted to become contaminated with toxic substances, don't "lead to risk."* The source of the risk is not fish or fishing. People's health is not jeopardized by eating fish – in fact, fish are widely recognized to be a healthful source of protein and other nutrients – people's health is put in jeopardy when risk-producers are allowed to contaminate the waters in which fish swim. It is true that humans are only exposed to these contaminants when they eat fish. But the IDEQ Risk Discussion Paper presents this discussion in a one-sided fashion, as if the risk results solely from consuming fish. This depiction is unfortunate, as it appears to fault people for eating fish.[36]

Second, this discussion seems to open up to question a matter on which Congress and the Idaho state legislature have already spoken: under the CWA, it *is* "the responsibility of the government to reduce [the] risk" associated with fishing, to the point that the nation's waters are again "fishable."[37]

Third, this discussion implicitly rewrites the relevant statutory approach – substituting a feasibility-based standard for the health-based standard under the CWA ("When regulatory efforts have reduced the risk associated with fish consumption to the extent possible ...."). But the CWA doesn't permit this; water quality standards require that pollution be controlled to the point that it is healthful – feasibility and cost aren't appropriately part of an agency's standard-setting efforts. The discussion then summarily answers the question it purported to ask: "individual responsibility still plays a role in managing risk associated with fish consumption." Here again, the IDEQ Risk Discussion Paper takes the opportunity to

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[34] IDEQ, Risk Discussion Paper, *supra* note 1, at 7-8.

[35] *Id.* at 7-8.

[36] See, O'Neill, *Risk Avoidance and Cultural Discrimination*, *supra* note 25; O'Neill, *No Mud Pies*, *supra* note 25.

[37] Federal Water Pollution Control Act (Clean Water Act), 33 U.S.C. § 1251(a)(2)(2012). The EPA has interpreted this goal to require a baseline "use" of "fishable/swimmable" waters. 40 C.F.R. § 131.2, § 131.4 (2012).

shift responsibility from risk-producers (and the government that is directed to regulate risk production) to risk-bearers.

C. Self-Relevance

The IDEQ Risk Discussion Paper's discussion of voluntariness raises another issue recognized to affect agency and other evaluators' judgments about risk: whether a risk is perceived to be "self-relevant." According to the risk literature, where evaluators are not themselves likely to have to bear a risk, they may view it as less serious or worthy of public response.[38] Conversely, where a risk is self-relevant, i.e., likely to be borne by and of concern to the evaluator, studies have shown that risks will be seen as more serious and worthy of public response.[39] While one can't be sure of the perceptions of particular agency or other evaluators, this effect is worthy of note in a public discussion of risk. There is cause for concern, in any case, where agency statements suggest that agency personnel will be unaffected by relatively greater risk –whether because they don't care to eat fish or because they see ready options for substituting other foods for fish.[40] The IDEQ Risk Discussion Paper, for example, takes it as a "given" that people can easily and healthfully omit fish from their diets and their lives ("given the availability of other healthy food choices, consuming large amounts of fish must be considered a voluntary risk").[41]

On the whole, the IDEQ Risk Discussion Paper's section titled "Voluntary versus Involuntary Risks" contains numerous unstated assumptions and incomplete or one-sided portrayals of the issues. Although it occasionally introduces countervailing considerations, it quickly dismisses these – with the result that the entire section appears to be less a balanced analysis and discussion and more an argument for a position already decided upon.

V. Risks Disproportionately Borne by Tribal People

The IDEQ Risk Discussion Paper is silent on another aspect of risk that is recognized in the risk literature to be important to a risk's acceptability: whether it is shared equally or whether it is borne disproportionately by a few. Such concerns for equity are particularly acute, moreover, if the "few" are members of an identifiable group that has historically been subjected to discrimination or colonization. Where, as here, members of the fishing tribes are among the most highly exposed and will thus disproportionately have to bear the risk, evaluations of risk raise issues of environmental justice.

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[38] See Walker Wilson, *supra* note 14, at 150.
[39] *Id.*
[40] See, e.g., Washington State Department of Ecology, Presentation, Lake Roosevelt Forum Conference, Spokane, WA, Nov. 20, 2013 (Ecology representative in a recent public presentation mentioned that she didn't eat much fish because she "didn't like the taste" and discussed this in contrast to people "who love fish" and therefore eat a lot of it). The existence of people who simply don't eat fish, and so will never be among those exposed to any contaminants permitted to reside in fish, is a feature of exposure via the fish consumption pathway that distinguishes it from some other important exposure pathways.
[41] IDEQ, Risk Discussion Paper, *supra* note 1, at 7.

Yet, public debate about risk is often couched in the abstract, in terms of "statistical lives," i.e., nameless, faceless probabilities. As Douglas MacLean observes, "[r]isk analysts have tended to focus only on the magnitude of the risk, however distributed. … If exactly one person will die each year, the $1(10^{-6})$ magnitude indicates our ignorance in advance about who it will be."[42] This theoretical ignorance allows the discussion about risk to proceed on the premise that everyone is equally likely to be among the unfortunate.

This requisite – that everyone is equally likely to have to bear the risk – is thought to be satisfied in one of two ways. First, everyone can be expected to experience roughly the same level of risk if their circumstances of exposure are roughly the same – that is, the physical, geographical, and other parameters that determine each individual's exposure don't vary that much from person to person. Alternatively, everyone can be thought to experience roughly the same *chance* of experiencing a relatively high or relatively low level of cancer risk if we don't know, in advance, on whom the greater risk will fall – it is a greater chance being taken by all of us, like a lottery.[43] But, as elaborated below, neither of these conditions holds true when we are talking about fish consumption.

As to the first, individuals' circumstances of exposure are emphatically *not* "roughly the same" where the exposure pathway involves fish consumption. In fact, fish intake is highly variable, with differences in people's contemporary intake spanning as many as three orders of magnitude. Some people eat no fish at all; others eat 1453 grams/day.[44] The 90th percentile intake rate for the general population is the source of the EPA's national default of 17.5 grams/day.[45] By contrast, the 90th percentile intake rate documented by recent surveys of the Suquamish and Lummi is 489 grams/day and 800 grams/day, respectively.[46] Note that these are contemporary, suppressed fish consumption rates (FCRs); if historical or "heritage" rates were considered the variability would be even more marked.

As to the second, we cannot pretend that everyone's chances of being subjected to a greater level of risk are roughly the same.[47] Here in the Pacific Northwest, we know *who* it is that depends on fish, *who* it is that is the most exposed. We know, then, *who* will be left to bear the risk if a state such as Idaho shifts to a less protective level: it will be tribal people. This is problematic as an ethical matter, and it changes the terms of the policy debate. We cannot pretend to be debating the appropriate risk level in the abstract, i.e., in terms of statistical lives. In the states of the Pacific Northwest, a determination that

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[42] Douglas MacLean, *Social Values and the Distribution of Risk*, in VALUES AT RISK 75, 78-79 (Douglas MacLean, ed., 1986).

[43] See discussions in Catherine A. O'Neill, *Variable Justice: Environmental Standards, Contaminated Fish, and "Acceptable" Risk to Native Peoples*, 19 STANFORD ENVTL L. J. 73-75 (2000)[hereinafter O'Neill, *Variable Justice*]; Catherine A. O'Neill, *Fishable Waters*, 1 AMERICAN INDIAN LAW JOURNAL 181, 255-260 (2013)[hereinafter O'Neill, *Fishable Waters*], *available at* http://www.law.seattleu.edu/Documents/ailj/Spring%202013/O'Neill-Fishable%20Waters.pdf.

[44] See O'Neill, *Fishable Waters*, *supra* note 43, at Table 1 (The 1453 grams/day figure is the value for intake by the maximum consumer surveyed in the Suquamish tribal study).

[45] EPA's most recent calculations assume a slightly greater fish consumption rate of 22 grams/day.

[46] O'Neill, *Fishable Waters*, *supra* note 43, at Table 1.

[47] Importantly, this fact also renders the use of probabilistic risk assessment techniques, such as Monte Carlo analysis, inappropriate for jurisdictions such as Idaho and Washington.

highly exposed subpopulations may be subjected to risk levels of 10^{-4} is effectively a determination that *tribal people* may be subjected to risk levels of 10^{-4}.

Previously, the state of Idaho had deemed "acceptable" a risk level of 10^{-6}.[48] This is the risk level that Idaho found tolerable *when it assumed that everyone was more or less equally likely to be on the receiving end of the risk of cancer* – when it employed the national general population default rate for fish intake in its calculations. Now, however, Idaho has been required to consider studies that demonstrate both that fish intake is highly variable and that tribal people are among the very highest consumers. Why, now, when EPA has instructed IDEQ to consider this data and to ensure that its standards are "adequately protective of the most highly exposed population"[49] (and when Idaho might be expected to increase its FCR) has IDEQ proposed to reconsider its longstanding cancer risk level? If Idaho now deems acceptable a tenfold increase in its risk level, it cannot deny the implication of this shift: namely, that Idaho believes it to be "okay" for risk-producers to transfer the costs of their processes to identifiable people, tribal people, in the form of increased cancer risk.

If Idaho's decisions regarding the risk level and other aspects of its water quality standards permit tribes to be disproportionately impacted, they may run afoul of commitments to environmental justice. EPA has indicated that it will take seriously its obligations to ensure environmental justice in discharging its duties and in overseeing states' administration of their programs. Executive Order 12,898 commits agencies of the federal government to further environmental justice and specifically mentions to need to protect "subsistence consumption of fish and wildlife."[50] Federal civil rights laws prohibit recipients of federal funds, including state environmental agencies, from administering their programs in a manner that discriminates against American Indians.[51] Moreover, EPA has recently emphasized its particular commitment to ensuring environmental justice for tribes, their members, and indigenous people. EPA's July 2014 *Policy on Environmental Justice for Working with Tribes and Indigenous Peoples* commits in this context to addressing disproportionate risks to human health and the environment.[52] EPA also commits to encouraging states to implement environmental justice principles when states' programs, policies, and activities may affect tribes and their members.[53]

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[48] U.S. Environmental Protection Agency, Region X, Technical Support Document: EPA's Disapproval of the State of Idaho's Revised Human Health Water Quality Criteria for Toxics Submitted on July 7, 2006 10 (May 10, 2012)[hereinafter EPA, Idaho Disapproval TSD].

[49] Letter from Michael A. Bussell, U.S. Environmental Protection Agency, Region X, to Barry Burnell, Idaho Department of Environmental Quality 2 (May 10. 2012)[hereinafter EPA, Idaho Disapproval Letter].

[50] Executive Order 12,898: Federal Actions to Address Environmental Justice in Minority Populations and Low-Income Populations (Feb. 11, 1994) (singling out the issue of "subsistence consumption of fish and wildlife" in section 4-4, the only subject matter issue receiving specific mention in the Executive Order).

[51] Civil Rights Act of 1964 § 106, 42 U.S.C. § 2000d (2012); 40 C.F.R. § 7 (2012).

[52] U.S. Environmental Protection Agency, Policy on Environmental Justice for Working with Tribes and Indigenous Peoples 1 (July 24, 2014), *available at* http://www.epa.gov/oecaerth/environmentaljustice/resources/policy/indigenous/ej-indigenous-policy.pdf.

[53] *Id.* at 4 (Principle 16).

VI. Risks That Impair Legally Protected Rights

The IDEQ Risk Discussion Paper also does not mention the fact that the tribes impacted by Idaho's determination regarding risk have legally protected rights to take fish. As I have discussed at length in an article published by the *American Indian Law Journal*, tribes' fishing rights, which are secured by treaties and other legal protections, can be undermined when the environments that support the salmon and other fish are permitted to be degraded, leading to depletion and contamination of the fish resource.[54] If IDEQ opts for a less protective risk level and thereby derives more lenient water quality standards, it may impair tribes' rights to harvest and consume fish. Presumably, the IDEQ Risk Discussion Paper does not take up this topic because it intends to engage the question fully at another opportunity. My comments, similarly, will not undertake a thorough discussion of the import of tribes' legally protected fishing rights (but will incorporate by reference my *American Indian Law Journal* article, a copy of which will be submitted to the rulemaking docket). Note, however, that courts have repeatedly recognized that if the waters are permitted to be significantly degraded, tribes' legally protected fishing rights can be eviscerated as surely as if tribal members had been barricaded from their fishing places. Idaho, thus, may simply not be free to choose a risk level that undermines or unduly burdens tribes' fishing rights.

VII. EPA Guidance on Risk in the Water Quality Standard Setting Context

States' water quality standard-setting efforts must be framed by tribes' legally protected fishing rights and must comport with the Clean Water Act. EPA has provided guidance for these efforts; EPA has also issued particular direction to Idaho for the effort at hand. The IDEQ Risk Discussion Paper correctly notes that EPA's AWQC Guidance provides some flexibility to states to account for local circumstances and other particularized considerations as they set their water quality standards. Among these circumstances are the presence of highly exposed groups and the applicability of particular legal obligations. In Idaho's case, EPA made clear in its disapproval letter that it understands local tribes to have higher rates of fish intake and so to be among the most highly exposed.[55] EPA also reminded Idaho that it "recommends that priority be given to identifying and adequately protecting the most highly exposed population."[56]

States have cited EPA guidance for the claim that water quality standards premised on less protective risk levels, e.g., 10^{-5}, would be "legitimate and approvable."[57] But EPA has qualified its willingness to entertain a range of risk levels in important ways. First, EPA has recognized – as it must – that its guidance must be considered by states as subsidiary to any applicable sources of law. This would

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[54] O'Neill, *Fishable Waters*, *supra* note 43.

[55] EPA, Idaho Disapproval Letter, *supra* note 49.

[56] *Id.* at 2.

[57] Lee Logan, *Washington Rejects EPA Push To Curb Additional Exposures In CWA Limits*, INSIDE EPA (Nov. 12, 2013)("State officials note that EPA guidance says states can use either risk level, as long as highly exposed populations are protected at least at a 1×10^{-4}, or 1 in 10,000, level. 'We were pretty careful that we didn't really show a preference for one or the other today,' [Washington State Department of Ecology Water Quality Program Director, Kelly] Susewind said. 'We think they're both legitimate and approvable.'").

include tribes' legally protected fishing rights; that is, the guidance cannot be read as authority to undermine these rights. Second, EPA has expressed concern for the *actual* risk posed to affected individuals, based on the best information available, when all of the parameters and circumstances are considered.

Additionally, EPA's AWQC Guidance must be interpreted in light of data and developments since it was published, in 2000.[58] Although there was then increasing awareness of the variability in fish consumption as among various subpopulations, EPA's guidance pre-dated the focused analysis of this issue provided by the National Environmental Justice Advisory Council (NEJAC) report in 2002.[59] As a consequence, statements in the guidance must be understood as a product of their time. Importantly, EPA's AWQC Guidance didn't contemplate fully the environmental justice issues raised by the fact that tribal people are among those most highly exposed to toxic contaminants in fish. In particular, the guidance's discussion of "subsistence" and "suppression" warrant comment.

These four issues are taken up in turn:

A. Tribal Fishing Rights

States cannot assume that EPA's AWQC Guidance has accounted for tribes' fishing rights, including rights secured by treaty and other legal agreements. Thus, while EPA's guidance outlines the considerations that will bear generally on EPA's decision whether to approve a state's water quality standards, and while EPA was surely aware at the time it published the guidance that tribes' fishing rights were implicated, EPA cannot be taken to have incorporated an analysis of how these standards intersect with tribal rights to harvest and consume fish. Nor could EPA, in guidance, purport to authorize state actions in contravention of the tribes' treaties and other agreements with the United States.[60] In fact, EPA is careful to make a disclaimer at the outset of its guidance to this effect: "This Methodology does not substitute for the CWA or EPA's regulations; nor is it a regulation itself. Thus, the 2000 Human Health Methodology cannot impose legally-binding requirements on EPA, States, Tribes or the regulated community, and may not apply to a particular situation based upon the circumstances."[61]

B. Actual Risk, When All Parameters are Considered

EPA has indicated that it will consider the actual risk that results to those affected when all of a state's selected parameters are considered, and has stated that its scrutiny will increase as a state's target risk level becomes less protective or less conservative, e.g., if it moves from 10^{-6} to 10^{-5}.[62] EPA has

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[58] EPA, AWQC Guidance, *supra* note 5.

[59] NATIONAL ENVIRONMENTAL JUSTICE ADVISORY COUNCIL, FISH CONSUMPTION AND ENVIRONMENTAL JUSTICE (2002).

[60] O'Neill, *Fishable Waters*, *supra* note 43, at 255-260.

[61] EPA, AWQC Guidance, *supra* note 5, at 1-2.

[62] U.S. Environmental Protection Agency, National Toxics Rule, 57 Fed. Reg. 60848-01 (1992) ("In submitting criteria for the protection of human health, States were not limited to a 1 in 1 million risk level (10^{-6})… If a State

emphasized that it will require "substantial support in the record," including an analysis of how the state's selected inputs to its risk assessment equation, when taken together, reasonably estimate the risk actually posed.[63] Among other things, EPA's statements suggest that states do not have unlimited flexibility to choose the least protective or least conservative values for most or all of the relevant variables, e.g., target risk level, FCR, human bodyweight, human lifespan – at least not, as here, where the result leaves people exposed to significant risk. As the IDEQ Risk Discussion Paper recognizes, moreover, people aren't actually exposed to one chemical at a time in the real world; rather, they are often exposed to multiple chemicals present in the water and, so, the fish: "If criteria for carcinogens are based on a risk of 1×10^{-6}, and if an individual is exposed to multiple carcinogens at their criteria concentrations, the total cancer risk experienced by that individual will be greater than 1×10^{-6}."[64] As IDEQ observes, "[t]his situation presents an argument for conservatism in setting criteria, favoring lower [i.e., more protective] risk levels."[65] This concern for the risks actually faced by those exposed counsels attention not only to estimates of cumulative impacts experienced by tribal members consuming at contemporary suppressed rates,[66] but also at historical or "heritage" rates, a concept discussed below under "suppression."

C. "Subsistence"

EPA's use of the term "subsistence" in its AWQC Guidance does not necessarily track a more particularized understanding of that term as it applies to Native peoples' lifeways. As set forth above, EPA uses the term "subsistence" both in describing the national default FCR for higher-consuming populations and in discussing the range of risk levels from which states might choose. Specifically, EPA indicates that states must ensure that, whatever risk levels they select, the resulting water quality

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selects a criterion that represents an upper bound risk level less protective than 1 in 100,000 (i.e., 10^{-5}), however, the State needed to have substantial support in the record for this level…. [Among other things,] the record must include an analysis showing that the risk level selected, when combined with other risk assessment variables, is a balanced and reasonable estimate of actual risk posed, based on the best and most representative information available. The importance of the estimated actual risk increases as the degree of conservatism in the selected risk level diminishes. EPA carefully evaluated all assumptions used by a State if the State chose to alter any one of the standard EPA assumption values.").

[63] *Id.*

[64] IDEQ, Risk Discussion Paper, *supra* note 1, at 7.

[65] *Id.* In a related vein, people aren't exposed to contaminants solely via the fish consumption pathway. For threshold pollutants, concern for an individual's total exposure counsels enlisting more protective assumptions for the relative source contribution (RSC). One would expect that a state seeking to depart from EPA's default assumptions for RSC in the direction of less protection to have to satisfy a heavy burden justifying this move – one that addressed the potential for tribal people's exposure to exceed threshold levels recognized to be safe.

[66] Studies of cancer risks from the multiple chemicals present in the Columbia River Basin suggest reason for concern. When one considers particular species or sites, the risk levels are sobering. For example, at a site between the John Day and McNary dams, a person consuming fish at contemporary levels documented in the CRITFC survey (389 g/day) has an excess cancer risk between 1 in 100 and 1 in 1000 for all four species surveyed (i.e., steelhead, fall Chinook, largescale sucker, and white sturgeon). EPA and CRITFC, Columbia River Basin Contaminant Survey, app. N, 2-3 and fig. 6-26. (2002), *available at* http://yosemite.epa.gov/r10/oea.nsf/0/C3A9164ED269353788256C09005D36B7?OpenDocument. This estimate of risk is for whole body samples and assumes a 70-year exposure duration.

standards do not pose a risk above 10^{-4} to those more highly exposed, such as “sportfishers” or “subsistence fishers.” However, EPA’s use of these terms here is generic. EPA did not (and arguably cannot) authorize states to impose disproportionately greater risks on tribal fishers by its reference to “subsistence fishers.” In fact, in the Technical Support Document (TSD) for the AWQC guidance, EPA’s use of the term “subsistence” is not consistent.[67] While the term often includes tribal populations alongside other higher-consuming populations, EPA clearly does not mean to refer only to tribal people or other American Indians/Alaska Natives in discussing “subsistence” fishers. Rather, EPA seems to use the word in its more general sense, i.e., to refer to individuals who simply eat a lot of fish, for whatever reason. Thus, for example, among the groups considered in the TSD’s discussion of “subsistence” are “Florida residents receiving food stamps,” and “high-end Caucasian consumers on Lake Michigan.”[68] By contrast, the term “subsistence” is a term of art in some contexts, and is understood by many American Indian and Alaska Native people to refer to a set of interwoven cultural practices and lifeways that includes but is not coterminous with heavy reliance on fish, wildlife, and other natural resources for food and other purposes. Given EPA’s general use of the term “subsistence,” its stated willingness to tolerate a less protective risk level for “subsistence fishers” cannot be taken to suggest that it has explicitly authorized less protective risk levels for tribal people or other American Indians/Alaska Natives. While EPA was clearly aware at the time it issued its guidance that tribal people were among those highly exposed groups and subpopulations consuming fish at the greatest rates, EPA never attempted to delineate precisely who it meant to include in the term “subsistence.”

D. Suppression

EPA’s AWQC guidance also pre-dated widespread recognition of the problem of “suppression,” which was highlighted by the NEJAC report in 2002.

“A ‘suppression effect’ occurs when a fish consumption rate (FCR) for a given population, group, or tribe reflects a current level of consumption that is artificially diminished from an appropriate baseline level of consumption for that population, group, or tribe. The more robust baseline level of consumption is suppressed, inasmuch as it does not get captured by the FCR.[69]

For tribal people in the Pacific Northwest, the forces of suppression, often perpetrated or permitted by federal and state governments, have included inundation of fishing places; depletion and contamination of the fishery resource; and years of prosecution, intimidation, and gear confiscation. By contrast, a baseline reflecting tribes’ historical or “heritage” rates would not be distorted by suppression effects. Scholars of risk assessment have developed methods for deriving quantitative estimates of these historical or “heritage” rates for tribes in the Pacific Northwest. For example, Barbara Harper, et al.

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[67] O’Neill, *Variable Justice*, *supra* note 43, at n.194 (cataloguing different uses of the term “subsistence,” and different groups included among those referred to as “subsistence fishers” in the TSD).

[68] *Id.*

[69] NEJAC, *supra* note 59, at 43-45.

concluded that "[h]istorically, the Spokane Tribe consumed roughly 1,000 to 1,500 grams of salmon and other fish per day."[70]

EPA's recent "Frequently Asked Questions" document remedies the AWQC's silence on this vital point by recognizing the issue of suppression.[71] EPA's recent approval of the Spokane Tribe's water quality standards, moreover, signals its support for addressing suppression by use of a FCR premised on historical or "heritage" fish intake rates.[72] Given that contemporary rates and practices reflect fish consumption at or close to its nadir – a point vividly illustrated by the Nez Perce Tribe's presentation on suppression during the October 2nd public meeting[73] – an FCR selected from the 90th or even the 99th percentile of contemporary consumption surveys will be considerably lower than fish intake consonant with a more robust fish resource and fuller exercise of tribal fishing rights.

In sum, EPA's AWQC Guidance cannot be taken to authorize states to promulgate water quality standards that expose tribal people disproportionately to elevated risk of cancer and that undermine rights to fish that are secured to tribes by treaty and other legal agreements.

Conclusion

Thank you for considering these comments and the document they incorporate by reference.

Respectfully submitted,

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[70] Barbara L. Harper, et al., *The Spokane Tribe's Multipathway Subsistence Exposure Scenario and Screening Level RME*, 22 RISK ANALYSIS 513, 518 (2002).

[71] U.S. Environmental Protection Agency, Human Health Ambient Water Quality Criteria and Fish Consumption Rates Frequently Asked Questions (Jan. 18, 2013), *available at* http://water.epa.gov/scitech/swguidance/standards/criteria/health/methodology/upload/hhfaqs.pdf.

[72] Letter from Daniel D. Opalski, Director, Office of Water and Watersheds, U.S. Environmental Protection Agency, Region X, to Rudy Peone, Chairman, Spokane Tribe of Indians (Dec. 19, 2013).

[73] Nez Perce Tribe, The Nez Perce Tribe and its Fisheries: "Our Fate and the Fate of the Fish are Linked," Powerpoint Presentation (Oct. 10, 2014) *available at* http://www.deq.idaho.gov/media/1118105/58-0102-1201-nez-perce-tribe-fisheries-presentation-100214.pdf.

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Washington State Department of
Health

PCB Chemical Action Plan

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PCB Chemical Action Plan

by

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Summary of Findings

PBT Rule and Chemical Action Plans

A Chemical Action Plan (CAP) identifies, characterizes, and evaluates uses and releases of specific persistent, bioaccumulative and toxic chemicals (PBT) or a group of chemicals and recommends actions to protect human health and the environment (173-333 WAC). PBTs are considered the “worst of the worst” chemical contaminants because they remain in the environment for a long time, and build up within organisms and/or the food chain.

This Polychlorinated Biphenyls (PCBs) CAP estimates releases of PCBs from various sources to air, land, and water. It also describes the physical and chemical properties of PCBs and why they are considered toxic to humans and other organisms. The recommendations are a set of actions to reduce and phase out uses, releases, and exposures in Washington in consideration of current management approaches. An economic analysis on the cost of recommendations and the most promising options is also included.

PCBs

From 1929 to 1979 about 600,000 metric tons of PCBs were commercially manufactured in the US. The 1976 *Toxics Substances Control Act* (TSCA) prohibited manufacture, processing, and distribution of PCBs. While TSCA is often referred to as a “ban” on PCBs, the law allowed some historical uses to continue, and set allowable levels of inadvertent production of PCBs in other products. PCBs are also regulated under additional state and federal laws, and they are not always consistent. For example, the level of PCBs that is allowed in products under TSCA is millions of times higher than what is allowed in water under the Clean Water Act. This leads to water permit holders being held responsible at the end of their pipe for PCBs that came from other products. Back in the late 70’s the total amount seemed small and the amount allowed in each product seemed low, but now we know that it’s high compared to levels that impact human health..

PCBs are synthetic compounds that consist of 209 possible arrangements (called congeners) of chlorines around a biphenyl molecule. PCBs were sold commercially as various mixtures of these different arrangements. They are also found in the environment as various mixtures, bioaccumulate as complex mixtures, and have been assessed for toxicity largely as mixtures. They are largely regulated as total PCBs. As a result, Ecology chose to evaluate available information on all PCB compounds.

Major sources

For the purposes of this CAP, sources are considered to be the original material, such as PCBs in transformers. PCBs move through pathways such as stormwater and expose people and wildlife.

Current PCB levels in Washington State represent both historical uses and ongoing manufacturing processes that create PCBs. A large reservoir of past uses of PCBs includes electrical equipment such as transformers and capacitors, and building materials such as caulk and paint. About 75 percent of PCBs produced before 1979 were used in transformers and capacitors, including small capacitors in lamp ballasts and appliances. The second largest pre-1979 use, about 10 percent, was as plasticizers, including in caulk.

PCBs are still unintentionally generated by combustion and by different manufacturing processes, such as for some pigments and dyes. While there is a lot of recent research on the generation of PCBs during production of pigments and dyes, little work has been done to shed light on other processes that are likely to generate and release new PCBs. Non-point releases, such as from consumer products, are becoming increasingly important to control and reduce overall PCB delivery to humans and the environment.

Pathways

Lower chlorinated congeners are more readily emitted to the air from sources such as old caulk and intact lamp ballasts. When lamp ballasts fail or caulk is disturbed, a greater amount of all of the congeners are released. Lower chlorinated congeners travel further in air compared to the higher chlorinated congeners, and eventually all the congeners are deposited onto surfaces. PCBs do not readily dissolve in water and they bind to particles. Particle-bound PCBs can be transported through stormwater and end up in sediment. The relative abundance of PCBs in sediment is seen in Puget Sound where 97 percent of the PCBs are bound to sediment (1440 kg), less than three percent are in organisms (40kg), and less than one percent (10 kg) are dissolved in water. Under certain conditions, such as in the Spokane River, PCBs are found in the water column rather than in sediments.

Stormwater is the largest delivery pathway to surface waters for PCBs statewide. Loadings from water treatment plants and atmospheric deposition are each less than 10 percent of the total, although atmospheric deposition is less well studied. There are smaller pathways, such as from salmon that accumulate PCBs while in the Pacific Ocean and then return to Washington to spawn. While the long-term goal is to prevent PCBs from entering stormwater, it is important to continue work on stormwater management, including piloting new technologies.

Exposure

Levels of PCBs in the environment are mostly declining, but PCBs are still widespread. Wildlife are exposed to PCBs in their diet, along with PCBs in water, soil, and sediments. PCBs accumulate in fatty tissues, including in animals eaten for food. PCBs in food are the most significant source of exposure for most people, and we are particularly concerned with levels of PCBs in fish we eat. People are also exposed to PCBs in air, water, soil, and house dust. Levels of PCBs in people have declined since the 1980s, but everyone in the US has detectable levels of PCBs in their bodies. PCBs remain in people and animals for different numbers of years, varying by type of organism and type of PCB congener. Because PCBs are more readily absorbed than excreted, they accumulate in the body over time.

Toxic effects

PCBs are persistent in the environment, build up in the food chain, and can cause adverse health effects in humans and wildlife including cancer and harm to immune, nervous, and reproductive systems. PCBs disrupt thyroid hormone levels in animals and humans, hindering growth and development.

Priorities for new actions

As shown in Table 1 below, there are a variety of PCB sources with different concerns (each estimate is presented in the order it is discussed in the section on Sources, Uses and Releases and how each is derived is explained in that section). The table divides sources into those that are a legacy of the historic uses before 1979 and those that are currently ongoing.

We are especially concerned about exposure to children in school buildings with old lamp ballasts and other PCB-containing building materials. It is uncertain how many pre-1979 lamp ballasts are still in use, but they should be removed both for their potential to expose people to PCBs when they fail and because newer lights are more energy efficient. A large reservoir of PCBs in old caulk and other building materials is slowly being released into the environment. Releases from building materials can be greatly accelerated during remodeling and demolition. There is an opportunity, through use of best management practices, to prevent releases of PCBs during remodeling and demolition.

Inadvertent generation of PCBs is a potentially large and important source of uncontained PCBs, but little is known about the processes that inadvertently generate PCBs and what products contain them. We do know PCBs are created as a byproduct of pigments and dyes, and this creates a regulatory burden for paper recycling, municipal treatment works, and other dischargers in Washington. Finding alternative pigments and dyes will reduce PCB releases to the environment and alleviate water treatment burdens on business and municipalities.

While about 75 percent of the PCBs produced prior to 1979 were used in electrical equipment, most of the transformers and large capacitors that contain PCBs have been identified and replaced. The remaining equipment is largely monitored for spills that are cleaned up, further reducing the impact to people and the environment.

Table 1. Summary of sources, reservoirs, releases, exposures and priorities.

Source	Legacy reservoir of PCBs	Annual releases of PCBs (kg/yr)	Potential exposure pathways and concerns	Is the release contained?	Priorities
Historic uses					
transformers	100-200 kg	< 2	Accidental spills, which are identified and cleaned up.	Yes	
large capacitors	20 metric tons	10 to 80	Accidental spills, which are identified and cleaned up.	Yes	
lamp ballasts	100-350 metric tons	400 to 1,500	Continual release of lower concentrations, with high concentrations released when the ballast fails.	Yes	**In school buildings as part of energy efficiency improvements.**
small capacitors	1-35 metric tons	3 to150	Disposal in landfills from a variety of old appliances.	Yes	
other closed uses		unknown		Yes	
caulk	87 metric tons	160	Continual release of lower concentrations into the air, with high concentrations released when materials are disturbed.	No	**Remodeling and demolition, especially in schools.**
other open uses		unknown		No	
Current generation					
pigments and dyes	N/A	0.02 to 31[a]	Continual release of lower concentrations, with higher concentrations released during recycling.	No	**Identify and promote safer alternatives.**
other inadvertent generation	N/A	900	Concerns about both continual releases and potential large releases.	No	**Identify processes and products first and then identify and promote safer alternatives.**
residential waste burning	N/A	199	Released to air and already addressed by current regulations.	No	
commercial marine vehicles	N/A	0.4	Released to air and already addressed by current regulations.	No	

a. This estimate is for PCB-11, although additional congeners are present.

Recommendations for New Actions

The goal of a CAP is to recommend actions to protect human health and the environment. Averting toxic exposures and avoiding future costs is the smartest, cheapest, and healthiest approach. The priority recommendations address the largest sources, largest reservoirs, uncontrolled sources, protect the most people, or protect especially vulnerable children, in the context of existing programs, costs, and available technology. Ongoing permitting, cleanup, and other actions are crucial to any efforts on PCBs. The following recommendations are for new actions in addition to our existing efforts to reduce PCBs.

The recommendations are based on an evaluation of the following factors associated with implementing the action: 1) environmental and human health benefits, 2) economic and social impacts, 3) feasibility, 4) availability and effectiveness of safer substitutes, and 5) consistency with existing federal and state regulatory requirements.

Historic PCB-Containing Building Materials

1. Identify PCB-containing lamp ballasts in schools and other public buildings. Encourage replacement with more energy efficient PCB-free fixtures.

Goal: Remove remaining PCBs lamp ballasts from schools and other publically owned buildings.

Before 1979 PCBs were widely used in fluorescent lamp ballasts. PCB-containing lamp ballasts still in use should be identified and replaced with more energy efficient lighting. These ballasts have outlived their useful lives and are at high risk for failing (dripping, smoking, and catching fire). Ballast failures can expose children to concentrated PCB oils and elevated PCBs in air. Low concentrations of lower chlorinated PCB congeners are continually released from lamp ballasts. When ballasts fail, high concentrations of a broader spectrum of congeners are released, so it is important to find and remove the lamp ballasts before they fail.

There is no easily accessible source of information on how many of our approximately 9,000 school buildings are of the age and construction type likely to have PCB-containing lamp ballasts. The first step is to conduct a survey on schools (and other public buildings as time and resources allow) to identify buildings most likely to contain PCBs based on age, type of construction and scope of any past remodeling. This data will be used to identify those buildings where PCB-containing light ballasts are likely still in use. Lamp ballasts with PCBs can then be identified through visual inspection.

Public money should be used to remove PCB-containing lamp ballasts from schools and other public buildings. Since 2009, the legislature has provided money to the Office of the Superintendent of Public Instruction (OSPI) and the Department of Commerce to support energy efficiency measures in schools and other public buildings. It makes sense to combine PCB removal with increasing energy efficiency rather than create a new program just for removal of PCB-containing ballasts. If the grant programs are not funded, the legislature should establish a fund to help offset the costs of replacing PCB-containing lamp ballasts. Schools with PCB-containing lamp ballasts will be provided with information about the importance of removing these ballasts and referred to OSPI (or Washington State Department of Health and other available resources) to replace these fixtures with more energy-efficient lighting. Environmental justice will also be considered in setting priorities for removing PCB-containing lamp ballasts.

2. Develop and promote Best Management Practices (BMPs) to contain of PCBs in building materials currently in use and those slated for remodel or demolition.

Goal: Reduce exposure to people from PCBs in historic building materials and prevent PCBs in building materials from getting into stormwater.

Ecology should work to prevent PCBs currently in building materials from being released into the environment.Historically, PCBs were used at high levels in some caulks and paints. Studies in other areas have shown the widespread occurrence of PCB-containing caulk in buildings from about 1950-1980, especially masonry buildings, and smaller sampling efforts in Washington support this conclusion. There is some information about PCB-containing building materials in the Duwamish basin and other information from source tracing, such as PCBs in sidewalk and building caulk in Tacoma.

Based on available data in Washington, other government programs, and the scientific literature, Ecology would develop BMPs for containing PCBs to prevent exposure during the life of the building and during remodeling or demolition. Lower concentrations of lower chlorinated congeners are continually released from caulk and paint, with higher concentrations of a broader spectrum of congeners released when the materials are disturbed. Ecology should also provide education and outreach on BMPs to local governments and those in the building trades.

While Ecology is working on BMPs, it should also support assembling existing information into a PCB Source Control Guidance Manual that can aid Local Source Control work to identify and control sources of PCBs. PCB source identification work has been performed by a number of urban waters programs around the Northwest. To date, the lessons learned from each of these programs has not been synthesized and summarized for the benefit of future pollution prevention efforts at the state and local level

3. Assess schools and other public buildings for the presence of PCB-containing building materials.

Goal: Reduce children's exposure to PCB-containing building materials in schools.
Goal: Prevent PCBs in building materials from getting into stormwater.

Many historical building materials, such as caulk and paint, have been found to contain high levels of PCBs. These materials are more common in industrial buildings, including schools, compared to residential buildings. It makes sense to focus on schools for testing and remediating these materials, as children are more sensitive to PCBs and the buildings are usually publically owned. Washington has not tested schools for PCBs, but other states have found high levels of PCB contamination in schools.

The first step in Recommendation #1 is to get information on how many of our approximately 9,000 school buildings are of the age and construction type likely to have PCB-containing materials. The information would be used to prioritize schools for testing, pending the availability of funding to either contain or remediate PCBs that pose a risk for children and teachers. A similar approach should be used to assess other public buildings once the assessment and remediation of schools is complete.

Ecology would initially focus on determining how many schools are likely to contain PCBs in historic building materials, narrow that list with visual inspections and then physical testing to determine the scope of the problem in Washington. This will determine how much time and money will be required for remediation and allow for long term planning, including funding. As Ecology learns more about PCB-containing building materials in Washington schools and other buildings, that information will be used to improve efforts to locate and remediate buildings. Environmental justice will also be considered in setting priorities for removing PCB-containing building materials.

Current Manufacturing Processes

4. Learn more about what products contain PCBs and promote the use of processes that don't inadvertently generate PCBs.

Goal: Reduce newly generated PCBs in manufacturing processes.

In 1982, the Environmental Protection Agency (EPA) identified 70 manufacturing processes likely to inadvertently generate PCBs. Little is known about most of this potentially large source of uncontained PCBs, including which congeners are produced. More information is known about PCBs in pigments and dyes, which are known to be released into the environment in stormwater, effluents from municipal treatment works, and effluents from pulp mills re-pulping post-consumer paper. Unpermitted non-point releases, such as from consumer products, are becoming increasingly important to control to reduce overall PCB delivery. Ecology should

work with EPA, manufacturers, the Northwest Green Chemistry Center, and other partners to identify products likely to contain PCBs, including a workshop focused on bringing together the supply chain to look for solutions to the current PCBs in products issue.
Ecology should test identified products and give that information to the Department of Enterprise Services (DES) to assist them in implementing new purchasing policies (RCW 39.26.280) that provide a preference for products that do not contain PCBs. Government purchasing of non-PCB-containing products is an effective way to both identify products that don't contain PCBs and encourage suppliers to remove PCBs from their products. Once products are identified that contain fewer PCBs, the information will be available shared with other purchasers, such as by schools, cities and residents. Ecology should support other government and business entities that adopt similar purchasing policies to drive the market-demand for PCB-free products.

Ecology, the Washington State Department of Health (DOH), and DES should work together to educate purchasers and vendors in the state supply chain by developing a focus sheet that concisely outlines the problem and its potential impacts on compliance with water quality standards, as well as impacts on people and the ecosystem.

Ecology should begin with directing an alternatives assessment for pigments and dyes, based on the Interstate Chemicals Clearinghouse (IC2) Alternatives Assessment Guide. Businesses and other concerned stakeholders would be asked to participate in the alternatives assessment process. If there are currently no alternatives that do not generate PCBs, Northwest Green Chemistry should work to develop alternative processes.

In addition to investigating which products contain inadvertently generated PCBs and finding safer alternatives, Ecology and DOH should petition the federal government to reform current regulations under the *Toxics Substances Control Act* (TSCA). TSCA generally allow a maximum of 50 ppm of inadvertently generated PCBs in most products, while Washington standards under the *Clean Water Act* (CWA) to protect human health that allow only 0.00000017 ppm (170 ppq or parts per quadrillion) PCBs in water bodies. The TSCA allowance is over 294 million times greater than the CWA criteria for human health and it is suspected that the TSCA allowance is likely a pathway for new PCBs to enter the environment, resulting in a paradox between these regulatory structures that must be resolved. The lower CWA standards are based on protecting human health and what we know about the health effects of PCBs. The higher allowable level of PCBs in products leads to higher levels of PCBs in water, and the responsibility of PCB levels in water are borne by downstream permittees that are not generating PCBs. Allowable levels of newly generated PCBs in products must be lowered by the federal government in order to meet water quality standards to protect human health and the environment.

Historic Electrical Equipment

5. Survey owners of historic electrical equipment, including transformers and large capacitors.
Goal: Confirm estimates of EPA-regulated electrical equipment with more than 500 parts per million (ppm) PCBs, learn what is known about electrical equipment with PCBs greater than 2ppm, and find out when such electrical equipment is estimated to be replaced.

Before 1979, the majority of PCBs were produced for use in electrical equipment. Major industries and electrical utilities indicate that they have either already removed these potential sources or have plans to do so. Ecology should survey the state's 61 utilities and other owners of electrical equipment to confirm that this is the case and provide technical assistance for proper replacement and disposal.

Federal regulations focused on transformers with more than 500 ppm PCBs. According to industry, many of these transformers and other electrical equipment have already been disposed of, but we do not have updated inventories of this equipment to reflect this progress and target technical assistance to remaining equipment. To avoid spills or other unexpected releases, the users should know which pieces of equipment have PCBs and have a plan to remove all such pre-1979 equipment from service or an estimate on when they will be replaced during regular maintenance. This inventory would not include small capacitors. This survey would not require additional testing or disposal of equipment by a certain date. Owners would be asked what they know about detectable levels of PCBs in their equipment, using the industry standard detection level (currently 2 ppm) and when they expect all the equipment to be replaced based on their current maintenance practices. A statewide inventory will allow the state to confirm current inventories and target efforts to prevent releases of PCBs from this equipment. Ecology should also work with EPA to update EPA's 1998 inventory and more accurately reflect the PCB transformers that are no longer in use.

PCB concentrations in many pieces of electrical equipment are high compared to other sources, so relatively rare leaks and spills can release a significant amount of PCBs into the environment. While PCB spills from electrical equipment are cleaned up, there are some spills where PCBs enter the waterways before being cleaned up, such as from transformers that are located close to storm drains or water bodies.

Ecology should also investigate the possibility of providing state financial assistance to businesses, state agencies, cities, municipalities, and schools to accelerate this process of collecting and properly disposing of equipment with concentrated PCBs.

Multi-Source

6. Expand environmental monitoring to identify any new areas requiring cleanup and investigate air deposition.

Goal: Find areas with highly concentrated PCBs and clean them up to prevent the wider release of PCBs.
Goal: Find about more about distribution of PCBs in the state to prioritize future actions.

Ecology should expand environmental monitoring of water, fish tissue, and sediment to identify PCB hot spots. For example levels of PCBs in fish were high enough to prompt Oregon and Washington to issue a joint consumption advisory for select species from Bonneville Dam upstream to McNary Dam. This led to the discovery of an historical landfill of electrical equipment on Bradford Island near Bonneville Dam on the Columbia River. This site is currently a federal Superfund site. A number of fish sampling efforts in the Columbia basin (CRITFC, Hanford Corridor Study and Ecology's Freshwater Fish Contaminant Monitoring Program) found other areas (mid-Columbia and Snake River) with elevated levels of PCBs and a number of hydroelectric facilities. To our knowledge, no work has been done to identify sources of contaminants in these areas. The proposed monitoring would conduct sampling to determine if contaminant sources exist in areas with a focus on hydroelectric facilities. Mapping and prioritization of historical information on the location of potential sites, such as landfills, industrial sites, railroad switching yards, etc., should also be used to find potential sites. Newly identified sites would be prioritized for cleanup using existing procedures.

In addition to identifying new hot spots, Ecology should continue its trend monitoring to show changes in PCBs in the environment and organisms over time.

Air deposition is a potentially significant pathway for PCBs to move into the environment. Ecology should investigate monitoring air deposition to assess the relative importance of this pathway.

7. Conduct a public educational campaign.

Goal: Provide information to residents about ways they can minimize exposure.
Goal: Raise awareness of the problems associated with current and past production of PCBs.
Goal: Educate residents to identify and addresses possible household sources of PCBs.

Ecology and DOH should work together with local entities to help residents, people who fish, schools, local governments, and businesses understand the risks associated with PCB exposure and ways to reduce risks, including the availability of safer alternatives.
The public educational campaign will include perspectives on public health risks and information on where exposures to PCB are most likely. It will also include advice on how individuals can

minimize their own exposures to PCBs. The campaign will include where to look for potential sources of PCBs in households, such as in old appliances, electrical equipment, and building materials. It will also teach the public how to safely remove and dispose of these materials to prevent PCB releases.

8. Conduct a study on which PCB congeners are present in Washington residents.

Goal: Learn more about PCB congeners to which Washington residents are exposed.
Goal: Find out more about the distribution of PCBs in Washington to prioritize future actions.

Within available resources, DOH should conduct biomonitoring of Washington residents for PCBs including PCB-11 and other inadvertently produced PCBs associated with dyes, pigments, and printing inks. This would be in tandem with learning more about what other processes produce certain PCB congeners. Use the data to better understand 1) the extent of total human exposure from multiple potential pathways and 2) the relative contribution of these congeners to human body burden of PCBs. In addition, use the data to estimate the statewide distribution of PCBs in Washington residents. This will provide a better baseline than national data, given the elevated levels of PCBs in local fish populations and relatively high fish consumption in a number of Washington communities and regions.

Summary of Recommendations

1. Identify PCB-containing lamp ballasts in schools and other public buildings. Encourage replacement with more energy efficient PCB-free fixtures.

2. Develop and promote Best Management Practices (BMPs) to contain PCBs in building materials currently in use and those slated for remodel or demolition.

3. Assess schools and other public buildings for the presence of PCB-containing building materials.

4. Learn more about what products contain PCBs and promote the use of processes that don’t inadvertently generate PCBs.

5. Survey owners of historic electrical equipment, including transformers and large capacitors.

6. Expand environmental monitoring to identify any new areas requiring cleanup and investigate air deposition.

7. Conduct a public educational campaign.

8. Conduct a study on which PCB congeners are present in Washington residents.

Implementation Steps

The recommendations outline a set of first steps in a long-term plan to reduce PCB releases and exposures. Due to the magnitude and diversity PCB sources, many of the approaches will take significant commitments of time and money to implement effectively. In addition, Ecology and DOH can support other agencies, but it is up to the other agencies to carry out some of the recommendations.

Ecology focuses on prevention, which is the smartest, cheapest, and healthiest approach to reducing PBTs. Our priority is actions that result in the biggest reduction in exposure to the most sensitive receptors. However, sometimes other opportunities to reduce PCBs may arise and it makes sense to reduce all sources of PCBs where possible.

Ecology will continue our existing programs on PCBs to the environment, such as cleanup, stormwater management, and permits. DOH will also continue their existing programs, including fish advisories and working with residents to reduce their exposures. Expanding or increasing programs will require additional funds. Ecology and DOH will work on acquiring additional funds, but cannot predict when we will be able to obtain them.

Ecology and DOH will seek resources to implement the following recommendations in:

FY16-17

Recommendation 1. Survey and assess PCB-containing lamp ballasts in schools and other public buildings. Encourage replacement with more energy efficient PCB-free fixtures.

Recommendation 2. Develop and promote Best Management Practices (BMPs) for containment of PCB-containing materials in buildings currently in use and those slated for demolition.

Recommendation 4. Learn more about what products contain PCBs and promote the use of processes that don’t inadvertently generate PCBs. This will begin with an alternatives assessment on PCBs in pigments. Use this information to continue to request reforms of PCB regulations under TSCA.

Recommendation 5. Survey owners of historic electrical equipment.

Recommendation 6. Expand environmental monitoring to identify any new areas requiring cleanup.

Recommendation 7. Collaborate with DOH to conduct an educational campaign.

FY18 -21

Recommendation 3. Assess schools and other public buildings for the presence of PCB-containing building materials. This work will start once school buildings have been surveyed (Recommendation 1) and the BMPs have been developed (Recommendation 2).

Recommendation 4. Learn more about what products contain PCBs and promote the use of processes that don't inadvertently generate PCBs. Ongoing work will focus on determining other processes that inadvertently generate PCBs and alternatives that do not generate PCBs. Ongoing work to support reforms of PCB regulations under TSCA.

Recommendation 6. Expand environmental monitoring to identify any new areas requiring cleanup. This project is expected to be finished in FY18.

Recommendation 7. Conduct an educational campaign. This education and outreach will be ongoing past FY17.

Recommendation 8: DOH will seek resources to design and conduct biomonitoring.

General Chemical Information

Summary

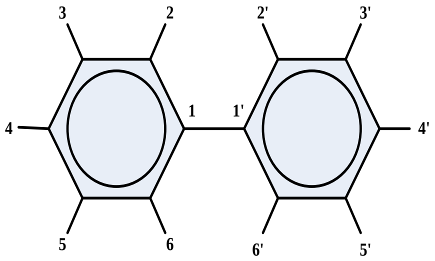

Figure 1. PCB Structure

Polychlorinated biphenyls (PCB) are a family of man-made chemicals consisting of two benzene rings joined together and containing one to 10 chlorine atoms attached to the benzene rings. There are 209 possible combinations of chlorine positions, called congeners. Depending upon the amount of chlorine present, PCBs appear as oily liquids to white crystalline solids and hard non-crystalline resins (HSDB, 2013). PCBs are hydrophobic and bind to particles. Due to their non-flammability, chemical stability, high boiling point and electrical insulating properties, PCBs were used in hundreds of industrial and commercial applications including electrical, heat transfer and hydraulic equipment; as plasticizers in paints, plastics, and rubber products; in pigments, dyes, and carbonless copy paper; and many other industrial applications (EPA 2013a).

From 1929 to 1979, 700,000 tons (or 1.4 billion lbs or 600,000 metric tons) of PCBs were commercially manufactured in the US (EPA 1997a). Most of the PCBs in the US were manufactured by Monsanto, which continued manufacture in the US until 1979 when US production stopped. PCBs were not typically manufactured as individual congeners but as mixtures called Aroclors. Individual Aroclor mixtures were manufactured using specific chemical processes imparting varying chemical characteristics such as vapor pressure, solubility, viscosity, amount of chlorination, etc. that produced a unique distribution of congeners in the different Aroclor mixtures. (HSDB, 2013) Much of the research and discussion on PCBs is centered upon the specific Aroclor mixtures. The 1976 Toxic Substances Control Act (TSCA) prohibited manufacture, processing, and distribution of PCBs. Some legacy uses of PCBs were allowed to continue.

PCBs are identified as persistent, bioaccumulative and toxic (PBT). Because of their persistence, PCBs continue to be found in the environment and contamination from legacy sources remains a problem. In addition, PCBs are not prohibited in some products at concentrations below 50 ppm. PCB concentrations of less than 50 ppm are considered to be “PCB-free.” Recent evidence has also indicated that PCBs may be found as contaminants in a wide range of consumer products because of the presence in several pigments and dyes (Hu *et al.* 2010, Rodenburg *et al.* 2010)

Physical and Chemical Properties of PCBs

A summary of typical characteristics for PCBs can be found in Table 2 (ATSDR 2000).

In general, PCB compounds range from heavy oily liquids to sticky resins, or melting crystalline solids depending upon the amount of chlorine present. These man-made compounds are odorless, colorless to light yellow or amber, and very stable and have relatively low volatility at ambient temperatures. PCBs were attractive in many applications because they resist breakdown at high temperatures or from aging, or oxidation. They persist in the environment since they do not easily biodegrade. PCBs are hydrophobic and thus do not dissolve well in water. As hydrophobic and very stable compounds, PCBs may volatilize from water despite their low vapor pressure. PCBs also easily adsorb onto organic particles in soils, sediments, biological systems, or water. (Panero *et al*. 2005) These organic particles can be transported long distances and has been shown as one of the reasons PCBs are distributed throughout the planet including remote areas.

Washington State's PBT Rule (WAC 173-333) defines persistence, bioaccumulation, and toxicity:

- The criterion for persistence is the half-life (the time it takes for half of the chemical to breakdown) of the chemical in water, soil, or sediment is greater than or equal to 60 days.
- The criterion for bioaccumulation is either:
  - The bioconcentration factor (BCF) or bioaccumulation factor (BAF) in aquatic species for the chemical is greater than 1,000.
  - In the absence of such data, the log-octanol water partition coefficient (log K_{ow}) is greater than five.
- In order for a chemical to be considered toxic, it must meet at least one of the following criteria:
  - Be a carcinogen, a developmental or reproductive toxicant, or a neurotoxicant.
  - Have a reference dose or equivalent toxicity measure less than 0.003 mg/kg/day.
  - Have a chronic no-observed-effect concentration (NOEC) or equivalent toxicity measure less than 0.1 mg/L or an acute NOEC or equivalent toxicity measure less than 1.0 mg/L.

Many but not all PCB congeners are persistent and bioaccumulative as defined in Washington's PBT Rule. Table 2 demonstrates persistence and bioaccumulation for a series of PCBs, one from each of the ten homolog groups as predicted by EPA's PBT Profiler (EPA 2012b). PCBs are often grouped by the total number of chlorine atoms and a group with the same number of chlorines is called a homolog. Washington's PBT characteristics are included in the bottom of the table (WAC 173-333). Table 4 (IPCS 1995) provides ranges of characteristics for congener groups and also includes Washington's PBT characteristics on the bottom of the table.

Although the information in Table 2 is modeled data, which should not be confused with analytical results, the PBT Profiler results do demonstrate some trends. In general, persistence as indicated by the half-lives increases as the number of chlorine atoms increase. The tendency to bioaccumulate increases until the molecular structure of the PCB becomes large enough that the amount of bioaccumulation plateaus and begins to decrease. This plateauing is a direct result of the size of the PCB molecule, which can restrict transport through cell walls. Although all PCBs shown meet Ecology's persistence criterion, some of the mono-substituted may have sufficiently low bioaccumulation factors (BCF) that they may not meet Ecology's PBT criteria.

Table 2. Summary of Typical PCB (Aroclor) Physical Characteristics (from ATSDR 2000)

Property	**Aroclor 1254**	**Aroclor 1260**	**Aroclor 1262**	**Aroclor 1268**
Molecular weight	328	357.7	389	453
Color	Light yellow	Light yellow	No data	Clear
Physical state	Viscous liquid	Sticky resin	No data	Viscous liquid
Melting point, ^{o}C	No data	No data	No data	No data
Boiling point, ^{o}C	365 - 390	385 - 420	390 - 425	435 - 450
Density, g/cm^3, 25 ^{o}C	1.54	1.62	1.64	1.81
Odor	Mild hydrocarbon	No data	No data	No data
Solubility:				
Water, mg/L	0.012, 0.57 (24^{o}C)	0.0027, 0.08 (24^{o}C)	0.052 (24^{o}C)	0.300 (24^{o}C)
Organic solvent(s)	Very soluble	Very soluble	No data	Soluble
Partition coefficients:				
Log K_{ow}	6.5	6.8	No data	No data
Vapor pressure, mm Hg at 25 ^{o}C	7.71×10^{-5}	4.05×10^{-5}	No data	No data
Henry's law constant, atm-m3/mol at 25 ^{o}C	2.0×10^{-3}	4.6×10^{-3}	No data	No data
Flashpoint, ^{o}C (Cleveland open cup)	No data	No data	195 ^{o}C	195 ^{o}C
Flammability limits, ^{o}C	None to boiling pt	None to boiling pt	None to boiling pt	None to boiling pt
Conversion factors				
Air (25 ^{o}C)	1 mg/m^3= 0.075 ppm	1 mg/m^3= 0.065 ppm	1 mg/m^3= 0.061 ppm	1 mg/m^3=0.052 ppm

Table 3. EPA PBT Profiler Estimates of Persistence and Bioaccumulation for Select PCB congeners

PCB	Congener number	CAS	Half-Life (days)				BCF[1]
			Water	Soil	Sed.	Air	
4-Chlorobiphenyl	PCB-3	2051-62-9	38	75	340	4.2	510
3,3’-Dichlorobiphenyl	PCB-11	2050-67-1	38	75	340	3.9	5,400
2,3,4’-Trichlorobiphenyl	PCB-22	38444-85-8	60	120	540	15	6,700
2,3’,5,5’-Tetrachlorobiphenyl	PCB-72	41464-42-0	180	360	1,600	13	27,000
2,2’,4,4’5-Pentachlorobiphenyl	PCB-99	38380-01-7	180	360	1,600	13	40,000
2,3,3’,4,4’,5-Hexachlorobiphenyl	PCB-156	38380-08-4	180	360	1,600	75	26,000
2,3,3’,4,4’,5,6’-Heptachlorobiphenyl	PCB-190	41411-64-7	180	360	1,600	130	12,000
2,2’,3,3’,4’,5,5’,6’-Octachlorobiphenyl	PCB-199	52663-75-9	180	360	1,600	290	5,900
2,2’,3,3’,4,4’,5,6,6’-Nonachlorobiphenyl	PCB-207	52663-79-3	180	360	1600	370	2,900
Decachlorobiphenyl	PCB-209	2051-24-3	180	360	1,600	880	12,000
WA PBT Characteristics			> 60	> 60	> 60		>1,000

BCF = Bioconcentration Factor; half-life = the amount of time it takes for the concentration of a chemical to diminish to half its original value; N/A = Not Applicable

Table 4. PCB Homolog Chemical properties (IPCS, 1995)

Congener Group	CASRN	Molecular weight (g/molecular)	Vapour Pressure (Pa)	Water Solubility (g/m^3)	log K_{OW}
Monochlorobiphenyl	27323-18-8	188.7	0.9-2.5	1.21-5.5	4.3-4.6
Dichlorobiphenyl	25512-42-9	223.1	0.008-0.60	0.06-2.0	4.9-5.3
Trichlorobiphenyl	25323-68-6	257.5	0.003-0.22	0.015-0.4	5.5-5.9
Tetrachlorobiphenyl	26914-33-0	292.0	0.002	0.0043-0.010	5.6-6.5
Pentachlorobiphenyl	25429-29-2	326.4	0.0023-0.051	0.004-0.02	6.2-6.5
Hexacholorbiphenyl	26601-64-9	360.9	0.0007-0.012	0.0004-0.0007	6.7-7.3
Heptachlorobiphenyl	28655-71-2	395.3	0.00025	0.000045-0.000	6.7-7
Octachlorobiphenyl	55722-26-4	429.8	0.0006	0.0002-0.0003	7.1
Nonachlorobiphenyl	53742-07-7	464.2	-	0.00018-0.0012	7.2-8.16
Decachlorobiphenyl	2051-24-3	498.7	0.00003	0.000001-0.000	8.26
WA PBT Criterion					>5

log K_{ow} = natural log of the octanol/water coefficient

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[1] EPA’s PBT Profiler defines the bioconcentration factor (BCF) as ‘… *a measure of the ability for a water-borne chemical substance to concentrate in fatty tissue of fish and aquatic organisms relative to its surroundings. EPA defines bioconcentration as the net accumulation of a substance by an aquatic organism as a result of uptake directly from the ambient water through gill membranes or other external body surfaces (60 FR 15366).’*

Stability of Ring Structure and Carbon to Halogen Bond

The chemistry and related stability of ring compounds was discussed extensively in the Polycyclic Aromatic Hydrocarbon (PAH) Chemical Action Plan (CAP) (Ecology, 2012a). Benzene (cyclohexa-1,3,5-triene)[2] is cyclohexane with three double bonds equally spaced throughout the molecule. Unlike compounds where the electrons forming the double bonds are localized around specific carbon atoms, the electrons in benzene's double bonds are equally shared among all six-carbon atoms. This is a defining characteristic of aromatic compounds. Benzene is typically represented by chemists as a six-carbon ring with a circle inside to represent the sharing of all electrons equally among the carbon atoms (Figure 2):

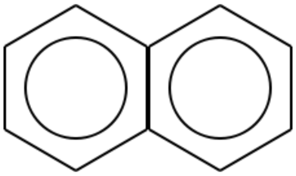

Figure 2. Chemical abbreviation for benzene

The most common theory currently accepted by chemists is that this sharing of electrons accounts for the thermodynamic stability of benzene and other aromatic compounds. Other theories have been promulgated (Cooper *et al.*, 1986) but are not currently widely accepted. Regardless, the increased stability of benzene and benzene-based compounds like PCBs can be attributed to the unique ringed structure and sharing of electrons throughout the molecule.

PCBs have another feature that contributes greatly to their stability. Carbon and chlorine form a very strong bond and the amount of energy needed to break apart the bond is higher than most other covalent bonds. The strength of this bond greatly increases the ability of PCBs to persist in the environment. Persistence is also related to the number of chlorine atoms with increasing degree of persistence with increasing chlorine mass. Comparatively, the mono- and di-chlorobiphenyls are less persistent than the larger congeners; however, all PCBs meet the definition of persistence (see Table 3 for examples).

Naming of PCB congeners

PCBs have a variable structure with two benzene rings joined together. Each benzene ring can have one to five chlorine atoms attached. The number and location of the chlorine atoms attached to the biphenyl ring determine the physical properties and characteristics of the PCB congener.

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[2] The formal name for benzene describes a cyclical compound (cyclo) with six carbons (hexa) and three (tri) double bonds (ene). The '1,3,5' indicates which carbon atoms contain the double bond. Given the structure, the double bonds are represented as between the carbons 1 & 2, 3 & 4 and 5 & 6, although in reality the electrons are equally shared with all of the carbons on the ring.

The position of the chlorine atoms are differentiated by using 2 through 6 for one benzene atom and 2’ (two prime) through 6’ (six prime) for chlorine atoms on the second benzene ring. The naming convention assumes that the two benzene molecules are joined together at the 1 and 1’ position. See Figure 3 for the carbon numbering.

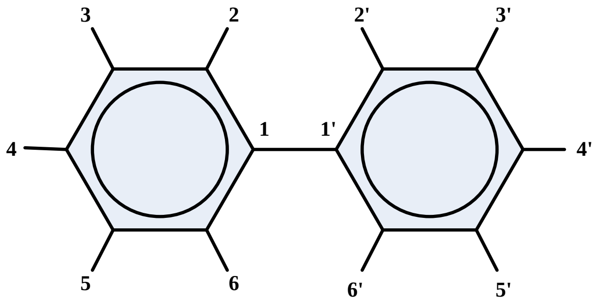

Figure 3. PCB Structure

This naming convention allows chemists to identify the structure of specific PCB congeners. For example, PCB-11 (3,3’-dichloro-1,1’-biphenyl or 3,3’-dichlorobiphenyl) contains two chlorine atoms in the 3 and 3’ position. Unless indicated otherwise, a hydrogen atom is located on the benzene ring in all the unmarked locations.

Because it is possible for the PCB-11 molecule to rotate around the 1-1’ carbon bond, there is no chemical difference between the above structure and 3,5’-dichlorobiphenyl (Figure 4).

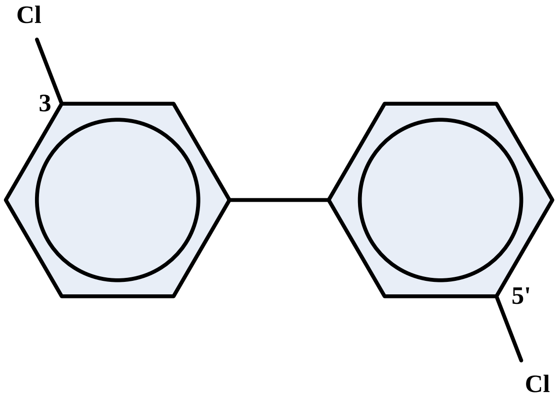

Figure 4. Alternate PCB-11 Structure

Several additional names potentially exist for PCB-11 including 5,3’-dichlorobiphenyl and 5,5’-dichlorobiphenyl. To prevent confusion, the naming convention uses the lowest numbers for these equivalent structures leading to PCB-11 being identified as 3,3’-dichlorobiphenyl. The naming convention can lead to confusion if an incorrect name is used.

Planar and non-planar PCBs

The issue of rotation can also have an impact on the relative toxicity of the PCB congeners. PCB congeners can either exist as planar where the two benzene rings are in the same plane or non-planar where the benzene rings are at 90 degree angle from each other.(ATSDR 2000). Planar and non-planar PCBs can have very different toxicity and this issue will be an important consideration in the relative toxicity of the PCB congeners discussed in subsequent sections.

A further naming convention using the terms ortho, meta and para are also used to identify the position of the chlorine atoms in a PCB molecule. If a chlorine atom is attached to the carbons adjacent to the 1 to 1’ bond between the two benzene molecules (positions 2, 2’, 6 or 6’), the chlorine atoms are said to be in the meta position. If the chlorine atom is attached to positions 3, 3’, 5 or 5’, they are in the ortho position. If they are attached to the 4 or 4’ position, they are in the para position. Table 5 and Figure 5 indicate the position of the chlorine atoms in a PCB congener using the three naming conventions:

Table 5. Table of location of chlorine atoms

Name	Carbon atom location
Meta	3,3’,5,5’
Ortho	2,2,6,6’
Para	4,4’

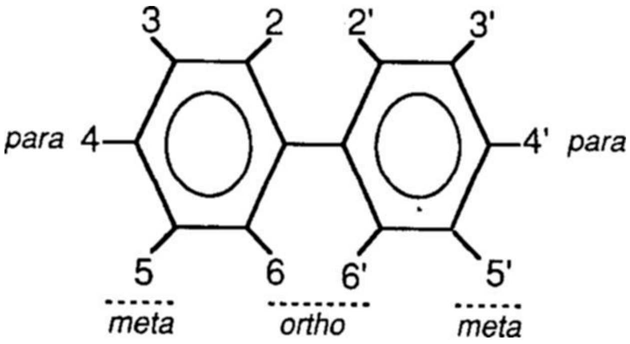

Figure 5. Location of meta, ortho, and para positions

The location of chlorine atoms plays an important role in the decomposition and toxicity of specific PCB congeners. Degradation reactions, for example, may selectively remove chlorine atoms from specific locations while PCB molecules with specific arrangements of chlorine atoms may have greater toxicity than related congeners.

Using the base structure (Figure 3), 209 different and unique PCBs (also known as congeners) can exist depending upon the number and position of chlorines involved. A list of these 209 congeners (EPA 2003) can be found in Appendix A. In addition to specific congeners, PCBs are also often grouped by the total number of chlorine atoms also called homologs (Table 4).

Historic Manufacturing

Historically, PCBs were not manufactured as specific congeners or homologs but as mixtures. Globally there was a wide range of product names for PCB mixtures (Appendix B). There were nine major mixtures in the US called Aroclors (Table 6). Prior to 1971, the Monsanto Chemical Company produced Aroclors 1016, 1221, 1232, 1242, 1248, 1254, 1260, 1262, and 1268. (ATSDR 2000). Figure 6 shows the amounts and uses of PCBs produced in the US between 1957 and 1971. (EPA 1987). Most Aroclor mixtures are named utilizing a code. Most begin with a 12 and the last two digits indicate the percentage amount of chlorine in the mixture. Therefore Aroclor 1254 contained 54% chlorine by weight, Aroclor 1216 contained 16% chlorine, etc. The only major Aroclor mixture that deviates from this system is 1016.

In 1971, Monsanto voluntarily restricted the uses of PCBs and subsequently produced only Aroclor 1016, 1242, 1254, and small quantities of Aroclor 1221. In 1974, the Monsanto Chemical Company produced slightly more than 40 million pounds (18 million kg) of Aroclor mixtures. Of the total volume of Aroclors sold in the United States for that year, the percentages of the market for each of the Aroclors were: Aroclor 1016, 64%; Aroclor 1242, 17.9%; Aroclor 1254, 17.9%; and Aroclor 1221, 0.1%. The estimated, cumulative production and consumption volumes (in millions of pounds) of PCBs in the United States from 1930 to 1975 were: total production, 1,400 (635 million kg); imports, 3 (1.4 million kg); domestic sales, 1,253 (568 million kg); and exports, 150 (68 million kg) (ATSDR 2000).

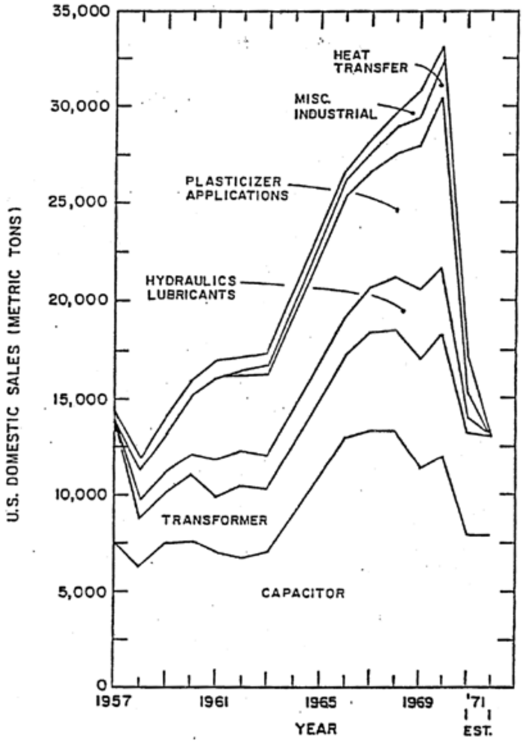

Figure 6. Monsanto Domestic sales of PCBs in the US by use (EPA 1987)

Table 6. Table of Aroclors (EPA 2013a)

CASRN	IUPAC Name
12674-11-2	Aroclor 1016
147601-87-4	Aroclor 1210
151820-27-8	Aroclor 1216
11104-28-2	Aroclor 1221
37234-40-5	Aroclor 1231
11141-16-5	Aroclor 1232
71328-89-7	Aroclor 1240
53469-21-9	Aroclor 1242
12672-29-6	Aroclor 1248
165245-51-2	Aroclor 1250
89577-78-6	Aroclor 1252
11097-69-1	Aroclor 1254
11096-82-5	Aroclor 1260
37324-23-5	Aroclor 1262
11100-14-4	Aroclor 1268
12767-79-2	Aroclor (unspecified)

These Aroclor mixtures can be fingerprinted depending upon the distribution of specific PCB congeners as indicated below (Figure 7) for the Aroclor 1260 mixture.

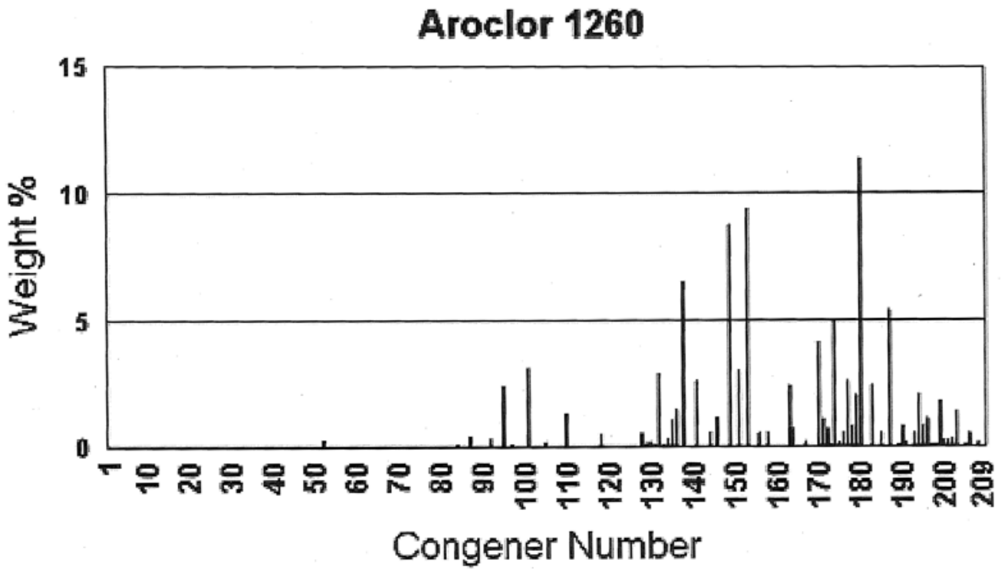

Figure 7. PCB Distribution in Aroclor1260 (EPA 2013e)

Additional fingerprints of common Aroclor mixtures can be found in Appendix C. Fingerprinting is an analytical technique that shows a distinct distribution of chemicals. Each Aroclor mixture has an identifiable distribution of PCB congeners. Aroclor fingerprinting is important as it can point toward potential PCB sources when contamination has been found in the environment. However, due to differential uptake by organisms, differential volatilization, and differential degradation, the congeners present in weathered mixtures in the environment will be different than the original congeners present in the Aroclors.

Analytical Methods

Because of their impact upon human health and the environment, considerable data exists on the presence of PCBs in a wide range of media. Historically, analytical methods were developed based upon the Aroclor fingerprints or values for total PCB concentrations. Recently, more sensitive and detailed congener-specific analyses have been developed to enable more detailed study of PCBs in the environment. PCBs are regulated under the Toxics Substances Control Act (TSCA), other Federal regulations (EPA 2013f) and state regulations such as Washington State's Dangerous Waste Regulations (WAC 173-303). Special analytical methods have been developed to meet these regulatory requirements. Methods range from traditional gas chromatography (GC), more recent high resolution GC mass spectroscopy, to new immunoassay techniques.

Numerous methods (Table 7 from National Environmental Methods Index) have been developed to analyze PCBs in a wide range of media using techniques with variable costs and detection levels. Although the list is not complete, it is indicative of the variety and type of methods currently available to test samples for PCBs. In Table 7 "Source" refers to the organization that developed the method, although it may now be required by different regulations.

For the purposes of this CAP, the three most commonly used analytical methods will be discussed in more detail:

- Aroclor methods
- Congener specific methods
- Screening methods

Table 7. Methods developed to test for PCBs in a wide range of media (NEMI, 2013)

Method Number	Source	Method Descriptive Name	Detection Level	Instrumentation	Relative Cost
530021	Abraxis	PCBs by Immunoassay, Lower Chlorinated, Magnetic Particle	5 ppb	IA	$
530011	Abraxis	Coplanar PCBs by Immunoassay, Microtiter Plate	14 ng/L	IA	$
530001	Abraxis	PCBs by Immunoassay, Higher Chlorinated, Magnetic Particle	0.1 ppb	IA	$
505	EPA-NERL	Pesticides and PCBs in Water GC-ECD	N/A	GC-ECD	$$$
525.3	EPA-NERL	Organics in Water Using GCMS	N/A	GC-MS	$$$
508A	EPA-NERL	PCBs by GC-ECD	N/A	GC-ECD	$$$
508.1	EPA-OGWDW/TSC	Chlorinated Pesticides, Herbicides, and Organohalides in Water by GC-ECD	N/A	GC-ECD	$$$
8082A	EPA-RCRA	Polychlorinated Biphenyls (PCBs) by GC	N/A	GC-ECD/ELCD	$$
508	EPA-TSC/NERL	Chlorinated Pesticides in Water Using GC-ECD	N/A	GC-ECD	$$$
ET013	Envirologix	PCB (polychlorinated biphenyl) immunoassay	.3 µg/g	IA	$
A00134	MWI	PCB (polychlorinated biphenyl) in water by immunoassay	.2 µg/L	IA	$
A00134/A00137	MWI	PCB (polychlorinated biphenyl) in soils/sediment by immunoassay	.5 µg/g	IA	$
130.10	NOAA NST	Organic contaminants in marine sediments by GC-ECD	.05 ng/g	GC-ECD	$$$
130.11	NOAA NST	Organic contaminants in marine animal tissues by GC-ECD	.05 ng/g	GC-ECD	$$$
SPMDs	USGS	Passive sampling of organic compounds in water, air, and soils/sediments by SPMDs	N/A	SPMD	$$$$
O-1104	USGS-NWQL	Organochlorine and organophosphorous compounds, dissolved	.01 µg/L	GC-ECD	$$$
O-3104	USGS-NWQL	Organochlorine and organophosphorous compounds, total recoverable	.01 µg/L	GC-ECD	$$$
O-5129-95	USGS-NWQL	Organochlorine Pesticides and Gross PCBs in Bottom Sediment by GC	50 µg/kg	GC-ECD	$$$$
525.2	EPA-NERL	Organics in Water Using GCMS	.11 µg/L	GC-MS	$$$
1668a (Tissue)	EPA-OGWDW/TSC	Chlorinated Biphenyls in Tissue by HRGC/HRMS	.011 ng/g	GC-MS	$$$$
1668a (Water)	EPA-OGWDW/TSC	Chlorinated Biphenyls in Aqueous Samples by HRGC/HRMS	112 pg/L	GC-MS	$$$$
1668a (Soil/Sediment)	EPA-OGWDW/TSC	Chlorinated Biphenyls in Soil, Sediment, and Mixed Samples by HRGC/HRMS	.011 ng/g	GC-MS	$$$$

ECD = Electron capture detector	MS = Mass Spectroscopy MWI = Modern Water Inc.	OGWDW = Office of Ground Water and Drinking Water
ELCD = Electrolytic conductivity detector	NERL = New England Regional Laboratory	RCRA = Resource Conservation and Recovery Act
GC = Gas Chromatography	NOAA = National Oceanographic and Atmospheric Association	SPMD = Semi-permeable membrane device
HRGC = High Resolution Gas Chromatography	NST = National Standards and Trends	TSC = Technical Support Center
HRMC = High Resolution Mass Spectroscopy	NWQL = National Water Quality Laboratory	USGS = United States Geological Survey

Aroclor detection

EPA developed specific methods to comply with TSCA and other applicable legislation. In order to meet the requirements of the Resource Conservation and Recovery Act or RCRA, EPA developed Test Methods for Evaluating Solid Waste, Physical/Chemical Methods, more commonly known as SW-846 (EPA 2012a). Included in SW-846 are two specific methods for analyzing PCBs in a wide range of media:

- Method 8082A: Polychlorinated Biphenyls (PCBs) by Gas Chromatography
- Method 8275A: Semi-volatile Organic Compounds (PAHs and PCBs) in Soils/Sludges and Solid Wastes Using Thermal Extraction/Gas Chromatography/Mass Spectrometry (TE/GC/MS)

Method 8082A is the more traditionally used as one of the earliest methods developed to meet regulatory requirements and is responsible for much of the legacy data reported as Aroclor mixtures or specific PCB congeners identified in the method. Method 8082A is '… *used to determine the concentrations of polychlorinated biphenyls (PCBs) as Aroclors or as individual PCB congeners in extracts from solid, tissue, and aqueous matrices, using open-tubular, capillary columns with electron capture detectors (ECD) or electrolytic conductivity detectors (ELCD).'* (EPA 2012a) The specific chemicals reported by this method (Table 8) are detected in the parts per billion (ppb) to parts per million (ppm) levels depending upon complexity of sample and matrix involved.

Congener detection

As technology improved and the need for congener specific analysis was identified, Method 1668C (USGS 2010) was developed. Method 1668 was created to analyze PCBs in water, soil, sediment, biosolids and tissue. It provides analytical results for '… *the 12 polychlorinated biphenyls (PCBs) designated as toxic by the World Health Organization (WHO): congeners 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189* [and] *the remaining 197 CBs* [chlorinated biphenyls]*, approximately 125 of which are resolved adequately on an SPB-octyl gas chromatographic column to be determined as individual congeners. The remaining approximately 70 congeners are determined as mixtures of isomers (co-elutions).'* (EPA 2010) Method 1668 requires the use of a high-resolution mass spectrometer for detection and, therefore, is considerably more expensive than Method 8082. Method 1668, however, is becoming more common as concerns have been raised about PCBs from non-legacy sources and potential degradation products from legacy Aroclor mixtures. Detection limits for Method 1668 can be in the low part per quadrillion (PPQ) levels in clean water to ppb levels or higher depending upon complexity of sample and matrix involved.

It is important to note that PCB detection methods have improved over time. Current methods provide detailed data on specific congeners while earlier methods provided data on specific PCB mixtures and homologs. This improvement of analytical methods can prove challenging as it is often difficult to compare data over time as the methods do not provide comparable information at similar detection levels.

Table 8. PCBs reported by Method 8082A (EPA 2012a)

Compound	CAS No.	IUPAC #
Aroclor 1016	12674-11-2	-
Aroclor 1221	11104-28-2	-
Aroclor 1232	11141-16-5	-
Aroclor 1242	53469-21-9	-
Aroclor 1248	12672-29-6	-
Aroclor 1254	11097-69-1	-
Aroclor 1260	11096-82-5	
2-Chlorobiphenyl	2051-60-7	1
2,3-Dichlorobiphenyl	16605-91-7	5
2,2',5-Trichlorobiphenyl	37680-65-2	18
2,4',5-Trichlorobiphenyl	16606-02-3	31
2,2',3,5'-Tetrachlorobiphenyl	41464-39-5	44
2,2',5,5'-Tetrachlorobiphenyl	35693-99-3	52
2,3',4,4'-Tetrachlorobiphenyl	32598-10-0	66
2,2',3,4,5'-Pentachlorobiphenyl	38380-02-8	87
2,2',4,5,5'-Pentachlorobiphenyl	37680-73-2	101
2,3,3',4',6-Pentachlorobiphenyl	38380-03-9	110
2,2',3,4,4',5'-Hexachlorobiphenyl	35065-28-2	138
2,2',3,4,5,5'-Hexachlorobiphenyl	52712-04-6	141
2,2',3,5,5',6-Hexachlorobiphenyl	52663-63-5	151
2,2',4,4',5,5'-Hexachlorobiphenyl	35065-27-1	153
2,2',3,3',4,4',5-Heptachlorobiphenyl	35065-30-6	170
2,2',3,4,4',5,5'-Heptachlorobiphenyl	35065-29-3	180
2,2',3,4,4',5',6-Heptachlorobiphenyl	52663-69-1	183
2,2',3,4',5,5',6-Heptachlorobiphenyl	52663-68-0	187
2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	40186-72-9	206

Screening methods

In response to a need by industry to test wastes quickly and cheaply, manufacturers developed screening methods to test specific waste types. These methods were reviewed and adopted by EPA into SW-846. Specifically, PCB applicable screening methods listed in SW-846 include:

- Method 9077: Test Methods for Total Chlorine in New and Used Petroleum Products (Field Test Kit Methods)
- Method 9078: Screening Test Method for Polychlorinated Biphenyls in Soil
- Method 9079: Screening Test Method for Polychlorinated Biphenyls in Transformer Oil
- Method 4020: Screening for Polychlorinated Biphenyls by Immunoassay

Method 9077 is *'… used to determine if a new or used petroleum product meets or exceeds requirements for total halogen measured as chloride. An analysis of the chlorine content of petroleum products is often required prior to their use as a fuel. The method is specifically designed for used oils, permitting onsite testing at remote locations by nontechnical personnel to avoid the delays for laboratory testing'* (EPA 2012a). It provides results ranging from 300 to 4,000 parts per million (ppm).

Method 9077, however, tests for total chlorine and cannot differentiate PCBs from other chlorinated species such as chlorinated solvents commonly used in industry. Methods 9078 and 9079 test specifically for PCBs. Method 9078 is '*…used to determine the amount of PCB (polychlorinated biphenyl) contamination in soils such as sand, gravel, loam, sediment, and clay, assuming that PCBs are the sole source of organic halogens in the sample.*' Detection levels range from 2 to 2,000 ppm PCBs. The method provides inaccurate results if other chlorinated species are present and should be used with caution. However, in those instances where PCB contamination is known, it provides a quick and easy method to determine the extent of contamination and is often used as a screening tool to limit the number of samples sent to a laboratory for more detailed analyses.

Method 9079 is '… used to screen hydrocarbon based electrical insulating fluids for polychlorinated biphenyls (PCBs) at preset levels of 20, 50, 100, or 500 µg/g [ppm].' The method is calibrated using Aroclor 1242 as a standard and results for other Aroclor mixtures may vary slightly. Method 4020 is '… a procedure for screening soils and non-aqueous waste liquids to determine when total polychlorinated biphenyls (PCBs) are present at concentrations above 5, 10 or 50 mg/kg.' Method 4020 only works on soils containing more than 0.625 ppm PCBs.

Used correctly, Methods 9078, 9079 and 4020 are specifically designed to help meet regulatory requirements. Simple PCB kits meeting the requirements of these methods include but are not limited to:

- Dexsil® Clor-N-Oil Field Test Kit[3]
- Dexsil® Clor-N-Soil Field Test Kit[4]
- Dexsil® Clor-d-tect Field Test Kit[5]
- EnSys Field Test[6]
- RaPID Assay Field Test[7]

In addition to these wet chemical methods, several simple instrumentation and detection kits are also available to test for PCBs including, but are not limited to:

- Dexsil® L2000DX PCB/Chloride Analyzer System (LP-2000)[8]
- Hach® PCB in Soil Pocket Colorimeter II Test Kit[9]
- Hach® DR 2700™ Portable Spectrophotometer[10]

These field test kits are useful as they allow detection for PCBs in the field by individuals with limited technical knowledge and expertise.

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[3] More information available at: http://www.dexsil.com/products/detail.php?product_id=2, accessed 7/2013.
[4] More information available at: http://www.dexsil.com/products/detail.php?product_id=4, accessed 7/2013.
[5] More information available at: http://www.dexsil.com/products/detail.php?product_id=29, accessed 7/2013.
[6] More information available at: http://www.tttenviro.com/store/ensys, accessed 7/2013.
[7] More information available at: http://www.tttenviro.com/store/rapid-assay, accessed 7/2013.
[8] More information available at: http://www.dexsil.com/products/detail.php?product_id=13, accessed 7/2013.
[9] More information available at: http://www.hach.com/pcb-in-soil-pocket-colorimeter-ii-test-kit/product-parameter-reagent?id=7640220978, accessed 7/2013.
[10] More information available at: http://www.hach.com/dr-2700-portable-spectrophotometer/product?id=7640439006&callback=bp, accessed 7/2013.

PCB Uses and Sources

Legacy

Historically, PCBs were used in closed systems such as electrical transformers and capacitors, partially closed uses such as heat transfer and hydraulic systems, and open systems such as surface coatings, adhesives, plasticizers, inks, insulating materials, and pesticides (UNEP 1999). PCBs were valued for their stability, inability to conduct electricity and anti-microbial effects. 60% of worldwide and 77% of US production was used in the production of transformers and capacitors and total worldwide production from 1929 to 1989 is estimated at 1.2 million tons (Tanabe 1988).

PCBs were intentionally added to some products in open applications where the PCBs are in direct contact with the environment and may be transferred from the product into the environment. Plasticizers were the most common use of PCBs in products such as PVC (polyvinyl chloride), neoprene and other chlorinated rubbers. PCBs have also been used in paints and surface coatings as flame retardants and adhesives as plasticizers (UNEP 1999). PCBs were often added to caulk and paint in the field at varying amounts.

Current levels in the environment are due to cycling of PCBs from these historical uses with additional releases of PCBs from legacy uses and new inadvertently produced by-products of chemical manufacture. Specific Aroclor mixtures were often used in specific applications (Table 9). Companies have found alternatives for most PCB uses.

Table 9. Historical Aroclor Uses (from ATSDR 2000)

End Use	Aroclor								
	1016	1221	1232	1242	1248	1254	1260	1262	1268
Capacitors	•	•				•			
Transformers				•		•	•		
Heat transfer				•					
Hydraulics/lubricants									
Hydraulic fluids			•	•	•	•	•		
Vacuum pumps					•	•			
Gas-transmission turbines		•		•					
Plasticizers									
Rubbers		•	•	•	•	•			•
Synthetic resins					•	•	•	•	•
Carbonless paper				•					
Miscellaneous									

End Use	Aroclor								
	1016	1221	1232	1242	1248	1254	1260	1262	1268
Adhesives		•	•	•	•	•			
Wax extenders				•		•			•
De-dusting agents						•	•		
Inks						•			
Cutting oils						•			
Pesticide extenders						•			
Sealants/caulks						•			

Inadvertent Generation in New Products

PCBs may be found as contaminants in a wide range of chemical processes involving chlorine and can be found in recycled materials contaminated with PCBs. As part of 1984 TSCA rulemaking on PCBs the EPA identified about 200 chemical processes that may inadvertently create PCBs and narrowed the list to 70 chemical processes that are likely to contain PCBs as contaminants from manufacturing processes (Panero *et al.* 2005, see Appendix D for list). Most of these chemical processes have not been evaluated to determine if PCBs are actually a reaction byproduct and present in the final product. In addition there are other processes that may inadvertently generate PCBs that are not on this list. Nor have these processes been analyzed to determine how inadvertently generated PCBs enters the consumer supply chain.

Recent studies on PCBs from pigment manufacturing have shown that PCBs can still be found in products. Many of these products contain PCBs as an impurity created during the production process. Inadvertent sources include contaminants or byproducts from manufacturing processes using chlorinated compounds either as a reaction component or solvent. As shown in Table 10, PCBs have been found in various pigments at substantial levels (EPA 1982b).

Table 10. PCBs congeners found in specific pigments (EPA 1982)

PCB Congeners	Pigment	Levels found (µg/g or ppm)
PCB-11	Diarylide yellow	70
PCB-209	Phthalocyanine green	40
Mix of penta- and hexa-	Phthalocyane blue	90*

*Total of PCB congeners

According to one of the rules created by EPA to implement TSCA (49 FR 28172) products may contain low levels of PCBs if the certain conditions are met (see section on Regulations).

Over the last few years, researchers have begun to test consumer products for the potential presence of PCBs. Numerous organic pigments and dyes exist that may contain PCBs as an unintentional byproduct including diarylides (yellow and orange), naphtharylamides (oranges and reds), phthalocyanines (blue), and basic dye complex pigments (reds, violets, blues and greens) (Christie 2013). In general pigments are insoluble in their application medium and dyes are soluble, with inks mostly being used for textile coloration and pigments having broader uses (Christie 2013, Guo *et al.* 2014). Many of these pigments fall into the broad category of azo compounds. An azo compound contains one or more double-bonded nitrogen atoms (R-N=N-R′) where R and R′ are organic additions with varying degrees of complexity. Azo compounds are very efficient at absorbing light and emitting the radiation in specific wavelengths, thereby providing specific colors. Chlorinated compounds are often used in azo pigments as they can greatly increase the lifetime of the resultant product.

Hu and Hornbuckle (2010) conducted sampling of consumer paints containing specific azo (yellow) and phthalocyanine (blue and green) organic pigments and found PCB levels ranging from 2 to 200 ppb in 15 of 33 consumer paints tested. PCB-11 is also found in printed materials (Table 11) from various locations around the world (Guo *et al.* 2014).

Table 11. PCB-11 worldwide concentrations from printed materials

Printed Material (Country)	**PCB 11 concentration (ng/g or ppb)**
Black and white printed newspaper (Georgia)	1.6
Black and white printed newspaper (Moldova)	9.7
Black and white printed newspaper (China)	15
Color newspaper (Georgia)	6.5
Color newspaper (Moldova)	16
Food packaging box (Czech Republic)	6.8
Food packaging box (Ukraine)	5.0

As an example of dyes contaminated with PCBs, diarylide yellow comprises approximately 25% of the 250 million tons of organic pigments produced yearly worldwide (Rodenberg 2012) and testing has shown PCBs and especially PCB-11 are produced during pigment manufacture. PCB-11 is part of the structure of diarylide yellow (Figure 8) as indicated in the red box. PCB-11 can be produced either as a byproduct during the manufacturing process or from degradation of the pigment.

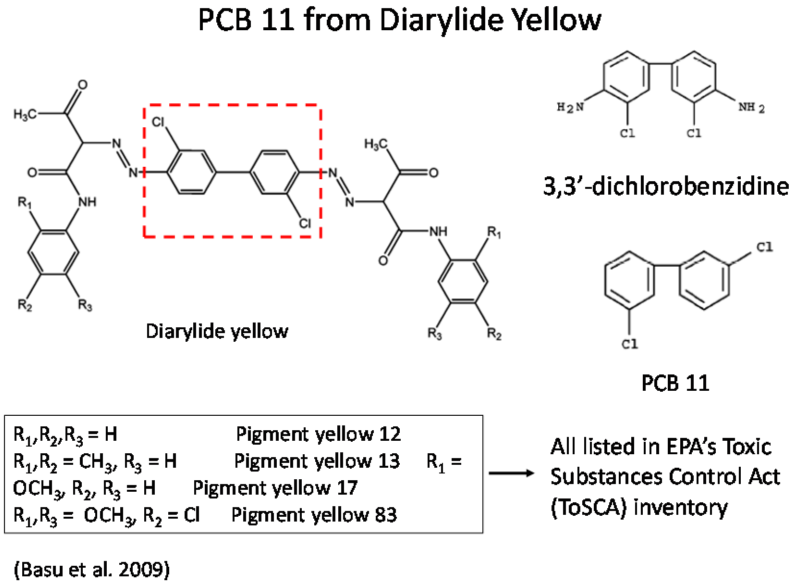


Figure 8. Diarylide yellow and PCB-11 (Rodenburg, 2012)

In addition to PCB-11, purification of the inorganic pigment titanium dioxide (TiO_2) produces larger molecular weight PCBs as a byproduct (Rodenburg 2012). Chlorine is reacted at high temperatures with titanium dioxide (TiO_2) ores containing other metal oxides such as rutile (TiO_2) or ilmenite ($FeTiO_3$) to form titanium tetrachloride ($TiCl_4$) which as a liquid is easily collected. $TiCl_4$ is then reacted with oxygen to make pure TiO_2 (UNEP 2007). During this product process, the larger molecular weight PCBs are created as a reaction byproduct.

Titanium dioxide can also be produced by a sulphate process that does not generate PCB contamination. The sulfate process uses 2.4-3.5 tons of concentrated sulfuric acid (H_2SO_4) per ton of titanium dioxide and the process creates large amounts of acid waste that must be further treated. The chloride process does not generate spent acids and, therefore, creates less waste to be dealt with. (UNEP 2007)

Research is continuing on alternatives to the organic azo pigments. The main challenges faced with replacements (Christie 2013) are:

- Required color performance
- Required degree of transparency or opacity
- Level of fastness or permanence to light, solvents, heat, chemicals, etc. demanded by specific applications
- Cost effectiveness
- Acceptable toxicological and environmental profile.

Research is continuing and alternatives have been identified which appear to address many of these concerns (Christie 2013).

Combustion

Lastly, PCBs are formed through incomplete combustion of products containing carbon and a chlorine source (ATSDR 2000). Given the widespread use of chlorinated compounds such as polyvinylchloride for packaging, tubing, and other applications, incomplete combustion forms a wide range of halogenated compounds such as chlorinated dioxins and furans and PCBs. Most municipal incinerators are not effective in destroying PCBs and it is recommended that PCB-contaminated waste be burned at temperatures above 1,100 degrees Celsius and that care is taken with the temperature, residence time and turbulence of the waste in order to guarantee complete combustion (UNEP 1999). There is only one municipal solid waste incinerator in Washington.

Methods of Manufacturing

PCBs were first mentioned in a publication in Germany in the 1880s. The Swann Chemical Company in Anniston, Alabama was the first US company to manufacture PCBs commercially by bubbling benzene through molten lead to create biphenyl with subsequent chlorination of the biphenyl. Monsanto purchased the Swann Chemical Company in 1935 (Erickson and Kaley 2011).

Much has been written about the methods in which PCBs were manufactured (Panero *et al.* 2005, ATSDR 2000, Pomerantz 1978). Similar methods were used to manufacture PCBs with the main variable being the starting materials of biphenyl and naphthalene. The manufacturing process for Aroclors involved the *'… chlorination of biphenyl with anhydrous chlorine in the presence of a catalyst, such as iron filings or ferric chloride. The degree of chlorination, which determines the nature of the Aroclor, was controlled by the chlorine-contact time (range, 12–36 hours) in the reactor.*' (ATSDR 2000)

Once the manufacturing process was complete, '*The crude product [was] blown with air, and a small amount of lime … added to remove hydrogen chloride and ferric chloride. The resulting chlorinated mixtures [were] batch-distilled to remove color and traces of hydrogen chloride and ferric chloride'* (HSDB 2013).

PCBs were also created using naphthalene which was *'… reacted to varying degrees with chlorine to produce a number of compounds designated by various trade names such as* ***Aroclor****'* (HSDB 2013).

Environmental Transformation and Degradation

Although very stable in the environment, the major pathways for degradation (ATSDR 2000) are:

- Vapor phase degradation with hydroxyl radicals
- Photolysis in water
- Aerobic biodegradation (preferentially less chlorinated congeners)
- Anaerobic microbial degradation (more highly chlorinated congeners favored)

PCBs in the atmosphere undergo complicated reactions (Figure 9) primarily with hydroxyl radicals created when water absorbs sunlight and separates into hydroxyl (•OH) and hydrogen (•H) radicals. Reactions with hydroxyl radicals are most prevalent. A radical is an atom or chemical that has a net charge of zero (neither negative nor positive) but has less than the preferred number of electrons in its outer shell. This instability causes a radical to be very reactive (ATSDR 2000).

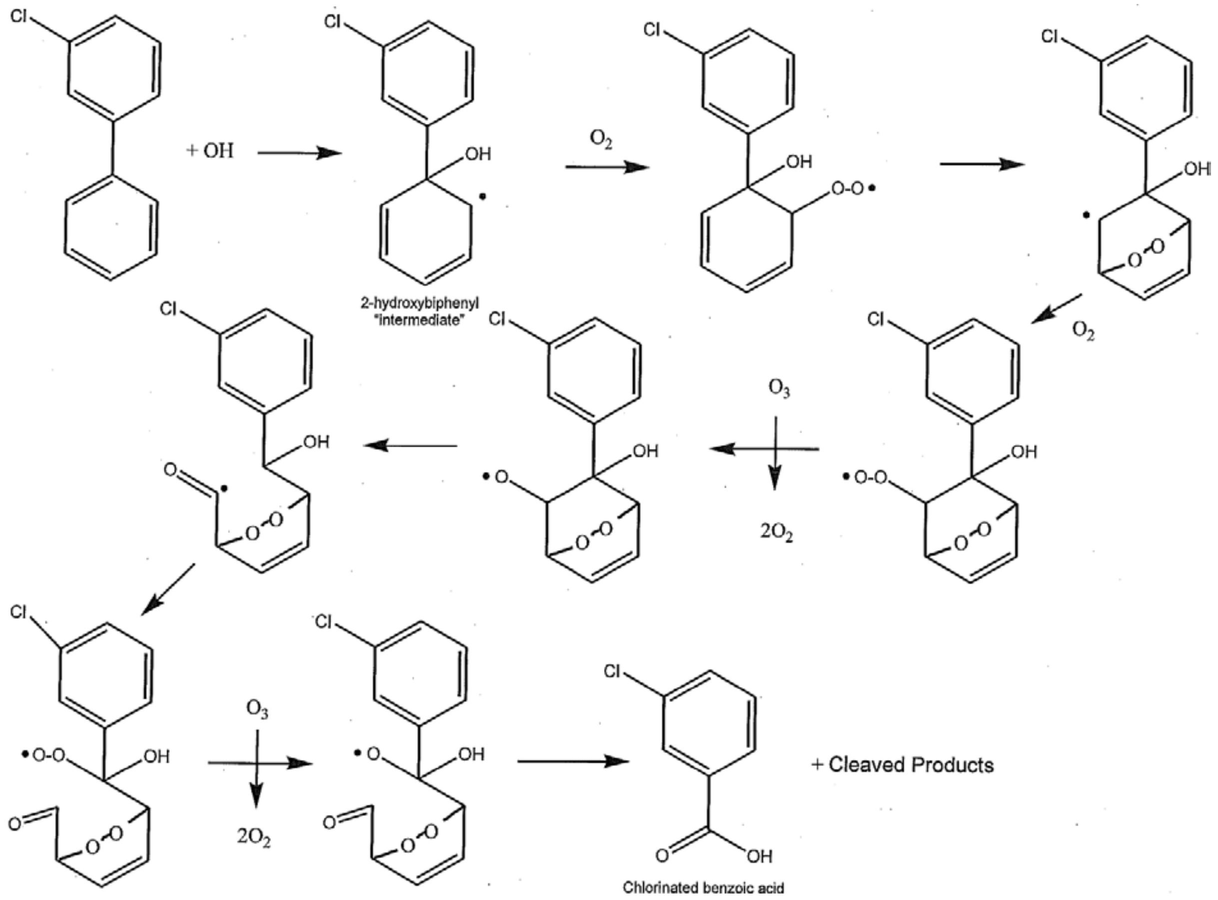
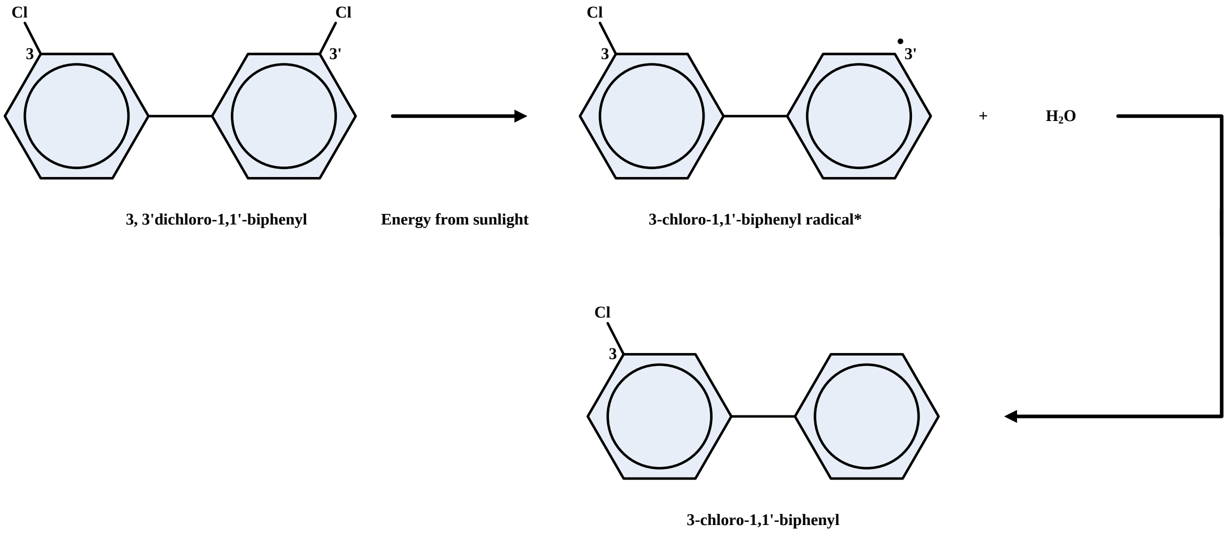


Figure 9. Hydroxyl photo-degradation pathways for PCBs in air (ATSDR 2000)

In water, photolysis is the primary pathway for degradation as other more common reaction mechanisms such as hydrolysis and oxidation do not appear to contribute substantially. In these reactions, a carbon to chlorine bond absorbs energy from sunlight and separates into PCB and chlorine radicals. The PCB radical reacts with water forming a stable PCB compound but with one less chlorine (Figure 10). This reaction is particularly important for the larger PCBs as the more chlorines present, the easier it is to cleave a carbon to chlorine bond. In large PCB molecules, cleavage occurs preferentially on the ring with the most carbons. (ATSDR 2000)



*Note: Although the radical is shown on the 3' position, the electron is actually dispersed throughout the benzene ring.

Figure 10. Photolysis of PCB-11

In sediment and soil, no abiotic process is known that significantly degrades PCBs. PCBs, however, have been found to degrade readily in both aerobic and anaerobic conditions. Both bacterial and fungal species have been shown to biodegrade PCBs using aerobic processes. Because of size restrictions, PCBs with 1 to 4 chlorine atoms are most likely to be degraded under aerobic conditions via a two step process (Figure 11). First, one of the two benzene rings is oxygenated and separated from the other ring. The remaining benzene ring is left as a chlorobenzoic acid. This combined process is called cometabolism.

After cometabolism has occurred, the remaining chlorobenzoic acid is further broken down into water and carbon dioxide (mineralization) in a series of reactions that continually add oxygen to the compound. Aerobic biodegradation of PCBs also occurs primarily in soil and surface sediments. Interestingly, PCBs with fewer chlorine atoms (1-3) degrade faster than those with more chlorine atoms. This causes a fractionating effect where less chlorinated species biodegrade first while those with higher levels of chlorine atoms are left behind for long-term build up in the environment. (ATSDR 2000)

Anaerobic degradation of PCBs is a much slower process compared with aerobic degradation and occurs primarily by reductive dechlorination where chlorine atoms are removed one after the other from a PCB molecule. At least eight distinct and complicated anaerobic pathways have been identified which may occur alone or in combination. Different pathways may favor chlorine in specific positions on the PCB molecule. (ATSDR 2000)

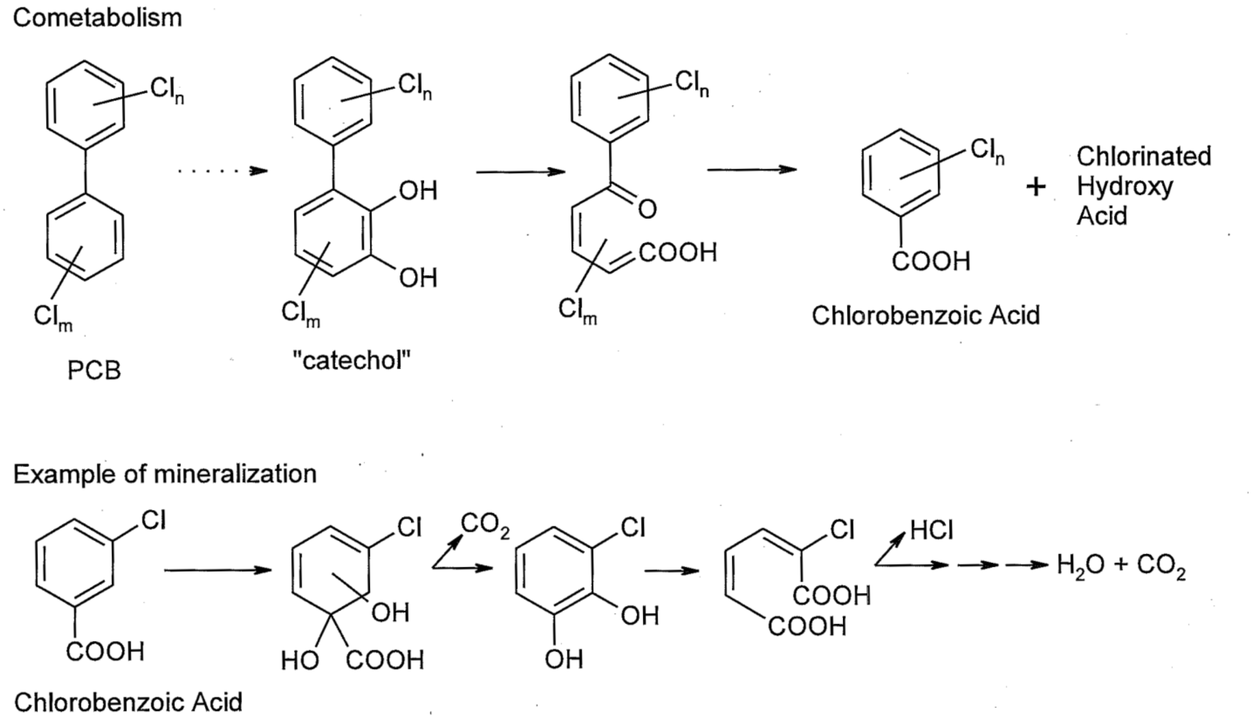


Figure 11. Pathways for Aerobic Degradation of PCBs (ATSDR 2000)

Production, Uses, and Releases

According to the PBT Rule (WAC 173-333) chemical action plans (CAPs) must include a section on “production, uses and releases” that contains information on the production of the chemical and estimates on the amount of the PBT used and released from all sources or activities in Washington.

From 1929 to 1979 the production of PCBs in the US was approximately 1.4 billion lbs (600,000 metric tons), with the largest use for electrical equipment (EPA 1997a). Monsanto, the primary manufacturer of PCBs in North America, voluntarily limited production to certain Aroclors in 1971 (ATSDR 2000) and commercial production was stopped by 1979 under TSCA. Washington’s portion was estimated based on population size to give an estimate of the expected uses in Washington State.

Table 12. Industrial Uses of PCBs (1929-1975) from EPA 1997

PCB Use	Pounds (millions)	Percentage of Total	Washington’s portion (millions of pounds)	Washington’s portion (metric tons)
Capacitors	630	50.3%	13	5,700
Transformers	335	26.7%	7	3,040
Plasticizer uses	115	9.2%	2	1,040
Hydraulics and lubricants	80	6.4%	2	730
Carbonless copy paper	45	3.6%	1	410
Heat transfer fluids	20	1.6%	0.4	180
Petroleum additives	1	0.1%	.02	9
Miscellaneous industrial uses	27	2.2%	.5	250
TOTALS	1,253	100.0%	25	11,400

EPA defines transformers and capacitors as closed uses. There are partially closed uses, such as hydraulic fluids, heat exchange fluids, and gas pipelines. There is a much larger variety of open uses as detailed in Table 13. In closed sources PCBs are contained, barring accidental spill or leakage. In partially contained sources PCBs are partially contained and there is some exposure to the environment. In open sources PCBs are exposed to environment with no containment.

Table 13. Examples of legacy uses of PCBs

Class	US consumption	Examples
Closed	75%	Industrial scale transformers, capacitors, voltage regulators
		Fluorescent light ballasts
		Consumer electrical items (fridges, televisions, washing machines)
		Manufacturing machinery (capacitors, transformers, associated switchgear)
Partially closed	10%	Hydraulic fluids
		Heat exchange fluids
		Gas pipelines
Open	15%	Plasticizer in paints, resins, synthetic rubber, surface coatings, wax
		Sealants, waterproofing compound, glues and adhesives
		Caulking compounds
		Pesticide extenders
		Pigments and dyes
		Carbonless copy paper
		Microscope immersion oil
		Sound proofing materials
		Window glazing

We do not have enough information to estimate the historic use of PCBs in all of these applications or how much is still in use in Washington.

We cannot estimate the amount of PCBs currently in Washington State from partially closed applications, i.e. hydraulic fluids, heat exchange fluids, and gas pipelines. Hydraulic fluids containing PCBs were used in industrial applications that required heat and/or fire resistance. From 1929 to 1975 about 6% of PCBs produced were used for hydraulic fluids and lubricants (EPA 1997a). About 2% of PCBs produced were used heat transfer fluids (EPA 1997a) during the same time period. PCB-based oils were also used in gas transmission compressors. The compressors were used to move natural gas through thousands of miles of pipelines across the U.S. PCBs were also used as a fine mist into underground metal gas pipes to slow corrosion and lubricate the pipelines. PCBs remain in the pipelines until remediated, contributing to environmental releases through leaks and spills.

Closed Legacy Uses

Transformers and Large Capacitors

PCBs have not been manufactured in the US for use in transformers and capacitors since 1979. Although many PCB-containing transformers have been retired, some remain in use. The estimated lifetime of transformers may be as long as 85 years and 20 years for capacitors (Ecology 2011b). While transformers and capacitors are considered "totally enclosed" under TSCA, there are leaks and spills from such equipment until it is retired and replaced.

Federal regulations classify transformers into three groups:

1. *PCB transformers* with > 500 ppm PCBs.
2. *Contaminated transformers* with 50-500 ppm PCBs.
3. *Non-PCB transformers* have less than 50 ppm PCBs.

Transformers are used to transform electricity from one voltage to another through electromagnetic induction. For example, they are used to convert a power generator's low-voltage electricity to higher voltage levels for transmission or to convert high voltages to lower voltages at the end user. Examples include pole-mounted, pad-mounted, and underground distribution transformers and larger transformers at substations. Utilities operated about 80% of mineral oil transformers (Panero *et al.* 2005). Transformers are also found in institutional, commercial, or other private facilities, including schools, mines, and railways.

Capacitors are passive electronic components used to store energy. They have many uses and come in many different sizes. This section includes large capacitors and small capacitors are discussed in the following section.

The size of transformers and capacitors vary and the amount of PCBs per unit also varies (see side bar for regulatory classification and the regulations section for more information). Mixtures of PCBs were marketed under different names in different areas. Askarel was the trade name used in the US for the blend of PCB and trichlorobenzene used for transformers. Askarel transformer fluid was typically 60-70% PCB by weight. Other transformers used mineral oil as the dielectric fluid. Transformers that were manufactured after 1979 are certified to be PCB-free by the manufacturer. Prior to that date some mineral oil transformers were contaminated with PCBs, often during manufacture when the same equipment was used for both Askarel and mineral oil fluids.

Capacitors were typically filled with nearly pure PCB oil and the largest capacitors contained as much as 35 kg PCB (Ecology 2011b). The typical large capacitor contained 31 lbs (14 kg) of PCBs (Federal Register Vol 47, No. 78).

A 1987 EPA report on sources of PCBs includes accidental release estimates from a 1982 study by the Edison Electric Institute and the Utility Solid Wastes Activity Group (USWAG/EEI) in response to an order issued by the US Court of Appeals for the District of Columbia Circuit. The

EEI/USWAG study gathered survey data from 98 of the 100 largest utilities on leaks and spills. It also estimated the number of pieces of equipment and concentrations of PCBs in the equipment based on data from utilities (USWAG/EEI 1982). This is the only study on leaks that was identified.

Estimate in Washington

An earlier report by Ecology overestimated PCBs in transformers and large capacitors in the Puget Sound basin (Ecology 2011b) in several ways. The earlier estimate relied on an EPA database of registered PCB transformers that was not up to date, estimates of unregistered PCB transformers, an assumption that the PCB transformers in Washington are Askarel transformers with 60% PCBs, and the leakage rates did not take into account the lower concentrations of PCBs in transformers currently in use. Conversations with public and private utilities in Washington, non-utility users of PCB transformers, and other experts led to much lower estimates here.

It is not appropriate to use national estimates or estimates from other regions for Washington State. Compared to other regions of the country, public utilities in Washington State have been the most progressive in testing equipment for PCBs and disposing of equipment with PCBs (Mark Pennell, personal communication).

Transformers

There are 252 registered transformers in the EPA PCB Transformer Registration Database with 121,053 kg of PCB oil in Washington (Appendix F). These are PCB transformers (>500 ppm) that were required to be registered with the EPA in 1998. The regulations do not require EPA to update the database when PCB transformers are taken out of use. We were able to contact most of the parties who registered transformers and found that most of the transformers had been disposed of (Table 14).

Table 14. PCB transformers in use in Washington

Still in use	**14**
Disposed of	228
Unknown	10
Total	252

The PCB transformer registry does not reflect the actual number of PCB transformers still in service and no current inventory of PCB equipment exists in the US (EPA/EC 2009). The EPA states that the database is “not particularly useful for determining the amount of PCB equipment that is remaining in service” (EPA/EC 2009). Various attempts have been made to estimate the number of units remaining in use. The Great Lakes Binational Toxics Strategy 2009 Biennial Report (EPA/EC 2009) estimates that 64,312 PCB transformers remained in use throughout the US while there are about 14,150 registered transformers in the EPA database. Scaled down from the national to the state based on 2010 population, an estimated 1,401 transformers remained in use in Washington in 2007. This estimate assumes that about 80% of the PCB transformers still in use were not registered with EPA. Based on conversations with public and private utilities in Washington, there is not that much unknown equipment in use. A more reasonable estimate would be about 20% of transformers are unknown and never registered. Some utilities know the PCB concentration of all their transformers.

Using the 14 known PCB transformers still in use from the EPA database, adding an additional 20% results in an additional three PCB transformers still in use in Washington. If the other 10 registered PCB transformers whose status is unknown are all still in use, the total number of PCB transformers (> 500 ppm PCB) in Washington could be as high as 27, including the additional 20% of unknown and never registered PCB transformers. Based on the sizes and PCB concentrations of some of the known PCB transformers still in use, we have estimated they each contain about 665 gallons of fluid that is 1500 ppm PCB. Askarel transformers were less than 0.2% of utility transformers nationwide (USWAG/EEI 1982) and were used even less frequently in Washington State. Askarel transformers contain 60-70% PCBs and the more common mineral oil transformers have a much lower concentration of PCBs. When national data was compiled for the USWAG/EEI study (1982) 90% of the mineral oil transformers had <50 ppm PCBs. As utilities have identified and removed PCB contaminated transformers, the average concentration of PCBs has become even lower.

There do not seem to be known Askarel transformers in Washington, but each leak/spill from an Askarel transformer has the potential to release a significant amount of PCBs. The USWAG/EEI study estimated that each leak/spill would release 0.56 - 64.5 lbs (0.25 – 29 kg) of PCBs.

There are still some transformers with lower levels of PCBs, mostly < 50 ppm, that are considered PCB contaminated (50-500 ppm) or non-PCB (1-50 ppm) in the regulations. These have never been required to be tracked or reported. Utilities in Washington have been testing transformers and many have been disposing of transformers with any detectable level of PCBs (>1-2 ppm). Based on the number of these transformers that are known to be still in use by utilities, the amount of older equipment that is untested and has unknown levels of PCBs, there are about 40,000 PCB contaminated (50-500 ppm) and non-PCB (1-50 ppm) transformers in the state that have a concentration of about 25 ppm PCBs. The transformers used in distribution lines on top of poles have about 20 gallons of fluid.

Large Capacitors

Less is known about the number of large PCB capacitors in Washington compared to the number of PCB transformers. Capacitors were never required to be tested or registered. The Puget Sound study (Ecology 2011b) used the national estimate of 1,293,000 large capacitors still in use from The Great Lakes Binational Toxics Strategy 2009 Biennial Report (EPA/EC 2009). Scaled down from the national to the state level based on 2010 population size, an estimated 28,162 large PCB capacitors could remain in use in Washington. However, based on conversations with some of the 61 public and private utilities, there are very few large PCB capacitors remaining in Washington State. PCB capacitors were never required to be inventoried and not all equipment is known, but utilities we asked were confident that all the large PCB capacitors had been removed from 10 to more than 20 years ago. About 15% of large PCB capacitors were owned by non-utilities (EPA 1987) and we have not tried to identify where those were used or if they are still in use. If we estimate 95% of the PCB capacitors have been disposed of, then there would be about 1,400 remaining still in use. We do not have any evidence for which non-utilities in Washington had PCB capacitors or how many of those capacitors are still in use. The estimate that 5% remain in use is based on the hypothesis that non-utilities have also removed capacitors over time, but have not removed all of them.

Other equipment not estimated

Utilities have other equipment that historically contained PCBs, such as reclosers, switches, circuit breakers, bushings, etc. This equipment contained much less PCBs compared with the transformers and capacitors (USWAG/EEI 1982). We have not attempted to estimate how much of the old PCB-containing equipment still remains in use. Some utilities have been testing and removing this equipment.

Leakage and Spillage Rates

In the Puget Sound study (Ecology 2011b) the leakage rates were based on PCBs/unit for Askarel transformers, which does not take into account the lower concentrations of PCBs in transformers currently in Washington.

The USWAG/EEI study (1982) found about 2% of all transformers and 0.77% large capacitors had moderate leaks or spills each year. The study includes lower rates for equipment that had > 50 ppm PCBs. They did not include equipment with < 50 ppm PCBs that is considered non-PCB in federal regulations, but we are including that equipment here. The average amount of PCBs leaked/spilled per event was 0.00004 – 0.005 lbs (18 mg – 2.27 g) for mineral oil transformers and 2 – 17.1 lbs (0.9 – 7.76 kg) for PCB capacitors. As mentioned earlier, the average amount of PCBs per leak/spill for Askarel transformers was 0.56 - 64.5 lbs (0.25 – 29 kg) of PCBs.

Leaks from transformers and large capacitors were estimated in San Francisco Bay (McKee *et al.* 2006) and NY Harbor (Panero *et al.* 2005). Both of these studies relied on the EPA transformer database that is not accurate for Washington.

This results in a total annual release estimate of < 2 kg for transformers and 10-80 kg for capacitors in Washington State (Table 15).

Table 15. Release estimate for transformers and capacitors

Equipment Type	PCB concentration	Number of units	Reservoir	PCB spill/leak rate (annual)	PCBs released per spill/leak (kg)	Annual PCB release
Transformers	>500 ppm	14 - 27	100-200 kg	2%	0.000018-0.00227	5 mg – 1.2 g
	1-500 ppm	40,000		2%		14 g- 2 kg
Large Capacitors	100 %	1,400	20 metric tons	0.77%	0.9- 7.76	10-80 kg

There is uncertainty in the number of electrical units still in use and the older data on leakage rates may not reflect current operating conditions. The estimates do not account for spill response, thus actual amounts of PCBs released to the environment may vary. Indoor spills in particular are likely to be contained and cleaned up. Additional emissions from direct volatilization from equipment are likely, but not estimated due to lack of information.

Opportunities for Reduction

- Status quo
    - 1998 EPA registry for known transformers > 500 ppm
    - Voluntary removal
- Develop a state inventory of equipment that includes the number of units and the amount of PCBs.
- Require utilities and other owners of transformers, capacitors, and other electrical equipment to develop and implement a plan for removal.
- Require complete change out to remove old units (with proper disposal) by a certain date.

Based on conversations with public and private utilities in Washington, they have been actively testing and removing equipment with PCBs beyond what is required by federal law. Several utilities in Washington (personal communication) and other states (Panero *et al.* 2005) use serial numbers of transformers to identify additional transformers that are likely to have PCBs, once one transformer in a batch has been identified as having PCBs. Many utilities are disposing of equipment with any detectable level of PCBs (> 1 ppm).

Small Capacitors

Capacitors containing less than three pounds of PCB oil are considered small capacitors. Small capacitors containing PCBs have been used in a number of items including motors, appliances, and light ballasts. Small capacitors generally contain 45-270g of PCBs in oil and lamp ballast capacitors contain 45-70 g PCBs (EPA 1982 proposed rule in the Federal Register).

Wisconsin found submersible well pumps that contain PCB filled capacitors and in 1992 estimated that 10,000 -15,000 of their 800,000 wells contained capacitors with PCBs (Wisconsin DNR 2001). This only applies to equipment that was manufactured before 1979. Wisconsin has recommendations for owners to identify such pumps and prevent exposure. It is unknown how common this is in Washington. There is a report of one contaminated well from a pump in Whatcom County in 1990 (Seattle Times January 8, 1990).

Estimate in Washington

Several studies estimate the number of small PCB containing capacitors remaining in use that are not light ballasts. In 1992 the University of Illinois estimated that 10-25% of US household appliances contained capacitors with PCBs (Panero *et al.* 2005). EPA (1982b) estimated that historically there were 870 million small capacitors in use throughout the US in 1977 in industrial machines and small appliances. EPA (1987) also estimated a 10% annual disposal rate in 1982. Scaling the national estimate to Washington based on population and applying annual disposal rates of 20% and 10% yields an estimate of 12,000 to 586,000 small capacitors still in use in 2010.

Globally, one third of all PCB production may have gone into lighting ballasts (Ecology 2011b). National estimates of lamp ballasts currently in use include 300 million (US Army 2001) and 500 million (Missoula County 2010). In 1998 EPA, citing an unnamed industry source, estimated that 1 billion small lamp PCB ballasts remained in use in the US (EPA 1998). Scaling this estimate to Washington based on population and applying annual disposal rates of 20% and 10% yields an estimate of 1.7 million to 6.2 million such ballasts still in use in 2010 in Washington.

While we have some information on the number of PCB containing capacitors collected in Washington as hazardous waste or moderate risk waste, the information is not complete enough to use for estimating the number of units still in use.

A range of 12,000 to 6.2 million non-lamp ballast small capacitors and lamp ballasts remains in use in Washington State. While small capacitors may contain 45-270 g PCB per unit, most of the remaining units are likely to be lamp ballasts, which typically contain 45-70 g PCB per unit. For the estimate we used 57.5g PCB/unit as an average. The assumed leakage rate is 4.2 kg/metric tons of PCBs, from the 1982 study on large capacitors (EPA 1982b). This results in an estimate of 400- 1,500 kg for lamp ballasts and 3-150 kg annually for other small capacitors (Table 16).

Table 16. Lamp ballasts and other small capacitors

Equipment Type	Basis for estimate	Number of units	PCBs (kg)	PCB spill/leak rate (annual)	Annual PCB release kg/yr)
Other small capacitors	Scaled from national estimate	12,000-586,000	690-33,695	4.2%	3-150
Lamp ballasts	Scaled from national estimate	1.7-6.2 million	97,750-356,500		400-1,500

There is uncertainty around both the estimate of how many small PCB capacitors remain in use and how much leaks each year. Additional emissions from direct volatilization from equipment are likely, but not estimated.

Opportunities for Reduction

- Status quo
  - Continued use of old capacitors, with expected 10-20% annual disposal/replacement rates
- Educate consumers on which lamps and small appliances are likely to contain PCBs, including appropriate disposal options
- Statewide program to remove small capacitors.
- Remove PCB-containing lamp ballasts in schools.
- Investigate the status of PCB containing materials in schools.

Removal of old light ballasts could be linked to changes for energy efficiency. EPA has national guidance for schools to replace PCB ballasts. NY settled a lawsuit in 2013 after widespread PCB contamination was found in schools and some fires (http://www.epa.gov/region2/pcbs/).

Open Uses

Caulk

PCBs were used in caulk and joint sealants to improve the flexibility, increase the resistance to erosion, and improve adherence to other building materials from the 1950s to the 1970s (Robson *et al.* 2010). Monsanto voluntarily stopped producing PCBs for open uses, such as caulk, in 1971 (ATSDR 2000). While the use of PCBs in open products above 50 ppm was banned in the US effective in 1979 under TSCA, materials that contain PCBs were not required to be removed. The use of PCB-containing caulk was a common practice in the 1970s and caulk formulations changed during the late 1970s (Herrick *et al.* 2004). The studies on PCBs in caulk have focused on buildings built from about 1950 to 1980 to include using up the existing stocks of PCB-containing caulk.

Sealants with high levels of PCBs have been found at varying levels in buildings in several studies. All of the studies found congener profiles consistent with Aroclors 1248, 1254, and 1260. In general, PCBs were used at 5-30% in caulk (Priha *et al*. 2005). PCBs can be lost from caulk through volatilization, as well as wash-off and erosion. PCBs in caulk are associated with higher levels of PCBs in indoor air and dust, and the external soil (Priha *et al*. 2005, Herrick *et al.* 2007, SAIC 2011). Larger amounts of PCBs may be lost during renovations or destruction. Certain removal practices can reduce the amount of PCBs released both to workers and the environment (Sundahl *et al*. 1999).

Herrick *et al*. (2004) found PCBs in schools and other buildings in the Boston area. In 13 of the 24 buildings sampled, PCBs were found at concentrations of 2 to 36,000 ppm. PCB levels in the air ranged from 111 to 393 ng/m^3 and in dust samples up to 81 ppm.

There was a nationwide comprehensive survey in Switzerland (Kohler *et al*. 2005). In this study, 1348 caulk samples from concrete buildings built between 1950 and 1980 were analyzed for

PCBs. Forty-eight percent of the caulk samples contained PCBs, from < 50 ppm up to 550,000 ppm (55%).

The amount of PCBs in caulk was estimated in Toronto, Canada (Robson *et al*. 2010, Diamond *et al*. 2010). This study was based on a smaller sample size and found PCB-containing caulk in 14% of 95 buildings at concentrations of 0.57 ppm to 82 ppm. In Toronto, institutional and commercial buildings and infrastructure (e.g., bridges and parking lots) made of concrete were most likely to have PCB-containing caulk. They detected PCB in caulk in one single family detached home. As expected, they did not detect PCBs in caulk in buildings built before 1945 or after 1980. Based on the number of concrete institutional and commercial buildings built between 1945 and 1980, the size of the buildings, the amount of caulk in a typical building, the percentage estimated to have caulk, and the average concentration of PCBs in caulk, the authors estimated 13 metric tons of PCBs are in caulk in Toronto. The authors further estimated that up to 9% of the PCBs in caulk had been lost via volatilization. The observed congener pattern is consistent with volatilization of lower chlorinated congeners and comparative enrichment of higher chlorinated congeners (Robson *et al*. 2010).

There was also a study of PCB in caulk in the San Francisco Bay area as part of implementing the TMDL (Klosterhaus *et al*. 2011, 2014). This report estimates PCBs in buildings and how much is released to runoff during renovation and demolition. PCBs were detected in 88% of the 25 samples from 10 buildings. The concentrations ranged up to 220,000 ppm (22%) with 40% of the samples exceeding 50 ppm. The median and range were similar to the studies in Boston and Switzerland. The mid-range estimate was 10,500 kg of PCBs in caulk in existing buildings, using a similar method as was used in the Puget Sound Study (Ecology 2011b, Klosterhaus *et al*. 2011, 2014). Information on the number of renovations and demolitions in the San Francisco Bay area each year was used to estimate that 0.04 kg PCB is released each year to stormwater from renovation and demolition. Washington does not have information on the number of commercial buildings of that age and construction type that are renovated or demolished each year.

As part of the Lower Duwamish Waterway (LDW) cleanup in Seattle, Science Applications International Corporation (SAIC) investigated PCBs in old caulk and paint in the LDW (SAIC 2011). This was part of an effort to find additional sources of PCBs in the cleanup area, especially since high levels of PCBs in paint, caulk, and other building materials had been found at the former Rainier Brewery and North Boeing Field. They detected Aroclors in 8 of 17 composite caulk samples from representative buildings with detected concentrations from 3 to 920 mg/kg. The focus was on industrial buildings from 1950-1977. As expected, they did not find PCBs in a sample from buildings built in the 1940s. Surprisingly, they reported another building in the Seattle area that was built in 1989 and contained PCBs in caulk up to 1000 mg/kg. The use of PCBs in caulk in North America has not been reported this late. The number of samples with detectable PCBs (47%) is in agreement with the larger comprehensive study in Switzerland (Kohler *et al*. 2005).

Estimate in Washington

The report on sources of toxic chemicals released in the Puget Sound Basin (Ecology 2011b) estimated 59 metric tons of PCBs are in building sealants in that area with about 110 kg released annually. This estimate was based on the number of existing masonry commercial buildings that were built between 1945 and 1980, the average size of those buildings and the distribution of PCB concentrations in caulk found in the more comprehensive survey by Kohler *et al*. (2005). This is likely to underestimate the amount of PCBs in sealants because it does not consider all uses in buildings, such as around windows, uses in residential buildings, or in other structures, such as bridges and sidewalks. The annual release estimate was based on a release rate coefficient of 0.0018/yr from long term loss rates in Robson *et al*. 2010.

The estimate for the Puget Sound Basin was based on detailed information about buildings in Pierce and Snohomish Counties and then scaled up to the rest of the study area by population. The estimated volume of masonry buildings built from 1945 to 1980 in Pierce and Snohomish Counties was 21,941,562 m^3. To estimate PCBs in caulk for the state we scaled up the volume of masonry commercial buildings that were built between 1945 and 1980 by population, leading to an estimate of 97,702,645 m^3 with 5,373,645 kg of caulk for the state.

The large study in Switzerland (Kohler *et al*. 2005) found 48% of the targeted buildings had PCB- containing sealants. Applying this to the state estimate on sealants leads to 2,573,976 kg of PCB-containing sealants. The PCB concentration ranges from Kohler *et al*. (2005) were applied to the estimated mass of PCB-containing sealants in Washington, yielding an estimate of 87 metric tons of PCBs in sealants in Washington with 157 kg released annually (Table 17).

Table 17. Estimates from caulk

sealant quantity (kg)	sealants with PCBs (kg)	PCB conc bin (mg/kg)	bin mid point	% for each bin	PCB quantity (kg)	Annual releases (kg)
5,373,645	2,573,976	20-50	35	0.121	11	
		50-100	75	0.0772	15	
		100-1,000	550	0.1899	269	
		1,000-10,000	5,500	0.1815	2,569	
		10,000-100,000	55,000	0.2316	32,787	
		>100,000	100,000	0.2003	51,557	
Total				1.0	87,208	157

In addition, PCBs are released into the environment during renovation and demolition of buildings that contain PCBs in caulk and other building materials. In order to estimate this we need to know how many buildings of that age and construction type are demolished or renovated in the state, which we do not know.

Opportunities for Reduction

- Status quo
    - PCBs may be found and remediated during source identification efforts.
- Require removal or remediation of all PCB containing caulks, statewide.
- Develop best practices for demolition and renovation.
- Investigate the status of PCB containing materials in schools.
- Remove or remediate PCB-containing caulk in schools.
- Partner with EPA and federal facilities to identify and remove caulk and other PCB-containing materials.

Many schools in Washington were built when PCBs were used in caulk, lighting ballasts, paint, and other building materials. EPA has information on PCBs in caulk and other building materials that includes how to test for PCBs and how to safely remove PCB-containing materials (http://epa.gov/pcbsincaulk/index.htm). Their outreach has been focused on schools, due to the sensitivity of developing children. There are reports of methods for removing PCBs where more than 99% of the PCBs in caulk were captured following the use of BMPs specifically aimed at preventing PCB releases (e.g., Sundahl *et al.* 1999).

Caulk, other building materials, and other historic uses of PCBs are found on Navy vessels and other military equipment (EPA 2006). This is an opportunity for Ecology to partner with EPA and federal facilities to expand PCB source identification and removal activities.

Inadvertent generation in new products

PCBs are no longer intentionally manufactured in the U.S. and the manufacture, processing, and distribution in commerce of PCBs at concentrations of 50 ppm or greater is not allowed. EPA promulgated a rule under TSCA in 1984 for inadvertent generation of PCBs that are not in closed or controlled manufacturing processes (49 FR 28172). The concentration of inadvertently generated PCBs in products must have an annual average of < 25 ppm, with a maximum of 50 ppm. In addition, EPA required manufacturers with processes inadvertently generating PCBs and importers of products containing inadvertently generated PCBs to report to EPA any process or import for which the PCB concentration is greater than 2 mg/kg for any resolvable PCB gas chromatographic peak. More details on TSCA are in the section on Regulations.

As part of this rulemaking on inadvertently generated PCBs, EPA generated a list of 200 chemical processes with a potential for generating PCBs (Appendix D) and narrowed it to 70 with a high potential to inadvertently generate PCBs. The list does not include every process that inadvertently generates PCBs and not everything on the list inadvertently generates PCBs. In general, PCBs can be produced when chlorine and carbon are present with elevated temperatures or catalysts.

The final rule also includes an estimated annual production of inadvertently generated PCBs of 100,000 lbs (45,400 kg). Scaled to population, Washington's share of that would be about 900 kg a year. Only 11% of the PCBs were estimated to enter products, or 100 kg annually in Washington. As the economy has grown over the last 30 years, the amount of inadvertently generated PCBs may also have grown. The 100,000 lbs was an estimate from a consensus proposal from the Environmental Defense Fund, Natural Resources Defense Council, and Chemical Manufacturers Association (now known as the American Chemistry Council) that included all inadvertent generation of PCBs, without being broken down into how much came from each process. Products that are mentioned include paints, printing inks, agricultural chemicals, plastic materials, and detergent bars. The 1982 economic analysis for this rule mentions 135 manufacturing processes that generate PCBs at less than 50 ppm from a Chemical Manufacturers Association survey. The economic analysis also includes a list from EPA of about 20 "end-products of manufacturing processes in which PCBs are incidentally generated."

In their rule on inadvertent production, EPA specifically mentions surfactants as the component of detergent bars that is likely to contain PCBs. EPA also mentioned PCBs are likely to be in surfactants in skin lotions and creams that are regulated by the FDA. We have no estimate for how many PCBs are inadvertently produced in surfactant.

Reports to EPA on inadvertent generation

As mentioned above, the 1984 rule under TSCA (49 FR 28172) requires manufacturers to report inadvertent generation of PCBs. There are 77 reports for inadvertently generated PCBs from 1994 to present (Table 18). Some information on each report is in Appendix E. There are additional reports included in the docket for related topics, such as requests to produce small amounts of PCBs for research purposes. A lot of the information in the reports has been redacted to remove confidential business information (CBI). In general the reports repeated the federal requirements while stating the company is in compliance and without giving specific information about the concentration of PCBs in the products or the total amount of the products. None of the reports were for facilities in Washington State.

Many reports include a statement that the materials may contain PCBs > 2 ppm, but likely do not and the reporter was being very conservative in reporting anything that might contain PCBs > 2 ppm. The reports assert that no products contained more than 50 ppm or more than 25 ppm for an annual average (which are the limits in rule).

Table 18. Reports to EPA on inadvertent generation 1994-present

Chemical or process	Number of reports
Pigments and dyes	53
GE silicones	8
Vinyl chloride production	3
Unique	6
Unknown	7
Total	77

Some of the reports in Table 18 in the category of pigments and dyes list individual pigments (yellow, red, green, blue violet and orange with Color Index (CI) numbers), some include a general description such as “imported dyes,” while others do not include any specific information, but come from a division of the company such as the “Pigments Division.”

Eight reports are from GE Silicones. There is no additional information on the products.

There are three reports from three different companies regarding vinyl chloride production, one of which was a unique incident involving diesel contamination. Geon stated they are reporting on 740 lbs of PCBs in 62,676,000 lbs of chemical feedstocks used in a vinyl chloride monomer manufacturing facility in Texas. The third report is for incidental PCBs generated in the chlorination step of a process stream to remove an impurity.

There are six reports from six different companies on unique compounds or processes. These reports are for trichlorobenzene manufacture, 2,4,6-Trichlorophenylhydrazine (2,4,6-TCPH), 2,6-Dichloro-4-Nitro Aniline, a pesticide intermediate, chlorothalonil production process, and one report for electrical capacitors. The one report for electrical capacitors also included the information that PCBs were at 3.9 ppm in 134 liters. The report on 2,4,6-Trichlorophenylhydrazine stated the concentration was usually 9-12 ppm.
Seven reports were for unknown compounds or processes. Four of these were completely redacted with a place holder stating there was a report. Two reports redacted the name of the company in addition to the compounds. One report was for two containers of a non-PCB product with 4 and 5 ppm PCBs.

Pigments and dyes

More details on generation of PCBs during manufacturing are provided in the earlier section on Chemistry. PCBs are known to be inadvertently generated in certain pigments and dyes, including diarylides (yellow and orange), naphtharylamides (oranges and reds), phthalocyanines (blue), and basic dye complex pigments (reds, violets, blues and greens) (Christie 2013). PCB-11 is thought to be primarily from pigment production and not from legacy uses of Aroclors (Hu and Hornbuckle 2010, Guo *et al.* 2014), and so is useful as an indication of inadvertent generation of PCBs, especially from pigments. Specifically, the ratio of PCB 11 to a characteristic dechlorination end product, PCB-4, indicates that dechlorination is not a significant source of PCB 11 where it has been examined (Rodenburg *et al.* 2010). PCB-11 has been seen to be strongly correlated with PCBs 35 and 77 (Rodenburg 2014). PCB-77 is one of the dioxin-like congeners.

Hu and Hornbuckle (2010) found PCBs in azo and phthalocyanine pigments, including PCB-11 and higher chlorinated PCBs 206-209. Previously PCB-209 was only thought to be found in ferric oxide as a by-product of titanium dioxide production (Panero *et al.* 2005). PCB-11 and PCB-209 have been found in Washington's environment and animals (Ecology EIM database).

Higher chlorinated PCBs are inadvertently generated during the production of the inorganic pigment titanium dioxide using the chlorine process (UNEP 2007). We were unable to locate estimates on the amount of PCB inadvertently generated in this process.

PCBs have been detected in general consumer products purchased in Washington. Individual congeners were detected between 1 and 45 ppb. Four congeners known to be associated with pigments (PCB 11,206,208, and 209) were selected for the initial study and were tested for in 74 samples from 68 products (Ecology 2014). The products included packaging, paper products, paint and colorants, and caulk. Ecology is in the process of reporting on the results of all the congeners for the same samples. As in previously published work (Hu and Hornbuckle 2010), we also see a wide selection of congeners in the consumer products.

Estimate in Washington

While different researchers have detected PCBs in pigments and consumer products, we don't have a good estimate for how much is released in Washington each year. Panero *et al*. (2005) estimated PCB-11 represents 5-20% of the PCBs entering NY harbor. Guo *et al.* (2014) estimated that between 5 and 7800 kg[11] of PCB-11 are produced worldwide each year from diarylide yellow in 2006. The US market consumes approximately 20% of global organic pigments (Guo *et al.* 2014). Washington is approximately 2% of the US population, which leads to an estimate for Washington's share of PCB-11 from yellow pigment of 0.02 and 31 kg per year. This is the amount of PCB-11 in products, with an unknown amount entering the environment.

The Color Pigments Manufacturers Association (CPMA) estimated that the total annual amount of these pigments (phthalocyanine and diarylide) imported or manufactured in the US is about 90 million lbs (41,000 metric tons). They further estimated inadvertently generated PCBs in these pigments with an upper bound of 1.1 tons per year and a more reasonable estimate of 1000 lbs

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[11] Jia Guo is an author on an earlier paper (Rodenburg et al. 2010) that estimated worldwide production of PCB 11 from diarylide yellow pigment production at 1.5 t in 2006. This estimate was revised in Guo 2013 and Guo et al. 2014.

per year (CPMA 2010). Using the lower annual estimate of 1000 lbs (450 kb), leads to an estimate of 9 kg per year in Washington, that is within the range of the estimate above.

There are permitted releases in Washington State that are pathways for PCBs to get from sources such as pigments in paper and other consumer goods into waterways. Looking at the PCBs in these permitted pathways illustrates that some PCBs from pigments are released in Washington, even thought the permittees are not the source of the PCBs. Paper mills that recycle paper and municipal wastewater treatment plants discharge small amounts of PCBs into Washington waterways. In 2012 the average PCB concentrations in the discharge of two paper recycling mills were 2,520 and 1079 pg/L, respectively. Based on flow rate, the estimated PCB loading for these two facilities is 28 g per year, with 3.8 g being PCB-11. It is suspected that dyes from clothing and other consumer products such as soaps, lotions, and creams are also contributing PCBs to municipal wastewater treatment plants. For example, the Spokane River Source Assessment (Ecology 2011c) estimated that the Spokane waste water treatment plant was discharging 194 mg of PCBs/day.

Opportunities for Reduction

<u>Inadvertent generation</u>

- Status quo
    - Continue to permit products containing less than 50 ppm (with discount factor for mono- and bi-chlorinated biphenyls- see Regulations section) to be sold in Washington.
    - Implement RCW 39.26.280- Preference for PCB free products by state agencies.
- Assess alternatives for pigments and dyes to identify the availability of safer materials.
- Encourage businesses to use alternate processes/materials that do not generate PCBs.
- Conduct research using green chemistry to develop new processes that do not inadvertently generate PCBs.
- Test products to determine the extent of PCBs in products likely to contain PCBs.
- Require labeling to educate consumers on what products contain PCBs and which ones do not.
- Petition EPA to revise the federal regulatory limit on PCBs in products to align with the federal Clean Water Act.

There are many processes that might inadvertently generate PCBs, but not much is known about most of them. If the state doesn't make progress in reducing inadvertent generation of PCBs, permitted facilities might be forced to close or eliminate environmentally beneficial processes (such as recycling), which are undesired outcomes.

Commercial and industrial releases

Toxics Release Inventory (TRI)

The federal Toxics Release Inventory (TRI) includes permitted estimated releases from facilities that discharge from certain industries. The TRI database is authorized under the federal Emergency Planning and Community Right to Know Act to aid in community planning in case of an emergency and to generally inform the public about releases of toxic chemicals.

PCBs are subject to reporting and listed with the general CAS number 1336-36-3 for all PCBs. Because PCBs are PBTs, there is a lower level for reporting and the reporting threshold is 10 lbs.

There are only two reporters of PCBs in Washington on the TRI. Burlington Environmental in King County and Perma-Fix Northwest in Benton County are both hazardous waste treatment and disposal companies. From 2007-2011 they reported an average off-site waste disposal of 25,000 pounds (Table 19).

Table 19. Table of TRI reports from 2007-2011

Year	Reporter	Offsite waste reported (pounds)
2007	BURLINGTON ENVIRONMENTAL INC	485
2007	PERMA-FIX NORTHWEST RICHLAND INC.	14,163
2008	BURLINGTON ENVIRONMENTAL INC	389
2008	PERMA-FIX NORTHWEST RICHLAND INC	710
2009	BURLINGTON ENVIRONMENTAL INC	565
2009	PERMA-FIX NORTHWEST RICHLAND INC	11,869
2010	BURLINGTON ENVIRONMENTAL INC	1,081
2010	PERMA-FIX NORTHWEST RICHLAND INC	61,554
2011	BURLINGTON ENVIRONMENTAL LLC	1,000
2011	PERMA-FIX NORTHWEST RICHLAND INC	31,543

National Emissions Inventory (NEI)

The National Emissions Inventory (NEI) is a comprehensive and detailed estimate of air pollutants from all air emissions sources. The NEI is prepared every three years by the EPA based primarily upon emission estimates and emission model inputs provided by State, Local, and Tribal air agencies for sources in their jurisdictions, and supplemented by data developed by the EPA. According to the most recent NEI for 2008, there were 439 lbs (199 kg) of PCBs released to the air in Washington State from residential waste burning and 0.8 lbs (0.4 kg) released from commercial marine vessels. In addition to these sources, the Spokane Regional Clean Air Agency reported about 1 lb of PCB emitted from the Waste to Energy facility in 2011.

To obtain emission estimates for residential waste burning, EPA applies emission factors to an assumed mass of residential waste burned at the county level across the country. Some of the key assumptions are that residential waste burning only occurs in rural counties, and roughly 28% of the waste generated in these counties is burned in backyard burn barrels. This estimate is very uncertain, but can only be improved with location specific information regarding local compliance with residential waste burning rules.

Summary

Table 20 below summarizes the estimates for uses and sources in Washington State found in this section.

Table 20. Summary of Uses and Releases for Washington State

Source		Historic reservoir	Annual releases (kg/yr)
Closed	transformers	100-200 kg	< 2
	large capacitors	20 metric tons	10-80
	lamp ballasts	100-350 metric tons	400-1,500
	small capacitors	1-34 metric tons	3-150
	other closed uses		unknown
Partially closed			unknown
Open	caulk	87 metric tons	160
	other open uses		unknown
	pigments and dyes		(PCB-11) 0.02-31
	other inadvertent generation		900
	residential waste burning		199
	commercial marine vehicles		0.4

Pathways and Environmental Fate

Current PCB levels represent both historical and ongoing loadings and cycling among environmental compartments. We consider sources to be the original use of the material, such as PCBs in transformers. PCBs move through pathways such as stormwater and expose people and wildlife.

Unfortunately, we don't know where much of the PCBs that were produced prior to 1979 are currently located. For example, small capacitors used large amounts of PCBs, but since they were used in unregulated appliances, we don't know how many are still in appliances in people's homes, how many were disposed of in municipal waste landfills (and how much PCBs have since leaked out of those landfills or volatilized), and how many were disposed of outside of landfills.

Pathways

Assessments of both Puget Sound (Ecology 2011a) and freshwater systems (Ecology 2011c, King Co 2013b) have found the largest pathway for PCBs to reach the aquatic environment statewide is stormwater. Figure 12 shows the estimated loadings to Puget Sound by pathway (Ecology 2011a) and Figure 13 shows the relative stormwater loadings by type of land cover. In the Puget Sound study, we looked at the concentration of PCBs and other chemicals in surface runoff from four land-cover types: commercial/industrial, residential, agricultural, and forest, field, and other undeveloped lands. The concentration of PCBs in surface runoff was higher from commercial/industrial areas, especially during storm events, but the total loading was lower since industrial/commercial lands occupy less area than other land covers. Overall loads from forests accounted for 83% of the load (Ecology 2011g).

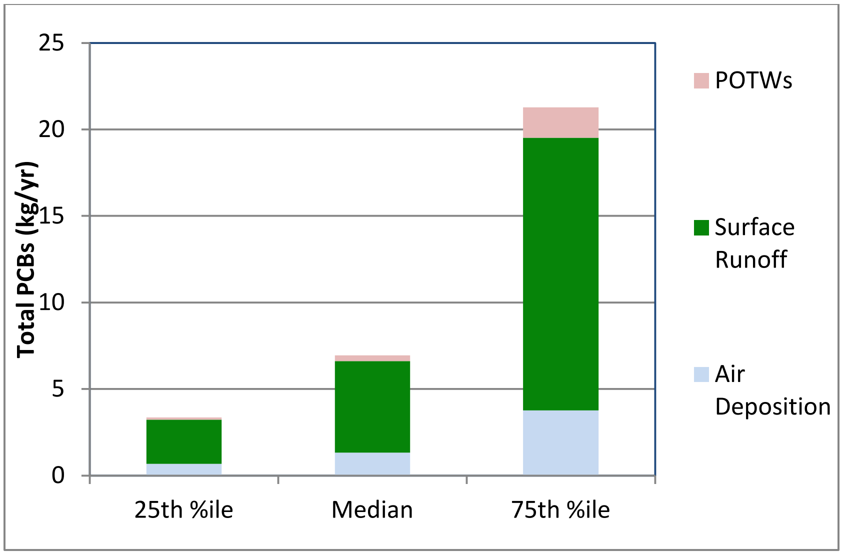


Figure 12. Total estimated PCB loading to Puget Sound by pathway (kg/yr)

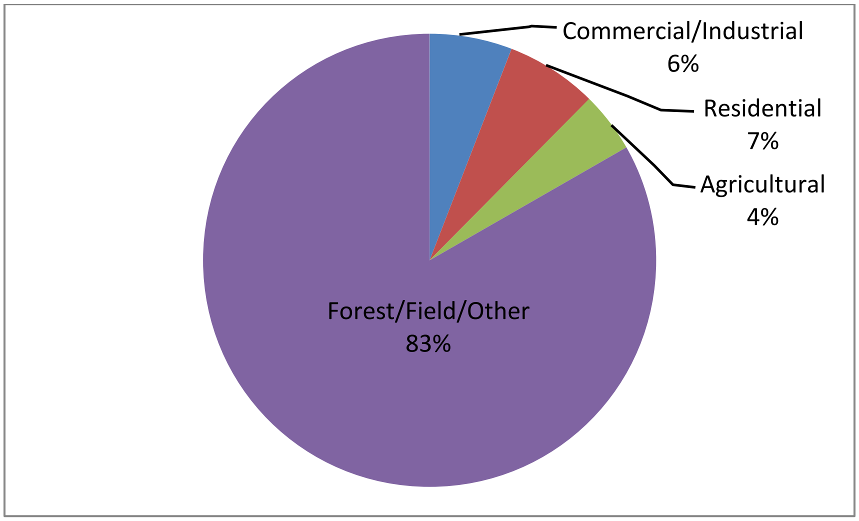


Figure 13: Relative contributions of different land use covers to PCBs in surface runoff in Puget Sound (Ecology 2011g)

Figure 14 shows the relative importance of different pathways in three freshwater systems in Washington. Publically owned treatment works (POTWs) are a smaller pathway in all the systems, with less than 10% of total loading. Direct air deposition was estimated to be the second largest pathway in the Puget Sound and Lake Washington study (Ecology 2010d, King County 2013b).There is a large and variable amount of PCB loading from unknown pathways.

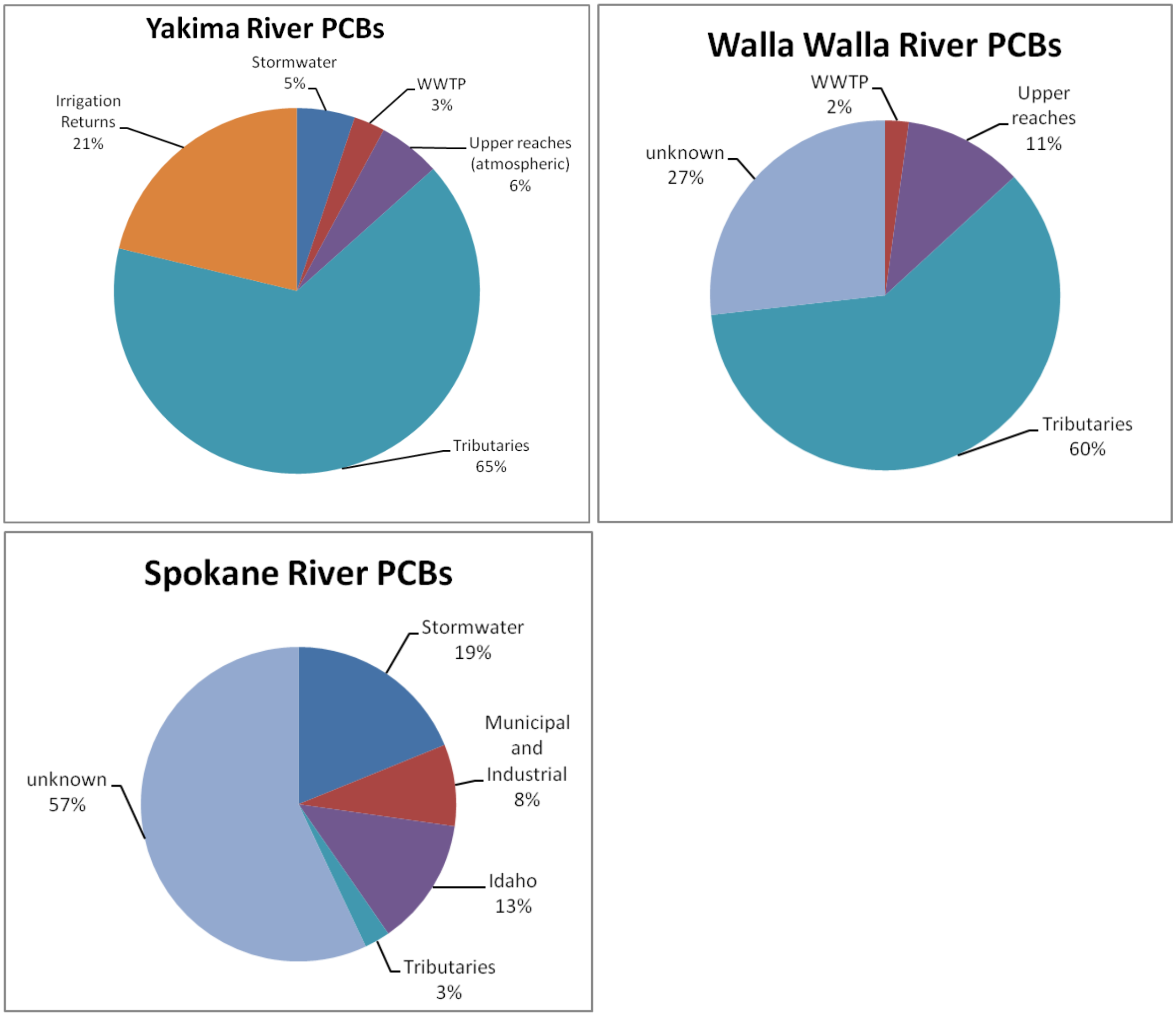


Figure 14. Relative importance of pathways assessed in freshwater systems

Salmon

Pacific salmon returning to spawn are another pathway for PCBs to enter Washington. Salmon have complex life histories and long-range migrations for feeding. The accumulation of PCBs in fish depends on many things including contaminated habitats, which food they eat and the levels of PCBs in food, lipid level, and age. Chinook salmon are 3-5 times more contaminated than coastal Chinook (West 2011). Most (99%) of the final weight of adult Chinook is achieved in salt water, both ocean and Puget Sound, and >96% of the PCBs in adult Chinook accumulated during their marine life history phase (O'Neill and West 2009). O'Neill estimated that 0.265 kg/yr PCBs enters Washington through this pathway (Ecology 2011a). This estimate comes from whole body PCB concentrations for five species of Pacific salmon and their estimated biomass.

Motor oil

The City of Spokane reported measurements of PCBs in motor oil in their 2013 Adaptive Management Plan for Reducing PCBs in Stormwater Discharges from the Wastewater Management Department. The concentration of total PCBs in four samples of motor oil ranged from 14 to 116 ppb with an average of 54 ppb. In the PAH CAP (Ecology 2012a), Ecology estimated that 9,737,812 kg of motor oil is released annually in Washington from drips and leaks and another 1,555,179 kg of used motor oil is released to the environment through improper disposal. Using the average concentration of PCBs in motor oil from sampling in Spokane (54 ug/kg) and the statewide estimate of drips and leaks from motor vehicles and improper disposal of used motor oil in Washington (11,292,991 kg), gives an estimate of 0.6 kg of PCBs per year from motor oil.

PCBs are not created in motor oil, so motor oil is not considered a source here. The PCBs in motor oil are likely contamination from an unknown source. The homologue pattern is different from the PCBs found in sediments from stormwater catch basins in Spokane, with a much larger percentage of mono and dichlorobiphenyls and smaller percentage of higher chlorinated congeners. Of course, the congeners in sediment have been weathered and would no longer match the profile of the original source.

Environmental Partitioning

The estimates we do have for the fate of all the PCBs produced are not current. Newer publications cite estimates in earlier publications. These estimates vary, but they agree that much of the PCBs that were produced up to 1979 are still in use. This is a large reservoir of PCBs that are slowly leaking out into the environment. For transformers in particular, we know that much of the PCBs in transformers have been removed since these estimates were made. Transformers and capacitors were the largest use of PCBs and have been targeted for PCB removal.

In 1997 EPA estimated the inventory of PCBs as of 1977 as "Of the 700,000 [short] tons of PCBs produced, 150,000 tons had been landfilled; 75,000 tons had entered the air, water, and soil; 25,000 tons had been incinerated; and 375,000 tons remained in electrical equipment. The remainder, approximately 75,000 tons, had been exported."

Converting this 1997 EPA estimate of short tons into metric tons leads to:

- 636,000 Produced (1927-1976)
- 568,000 Used (68,000 exported)

- 340,000 Remaining in use (60%)
- 228,000 Disposal/environment
- 132,000 Landfill (23%)
- 68,000 Environmental media (12%)
- 28,000 Incinerated (5%)

Table 21 shows estimates for the status of PCBs. The estimates in Keeler (1993) for the status of PCBs in the US as of 1982 are similar to those of the EPA above. The Canadian government estimated PCBs in Canada as of 1992 (CCME 1995) and Tanabe (1988) estimated similar percentages worldwide in 1985 (Table 21). All of these estimates include a large percentage of PCBs still in use and a small percentage destroyed by incineration. Unfortunately, we do not have more current estimates of PCB stocks and many of the PCBs that were in use at the time of these estimates have been taken out of use for disposal.

Table 21. Estimates for the status of PCBs (in metric tons)

Status	**US 1977 (EPA 1997)**	**US 1982 (Keeler *et al* 1993)**	**Canada 1992 (CCME 1995)**	**Global 1985 (Tanabe 1988)**
Produced	636,000	640,000		1,200,000
Used	538,000	582,000 (91%)	40,000	1,200,000
Remaining in Use	340,000 (40%)	346,000 (54%)	15,000 (38%)	780,000 (65%)
Landfill/Storage	132,000 (23%)	134,000 (21%)	6000 (15%)	
Environment	68,000 (12%)	70,000 (11%)	12,400 (31%)	370,000 (31%)
Incinerated	28,000 (5%)	19,000 (3%)	6,200 (16%)	50,000 (4%)

Tanabe (1988) also broke down the global PCBs in the environment into different media (Table 22). Not shown in Table 22 is that the largest global reservoir of PCBs is ocean water (while PCBs are not very soluble in water, the vast quantities of oceans worldwide hold more than half of the PCBs in the environment). Table 22 only includes estimates from the terrestrial and coastal waters and not ocean water.

Table 22. Global PCBs from Tanabe 1988 for PCBs on land, rivers and coastal waters

	PCB loads (metric tons)	**percent**
air	500	0.35
river and lakewater	3,500	2.45
seawater	2,400	1.68
soil	2,400	1.68
sediment	130,000	90.85
biota	4,300	3.00
Total	**143,100**	**100**

The estimates from Tanabe 1988 in Table 22 agree well with the model for Puget Sound (Ecology 2009b) and Lake Washington (King County 2013). For Puget Sound it was estimated approximately 97% (1440 kg) of the total mass of PCBs currently in the aquatic ecosystem of Puget Sound is contained in the active sediment layer (top 10 cm), <1% (10kg) is stored in the water column, and <3% (40 kg) is stored in the biota. O'Neill and West (2007) estimated PCBs in biota using PCB concentrations and biomass. Their total estimate is less than 40 kg of PCBs in Puget Sound biota.

Wildlife Health

Introduction

PCBs have similar effects in wildlife, people, and model organisms used to study people in laboratory experiments. Thus, this section and the section on Human Health have many similarities, although the health effects that are measured often differ between people and wildlife. Negative effects of PCBs in wildlife are of concern because of the effects on animal populations, because animal populations may be sentinels for human health, and because we are also part of the food chain and are exposed to PCBs through our diet.

PCBs can be acutely toxic to wildlife, but most of the impacts occur due to chronic exposure. Data are most prevalent on mortality, reproduction, development, and endocrine effects. In addition to toxicity endpoints, the induction of enzymes and genes are also used to assay PCB effects. Other endpoints include cancer, immunological, neurological/behavioral, and hepatic effects. Experimental studies are often performed for certain endpoints because of correlations seen in the field with PCB levels and specific endpoints.

Wildlife is exposed to varied mixtures of PCBs in the environment. Different PCB congeners behave differently in the environment; they preferentially partition into different media and they are preferentially degraded and bioaccumulated. In addition, there are multiple sources of PCBs that contain different mixtures of congeners that release PCBs into the environment. Therefore, the actual environmental conditions are different than many of the laboratory studies on specific Aroclor mixtures or specific congeners.

Sensitivity to PCBs varies among species and within species. For example, fish are most susceptible in early life stages. Inter-species variation is also due to different lipid levels, because PCBs are lipophilic and sequestered in lipid-rich tissues. PCBs are biomagnified up the food chain, so organisms on higher trophic levels have higher concentrations of PCBs. There are other differences among species that affect PCB metabolism. For example, invertebrates lack the enzyme systems that react with dioxin-like PCBs.

Although environmental levels of PCBs have declined substantially since they first came under regulation in the 1970s, the rate of decline has slowed in recent years and significant contamination continues to be widespread in Washington State..

Puget Sound is a regional hot spot for PCBs compared to the Pacific coast and British Columbia. Within Puget Sound, the most contaminated areas are in the main basin, especially Seattle’s Elliott Bay and Tacoma’s Commencement Bay, and, to a lesser extent, Everett Harbor and the Bainbridge Basin. Puget Sound’s food web, from plankton on up to harbor seals and killer whales, has significant PCB contamination. Herring and Chinook salmon are notably affected.

Hot spots for PCBs are also present in Washington rivers and lakes. Based on PCB levels in resident fish species, major waterbodies of most concern are the Wenatchee River, Lake Washington, the Columbia River, and the Spokane River.

Health effects in wildlife

Table 23 (from ATSDR 2000) summarizes PCB effects seen in laboratory experiments and field studies with wildlife species. Most species have mortality at high doses.

Adverse effects in birds include:

- Reduced egg hatchability and live births
- Reduced avoidance response
- Altered mating, reproductive, parenting, and nesting behavior
- Suppression of immune response

Adverse effects in fish include:

- Reduced hatchability in eggs
- Altered muscle coordination
- Depressed immune system with increased susceptibility to infections
- Loss of fins and tails in flatfish

Adverse effects in mammals include:

- Loss of embryos and fetuses and reduced live births
- Alteration in the immune system in mink, sea lions, and seals
- Tumors and deformities of skeleton and skin in seals

Table 23. PCB Hazards in Wildlife with references noted in original (ATSDR 2000 Table 3-6)

Adverse effect	Wild mammals				Birds			Reptiles	Amphibians		Fish	
	Primate	Mustelid	Cetacean, pinniped	Other	Piscivore	Galliform	Other	Turtle	Frog	Toad	Freshwater	Marine
Mortality	OE1	OE1		OE3	OE3	OE1	OE3		OE1	OE1	OE1	OE3
		OE3				OE3			OE3	OE3	OE2	
		OE4									OE3	
Systemic effects												
Respiratory		OE4				OE3						OE3
Cardiovascular		OE3	OC4			OE3						
		OE4										
Gastrointestinal	OE1	OE1	OC4			OE3						
	OE3											
Hematological		OE4									OE3	
Musculo-skeletal						OE3					OE3	
Hepatic	OE3	OE1			OE3	OE1	OE1				OE3	OE3
		OE3				OE2	OE3					
		OE4				OE3						
		OE5				OE5						
Renal		OE4	OC4			OE3					OE3	
Endocrine	OE3	OE3	OE3		OE3	OE3	OE1				OE3	OE3
		OE4	OE4		OC4		OE3					
		OE5	OC4									
Dermal/ocular	OE1	OE3	OC4								OE3	
	OE3											
Body weight	OE1	OE1				OE2	OE1			OE3	OE3	
	OE3	OE3										
Metabolic		OE5					OE1		OE2		OE3	

Adverse effect	**Wild mammals**				**Birds**			**Reptiles**	**Amphibians**		**Fish**	
	Primate	Mustelid	Cetacean, pinniped	Other	Piscivore	Galliform	Other	Turtle	Frog	Toad	Freshwater	Marine
Enzyme induction		OE1				OE1	OE1				OE1	OC4
		OE3				OE2	OE2				OE3	
		OE4				OE3	OE3					
		OE5										
Blood chemistry	OE1	OE5				OE3	OE1				OE3	
Immunological/ lymphorecticular	OE1	OE4	OE4		OC4	OE1	OE1				OE3	OE3
	OE3		OC4			OE3	OE3					
Neurological/ behavioral	OE2	OE2		OE3		OE3	OE3				OE2	OE3
	OE3	OE4									OE3	
Reproductive	OE3	OE1	OC4	OE3	OC4	OE3	OE1	OC4			OE3	OE3
		OE3	OE4				OE3				OC4	OC4
		OE4										
		OE5										
		OC4										
Developmental	OE3	OE3		OE3	OE1	OE1		OE4	OE1	OE1	OE3	
		OE4			OC1	OE3			OE3	OE3	OC4	
					OC4							
Egg shell					OC4	OE3	OE3					
							OC4					
Genotoxic							OE3					

O= observed effect E= experimental observation C= correlational field observation
1 = dioxin-like PCB congener (AhR binder; planar; chlorine para-substituted and non- or mono-*ortho*-substituted)
2 = non-dioxin-like PCB congener (poorly binds to AhR; non-planar; chlorine di-, tri-, or quatro-*ortho*-substituted)
3 = commercial PCB mixture (e.g., Aroclor 1016)
4 = “weathered” (i.e., environmentally degraded and/or metabolized) PCB mixture, usually in combination with other chemicals (e.g., PCBs in wild-caught fish)
5 = unspecified PCB

Examples of major endpoints

Reproduction and development

PCBs affect reproduction and development in different species. Mink are particularly sensitive to the reproductive effects of PCBs (Eisler 1986). Farm-raised mink fed a diet of PCB-contaminated fish from the upper Hudson River at the same levels wild mink are exposed to PCBs in food, showed effects on reproduction and offspring growth and mortality (Bursian *et al*. 2013). Females with higher levels of PCBs had fewer live kits per litter. Kit mortality increased over time, with no kits surviving in the animals fed higher levels of PCBs. The surviving kits also had lower body masses after 6 weeks. The effects on reproductive performance were similar to those seen in earlier studies on mink fed contaminated fish from Saginaw Bay (Heaton *et al*. 1995a).

Immune System

PCBs are linked to increased disease susceptibility in several species. Captive harbor seals exhibit negative effects on their immune system after being fed PCB-contaminated herring, as assayed by immune cell function and response (Ross *et al*. 1996). This research was undertaken to understand factors contributing to virus-caused mass mortalities of marine mammals, especially when attributed to a virus that does not always cause mass mortalities. The results suggest higher levels of PCBs contribute to higher virus-caused mortality.

Cancer

PCBs have been shown to cause cancer in laboratory animals (ATSDR 2000) and are considered to be probable human carcinogens by EPA. PCBs are thought to cause cancer indirectly, rather than by direct alterations to DNA.

Cancer is less well studied in wild populations compared to laboratory species, partly due to lower incidence. However, beluga whales in the St. Lawrence estuary and Hudson Bay have been found to have a high incidence of cancers and high levels of PCBs (Mikaelian *et al*. 2003). There is also evidence linking cancer in St. Lawrence estuary belugas to PAHs from nearby point sources (Martineau *et al*. 2002), illustrating the difficulties in pointing to a specific group of chemicals in these marine mammals with many different industrial contaminants. PCB levels in California sea lions have also been found to be significantly associated with death from cancer (Ylitalo *et al*. 2005).

Mechanisms of action

Endocrine

PCBs interfere with estrogen and thyroid hormone levels. Studies on PCB endocrine disruption have been done in the laboratory with model animals and cell cultures (see section on Human Health). The endocrine system regulates all biological processes, although endocrine disruption is often used just to refer to the disruption of thyroid hormones and the sex hormones estrogen and androgen. These hormones are important for growth and development, especially of the brain and nervous system and reproductive systems. While hormones are important throughout the life cycle, they are particularly important during fetal development. Hormones are signaling molecules that function at low levels, and compounds that either mimic or block natural hormones may have effects at low levels.

Ah-receptor dependent

Similar to dioxins, non-ortho (co-planar) and mono-ortho PCBs can bind to the aryl hydrocarbon (Ah) receptor. Subsequent to binding of the Ah receptor, there are changes in gene expression (e.g., induction of cytochrome p450 CYP1A1/1A2) leading to toxic responses. Induction varies by degree and pattern of chlorines and is the basis for the World Health Organization (WHO) toxic equivalency factors (TEFs) for dioxins and dioxin-like PCB congeners. These have been reviewed and modified several times. In 2005 WHO updated the TEFs for humans and mammals to replace the 1998 values (Van den Berg *et al*. 2006, see Table 29 in the Human Health Section). The adverse effects for these compounds are mediated through the Ah receptor and the relative potencies are compared to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD). Consensus TEFs for wildlife were developed in 1998 (Van den Berg *et al*. 1998). The WHO working group harmonized the TEFs across different taxa as much as they could, but there are large differences in responses among different taxa.

Table 24. WHO TEFs for fish and birds (Van den Berg *et al*. 1998)

Type	**Congener**	**TEFs**	
		Fish	Birds
Co-planar PCBs	3,3',4,4'-TCB (77)	0.0001	0.05
	3,4,4',5-TCB (81)	0.0005	0.1
	3,3',4,4'-5-PeCB (126)	0.005	0.1
	3,3',4,4',5,5'-HxCB (169)	0.00005	0.001
Mono-ortho PCBs	2,3,3',4,4'-PeCB (105)	<0.000005	0.0001
	2,3,4,4',5-PeCB (114)	<0.000005	0.0001
	2,3',4,4',5-PeCB (118)	<0.000005	0.00001
	2',3,4,4',5-PeCB (123)	<0.000005	0.00001
	2,3,3',4,4',5-HxCB (156)	<0.000005	0.001
	2,3,3',4,4',5'- HxCB (157)	<0.000005	0.0001
	2,3',4,4',5,5'- HxCB (167)	<0.000005	0.00001
	2,3, 3',4,4',5,5'- HpCB (189)	<0.000005	0.00001

PCBs in Washington's Environment

Air and Soil

Due to long-range regional and global atmospheric transport, PCBs are present in all parts of the environment. Most of the PCBs in air come from volatilization of PCB-contaminated soil and surface water. In the atmosphere, PCBs are primarily associated with the gaseous phase; approximately 10% is adsorbed to particulates, especially the higher chlorinated forms. Less chlorinated compounds travel farther than highly chlorinated compounds, which tend to stay closer to the source of contamination. Atmospheric deposition is, in turn, the dominant source of PCBs to most soil and water surfaces. The cycle of persistent compounds like PCBs depositing onto soil and then revolatilizing back into air is often referred to as the grasshopper effect.

PCB levels in the atmosphere have been decreasing slowly since the late 1970s when EPA began restricting their use (EPA Integrated Atmospheric Deposition Network http://www.epa.gov/glnpo/glindicators/air/airb.html, Venier and Hites 2010, ASTDR 2000). Due to residual sources in the U.S. and long-range transport from other countries, PCB levels in air may be leveling off after a period of decreasing.

Air in rural and remote locations has lower PCB levels than urban air, which is a source to nearby environments. Nationally, average total PCB concentrations at background locations (not near known sources of PCBs) are in the approximate range of one to several hundred pg/m^3 (Hornbuckle and Robertson, 2010). In contrast, concentrations near Lake Superior in the 1970s were well over 1,000 pg/m^3 due to influences from upstate New York and the East Coast (EPA Integrated Atmospheric Deposition Network)

The limited data available for PCBs in Washington's air has been from measurements of wet (rainfall) and dry (particulate) deposition rates to water and land surfaces (ng/m^2/day) rather than mass per unit volume. In 2008, Brandenberger *et al*. (Ecology 2010d) recorded PCB fluxes to the Puget Sound basin at seven stations from Nisqually River to Padilla Bay. The median flux across all stations and rain events was 0.51 ng/m^2/day. Similar results were obtained for most areas, except Tacoma's Commencement Bay had a median of 1.8 ng/m^2/day. Brandenberger *et al*. concluded that PCB deposition rates to the Puget Sound basin were similar to background sites in New Jersey (0.82 ng/m^2/day; 1999-2000), but lower than Chesapeake Bay (9.0 ng/m^2/day; 1990-1991) and Jersey City, NJ (11 ng/m^2/day; 1999). King County recently completed an air deposition study at six stations in the Lower Duwamish/Green River watershed (King County 2013a). Station locations included three in the Lower Duwamish River area, two in downtown Kent and one in Enumclaw. Median fluxes of total PCBs ranged from 1.1 ng/m2/day in Enumclaw to 16.9 ng/m2/day in the South Park neighborhood of the Lower Duwamish River. King County has also measured air deposition to Lake Washington at one station near Sand Point; a median flux of 1.39 ng/m2/day was estimated from these measurements (King County 2013b).

The Western Airborne Contaminants Assessment Project for national parks in the western U.S. included Mount Rainier, Olympic, and North Cascades parks (Landers *et al*. 2008). The results, however, are of limited use for present purposes in that only eight PCB compounds were analyzed and detection frequency was low.

Meijer *et al*. (2003) estimated that the contemporary PCB burden in background soils is about 2% of the known production volume. PCB levels in U.S. background soils generally average from several hundred to several thousand ppt dry weight (Hornbuckle and Robertson, 2010). An EPA nation-wide survey of soil at 27 remote or rural sites in 2003 put the average total PCB concentration at 3,089 ppt (EPA 2007). The single Washington site sampled during the study – Lake Ozette on the northwest coast – had 2,419 ppt.

With the exception of site-specific determinations for contaminated sites, the PCB background in Washington soils has not been well characterized. Relatively more is known about PCBs in Washington's marine and freshwater environment, as discussed below.

Marine and Fresh Waters

Historical vs. Recent Trends

PCB levels in Washington's marine and fresh waters have decreased substantially since peaking in the 1970s. This has been attributed to EPA's restrictions and bans on PCBs in the late 1970s and early 1980s, similar actions in Canada and other countries, contaminated site cleanups, improved wastewater treatment, losses through volatilization and metabolism of lighter compounds, and deep burial in aquatic sediments.

Although historical declines have been documented, there are components of Washington's marine and freshwater ecosystems where a decreasing trend is no longer evident. In most cases, the time-trend for PCBs can be characterized by an initial rapid decline after the ban, followed by a slowing and, ultimately, low to negligible rate of decrease over recent years, waterbodies benefitting from cleanups being a notable exception. Current PCB levels continue to be a concern for the health of fish, wildlife, and humans in Washington.

The effect of the 1970s and 1980s regulations can be clearly seen in age-dated sediment cores from Puget Sound and Lake Washington (Figure 15). A rapid increase in concentrations occurred in both Puget Sound and Lake Washington into the mid-1970s, reaching 35 ppb and 250 ppb, respectively, in sediments at these two locations. Concentrations in the recent past declined to around 10 to 20 ppb. (Detections shown prior to the mid-1930s when PCBs were first introduced are analytical noise.) A sediment core from Lake Spokane (lower Spokane River) in eastern Washington showed a similar steep decline in the 1960s and 1970s, followed by a gradual reduction over a 20-year period from approximately 1980 to 2000 similar to what was observed in Lake Washington (Ecology 2011c).

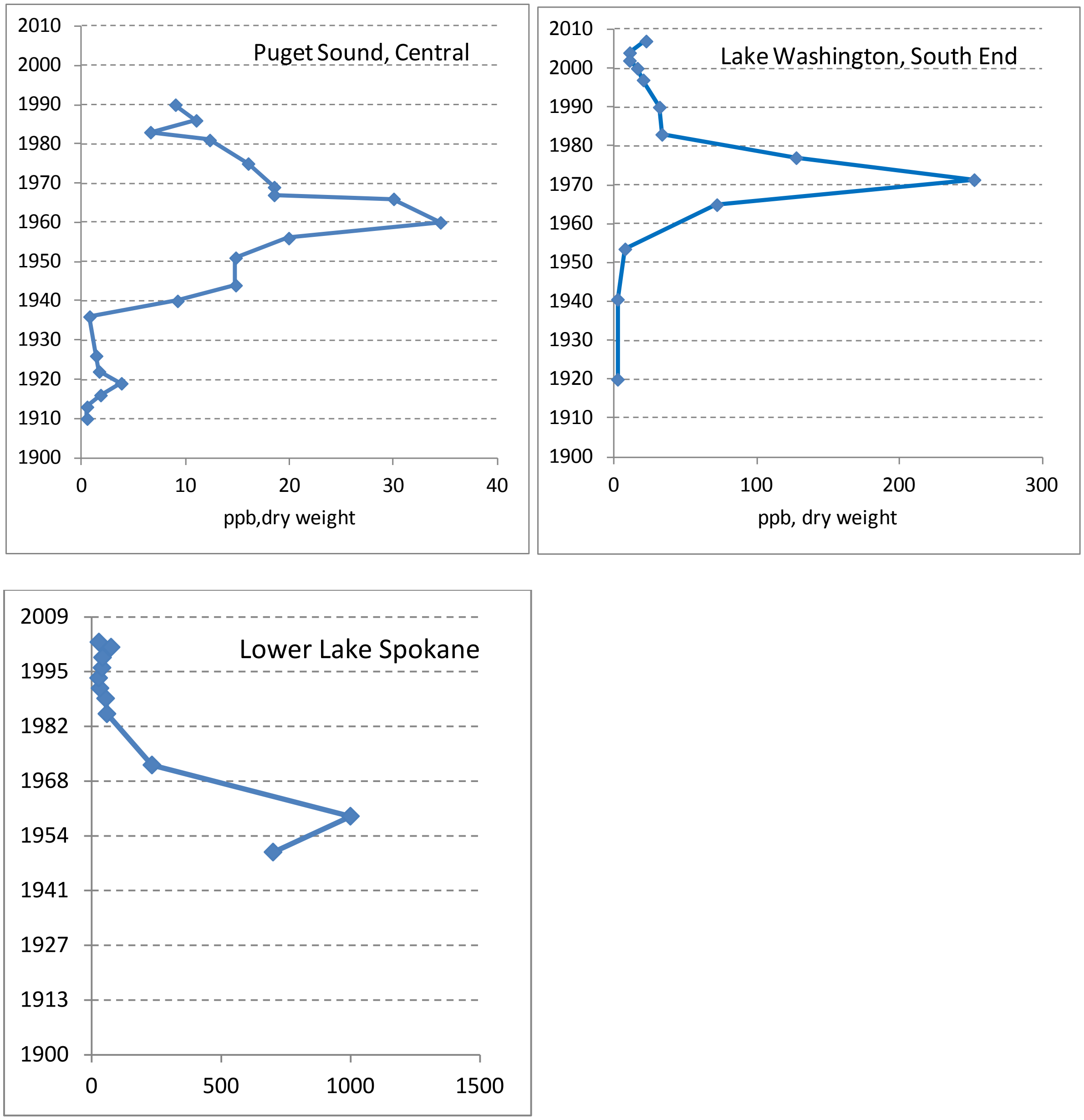
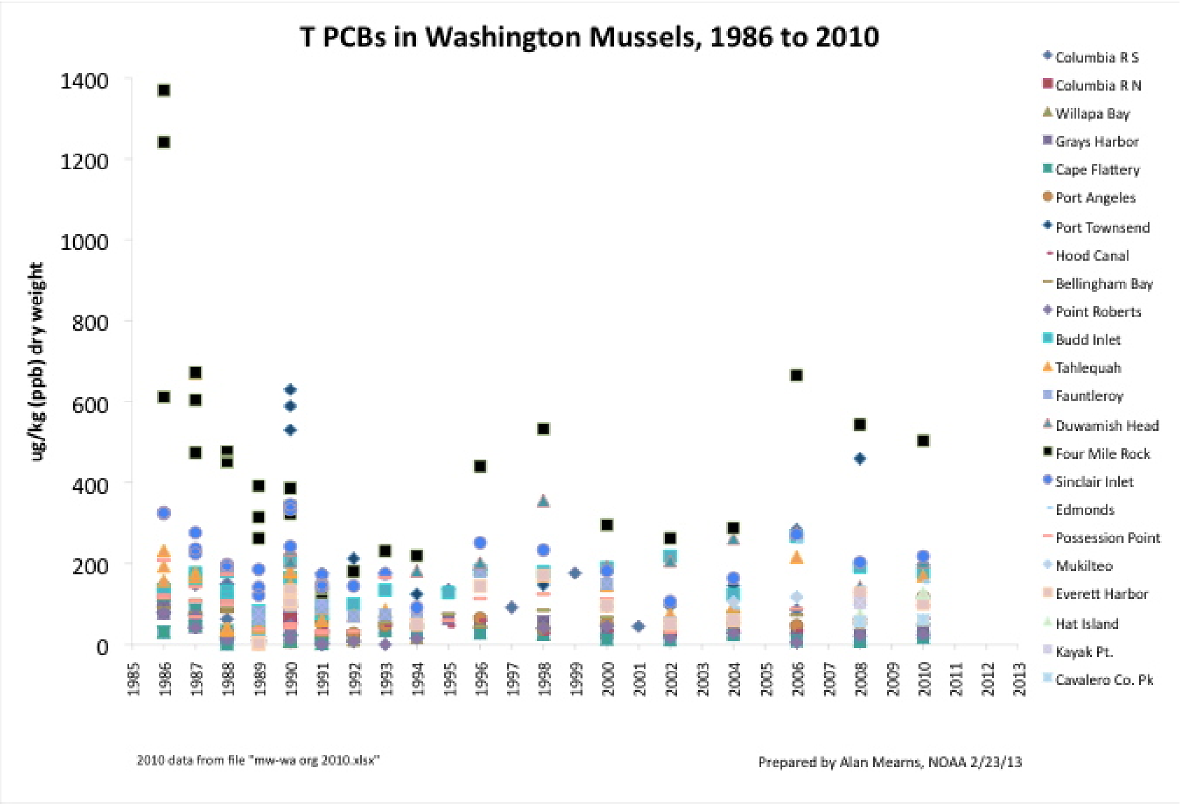


Figure 15. Total PCBs in Age-Dated Sediment Cores from Puget Sound, Lake Washington, and Lake Spokane

NOAA's Mussel Watch has monitored PCBs in marine mussels from the mouth of the Columbia River to Puget Sound since 1986. Results show that PCBs have been declining slowly, although somewhat erratically, in Washington's marine waters (Figure 16). After a steep decline in the mid-1980s, there have been two spikes of unknown origin, most obvious at the regional hot spot: Four-mile Rock in Elliott Bay. PCB levels in Puget Sound mussels remain well above national median concentrations (Mearns 2013, O'Connor and Lauenstein 2006).



2010 data from file "mw-wa org 2010.xlsx"

Prepared by Alan Mearns, NOAA 2/23/13

Figure 16. Total PCB Concentrations in Marine Mussels from Columbia River to Puget Sound: 1986 – 2010

(Data from NOAA National Mussel Watch Program, prepared by Alan J. Mearns, NOAA, Seattle)

Compared to historical levels, PCBs have declined in Puget Sound harbor seals (1972 to 1997) and killer whales (1993-1995 vs. 2004-2006) that inhabit or transit Puget Sound (Calamabokidis *et al*. 1999, Krahn *et al*. 2007, Hickie *et al*. 2007). Despite these declines, levels of PCB sassociated with health impacts are observed in seals from this region (Strait of Georgia) and most Southern Resident killer whales exceed health effects thresholds for PCB residues (Cullon *et al*. 2009, Hickie *et al*. 2007, Krahn *et al*. 2009). The PCB decline in these animals has been slowed by continued atmospheric delivery of PCBs from other parts of the world and internal cycling (Johannessen *et al*. 2008). Figure 17 shows the PCB changes observed in the blubber of South Puget Sound harbor seals up until 1997.

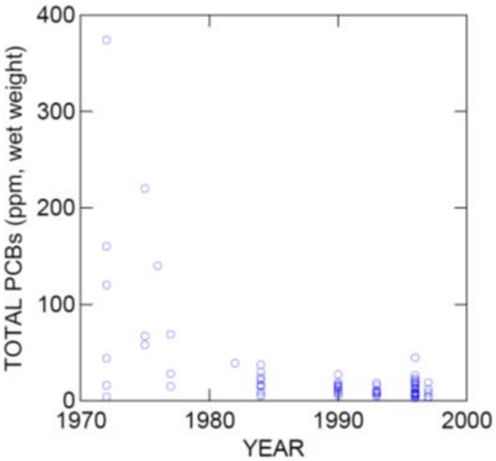


Figure 17. Historical Decline in PCB Levels among South Puget Sound Harbor Seals: Blubber Samples 1972-1991 (Calambokidis, 1999)

Monitoring by the Washington Department of Fish and Wildlife (WDFW) for the Puget Sound Estuary Monitoring Program (PSEMP) gives a mixed picture of PCB time-trends in edible tissues of marine fish (West 2011, West *et al*. 2011). Levels have declined in coho salmon from central Puget Sound, but only until the 1990s. Trend data do not exist for Chinook salmon. There is no evidence of a PCB decline in four stocks of Puget Sound herring. English sole from all urban and non-urban monitoring locations except Sinclair Inlet show no significant change in PCB concentrations over the past 20 years (Figure 18). There is some evidence of improvement in Sinclair Inlet in recent years, which has been attributed to reduced stormwater loading, and dredging and capping of contaminated sediments in 2000-2001 (O’Neill *et al*. 2011). Sole are a bottom-living species that demonstrate the link between PCBs in sediment and biota.

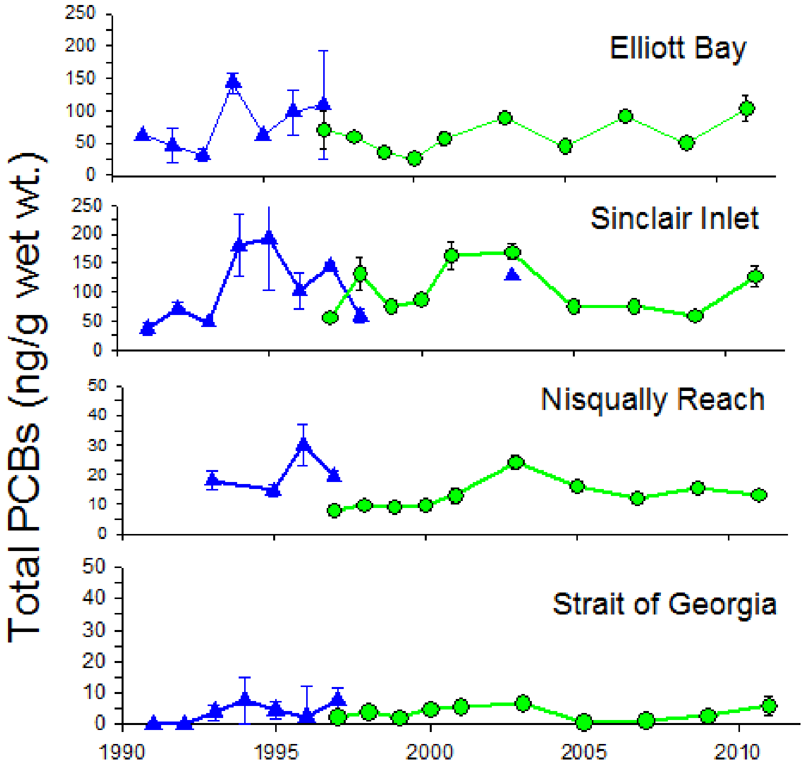


Figure 18. 20-year Time Series for PCBs in English Sole from Selected Urban and Non-urban Locations in Puget Sound
(prepared by James West, WDFW; parts per billion; symbol shift indicates change in analytical method)

Long-term trend data are limited for Washington freshwater fish. PCB levels have declined in Spokane River fish since the early 1990s due to cleanup of hazardous waste sites, excavation and capping of contaminated river sediments, and reduced discharge from industrial and municipal treatment plants (Ecology 2011a, EPA 2009).

The U.S. Geological Survey (USGS) analyzed changes in PCB residues in whole fish samples collected from 16 stations in the Columbia River Basin between 1970 – 1986, and in 1997 (Hinck *et al*. 2004). Not surprisingly, a number of sites had lower levels in 1997 than in the 70s or 80s. USGS observed, however, that criteria for the health of fish or wildlife were still exceeded and concluded that PCBs remain a cause for concern. Historical declines have also been documented for PCBs in Columbia River otter, mink, eagle, and osprey (EPA 2009), all of which prey largely on fish. Here again, the comparison is primarily with samples collected in the 70s and 80s.

Current Levels and Spatial Patterns

Water

Due to the extremely low solubility of PCBs and the high cost of analysis, there have been few attempts to measure concentrations in the water column. As part of the Puget Sound Toxics Loading Analysis (PSTLA), Ecology analyzed PCBs at four sites in Puget Sound, three sites at the eastern end of the Strait of Juan de Fuca, and five major Puget Sound rivers in 2009-2010 (Ecology 2011d). Average concentrations in the Strait of Juan de Fuca (20 ppq) and Puget Sound (31 ppq) were similar to that previously reported for the Strait of Georgia (42 ppq) by Canadian researchers (Dangerfield *et al*. 2007). The mean total PCB concentration in the five Puget Sound rivers was 16 ppq. The Puyallup and Stillaguamish Rivers tended to have higher concentrations (up to approximately 40 and 60 ppq, respectively) than the Skagit, Snohomish, or Nooksack (less than 20 ppq). King County (2013b) measured PCB concentrations in Lake Washington, the Ship Canal, Sammamish and Cedar rivers, and three major tributaries to the Lake. The total PCB concentrations in Lake Washington ranged from an average of 54 ppq during mixed conditions to 62 ppq in the hypolimnion and 229 ppq in the epilimnion during stratified periods. Concentrations in the two rivers were similar and together averaged 90 ppq. Ship Canal concentrations averaged 108 to 295 ppq. Concentrations in tributaries were higher than the Ship Canal, ranging from a mean of 451 ppq during baseflow conditions to 2,985 ppq during storm events. Williston (2009) and Gries and Sloan (Ecology 2009a) report concentrations in the Green/Duwamish River, up to 2,360 and 1,600 ppq total PCBs, respectively.

A surface water study conducted for Puget Sound Toxics Loading Analysis (PSTLA) in 16 Puyallup and Snohomish county streams found higher PCB levels during storm-events than for baseflow conditions (Ecology 2011g). The median total PCB concentration was 348 ppq for storm-event samples compared to 227 ppq for baseflow samples. Washington’s human health water quality criterion for PCBs is 170 ppq . Of the 70 samples analyzed, approximately 1/3 exceeded the criterion, primarily storm event samples. Except for a single sample, Washington’s aquatic life criteria (ranging from 0.014 to 10.0 ppb) for PCBs were not exceeded

PSTLA estimated the total PCB load to Puget Sound and the U.S. portions of the Straits of Georgia and Juan de Fuca was 3-20 kg per year (Ecology 2011a). Surface water was identified as the major PCB pathway, accounting for 74-76% of the total load (2.55–15.77 kg/yr). The concentration of PCBs in surface runoff was higher from commercial/industrial areas, especially during storm events, but the total loading was lower since industrial/commercial lands occupy less area than other land covers. Overall loads from forests accounted for 83% of the stormwater load (Ecology 2011g). Atmospheric deposition and publicly owned treatment works (POTWs) accounted for 18-20% (0.68-3.76 kg/yr) and 4-8% (0.126-1.75 kg/yr), respectively, of the loading (Ecology 2011a). PSTLA concluded there was insufficient data to estimate PCB loading from groundwater (Ecology 2011f).

Ecology and USGS have used passive sampling techniques (e.g., semi-permeable membrane devices (SPMDs)) to concentrate and estimate PCB concentrations in other freshwater areas (McCarthy and Gale 1999, Ecology 2012b, Ecology 2011a, Ecology 2005, Ecology 2004, Ecology 2010b, Ecology 2011m). Rivers, their tributaries, and lakes that have exceeded the human health criterion to the greatest extent are:

- Columbia River: Wenatchee River, Willamette River (Oregon), Lake River
- Spokane River: (major pathway is stormwater)
- Yakima River: Granger Drain and Sulphur Creek Wasteway (stormwater also a major pathway)
- Walla Walla River: Garrison Creek
- Lake Washington

Water Quality Assessment list

There are 158 303(d) listings for PCBs in Washington’s 2012 Water Quality Assessment. The federal Clean Water Act, adopted in 1972, requires that all states restore their waters to be “fishable and swimmable.” Washington's Water Quality Assessment lists the water quality status for water bodies in the state. The assessed waters are grouped into categories that describe the status of water quality. The 303(d) list comprises those waters that are in the polluted water category, for which beneficial uses– such as drinking, recreation, aquatic habitat, and industrial use – are impaired by pollution.

For the water body segments found to be impaired (category 5) Ecology conducts a total maximum daily load (TMDL) analysis and develops a cleanup plan for meeting water quality standards. Table 25 shows the PCB listings. There are 158 listings for PCBs in category 5. There are ten listings for PCBs in category 4a, which means a TMDL is completed and a plan is in place to meet water quality standards. Sinclair Inlet is in category 4b, which is similar to category 4a, but there is a pollution control plan instead of a TMDL. Fifty-six waterbody segments across the state are in category 2, meaning there is some evidence for elevated levels of PCBs, but there is not sufficient evidence to list it as impaired.

Figure 19 is a state map of the category 5 and 4 listings along with the Washington DOH fish consumption advisories. DOH guidance to develop fish consumption advisories differs from Ecology’s procedures to identify impaired waters. See the section on Human Health for a more detailed explanation. While the details of how a water body segment is considered impaired differs from how a fish consumption advisory is developed, there is a lot of overlap as seen in Figure 19.

Table 25. Water Quality Assessment for PCBs

Number of Waterbody Segments	Category	Description
158	5	Polluted waters that require a TMDL
11	4a and 4b	Polluted water that have a plan in place
56	2	Waters of concern

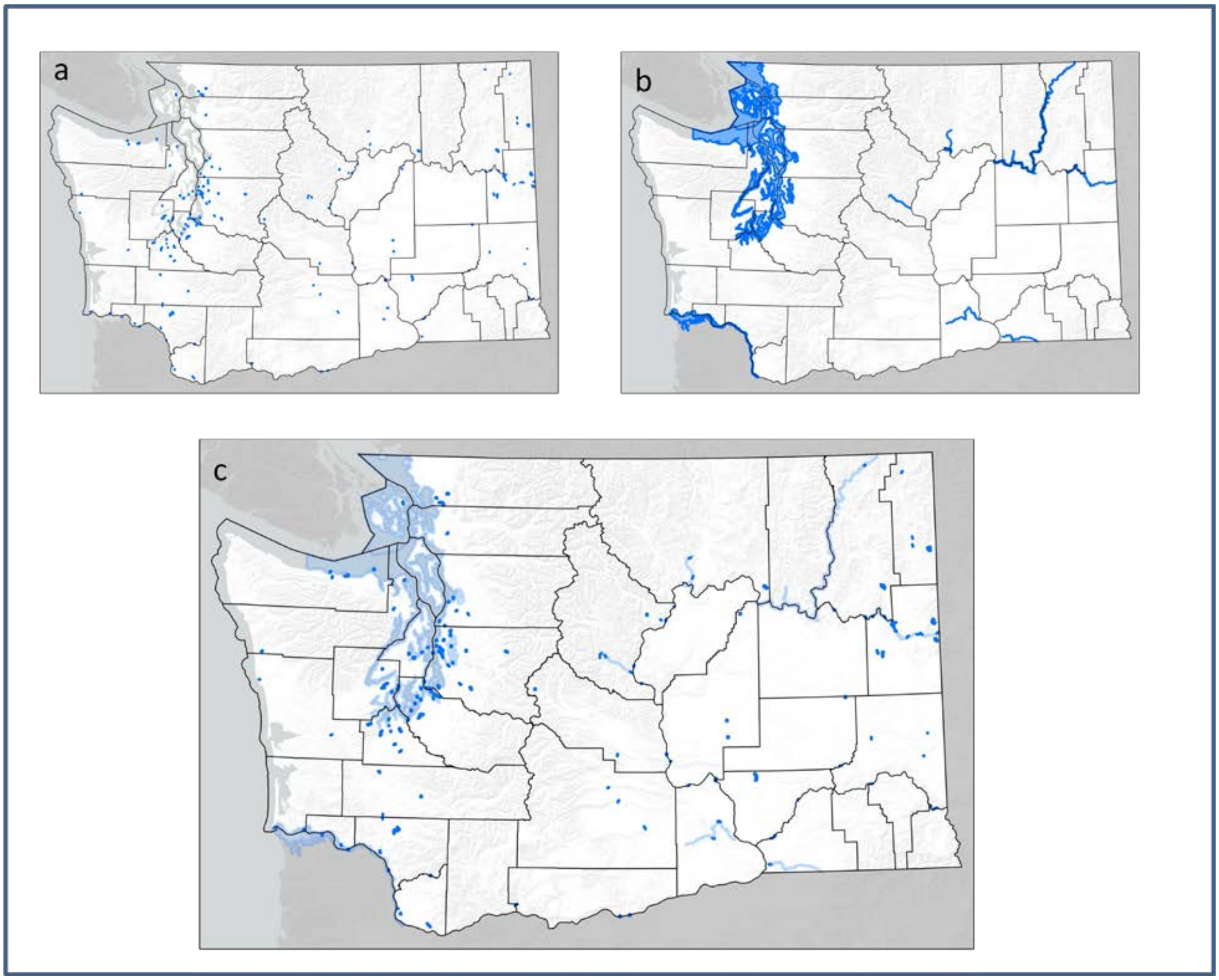


Figure 19. a. Category 4 and 5 303 (d) listings (blue dots). b. Waterbody-specific fish consumption advisories (blue lines, see section on Human Health). c. Overlap of water quality listings and fish consumption advisories.

Marine Sediments

PCBs have been extensively monitored in the marine sediments of Washington. Ecology has PCB data for 630 random sediment monitoring sites in Puget Sound and vicinity up through 2011, collected for PSEMP (e.g., Ecology 2013a).The highest levels are found in urban bays - Elliott Bay, Commencement Bay, Everett Harbor, and the Bainbridge Basin - with concentrations generally diminishing with distance from the shoreline (Figure 20). Particularly high PCB concentrations occur in the Duwamish and Hylebos Waterways in Seattle and Tacoma. Much lower levels are typically encountered in other marine areas.

Total PCBs in Washington marine sediments range from approximately 5 to 2,000 ppb (dry weight), with an overall median of approximately 15 ppb for the greater Puget Sound area. Ninety percent of PSEMP stations have a total PCB concentration below 40 ppb. Less than one percent exceed Washington State sediment quality standards (12 ppm, organic carbon normalized). For perspective, the mean concentration reported for total PCBs in bottom sediments from the Duwamish Waterway Cleanup site is 1,100 ppb, with maximum concentrations as high as 220,000 ppb, dry weight (EPA 2013g).

Freshwater Sediments

The PCB data on freshwater sediments are limited to studies focused on specific waterbodies or cleanup sites rather than large-scale monitoring programs as in Puget Sound. Spatial patterns for PCBs in Washington rivers and lakes are best illustrated with the more comprehensive fish tissue data, discussed below.

A regional freshwater sediment study by Ecology assessed the background for PCBs in northeast Washington (Ecology 2011h). Sediment samples were collected from fifteen lakes and one river thought to be minimally impacted by local human activity. Median and 90th percentile total PCB concentrations were 2.5 and 6.3 ppb, respectively. Atmospheric deposition is assumed to be the predominant PCB source to these waterbodies. By way of comparison, PCB levels in surface sediments of urban/industrial waterbodies such as Lake Washington and Lake Spokane approach 50 - 60 ppb (Ecology 2010a, Ecology 2011c).

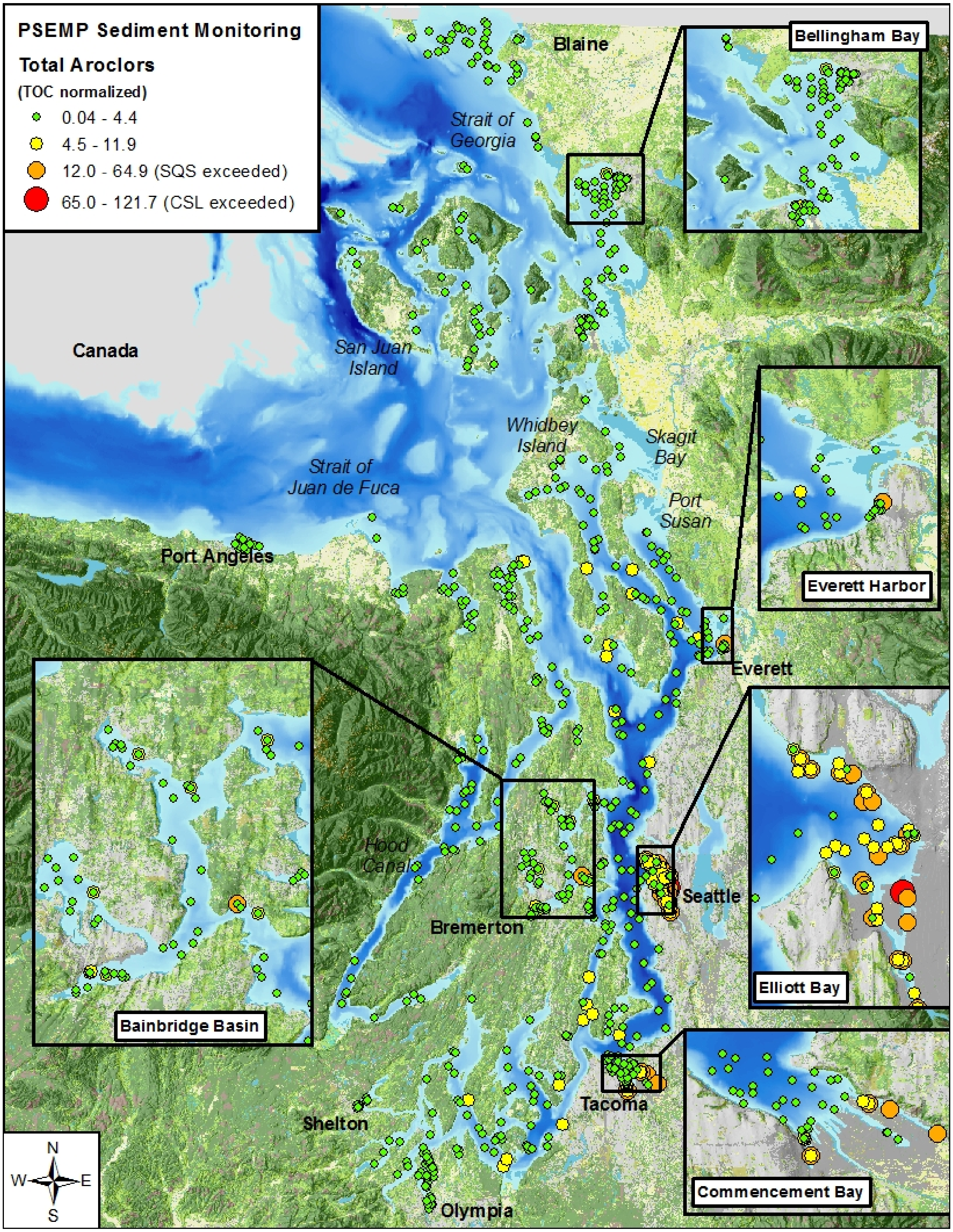
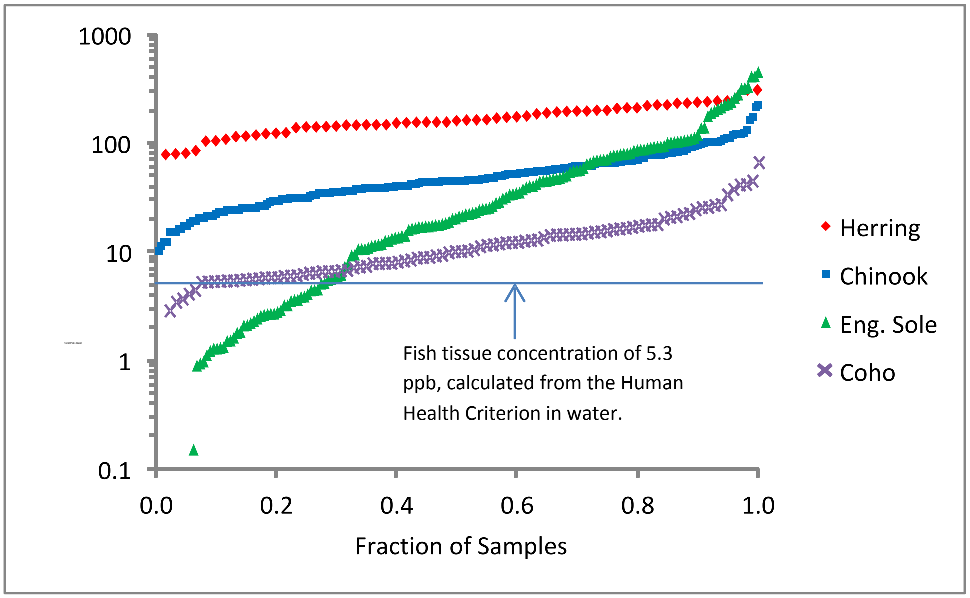


Figure 20. Distribution of PCBs in Marine Sediments from Puget Sound to Strait of Georgia (prepared by Sandra Weakland, Ecology Environmental Assessment Program; parts per billion, normalized to total organic carbon).

Fish

WDFW has monitored PCB levels in Puget Sound marine fish since 1992. Figure 21 summarizes the total PCB concentrations measured in edible tissues of four important marine and anadromous species. The highest concentrations have been observed in herring (whole fish), followed by Chinook salmon, English sole, and coho salmon, in that order. Median and 90th percentile concentrations for these species are 159/234 ppb (herring), 44/95 ppb (Chinook), 23/135 ppb (sole), and 10/26 ppb (coho). The fish tissue equivalent of Washington's human health water quality criterion for PCBs is 5.3 ppb. For more information on exposure to people from fish and fish advisories, see the section on Human Health. All samples of herring and Chinook, and most (70 – 80%) of the English sole and coho have exceeded the criterion.



Fish tissue concentration of 5.3 ppb, calculated from the Human Health Criterion in water.

Figure 21. PCBs in Edible Tissues of Four Species of Puget Sound Fish (1992-2010 data provided by James West, WDFW; N =60 – 210)

Puget Sound herring are 3 to 9 times more contaminated with PCBs compared to Strait of Georgia herring (West *et al*. 2008). The high concentrations in this pelagic species suggest continued input of PCBs to the water column, rather than direct uptake from contaminated sediments (O'Neill *et al*. 2011). WDFW's herring studies show Puget Sound is a regional hot spot for PCBs in the food web on the Pacific coast (Figure 22). Within Puget Sound there is a gradient of decreasing PCB levels in plankton and several pelagic fish species moving away from urban areas (West *et al*. 2011).

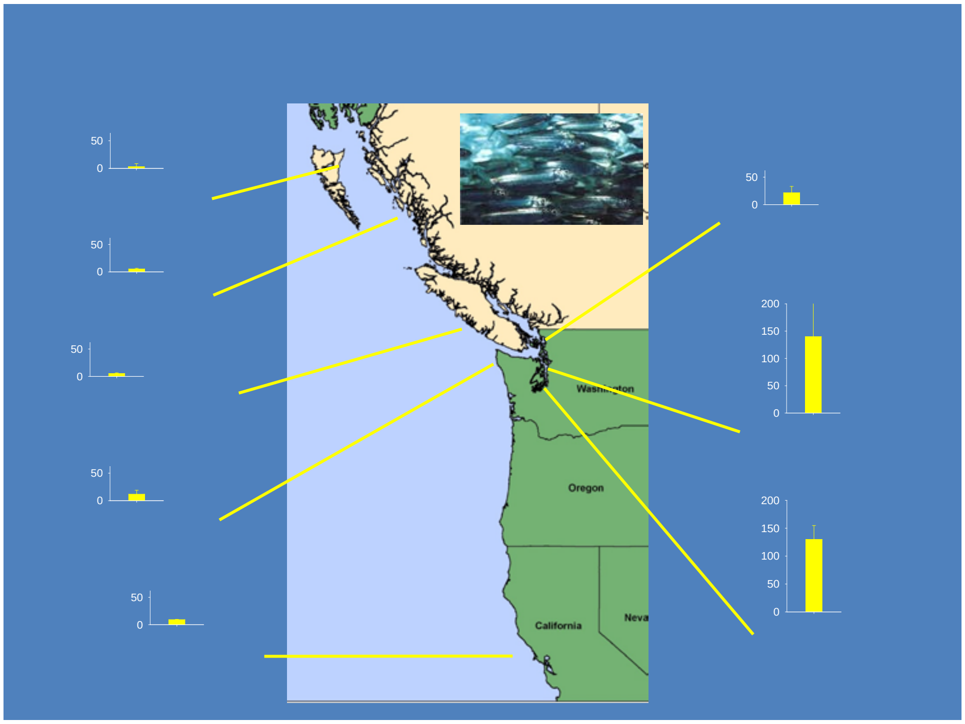


Figure 22. PCB Spatial Patterns in Herring: British Columbia, Washington, and California (prepared by James West, WDFW, and Sandie O’Neill, NOAA-NWFSC)

A high percentage of Chinook are resident in Puget Sound, with 29% of sub-yearlings and 45% of yearling out-migrants displaying resident behavior (O’Neill and West, 2009). Puget Sound Chinook are 3 to 5 times more contaminated than coastal Chinook (West 2011). According to O’Neill *et al*. (2011), 23-100% of juvenile Chinook from Puget Sound urban bays and 19% of returning adult Chinook have PCB levels above effects thresholds. The lower levels in coho are more reflective of combined oceanic and Puget Sound conditions.

English sole show a strong north-south gradient in PCB concentrations, increasing from the Strait of Georgia into Puget Sound (Figure 23). Sole from urban bays, especially the Duwamish River estuary, have much higher PCB levels than fish from non-urban locations. The degree of contamination in Puget Sound sole is positively correlated with PCB levels in the sediments (West 2011). Adverse effects on reproduction, growth, and immune response in English sole and other fish species have been attributed to the elevated levels of PCBs and other legacy contaminants in Puget Sound embayments (Collier 2009).

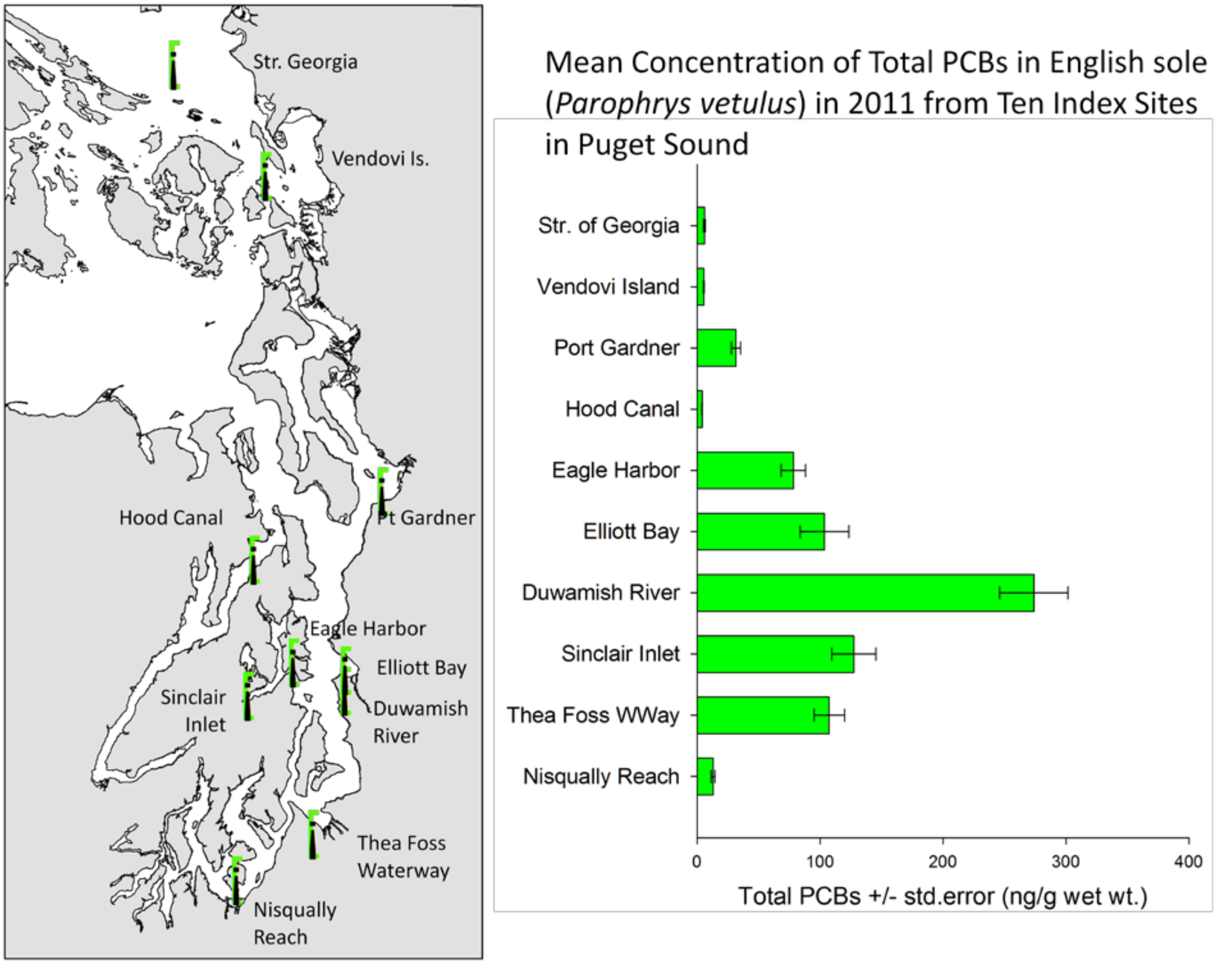


Figure 23. PCB Spatial Patterns in English Sole Muscle: Puget Sound and Vicinity (prepared by James West, WDFW; parts per billion).

A Pacific coast survey by the National Marine Fisheries Service showed outmigrating juvenile Chinook salmon typically have 2 to 5 times higher concentrations of PCBs and other contaminants compared to outmigrating coho (Ecology 2007). Of the 12 estuaries sampled, PCBs were highest in Chinook from the Duwamish River and Columbia River. The average PCB content in juvenile Chinook from these areas was near or above effects thresholds for growth and survival. For the Columbia River, the tidal freshwater portion of the estuary between Portland and Longview appears to be an important source of contamination. PCB levels were higher in fish that feed and rear in the lower river as opposed to those that migrate more rapidly through the estuary (Arkoosh *et al*. 2011, Johnson *et al*. 2013).

Figure 24 summarizes data obtained since 1997 on total PCBs in muscle tissue of resident freshwater fish collected throughout Washington, drawn from studies by EPA, Ecology, and others (CH2MHill 2007, Delistraty 2013, EPA 2002, Ecology 2004, Johnson *et al*. 2007, Ecology Washington State Toxics Monitoring Program, e.g., Seiders *et al*. 2012). The statewide natural background for PCBs in edible fish tissue has been estimated at 6.5 ppb (90th percentile;

Johnson *et al*. 2010). Large areas of the state have relatively low PCB levels (<20 ppb) not greatly above background. However, elevated to high concentrations are also commonly encountered, especially in the Columbia River, some of its major tributaries, and the Seattle urban area. In approximate decreasing order, the following ten rivers and lakes are reported to have the highest total PCB concentrations in fish muscle samples (1,100 to 60 ppb, site average):

- Wenatchee River
- Lake Washington
- Middle Columbia River
- Green Lake (Seattle)
- Spokane River
- South Fork Palouse River
- Upper Columbia River
- Walla Walla River
- Lower Columbia River
- Snake River

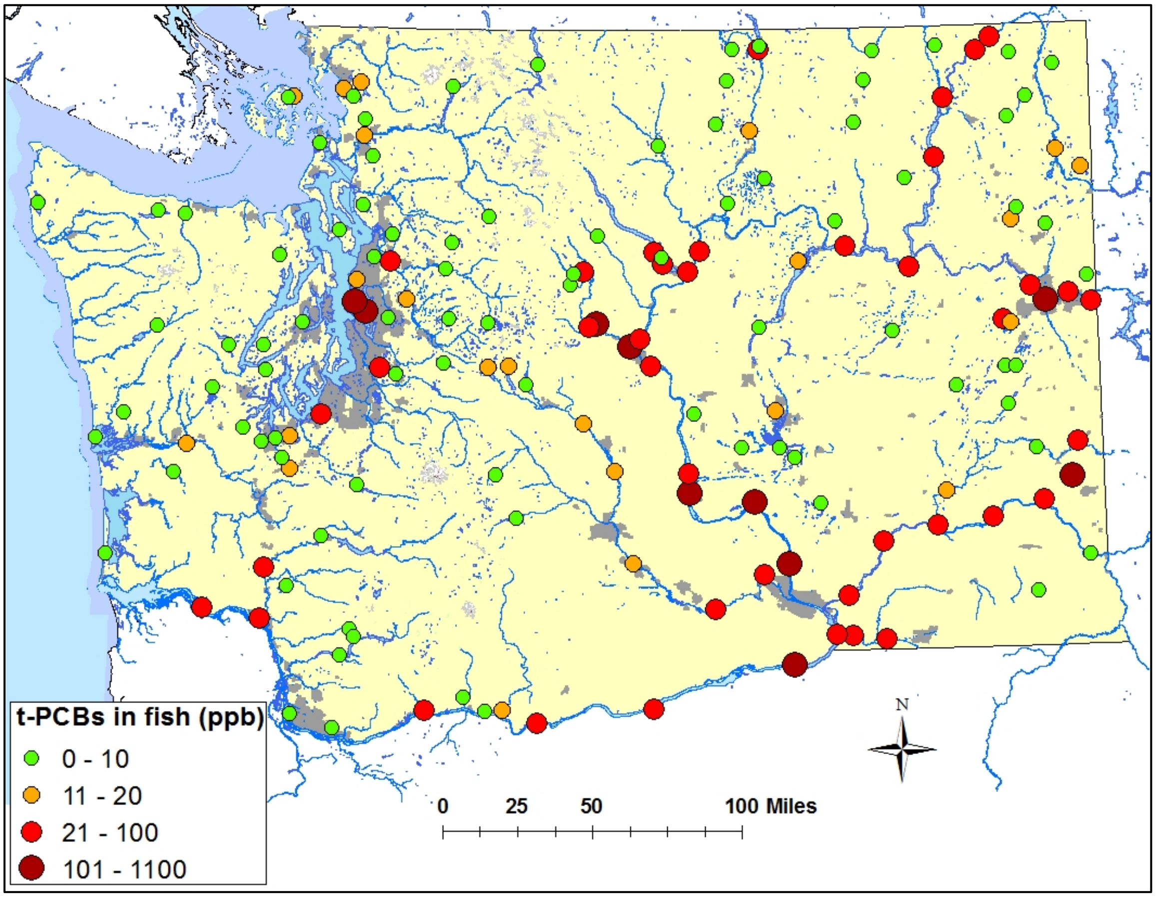


Figure 24. Average PCB Concentrations in Fish Muscle Samples from Washington Rivers and Lakes (prepared by Keith Seiders, Ecology Environmental Assessment Program; 1997-2010 data, N = 587).

Figure 27 in the section on Human Health shows statewide data on PCB levels in fish tissues (fillet). There is a line at 5.3 ppb, which is equivalent to the human health water quality criterion, and a line at 23 ppm, which DOH uses as a screening level for advisories. About 60% of samples exceed the 5.3 ppb level calculated from the water quality criterion. The median concentration statewide for total PCBs in freshwater fish is approximately 10 ppb. Ninety percent of samples are less than 85 ppb.

High mountain lakes experience enhanced atmospheric deposition of PCBs due to colder temperatures and greater amounts of precipitation (Blais *et al*. 1998, Gillian and Wania 2005). USGS analyzed PCBs in fish from 14 pristine Washington lakes over 3,000 ft. elevation (Moran *et al*. 2007). Although a relatively insensitive analytical method was used, total PCB concentrations of 17 – 20 ppb were found in approximately 20% of the tissue samples.

Mammals and Birds

Southern Resident killer whales are among the world's most PCB-contaminated marine mammals, which has been implicated in a range of negative health effects (Alva *et al*. 2012). It is estimated that Southern Residents have 4 to 7 times the daily PCB intake compared to Northern Residents (Cullon *et al*. 2009). Salmon, especially Chinook, comprise most of their diet and are thus the major source of contamination (Cullon *et al*. 2009). Transient killer whales have higher levels of PCBs compared to either resident population (Ross 2006) and are in a higher trophic level, eating marine mammals (Ross *et al* 2000). PCBs have been seen to cause reproductive impairment, immunotoxicity, skeletal abnormalities, endocrine disruption, and negative effects on population growth rate in marine mammals (Alva *et al*. 2012). While there are no established health effects thresholds for PCBs in killer whales, the levels in Puget Sound killer whales is above the health effects threshold for harbor seals that is based on immune system and endrocrine endpoints and predicted to remain high for decades, mostly driven by the long half lives of PCBs (Hickey *et al*. 2007).

Total PCBs in blubber biopsy samples collected in 1993-1997 averaged 146 ppm (lipid weight) in Southern Resident males vs. 37 ppm in Northern Resident males and 251 ppm in Transient males (Ross *et al*. 2000). Southern resident males sampled in 2004/2006 averaged 62 ppm (Krahn *et al*. 2007). Females off-load PCBs during calving and lactation, resulting in lower concentrations than males, by about a factor of three in the 1993-1997 study. PCB levels rise quickly in nursing calves, then the levels fall as the PCBs are diluted with growth, with levels in males increasing with age and females increasing until the onset of reproduction (Hickey *et al*. 2007).

A north-south gradient in increasing PCB levels has also been observed in harbor seals, which are non-migratory. Ross *et al*. (2004) reported average total PCB concentrations in adult seal blubber of 1.1 ppm in Queen Charlotte Strait, 2.5 ppm in the Strait of Georgia, and 18 ppm in Puget Sound. More recently, WDFW analyzed the blubber of 24 seal pups at four locations in Puget Sound (Ecology 2011i). Total PCBs ranged from 1.0 to 9.4 ppm. Concentrations increased following the order Hood Canal < Whidbey Basin < South Sound < Main Basin (Figure 25). As with killer whales, indicators of adverse health impacts have been observed in harbor seals and linked to PCBs or other persistent organic pollutants (Cullon *et al*. 2012, Ecology 2011i, Mos *et al*. 2010).

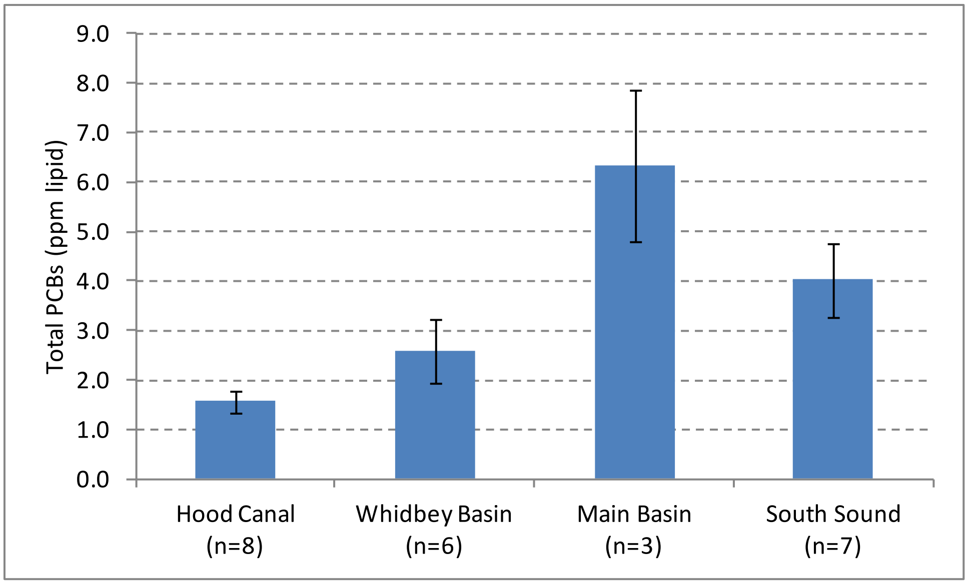


Figure 25. PCBs in Blubber of Puget Sound Seal Pups, 2009 (from Ecology 2011i; mean and standard error)

USGS and USFWS have analyzed PCBs in lower Columbia River wildlife. Their most recent data (1990-2004) show average wet weight concentrations of 0.2 ppm in mink livers, 0.5 ppm in otter livers, 0.8 ppm in osprey eggs, and 5.4 ppm in eagle eggs (Henny *et al*. 1996, Grove *et al*. 2007, Henny *et al*. 2007, Buck *et al*. 2005).

Another USGS study reported mean total PCB concentrations of 0.8 ppm in osprey eggs collected from the lower Duwamish River in 2006-07, but noted these birds had been feeding largely on hatchery raised salmon smolts (Johnson *et al*. 2009). The significance of hematological and biochemical differences observed between eggs from the Lower Duwamish and a reference site (upper Willamette River) was unclear. The same study reported 2.6 ppm, 1.9 ppm, and 0.7 ppm total PCBs, respectively, in osprey eggs collected from Lake Washington, the Lower Duwamish River, and Snohomish River estuary in 2002-2003.

Cleanup Sites

There are 336 hazardous waste sites listed in Ecology’s Integrated Site Information System (ISIS) as having confirmed or suspected PCBs. Many sites had multiple media with confirmed or suspected PCB contamination. Of the sites with confirmed or suspected PCBs in sediments, all but 15 also had soil with confirmed or suspected PCB contamination.

Table 26. PCB clean up sites

Medium	# of Sites	Confirmed	Suspected	Remediated	Below CUL
Soil	295	165	99	11	20
Groundwater	173	60	109	2	2
Sediment	62	47	11	1	3
Surface Water	89	19	64	6	0
Air	18	3	14	0	1

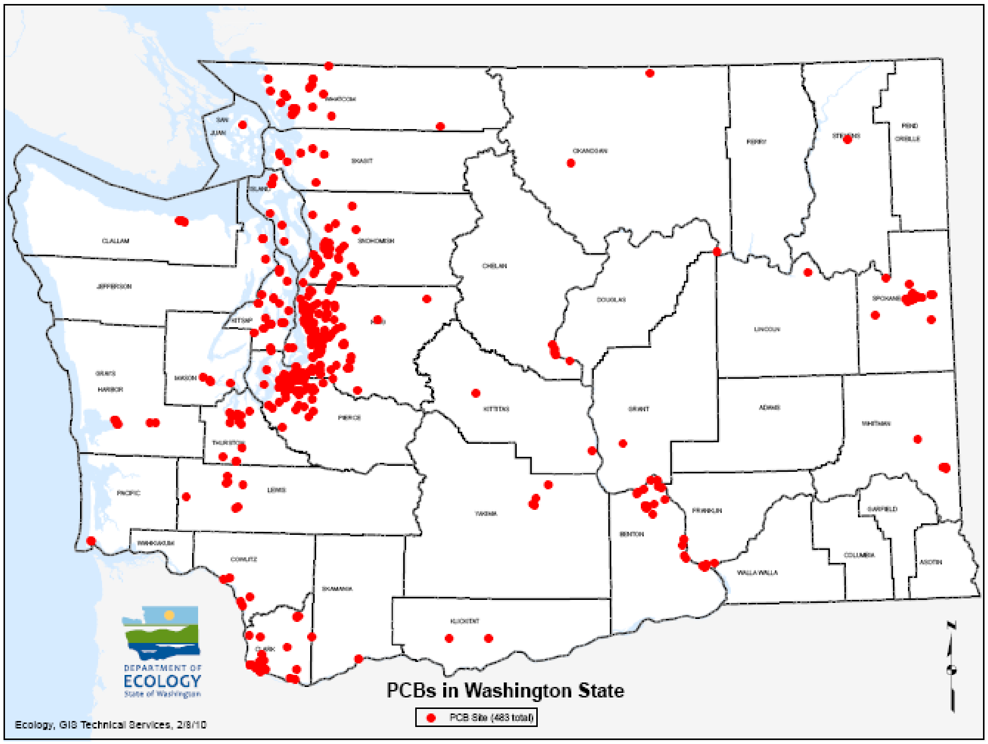


Figure 26. Figure of 483 PCB cleanup sites in Washington in 2010

Waste

Landfills

All Shred Residue

The material that remains after other parts are removed from automobiles at end of life for reuse or proper disposal is sometimes shredded and nearly all is used as landfill cover. PCBs may be present in automobiles in hydraulic fluids, plastics, voltage regulators, electric motors, switches, small capacitors, and light ballasts, leading to PCBs in the shredder waste. The all shred residue (ASR) is not just from end of life vehicles, but also includes consumer goods and commercial scrap. Feedstock sources may come from in or out of state. This material is difficult to sample, due to its heterogeneity in waste streams, metal extraction processes, and changes in manufacturing materials.

PCB concentrations in untreated shredder waste have been measured between 0.59 – 129 mg/kg and treated shredder waste contains 2.6 - 45.1 mg/kg (McKee *et al*.2006). McKee *et al*. further estimated that there is 270,000 tons of shredder waste from automobiles and appliances in California, leading to an estimate of 30-6,970 kg of PCBs (Average = 3,500 kg) in untreated shredded waste and 140-2,440 kg of PCBs (Average = 1,300 kg) of treated shredder waste in the Bay Area.

We have some information on PCBs in shredder residue in Washington in a 2013 report prepared for EPA Region 10 (Toeroek 2013). In this study Ecology and EPA sampled four facilities for PCBs (using Method 8082) and other analytes. There were different sampling protocols to look at variation, and all included a 9.5 mm sieve. Based on observations during the sampling, end of life vehicles were only shredded during two of the four days sampling occurred and all samples included consumer goods and commercial scrap. PCB concentrations ranged from 0.4 - 59 ppm (mg/kg) for the Aroclors tested, with a mean of 19 ppm, which is similar to the other results mentioned above.

Across Washington, shredders generate over 500 tons of ASR daily (Full Circle Environmental and Ecology 2011) and landfills reported disposing of 230,157 short tons (208,983 metric tons) of ASR in 2012. Using the range of concentrations found of 0.4 - 59 and the average concentration found of 19 ppm, results in an estimated 80 to 12,000 kg (12 metric tons) with an average of 4,000 kg (4 metric tons) of PCBs in 208,983 metric tons of ASR disposed of in Washington landfills each year.

Electrical equipment

Under federal TSCA, non-leaking small capacitors are allowed to be disposed of as solid waste. However, Washington State applies its own regulations to PCB electrical equipment containing 2 ppm PCBs or more and to materials contaminated to 2 ppm PCBs or more by electrical equipment. Unless the waste meets specific exclusions under the Washington State PCB rule (WPCB), these items must be properly disposed of and reported to Ecology as WPCB waste by the generator in annual data source reports. The waste code is for proper disposal and does not include more information on concentrations.

From 2009 – 2012, 10,577 MT of PCB-contaminated waste were reported under WPCB (Table 27).

Table 27. PCB-contaminated waste reported under WPCB

Year	WPCB (MT)
2009	2,322
2010	2,616
2011	1,310
2012	4,310

PCB-ballasts are considered moderate risk waste (MRW) when they are disposed of by households (household hazardous waste) and small businesses (small quantity generator waste). The weights reported in Table 28 include ballasts and shipping containers and we have not corrected for that due to the unknown weights of the shipping containers. Estimating that each ballast weighs 1.6 kg, and contains 44g of PCBs the estimated amount of PCBs ranges from 340 to 560 kg each year. Most of these PCB-ballasts go to a hazardous waste landfill outside of Washington State.

Table 28. PCB-ballasts collected as MRW (2010-2012)

Year	PCB-ballasts MRW (lbs)	PCBs (kg)
2010	32,871	410
2011	44,996	560
2012	26,885	340

Biosolids

Biosolids fall under the Clean Water Act (CWA) and the 503 rule (40 CFR 503). There are no requirements to monitor PCBs in biosolids, nor a regulated level of PCBs in biosolids. EPA(1995) made a “policy decision to delete all organic pollutants from land application and surface disposal sections of the final Part 503 rule because these pollutants met one of the following criteria: (1) the pollutant has been banned or restricted for use in the United States or it is no longer manufactured for use in the United States; (2) the pollutant is not present in biosolids at significant frequencies of detection based on data gathered in the NSSS [the National Sewage Sludge Survey], or (3) the limit for a pollutant from the biosolids exposure assessment is not expected to be exceeded in biosolids that are used or disposed based on data from the NSSS.”

EPA continues to investigate pollutants in sewage sludge and uses risk analysis to ascertain which pollutants require monitoring and regulatory levels.

While neither federal nor state regulations require testing of biosolids for PCBs, some jurisdictions such as King County, have tested for PCBs in biosolids (King County 2007). In 2006 Aroclor 1248, 1254, and 1260 were detected and other Aroclors were not detected at the West Point Treatment Plant. Only Aroclor 1254 was detected at the South Treatment Plant. Historically PCB Aroclors have been detected in King County biosolids at the ppb level. We do not have enough information to estimate the amount of PCBs in biosolids in Washington.

PCBs have also been measured in a few samples of Canada Goose guano in the Lower Duwamish area. In four composite samples the total Aroclor PCBs ranged from 28-103 ppb with an average concentration of 58 ppb. This is generally indicative of the ubiquitous nature of PCBs in Washington. This is not enough information to estimate the amount of PCBs in goose guano in Washington.

Environmentally Significant PCBs

As a result of partitioning, transformation, and bioaccumulation, PCB mixtures in the environment do not resemble the commercial products. About half of the 209 possible congeners account for most of the environmental contamination. Based on toxicity, prevalence, and relative abundance, less than 40 congeners are most commonly found in the environment.. Twenty-five of these account for 50-75% of the total PCBs in biological tissues (McFarland *et al*. 1989).

In Washington and elsewhere, PCBs with three to six chlorines (tri- through hexachlorobiphenyls) are the dominant compounds found in environmental samples. Less chlorinated compounds are more volatile and more readily metabolized and eliminated from organisms. Highly chlorinated compounds are relatively less abundant, more tightly bound to sediment particles, and taken up poorly by fish and other aquatic animals.

Twelve PCBs have a co-planar configuration that imparts dioxin-like toxicity (Tables 20 and 25). These compounds are frequently detected in Washington's environment. PCB-118 is the co-planar most often encountered in the highest concentrations (Cleverly *et al*. 1996, Ecology 2011i most toxic co-planar, PCB-126, is typically present at the lowest concentrations.

Although their toxicity is low relative to dioxin - a tenth or less – these PCBs can impart a significant fraction of the total dioxin toxicity equivalents (TEQ), particularly in higher animals. Co-planar PCBs accounted for up to 89% of the total TEQ in harbor seal pups collected from Puget Sound in 2009 (Ecology 2011i). PCB-118, -105, and -156, in that order, were the major contributors to the TEQ. Cullon *et al*. (2009) reports that PCBs explained the majority of the TEQ in adult salmon from British Columbia and Puget Sound waters. In contrast, the PCB TEQ in Washington freshwater fish is typically much lower than the dioxin TEQ, by an order of magnitude or more in most cases (CH2MHill 2007, USDOE 2010, Ecology 2010c).

PCB-11, a non-legacy PCB, has been identified in Columbia River water and clams (Ecology 2005, McCarthy 2007). In some samples, PCB-11 was either the most or second-most abundant congener detected. PCB-11 is neither associated with historical commercial PCB products nor a breakdown product of the commercial mixtures. The source has been traced to pigments currently used in paint (Hu and Hornbuckle, 2010). Reports of PCB-11 in environmental samples have become widespread in North America (Litton 2006, Grossman 2013). PCB-11 is considered a significant source of contamination to air, soil, and water (Hornbuckle and Robertson 2010).

Model Predictions

Ecology modeled the long-term fate and bioaccumulation of PCBs in Puget Sound (Ecology 2009b). Pertinent findings from Ecology's box model include the following:

- Approximately 97% of the total mass of PCBs in the aquatic ecosystem of Puget Sound is in the active sediment layer (top 10 cm), <1% is in the water column, and <3% is in the biota.
- Decreases in PCBs in sediment and biota are possible by the year 2020 in the urban bays due to burial and transport of sediments**.**
- Increases in PCBs in sediment and biota are possible by the year 2020 in the larger basins.
- Considering the wide range of uncertainty in loading from outside of the Puget Sound basin, it is possible the mass of PCBs in the aquatic ecosystem of Puget Sound may either increase or decrease over time at the current loading levels.

Ecology is currently re-evaluating this model using more recent data.

A delayed response to environmental declines in PCB levels is predicted for long-lived species like killer whales (~50-year lifespan). According to an individual-based model, Southern Resident killer whales may not fall below PCB effects thresholds for several more generations (Mongillo *et al*. 2012, Hickie *et al*. 2007).

Food web models for Lake Washington and the Spokane River conclude that PCB residues in fish are driven mainly by levels in the sediments in Lake Washington and are an important factor in the Spokane River (Ecology. 2010a and 2011c). Thus, recovery in these water bodies may respond slowly to reductions in external PCB loading.

Results of a mass balance model for the Willamette River basin in Oregon demonstrated that the PCB levels observed in fish could be due entirely to atmospheric deposition from global legacy sources (Hope 2008). In this analysis, PCB sources within the Willamette basin were assumed to be insignificant. Portland was discounted because it occupies a short segment of the lower river isolated by Willamette Falls.

Local Hazard Assessments

Spokane River

The Spokane River has elevated levels of PCBs in Washington. Ecology calculated hazard quotients (HQs) for different endpoints and receptors in six different parts of the river for aquatic life and fish-eating wildlife (Ecology 2001). HQs are ratios of the level of PCBs in the environment and the level at which no adverse effects are expected. If the HQ is greater than 1, then adverse health effects are possible, but do not necessarily occur.

The primary ecological hazards identified were:

1. Possible adverse effects on the sustainability of salmonid populations and fish-eating mammals, primarily in the reach between Trentwood and Nine-Mile Dam.

PCB levels in salmonid fish tissue were high compared to the National Marine Fisheries Service (NMFS) effects threshold for sublethal effects (HQs of 2.2-4.1).

The levels of PCBs in water were high compared to the Great Lakes criterion for reproductive effects on fish-eating mammals (HQs of 4.5-17). Also the PCB levels in fish tissue were high when compared to the British Columbia guideline for reproduction in fish eating wildlife (HQs of 2-6) and compared to the lowest observable effects levels (LOEL) for mink and otter reproduction (HQs of 1.5-2.4).

2. Possible adverse effects on benthic invertebrates in the Trentwood to Monroe Street Dam reach in areas where PCBs have been concentrated in fine-grained sediments, such as behind Upriver Dam. The levels of PCBs in sediment were high compared to the threshold effect for abundance and diversity of benthic invertebrates (HQs of 2.5-9.8)

Puget Sound

As part of the larger, multi-year, multi-agency, project to study toxic chemicals in Puget Sound, the Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011 (Ecology 2011a) included a hazard evaluation to estimate the relative hazard posed by the 17 chemicals of concern assessed in the Puget Sound studies. This was not a risk assessment, but part of the effort to prioritize efforts on pollutants in Puget Sound. The hazard evaluation was for the entire Sound, not for hot spots where there are higher levels of contamination, such as in cleanup sites. PCBs were ranked as having the highest level of concern for a range of media and receptors in both the freshwater and marine aquatic environments.

PCBs were found to be at the highest level of concern (Priority 1) for the five categories assessed. The category and the reason PCBs were placed in Priority one is given below for each.

- Aquatic life through surface water exposure. The 90th percentile of observed concentrations of Aroclors was above the chronic Water Quality Standard (WQS) for aquatic species in freshwater.
- Benthic organisms through sediment exposure. The 90th percentile of the observed concentrations exceeds the Sediment Quality Standard for Aroclors for freshwater (FP-SQS) and offshore marine sediment (SQS).
- Tissue Residue Effects. The 90th percentile of observed concentrations was above the 10th percentile of the effects concentration for non-decapod invertebrates for both Aroclors and total congeners.
- Wildlife through seafood consumption. Using Great Blue Heron, Osprey, River Otter, and Harbor Seal as representative species, the daily dose was more than the lowest effect dose divided by10.
- Human health through seafood consumption. The 90th percentile of observed tissue concentrations was above National Toxics Rule (NTR) water quality criteria for freshwater, nearshore marine, and offshore marine for both Aroclors and congeners for bivalves, fish and invertebrates. Tissue criteria were back calculated from the NTR based water quality criteria.

Environmental data from January 2000 to July 2010 were collected from a variety of sources, and the largest source of data was Ecology’s Environmental Information Management (EIM) system. Information on effects levels were found in the scientific literature and regulatory benchmarks. Comparisons to effects levels, criteria, and guidelines were done separately for both PCB Aroclor and congener data. Box Plots showing these comparisons can be found in the Assessment report Appendices D3-D7 (https://fortress.wa.gov/ecy/publications/summarypages/1103055.html).

Human Health Effects of PCBs

Introduction

There is a very large body of toxicological and epidemiological research on the health effects of PCBs. Research conducted in experimental animals has shown that PCBs can cause a wide variety of adverse health effects including, immune suppression, adverse reproductive effects, abnormal motor and cognitive development, injury to the liver and other internal organs, changes in the endocrine system, and cancer. Epidemiological studies have found evidence of similar adverse effects on human development and behavior, reproduction, immune function, and cancer (ATSDR 2000). The International Agency for Research on Cancer (IARC) recently changed their classification of PCBs and dioxin-like PCBs from “probable human carcinogens” to “human carcinogens” to recognize that there is now sufficient evidence in humans and animals. (Lauby-Secretan *et al*. 2013).

The primary historic uses of PCBs were banned in 1979 but existing electrical equipment and other closed systems containing PCBs were permitted to remain in use (40 CFR part 761). Caulk, joint sealant, paint, and other building materials sold before 1979 may have contained PCBs and could still be in place. PCBs have continued to slowly escape from these historic sources with rapid releases during leaks, fires, and building demolition.

Historically, PCB oils and equipment containing those oils have been disposed in ways that allowed PCB release into the environment. A number of waste sites in Washington have been identified as contaminated with PCBs. Many have been cleaned up but additional clean-up remains. Because many PCBs are persistent and bioaccumulative, these legacy sources of PCBs continue to cycle through air, water, soil, sediments, and biota. Although general environmental levels have dropped dramatically since 1979, human exposure to legacy PCBs in fish, other foods, and air will continue into the future.

In addition, PCBs are inadvertently formed during current production of certain pigments and ink (Hu and Hornbuckle 2010). These PCBs have been detected in colored papers, cardboard, plastics, and textiles and may be released to the environment during manufacturing, use, disposal, or recycling of consumer products (Litten *et al*. 2002, Rodenburg *et al*. 2010). PCB 11 has emerged as a useful indicator of these new sources of exposure since commercial PCBs mixtures did not contain more than trace amounts of PCB 11 (Grossman 2013).

There are 209 possible configurations of chlorine substitutions of the biphenyl molecule. Each possible configuration (called a congener) is assigned a number and most can now be quantified analytically. Because congeners vary in their toxicity and their resistance to metabolism, risk assessment of PCB mixtures is complex. One of the earliest discoveries in the search for common mechanisms of toxicity was that certain congeners have dioxin-like ability to bind to the aryl hydrocarbon (Ah) receptor. Dioxin-like PCBs are planar or nearly planar in structure (Van den Berg *et al*. 1998). Their interaction with the Ah receptor is dose-dependent, saturable, and induces hepatic enzymes, including aryl hydrogen hydroxylase (also called cytochrome p450 CYP1A1) and 7-ethoxyresorfin *O*-deethylase (CYP4501A2) (Seegal 1996). These enzymes metabolize (or breakdown) a number of environmental chemicals but also act on important endogenous chemicals such as hormones, retinoids, and neurotransmitters (ATSDR 2000).

Several toxic responses have been well correlated with the Ah-receptor binding affinity, including body weight reduction, hepatotoxicity, and thymic atropy (Seegal 1996). Laboratory animals exposed solely to coplanar PCBs have shown reproductive problems, increases in brain levels of biogenic amines including dopamine, alterations in fetal and neonatal plasma thyroid hormone levels, and neurobehavioral effects (Brouwer *et al*. 1995). Available evidence in lab animals suggests that the maturation of the immune system is especially vulnerable to adverse effects of dioxin-like compounds (Holladay and Smialowicz 2000). Toxic equivalency factors (TEF) for 12 PCB congeners have been developed to facilitate human health risk assessment of mixtures of dioxin-like chemicals (Van den Berg *et al*. 1998, 2005, see Table 24 and Table 29).

EPA and ATSDR have established health recommendations concerning oral intake and inhalation of PCBs. These screening levels help public health agencies and communities identify exposures of concern. The three primary pathways of current PCB exposure for the U.S. general public are diet, indoor air, and ambient air. In addition, ingestion of PCB residues in house dust could be a significant contributor to exposures in toddlers (ATSDR 2000, Harrad *et al*. 2009). Although background exposures in the U.S. population appear to have dropped below levels of concern, special populations at higher risk for exposure exist and require attention. These include people who work around PCB contaminated equipment or materials, who consume fish and seafood from contaminated waters, or who live or work in a building with PCBs in building materials or fluorescent light ballasts.

Historical Episodes of PCB Poisoning

Acute poisoning to PCBs has been documented in people following accidental food contamination and workplace accidents. These incidents underscore why people must be protected from direct contact with concentrated PCB liquids or materials during source removal, transport, and disposal. There have been two episodes of mass human poisoning by inadvertent community-wide consumption of PCB contaminated rice oil. One occurred in Japan in 1968 and the other in Taiwan in 1978-79. Affected people in Japan were diagnosed with “Yusho” which means “the oil disease” and in Taiwan with “Yu-Cheng,” the term for “oil disease” in Taiwanese. About 1700 adults were acutely affected in the Japanese incident. Exposure of adults resulted in increased skin pigmentation, severe acneform eruptions, swelling of the meibomian gland with eye discharge, thickening of the nail bed, numbness in extremities, and respiratory disease (Urabe and Asahi 1985, Ikeda 1996, Nakanishi *et al*.1985). Affected women who were pregnant at the time gave birth to children with physical abnormalities (dental disorders, hyperpigmentation of skin) and severe neurodevelopmental problems (Seegal 1996). Exposure monitoring for PCBs in blood of Yusho patients did not begin until five years after the onset of disease.

About 2000 persons were initially affected with illness in the Taiwan incident. Symptoms included hyperpigmentation, acneform eruptions, swelling of eyelids and increased discharge from the eyes, as well as systemic complaints (Lu and Wu 1985). Women who were pregnant at the time of exposure gave birth to children who showed hyperpigmentation, nail deformities, conjunctival discharge and swelling. Eight of the 39 infants born with hyperpigmentation during the four years following the incident died (Hsu *et al*. 1985). A cohort of most of the Taiwanese children (*n* =118) born to affected mothers up to six years past the incident has been followed and tested annually for cognitive deficits. A comparison population with the same number of children was matched on maternal age, child's birthdate, gender, and neighborhood of residence. Blood PCB levels of Yu Cheng patients were measured early in the disease outbreak and were high (44.4% of 613 patients sampled had blood levels of PCBs between 51-100 ppb). The highest value reported was 1156 ppb (Hsu *et al*. 1985). Yu-Cheng offspring have been shown to have persistent cognitive deficits, lower IQ, and higher rates of problem behaviors compared to neighborhood controls (Lai *et al*. 2002).

Immune effects were also reported in both groups of poisoned people including an increase in respiratory and skin infections and changes in immune parameters such as immunoglobulins and T cells (Lu and Wu, 1985). Studies also detected depressed responses to tuberculin tests (Lu and Wa 1985, Nakanishi *et al.* 1985). Infants born to mothers who had Yu-Cheng disease had more episodes of bronchitis or pneumonia during their first six months of life and had higher frequencies of ear infection and respiratory tract infection in a six-year follow-up (Yu *et al*. 1998).

There is debate about the degree to which these two mass poisonings are relevant to current assessments of PCB exposure (Schantz 1996, Seegal 1996). The levels of PCB intake were very high compared to environmental sources. PCBs in both incidents had been repeatedly heated to high temperatures in a heat exchanger before contamination of rice oil occurred. The high temperatures changed the chemical composition of the oil creating high concentrations of polychlorinated dibenzofurans (PCDF) and other toxic compounds. Many of the developmental and physical effects seen in these populations, however, are also observed in monkeys dosed with pure PCB mixtures.

Developmental abnormalities have also been observed in occupationally exposed populations. A seven-year follow-up study of capacitor manufacturing workers in Japan and their children evaluated effects on children born to mothers who had PCB blood and breast-milk levels that were 10-100 times the normal background and markedly higher than the blood of Yusho patients. Forty children were examined once a year for five years and none were diagnosed with PCB poisoning. Some of the children were found to have decay of nails, gingival pigmentation, mottled enamel, and dental caries that were typical symptoms in Yusho but were less serious in this study population (Hara 1985).

Endpoints of Human Health Concern

Cancer

There is clear evidence that commercial PCB mixtures cause cancer in animals in a dose-dependent manner (EPA 1996b, ATSDR 2000). Cancers observed primarily involve thyroid and liver tissue. Studies of workers exposed to commercial PCB mixtures found increases in liver and bile-duct cancers and malignant melanoma across multiple human studies (NTP 2011, Lauby-Secretan *et al*. 2013).

EPA and NTP consider PCBs to be probable human carcinogens and recently International Agency for Research on Cancer (IARC) strengthened its classification of PCBs to “human carcinogens” based on new evidence of melanoma in epidemiological studies (Lauby-Secretan *et al*. 2013). EPA concluded that the types of PCBs likely to be bioaccumulated in fish and bound to sediments are likely to be the most carcinogenic PCB mixtures (EPA 1996b).

PCB mixtures and individual congeners can act as tumor promoters (ATSDR 2000, WHO 2003, EPA 1996b). Tumor promotional activity has been observed by congeners that are aryl hydrocarbon agonists (dioxin-like congeners), that induce cytochrome P450 1A and 2B isozymes, and induce P450 CYP2 and CYP3 families of enzymes and have a phenobarbital pattern of enzyme induction (ortho-substituted congeners). Oxidative stress and disruption of intercellular communication have also been proposed as mechanisms for cancer promotion (EPA 1996b, WHO 2003).

Certain PCB congeners have been shown to be direct tumor inducers as well. In a series of recent investigations, the National Toxicology Program (NTP) demonstrated that dioxin-like congeners, PCB 118 and PCB 126, were able to induce lung, liver, bile duct, oral, and uterine cancers in rodents (NTP 2006). Certain non-dioxin-like congers that are likely to be in air, such as PCB 3, also appear to undergo metabolic activation in rodents to reactive species that are genotoxic and can initiate DNA and chromosomal damage (Xie *et al.* 2010; Ludewig and Robertson, 2013). Few studies of genotoxicty in humans have been conducted but these have been negative for environmental PCB exposures (Ludewig and Robertson, 2013).

Immune Effects

Numerous immune effects have been measured in laboratory animals exposed to PCBs (ATSDR 2000). Changes in the immune system were selected by both ATSDR and EPA as the most sensitive non-cancer endpoint for chronic exposure to PCBs. In the critical study chosen by these agencies (Tryphonas *et al*. 1989, 1991a, 1991b), PCB-treated rhesus monkeys had a dose-related reduction in antibody response to an injected antigen (sheep red blood cells). The diminished production of immunoglobulins IgM and IgG, in response to antigen was statistically-significant at two time points during the study (27 months and 55 months) at the lowest dose of Aroclor 1254 tested (5 µg/kg/d). No differences in baseline serum concentrations of IgG, IgM, or IgA were evident. Both EPA's Reference dose for chronic exposure and ATSDR's minimal risk level were derived from findings of this study at the 5 ug/kg/d dose level (EPA 1994, ATSDR 2000). Body burdens after 25 months of dosing at 5 µg/kg/d were reported to be 10.3 ppb in blood (8.4 ppm in blood when expressed on a lipid basis) and 2.2 ppm in adipose tissue (7.5 ppm in adipose when expressed on a lipid basis) (Arnold *et al*. 1993a, 1993b).

The experiment above continued into a breeding phase to measure reproductive and developmental outcomes in the rhesus monkeys. Female monkeys treated for 37 months with Aroclor 1254 were bred to unexposed males. Among other things, offspring were tested for immunological function. A reduction in IgM titres to sheep red blood cells were statistically significant for the 5 µg/kg/d dose level at multiple post-natal time points (Arnold *et al*. 1999).

Statistically significant but clinically mild developmental abnormalities were observed in offspring at the 5 µg/kg/d dose level including nail and nail bed changes, and inflammation and/or enlargement of the tarsal glands. There were also adverse reproductive effects that were elevated but did not reach statistical significance at the 5 µg dose (Arnold *et al*. 1999).

A number of epidemiological studies have reported immune effects associated with human environmental exposures to PCBs. Inuit children with high exposure to PCBs and other organochlorines, were reported to have higher incidence of ear infection and lower (but not upper) respiratory tract infection during the first five years of life (Dallaire *et al*. 2006). Early life exposures to PCBs have also been associated with reduced response to childhood vaccinations in European children (Weisglas-Kuperus *et al*. 2000, Heilman *et al*. 2006) and reduced size of the thymus (Park *et al*. 2008).

Neurological and Neurodevelopmental Effects

Extensive animal research on neurologic impacts of PCB shows adverse effects in adults and the young with fetal and early postnatal periods being the most sensitive in producing adverse effects in rodents and monkeys (Brouwer *et al*. 1995). Hyperactivity and learning and memory impairments are very sensitive to developmental PCB exposure in non-human primates (Bowman *et al*. 1978, Schantz *et al*. 1991). ATSDR based their health advice for oral PCB exposure over intermediate durations on learning and memory impairments observed after postnatal exposure to a PCB in male rhesus monkeys (ATSDR 2000). In this study, Rice *et al*. created a congener mixture that represented 80% of the congeners present in breast milk in Canadian women and administered it for 20 weeks post-natally at a dose estimated to be equivalent to nursing from a mother with 50 ppb PCB in breast milk (7.5 µg/kg/d). Behavioral tests were conducted 3-5 years later. Treated monkeys were slower to learn new responses, adapt to new response patterns, and inhibit a response that was previously rewarded (Rice 1999). This study also measured effects at the lowest and only dose tested.

A number of studies have reported developmental effects in children although PCB levels in cord blood at birth or in the mothers were significantly higher than current body burdens in most populations (Jurewicz *et al.* 2013). Some studies, such as the Michigan Maternal/Infant Cohort and the Oswego Newborn and Infant Development Project, compared pregnant women who consumed Great Lakes fish to mothers who did not. Other studies, such as the North Carolina Breast Milk and Formula Project and the Dutch PCB/Dioxin Study, focused on mothers in the general population. These studies reported a range of subtle neurobehavioral effects such as abnormal newborn reflexes, cognitive and memory deficits including decreased IQ and changes in physical activity that were associated primarily with *in utero* not lactational PCB exposure (Jacobson and Jacobson 1996, Stewart *et al*. 2008, Brouwer *et al*. 1995). Although some studies have measured effects of lactational exposure to PCBs via breast milk, breast feeding appears to have a net positive effect on children with regard to mental and physical development (Anderson *et al.* 1999, Boersma and Lanting 2000, Pan *et al*. 2009). Fish consumers had higher proportions of PCB congeners with 7-9 chlorines and, in one analysis, these congeners were shown to be more closely associated with the neurobehavioral effects observed (Stewart *et al*. 1999). In another study, maternal body burden of dioxin-like mono-ortho substituted congeners (PCBS 118 and 156) were most strongly associated with neurobehavioral outcomes in children (Park *et al*. 2010).

Several researchers have proposed potency schemes for neurotoxicity of PCB mixtures that include both dioxin-like and non-dioxin-like PCB congeners. One neurotoxicity equivalency scheme is based on congener potency in interfering with intracellular signaling pathways and calcium ion modulation in the nervous system (Simon *et al.* 2007). Another is based on congener potency in altering brain dopamine and blood thyroid hormone levels (Yang *et al.* 2009). Although these schemes are not sufficiently developed for risk assessment, they point to the need to expand beyond TEQ of dioxin-like congeners when assessing potential neurotoxicity of PCB mixtures.

Reproductive Effects, including Birth Defects

Reproductive effects of PCBs have been demonstrated in a variety of animal species including non-human primates (ATSDR 2000). Oral PCB exposures reduced birth weight, conception rates and live birth rates of monkeys exposed during preconception and gestation (Arnold *et al*. 1995). Similar results have been observed in rodents. Developmental exposures in rodents resulted in lasting changes in reproductive tissue that were measurable at puberty and into adulthood (WHO 2003).

Investigations of reproductive effects have also been carried out in human populations exposed to PCBs. Longer time-to-pregnancy for couples trying to get pregnant has been reported to correlate with higher levels of certain PCB congeners (Axmon *et al*. 2005, Buck Louis *et al*. 2013). Obvious growth retardation was observed in offspring following the Yusho and Yu-Cheng incidents. Children born to women who worked with PCBs in factories showed decreased birth weight and a significant decrease in gestational age with increasing exposures to PCBs (Taylor *et al*. 1989, Hara 1985). Studies in fishing populations believed to have high exposures to PCBs also suggest similar decreases in either birth weight or gestational age or both (Fein *et al*. 1984, Rylander *et al*. 1995). Govarts *et al*. 2012 reported that birth weight decreased with increasing cord serum concentration of PCB-153 after adjustment for potential confounders in 12 of 15 study populations in Europe. Several studies have observed persistent deficits in physical growth into childhood following prenatal exposure to PCBs, particularly in girls (Jacobson and Jacobson 1997, Lamb *et al*. 2006, Blanck *et al*. 2002). Other investigations did not detect lower weight or shorter gestation times in other populations, including fish consumers (Rogan *et al*. 1986, Patandin 1999, Dar *et al*. 1992, Buck *et al*. 2013, Cupul-Uicab *et al*. 2013).

Endocrine Effects

There has been significant discussion and research on the effects of PCBs on the endocrine (or hormone) system. A number of PCB congeners and their metabolites display weak estrogenic, antiestrogenic effects, or antiandrogenic effects (Goncharov *et al.* 2009, Hamers *et al*. 2011, Brouwer *et al*. 1999, Birnbaum 1994). Certain PCBs and their metabolites appear able to directly interfere with estrogen or androgen receptors and bind directly to the estrogen receptor while others may act indirectly by inducing enzymes that then change estrogen metabolism (Brouwer *et al*. 1999, Kester *et al*. 2000, Hamers 2011).

The strongest evidence is for disruption of thyroid hormone levels. Hydroxylated PCB metabolites are structurally similar to the thyroid hormone thyroxine (T4) and may interfere with hormone receptor binding directly. They have been shown to competitively displace binding of thyroid hormone (T_4) to transthyretin in rodents and to disrupt the normal delivery of thyroid hormone from maternal plasma to the rodent fetus *in vivo* (Porterfield 2000, Brouwer *et al*. 1999). In addition, PCBs and their metabolites may act indirectly by interfering with thyroid hormone metabolism. For example, activation of the Ah receptor by dioxin-like PCBs, induces production of the enzyme uridine diphosphoglucuronyl transferase that metabolizes T4 and may accelerate T4 clearance from the liver (Porterfield 2000, Koopman-Esseboom *et al.*1994). Thyroid hormones are essential for regulating metabolism and normal growth and brain development. They also promote normal cardiovascular, reproductive and nervous system functioning.

Dioxin-like PCBs have been associated with changes in thyroid hormone levels in infants (Koopman-Esseboom *et al*. 1994, Nagayama *et al*. 1998, Pluim *et al*. 1992). A more recent study by Wilhelm *et al*. 2008 looked for but did not find any decrease in thyroid hormones related to dioxin-like PCBs or total TEQ at current exposure levels in Germany. Chevrier *et al*. 2008 reported that blood concentration of thyroid stimulating hormone in newborns was associated with non-dioxin like PCB congeners (PCBs 99, 138, 153, 180, 187, 194, and 199). Similar results, higher TSH and lower free T4 levels with increasing PCB levels, were reported in adolescents, although only in the group that had not been breast-fed (Schell *et al*. 2008).

Established Health Guidelines for PCB Mixtures

EPA's approach to cancer risk assessment of PCB mixtures

EPA uses a tiered approach to cancer risk assessment for PCB exposure. EPA recognized that selective bioaccumulation in the environment creates dietary PCB mixtures that differ markedly from Aroclor mixtures produced commercially. The tiers attempt to address the likely differences in toxicity and persistence of PCB mixtures. The tiers also reflect that PCB mixtures comprised mostly of congeners with more than four chlorines are more persistent and more carcinogenic than more lightly-chlorinated, less persistent congeners.

EPA recommends that risk assessors use a cancer slope factor of 2.0 per mg/kg/d for PCB mixtures present in the food chain. This most potent assumption should also be applied to protect all early life exposures. A less potent assumption of 0.4 per mg/kg-d can be used in assessments of ingestion of water soluble congeners and inhalation of evaporated congeners. A third tier (cancer slope equal to 0.07 per mg/kg-day) is provided for assessment of exposure to PCB mixtures with less than one half percent congeners of four or more chlorines. (Cogliano 1998)

Dioxin-like congeners and Toxic Equivalency (TEQ) approach.

Mixtures of PCBs congeners that have dioxin-like toxic effects can be evaluated by their toxicity relative to the most toxic dioxin, 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD). Dioxins and dioxin-like compounds have a common mechanism of action mediated by the aryl hydrocarbon receptor (AhR) but differ in their potency. Their potency relative to TCDD is reflected in their Toxic Equivalent Factor (TEF) developed by the World Health Organization for 7 dioxins, 10 dibenzofurans, and 12 PCB congeners. TEFs for PCBS are shown in Table 29 (Van den Berg *et al*. 2006). The most potent PCB congener has a TEF =0.1 which means that it is 1/10 as toxic as TCDD. The TCDD Toxicity Equivalent (TEQ) of a mixture can be calculated by summing the individual congener concentration and their TEFs as shown in the equation below.

$$TEQ = \Sigma\, (TEF_i \cdot C_i)$$

Where:

TEQ = TCDD toxicity equivalance

TEF_i = Toxicity equivalency factor for an individual congener

C_i = Concentration of individual congener

Table 29. Toxicity Equivalence Factors for PCB Congeners

Class	Congener	Mammal Toxicity Equivalence Factor (TEF)
Co-planar PCBs	3,3',4,4'-TCB (77)	0.0001
	3,4,4',5-TCB (81)	0.0003
	3,3',4,4'-5-PeCB (126)	0.1
	3,3',4,4',5,5'-HxCB (169)	0.03
Mono-ortho PCBs	2,3,3',4,4'-PeCB (105)	0.00003
	2,3,4,4',5-PeCB (114)	0.00003
	2,3',4,4',5-PeCB (118)	0.00003
	2',3,4,4',5-PeCB (123)	0.00003
	2,3,3',4,4',5-HxCB (156)	0.00003
	2,3,3',4,4',5'-HxCB (157)	0.00003
	2,3',4,4',5,5'-HxCB (167)	0.00003
	2,3,3',4,4',5,5'-HpCB (189)	0.00003

Source: TEFs recommended by World Health Organization (Van den Berg et al. 2006).

The EPA recently set the daily level of exposure considered safe for humans over a lifetime (also known as the Reference Dose or the RfD) at 0.7 picograms per kilogram of body weight of TCDD or toxicity equivalent as calculated by the equation above (EPA 2012d). This number is believed to protect against the most sensitive non-cancer endpoints observed. It is based on observations of health effects at 20 pg/kg/d in two studies conducted after an industrial accident in Seveso, Italy. The follow-up studies detected reduced sperm counts in men exposed in childhood and increased thyroid hormones in infants of mothers exposed during pregnancy (Mocarelli *et al*. 2008, Baccarelli *et al*. 2008). An earlier health guideline for acceptable daily intake of dioxin-like compounds, set by the World Health Organization/United Nations in 2001, was based on older data. The newer EPA RfD is three times lower (see Table 30). EPA is currently developing its health guidance for dioxin cancer assessment.

Other Health Guidance

There are a number of health standards that address exposures to PCBs in people. For assessment of dietary intake, the Washington State Department of Health (Health) uses the EPA RfD and TEFs as described in WHO 2005 for non-cancer endpoints and an upper bound cancer slope factor of 2.0 per mg/kg/d for cancer.

The EPA standard for PCBs in drinking water is 0.5 µg/L and the FDA adopted the same standard for bottled water (see Table 30). The FDA has also set residue limits for PCBs in various foods to protect against harmful health effects including a maximum of 0.2 mg/kg in infant and junior foods, 0.3 mg/kg in eggs, 1.5 mg/kg in milk and other dairy products (fat basis), 2 mg/kg in fish and shellfish (edible portions), and 3 mg/kg in poultry and red meat (fat basis). The FDA regulatory action level for PCBs in fish is much higher than human health risk levels established under the National Toxics Rule or used by Health to assess fish. The FDA action level reflects FDA's higher tolerance for PCBs in food and the agency's emphasis on the net benefit to consumers of eating fish, despite contamination.

The National Toxics Rule sets water quality criteria for PCBs to protect human health over a lifetime of drinking water and eating fish from surface water. The NTR includes an equivalent fish tissue criteria (5.3 ppb) for PCBs. This serves as an "equivalent" measure of water quality in that fish living in water at the surface water criteria (0.00017 µg/L) should not exceed 5.3 ppb PCB in tissue. While Health supports Ecology's use of the NTR criteria for identifying problems and controlling water pollutant sources, Health does not use the NTR criteria to establish fish consumption advisories.

Health establishes fish advisory screening levels for mercury, PCBs, and other contaminants by using an approach similar to that outlined in EPA's Guidance for Assessing Chemical Contaminant Data for use in Fish Advisories Vol. 1-4 (EPA 2000b). This guidance provides a framework for state development of fish consumption advisories, based on best available science, and established procedures in risk assessment, risk management, and risk communication. Health's current screening level for PCBs also incorporates recommendations from the American Heart Association that people should consume two meals of fish per week to gain the known health benefits associated with fish consumption. The Health screening value of 23 ppb is derived from assumptions of two meals/week consumption rate, average body weight of a person, and EPA's RfD for protection of human health effects. Health uses this value to identify populations of local fish that may need a fish advisory. This is a starting point for state advisories which need to balance the many health benefits of fish with the possible risks of PCB ingestion. More on information about how Health develops PCB fish advisories is at: http://www.doh.wa.gov/AboutUs/ProgramsandServices/EnvironmentalPublicHealth/EnvironmentalHealthSafetyandToxicology/FishAdvisories.

For inhaled PCBs, EPA calculated indoor air guidance for schools ranging from 0.07-0.60 µg/m3 depending on the age of the children (see Table 30). These EPA calculations account for additional average exposures through diet, water, air and other sources and would maintain children's exposure levels below the RfD of 0.02 µg/kg/d (EPA 2012c). Residential indoor air levels would need to be lower to reflect the longer hours spent at home.

Public health guidance for PCBs in ambient air are lower still and assume exposure 24 hours a day and seven days a week. For inhalation of evaporated congeners, EPA IRIS assessment considers a concentration of 0.01µg/m^3 to be conservative protection against cancer risk (EPA 1997b). A variety of higher occupational inhalation exposure limits were established more than 10 years ago, assume exposures to adults only, and assume exposure during a standard work week (see Table 30).

Table 30. Established health regulations/guidance for PCBs

	Exposure Limits	Agency	Reference
Total oral daily intake-chronic	Oral Reference dose (RfDs 0.07 µg/kg/day (Aroclor 1016) 0.02 µg/kg/day (Aroclor 1254)	EPA, 2000	EPA, IRIS http://www.epa.gov/iris/subst/0462.htm http://www.epa.gov/iris/subst/0389.htm
Total daily intake of dioxin-like chemicals (TEQ)	0.7 pg/kg/day	EPA, 2012	EPA, Dioxin Assessment http://www.epa.gov/iris/subst/1024.htm
Total daily intake of dioxin-like chemicals (TEQ)	2.3 pg/kg/day	WHO 2001	World Health Organization, Joint Expert Committee on Food Additives
Minimal Risk Level for chronic oral intake	MRL is 0.02 µg/kg/d (based on Aroclor 1254)	ATSDR, 2000	Toxicological Profile for PCB http://www.atsdr.cdc.gov/toxprofiles/tp17.pdf
PCBs in specific foods	0.2-3.0 ppm in various categories of food.	FDA 1996	21 CFR 109
PCBs in edible fish entering interstate commerce	2.0 ppm	FDA, 2011	http://www.fda.gov/downloads/Food/GuidanceRegulation/UCM251970.pdf
PCBs in fish	Screening level of 23 ppb in fish tissue (fillet)	DOH	See DOH website http://www.doh.wa.gov/CommunityandEnvironment/Contaminants/PCBs
PCBs in fish	5.3 ppb Fish tissue equivalent for	NTR, 1999	National Toxics Rule 64 FR 61182 http://www.gpo.gov/fdsys/granule/FR-1999-11-09/99-25559

	surface water quality criteria		
Drinking water	MCL is 0.5 µg/L (or 500 ppt). The MCL Goal is set at zero.	EPA1992 (reviewed 2010)	http://water.epa.gov/drink/contaminants/basicinformation/polychlorinated-biphenyls.cfm
Bottled water	0.5 µg/L	FDA, 1999	21 CFR 165.110
Ambient Air	0.01 µg/m^3 based on cancer risk of 1 in 1,000,000	EPA, 2000	EPA IRIS http://www.epa.gov/iris/subst/0294.htm
Indoor air – schools*	0.07-0.60 µg/m^3 depending on age of children present.	EPA	http://www.epa.gov/pcbsincaulk/maxconcentrations.htm
Occupational air	1.0 µg/m^3	NIOSH, 2000	Http://www.cdc.gov/niosh/npg
Occupational air	1,000 µg/m^3 (Aroclor 1254); 500 µg/m^3 (Aroclor 1242)	OSHA	1998-29 CFR 1910.10003

PCB Exposures in People

PCBs are absorbed primarily from the diet and air, accumulate in fatty tissues, and are excreted, often very slowly, from the human body. PCBs are readily absorbed from the gastrointestinal tract. Absorption efficiency across the gut is higher with more chlorinated congeners. Once in the blood stream, PCBs are rapidly cleared and initially accumulate in liver and muscle tissue. In general, PCBs then redistribute to adipose tissue and skin but this varies depending on the congener (ATSDR 2000). For instance, PCB 126 is a coplanar PCB and binds very tightly to CYP1A2 and subsequently concentrates in the liver of rodent. Other highly persistent PCBs (e.g., PCB 153) are stored primarily in the adipose tissue and skin. There are also gender differences in storage of PCBs (Feeley and Jordan 1998). Metabolism of PCBs involves metabolic enzymes called cytochrome P$_{450}$ enzymes (CYP). Specific subtypes involved in PCB metabolism are CYP1A1 and 1A2, CYP2B1 and 2B2, and CYP3A. Metabolism can lead to biologically active arene oxides and hydroxylated and methysulfonyl metabolites. Elimination of PCBs from the body is largely dependent on biotransformation of congeners to more polar metabolites. Half-lives for PCBs congeners in humans are estimated at 1.4-4.9 years for lightly chlorinated PCB 28 and 10-15 years for the more chlorinated congeners such as PCB 153, 170, and 180 (Ritter *et al*. 2011). Longer half-life estimates have also been reported (ATSDR 2000, Milbrath *et al*. 2009). Because PCBs are more readily absorbed than excreted, they accumulate in the body over time.

PCB Body Burden in the General U.S. Population

PCBs are widely detected in adipose tissue and blood of people (Lordo *et al*. 1996, CDC 2009). PCB in serum is a common indicator of body burden since there is a dynamic equilibrium between PCBs stored in fat and PCBs circulating in blood. Most studies of PCBs in serum report lipid-adjusted measurements to correct for short-term fluctuation in circulating lipids.

Since the 1980s, body burdens of PCBs in the U.S. have declined by more than 80% and continue to decrease (CDC 2009, Longnecker *et al*. 2003, Woodruff *et al.* 2011, Xue *et al.* 2014). Recent estimates of body burden of PCBs in the U.S. general population are available from the National Health and Nutritional Examination Study or NHANES (CDC 2009). This survey sampled serum for 34 PCB congeners in 2001-2002 and added four dioxin-like PCB congeners in 2003-2004 for a total of 38 congeners. In the 2003-2004 survey, 100% of the 1866 participants sampled had detectable levels of PCBs in their serum (Patterson *et al*. 2009). The primary congeners detected were PCB 153, 180, and 138/158. PCB 28, 74, 118, 170, and 187 were also frequently detected (Table 31 lists NHANES results). The sum of 35 PCB congeners in participants had a mean of 134.4 ng/g blood lipid (0.820 ng/g whole weight blood). Five percent of the participants sampled had a sum of PCB in blood higher than 530.7 ng/g lipid weight (3.531 ng/g whole weight) (Patterson *et al*. 2009). The TEQ of total dioxins, furans and dioxin-like PCB in the NHANES 2001-2002 survey was calculated to be 30.4 pg/g lipid for the general population. The nine PCB congeners with dioxin-like properties contributed 38-41% of the TEQ depending on the age of the participant (Ferriby *et al*. 2007). Both studies showed that increasing PCB body burden is strongly associated with increasing age.

Table 31. Sum of 35 PCB congeners in 2003-04 NHANES survey (Patterson *et al*. 2009)

Age of participant	50th percentile for population (CI)	95th Percentile for population (CI)	Population sample size
12-29 years	51.2 (48.2-56.1)	139.0 (110.8-164.3)	585
20-39 years	75.4 (71.2–81.7)	226.5 (170.6-300.5)	452
40-59 years	174.4 (159.9-201.9)	470.7 (373.5-650.9)	383
60+ years	334.5 (308.7-351.8)	929.4 (752.2-1167.9)	446
Total (all ages)	131.8 (121.8-145.5)	530.7 (498.4-570.2)	1866

Serum levels in ng/g blood lipid
CI is the 95% confidence interval for the estimate of percentile in the study population.

Greater body burdens of PCB congeners that readily bioconcentrate have been observed in avid consumers of seafood or land and marine mammals that eat a diet rich in fish. (Dewailly et al. 1993; Ayotte *et al*. 1997, Muckle *et al.* 1998, Fängström *et al.* 2002, Grandjean *et al.* 2001).

Lightly chlorinated PCB congeners (congeners 1-52) may be elevated in people exposed to PCBs in ambient or indoor air. Disposal and recycling workers may have elevated exposures to the full spectrum of PCB congeners in the initial Aroclor mixtures.

PCB Exposures to Developing Children

Fetal and early life are considered the most sensitive life stage for PCB toxicity so it is important to understand early life exposure to PCBs. PCBs cross the placenta of experimental animals and humans. When PCB level is adjusted for percent lipid in the blood, there is no difference between PCB blood levels in maternal serum and cord blood at the time of birth (Jacobson *et al*. 1984).

PCBs stored in fat and fatty tissues are mobilized as serum lipids increase during the normal course of pregnancy. Median concentration of total serum lipids increased 43% and median serum levels of PCBs increased 34% between the first and third trimesters in a study of 67 women. The increase was completely explained by the increase in lipids; when results were lipid normalized there was no difference in µg PCB/g serum lipid (Longnecker *et al*. 1999).

The body burden of PCBs accumulated over a lifetime is thought to be the primary determinant of circulating levels of PCB during pregnancy; however, a study reported by Humphrey in 1989 demonstrated that a single meal could conceivably expose a fetus to a transient peak of PCBs. In his measurements following a meal of Great Lakes fish (fish contained 4-10 ppm PCB) he observed a short-term spike of 250-500% above serum baseline in healthy volunteers. Most fish consumed in the U.S. today are about 1000 times less contaminated than the fish used in Humphrey's experiment.

Breast milk has healthy fat to support the rapid growth of babies. PCBs can accumulate in fat and be transferred to babies during nursing. The PCB body burden of children at four years old is strongly related to the level of contamination in milk and duration of lactation (Swain 1991, Patandin *et al*. 1999, Walkowiak *et al*. 2001). Blood levels of PCBs in Japanese children of occupationally-exposed parents also showed a strong correlation with length of lactation (Hara 1985). By some estimates, human infants can receive up to 10-12% of their lifetime dose from nursing (Birnbaum and Slezak, 1999).Breast milk samples from 40 first-time mothers from the Pacific Northwest of the U.S. and Canada were analyzed for PBDEs and PCBs (She *et al*. 2007). Total PCBs were calculated by summing values of 82 separate PCB congeners and ranged from 0.049 to 0.415 mg/kg lipid. This is 10-100 times lower than the levels documented in breast milk in the 1980s in a North Carolina study and further evidence that PCBs in U.S. breast milk have declined since PCB production ceased (Pan *et al*. 2009, Zietz *et al*. 2008). PCB in breast milk should not deter women from nursing because studies have shown that even breast milk with PCBs has a net benefit on cognitive and motor development of children compared to formula-fed children (see DOH Recommendation for Breast Feeding).

Sources and Pathways of Human Exposure

PCBs in Food

PCBs in food are the most significant source of exposure for most people. Recent studies on fish indicate concentrations of PCBs can be in the 10 to 100 parts per million in fish (especially freshwater fish). High levels are typically found in top predator fish, in bottom-feeding fish such as carp and large scale suckers, and in fish living near known sources of PCB contamination. Meat and dairy products are generally much lower in PCBs with concentrations in the low parts per billion (see Table 34). A recent analysis of 2001-2004 NHANES data looked at food consumption patterns in a general U.S. population relative to 30 PCB congeners measured in their serum (Xue *et al.* 2014). The study found a strong correlation between serum PCB and reported fish consumption but no measurable correlation with consumption of meat or milk.

PCBs in Freshwater Fish Species from Washington State

PCBs can be highly concentrated in the fish of waters contaminated with even low levels of PCBs. The Washington State Department of Ecology routinely conducts fish tissue monitoring as part of its Washington State Toxics Monitoring Program (WSTMP). Between these two programs, thousands of fish have been sampled from hundreds of sites across Washington State. Figure 27 displays the distribution of total PCB tissue concentrations from fish collected across Washington State from several sources.

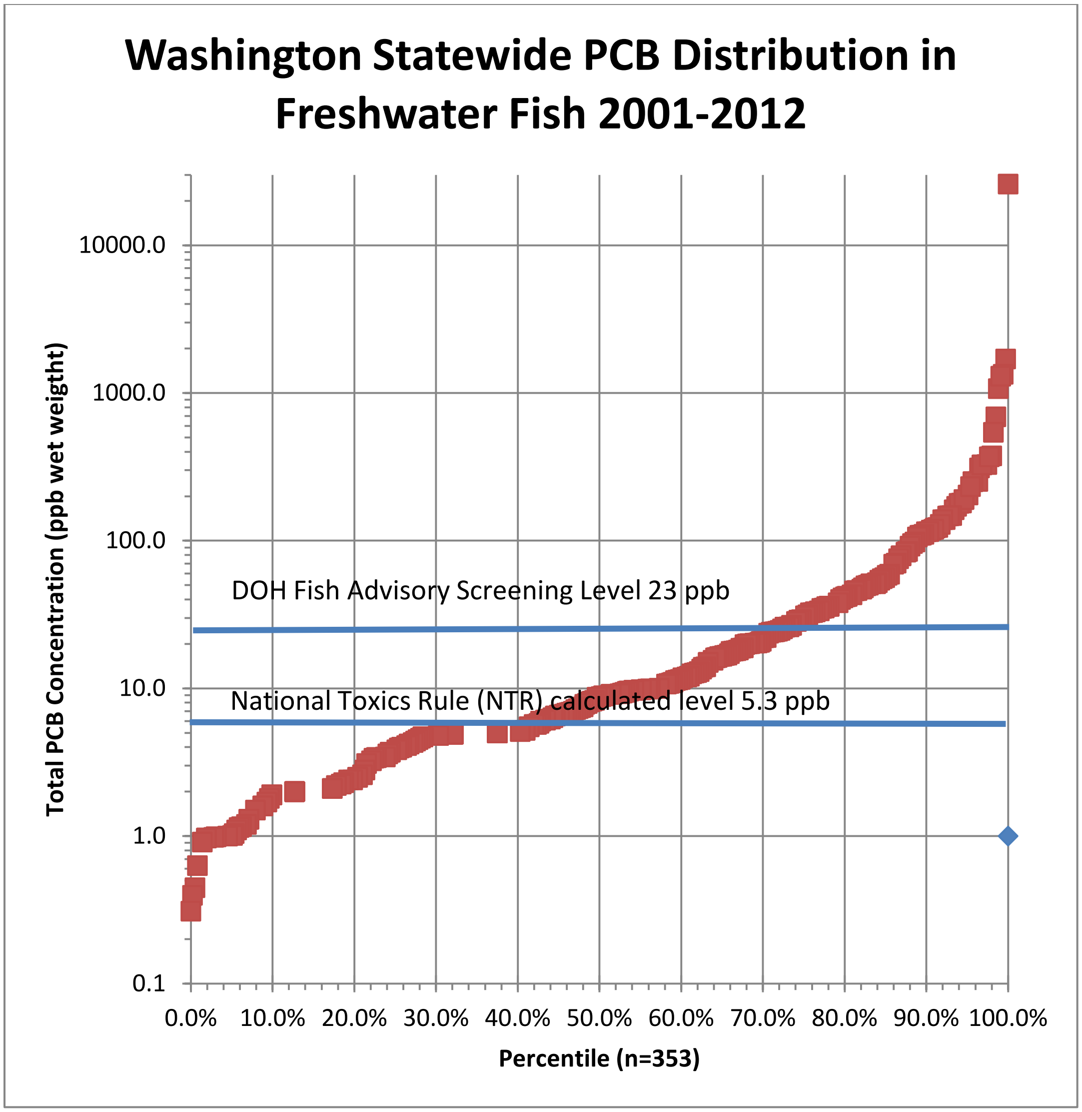


Figure 27. Washington Statewide PCB Distribution in Freshwater Fish Fillets 2001-2012. Data sources: 2001-2010 total PCB fish tissue concentrations extracted from Ecology's EIM database (Seiders 2012), EPA's Upper Columbia River site investigation as reported by Health (WDOH 2012), U.S Department of Energy's 2012 assessment of contaminant data in the Mid-Columbia River, and fish tissue data provided to Health by the U.S. Army Corps of Engineers near Bradford Island and the Bonneville Dam on the Columbia River (unpublished data). NTR and Health screening levels for PCB concentration in fish tissue are displayed for reference.

The data set displayed in Figure 27 includes 353 total PCB values that range from non-detects to greater than 26,000 ppb, with a median of 8.7 ppb. The maximum detection is from a single bass collected near the Bonneville Dam in the Columbia River.

PCBs in Commercially Available Fish in Washington State

Limited data on PCBs in commercially available fish are also available for Washington State. The primary source of this data is a Washington Department of Health 2005 study of contaminants in canned tuna and other frequently consumed store bought fish purchased in

Washington State grocery stores (McBride *et al*. 2005). In this study, PCBs (based on Aroclors concentrations) were detected in store-bought halibut, red snapper, and salmon in at least 10% of the samples collected. Salmon had the highest average PCB concentrations (31.5 ppb PCBs, total Aroclors). Additional data from the Washington State Department of Fish and Wildlife on PCB levels in Puget Sound Chinook and coho salmon were also included for this assessment (WDOH 2006). A comparison of PCB concentrations in store bought and Puget Sound commercially available fish can be seen in Figure 28. Of all fish species, PCB concentrations were highest in Chinook salmon collected in Puget Sound. PCB levels in Chinook salmon returning to Puget Sound waters typically have higher concentrations than coastal salmon or Alaskan Chinook. The higher concentration in Puget Sound Chinook and resident Blackmouth is believed to be due to residence time in areas such as Puget Sound that have greater PCB loads. DOH recommends that women of childbearing age and young children should eat no more than one meal per week of Puget Sound Chinook salmon. Most fish species collected from grocery stores were below Health’s general screening level of 23 ppb.

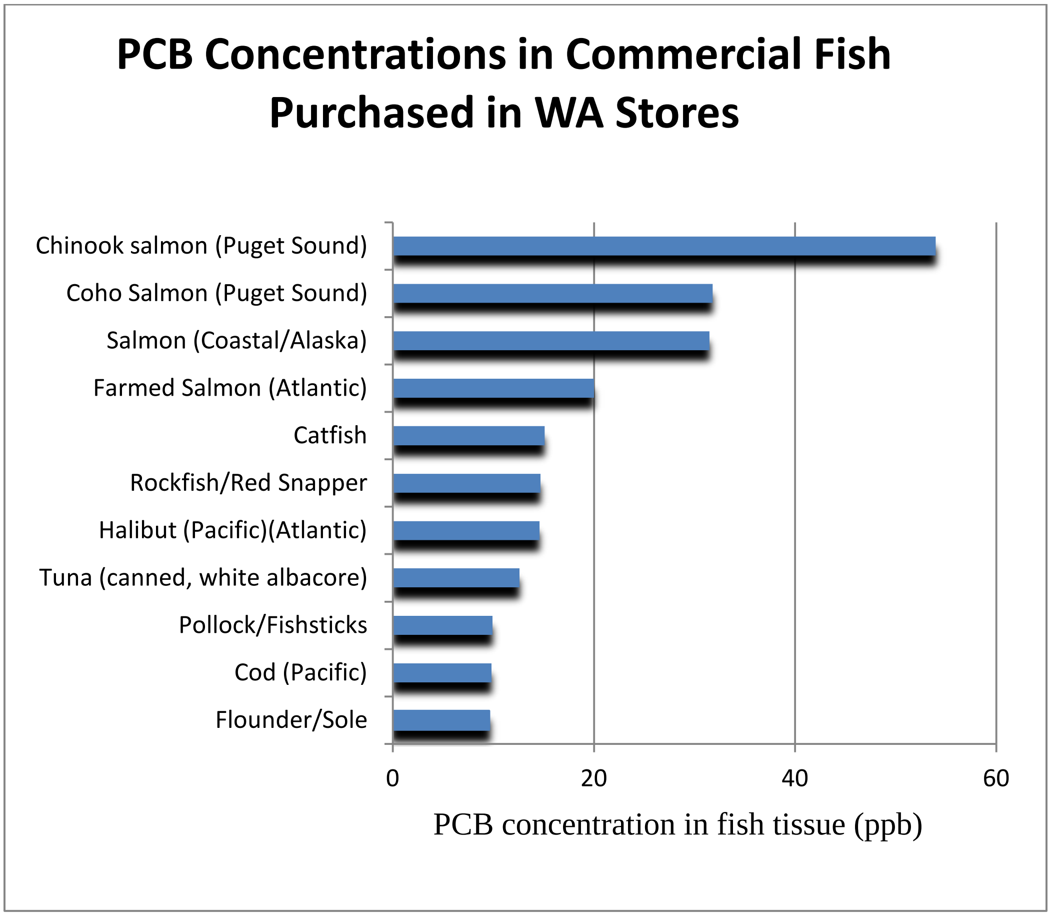


Figure 28. Mean PCB concentrations (total Aroclors) in fish collected from markets and grocery stores in Washington State and from Puget Sound. Data Source: McBride et al. 2005.

Other Dietary Sources of PCBs

Humans may be exposed to small but detectable quantities of PCBs in meat, dairy products, and other foods. PCB concentrations in fish, meat, and dairy products vary widely depending on where they are grown and how they are processed or cooked. Sampling for PCB concentrations in FDA's Market Basket studies between 1991 and 2003 showed PCB levels are far below FDA limits in a variety of prepared dishes. This section summarizes the limited data available from various U.S. and international sources.

The Total Diet Study (TDS), sometimes called the market basket study, is an ongoing FDA program that determines levels of various contaminants and nutrients in foods. http://www.fda.gov/downloads/Food/FoodScienceResearch/TotalDietStudy/UCM184304.pdf. A unique aspect of the TDS is that foods are prepared as they would be consumed (table-ready) prior to analysis, so the analytical results provide the basis for realistic estimates of the dietary intake of these analytes. TDS Market Basket surveys are generally conducted four times each year, once in each of four geographic regions of the countries. Food samples are purchased from supermarkets, grocery stores, and fast food restaurants in three cities in the region and are shipped to a central laboratory. The foods are then prepared table-ready and the three samples are combined to form a single analytical composite for each food. For each survey, samples of food are collected over a 5-week period. Table 32 presents data collected from 1991 through 2004 for PCBs in 26 separate food items. Total PCB concentrations are expressed as Aroclor equivalents, rather than as the sum of congener-specific measurements. Mean PCB concentrations ranged from 0.09 ppb for chicken potpie to 24.4 ppb for salmon.

PCB concentrations in foods from the market basket survey are much lower than previously reported by the Puget Sound Action Team in 2007 and cited by Ecology's Toxics Cleanup Program (Ecology 2012d). PCB levels in foods reported by the Puget Sound Action Team were based on very small sample sizes of one or two. FDA data presented in Table 32 are based on average samples sizes of 40 resulting in more robust, representative PCB levels. The state of origin of the food sampled is not available.

Table 32. Measured PCB Levels as Reported by U.S.FDA

Food Description	Sample Size	Results		
		Concentration (ppb)		Detection Frequency %
		Mean	Maximum	
Chicken potpie, frozen, heated	44	0.09	4	2.3
Candy, caramels	40	0.15	6	2.5
Beef roast, chuck, oven-roasted	44	0.23	10	2.3
Pork roast, loin, oven-roasted	44	0.23	10	2.3
Lamb chop, pan-cooked w/ oil	44	0.23	10	2.3
Chicken, drumsticks and breasts, breaded and fried, homemade	40	0.23	9	2.5
Corn/hominy grits, enriched, cooked	44	0.23	10	2.3
Cornbread, homemade	44	0.23	10	2.3
Biscuits, refrigerated-type, baked	44	0.23	10	2.3
Raisins	44	0.23	10	2.3
English muffin, plain, toasted	44	0.23	10	2.3
Veal cutlet, pan-cooked	40	0.25	10	2.5
Crackers, butter-type	44	0.25	11	2.3
Pork chop, pan-cooked w/ oil	44	0.45	20	2.3
Meatloaf, beef, homemade	44	0.45	20	2.3
Beef (loin/sirloin) steak, pan cooked with added fat	40	0.5	20	2.5
Pancakes made from mix with addition of egg, milk, and oil	40	0.5	20	2.5
Baby food, vegetables and chicken	44	0.68	30	2.3
Brown gravy, homemade	40	0.75	30	2.5
Tuna, canned in oil, drained	40	1.0	40	2.5
Eggs, fried with added fat	40	1.23	39	5.0
Chicken breast, oven-roasted (skin removed)	44	1.36	30	4.5
Popcorn, popped in oil	40	1.7	30	10.0
Butter, regular (salted)	44	3.18	120	4.5
Catfish, pan-cooked w/ oil	4	4.25	17	25.0
Salmon, steaks/fillets, baked	24	24.38	55	91.7

Table 32 summarizes PCB analytical results of food from the Food and Drug Administration's Total Diet Study program. The information pertains to Total Diet Study market baskets 1991-93 through 2003-04. Statistics were calculated using value of zero for results below the detection limit. This document is available on the internet at: http://www.cfsan.fda.gov/~comm/tds-res.html.

In addition to the U.S.FDA information, Arnold Schecter and colleagues at the University of Texas in Dallas have analyzed PCB concentrations in foods in the U.S. over the past 15 years. These studies have focused on common foods in the American diet that were collected throughout the country. The following summarizes those individual studies and a compilation of the data is presented in Table 33.

- Schecter *et al*. (1997) pooled food samples collected from grocery stores across the U.S. and measured 15 different PCB congeners, including eight coplanar PCBs, three mono-ortho PCBs, and four di-ortho PCBs. A total of 90 individual food specimens were pooled into 12 different food types (e.g., cheese, beef) and analyzed for dioxins, dibenzofurans, and PCBs. Based on a conversion of the measured congeners, the authors conclude that PCBs contribute significantly to total TEQ values in eight out of 12 sample types.
- Schecter and Li (1997) measured dioxin-like PCBs in U.S. fast food purchased at five cities across the US. Samples were pooled by type and tested for seven mono- and di-ortho PCBs. Total PCB levels ranged from 0.957 ppb (McDonald's Big Mac) up to 1.180 ppb (Pizza Hut Personal Pan Supreme with anchovies). The authors estimate that fast food accounts for roughly 16.7-52.7% of the total daily TEQ of dioxin-like compounds.
- Schecter *et al*. (1998) analyzed both cooked and uncooked samples of beef, bacon, and catfish from a supermarket in Binghamton, New York. A total of five cooked, and four uncooked samples of each type of meat were analyzed for dioxins, dibenzofurans, and three dioxin-like PCBs (77, 126, 169). Total PCB concentration for each food type ranged from 1.028 ppb (wet weight) (uncooked hamburger) to 5.370 ppb (cooked bacon). Broiling each type of sample resulted in a 50% decrease on average in total PCDD, PCDF, and coplanar PCB TEQ. However, broiling of hamburger resulted in an increase of total coplanar PCB concentration of 10.4%; broiling bacon resulted in an increase of 75.7% of total coplanar PCB concentration. The authors conclude that final concentrations (pg TEQ/kg) of PCDDs, PCDFs, and coplanar PCBs in broiled foods cannot be accurately predicted from raw samples due to variances in cooking method.
- Schecter *et al*. (2002) analyzed a total of 72 meat baby food samples purchased from grocery stores across the U.S. (Illinois, Nebraska, California, Georgia, New York, Pennsylvania, and Maryland). Three to six samples (200 g each) were purchased from each state, and analyzed for dioxins and three coplanar PCB congeners (77, 126, and 159). Total PCBs for each food sample type range from 0.579 ppb (wet weight) (lamb) to 2.280 ppb (chicken). Converted TEQ concentration for total PCBs for each food type ranged from 17.6 (lamb) to 95.9 (beef). The authors note that for the turkey, beef, lamb, and ham samples, total PCBs contributed more to total TEQ values than did the PCDD/PCDF values.

- Schecter *et al*. (2010) study expanded their previous studies of persistent organic pollutants contamination, including PCBs in composite U.S. food samples collected in Dallas, Texas. The study showed that PCBs were not detected by congener analysis in any meats except hamburger, in any fish except salmon and canned sardines, or in any dairy products or eggs.

Overall, Schecter and colleagues have shown that the PCB levels in foods common in the U.S. are typically low relative to freshwater fish species collected in Washington State. PCB levels in foods other than fish are generally in the low single digit parts per billion range whereas freshwater and marine fish species are generally one, two, and sometimes three orders of magnitude higher. Even when beef, chicken, and pork consumption rates are greater than fish consumption rates, dietary exposure to PCBs is dominated by the consumption of fish.

Table 33. Level of PCBs in U.S. Foods (1994 – 2009)

Location (date)	Type of Sample	PCB congeners	Food (sample size)	Total PCB concentration ppb (wet weight)	Reference
Across U.S. (1995)	Grocery Stores	15 total, including 8 coplanar, 3 mono-ortho, and 4 di-ortho PCB congeners	Beef (5 pooled)	Total coplanar 0.0428 ng/g; mono-ortho 0.344 ng/g; di-ortho 0.593 ng/g **Total 0.980 ppb**	Schecter et al. 1997
			Chicken (4 pooled)	Total coplanar 0.132 ng/g; mono- ortho 0.403 ng/g; di-ortho 0.505 ng/g **Total 1.04 ppb**	
			Pork (5 pooled)	Total coplanar 0.182 ng/g; mono-ortho 0.375 ng/g; di-ortho 0.322 ng/g **Total 0.879 ppb**	
			Hot dog/bologna (2 pooled)	Total coplanar 0.156 ng/g; mono-ortho 1.500 ng/g; di-ortho 1.871 ng/g **Total 3.027 ppb**	
			Eggs (3 pooled)	Total coplanar 0.0006 ng/g; mono-ortho 0.064 ng/g; di-ortho 0.147 ng/g **Total 0.212 ppb**	
			Cheese (5 pooled)	Total coplanar 0.0017 ng/g; mono-ortho 0.240 ng/g; di-ortho 0.342 ng/g **Total 0.584 ppb**	
			Butter (2 pooled)	Total coplanar 0.004 ng/g; mono-ortho 1.150 ng/g; di-ortho 2.080 ng/g **Total 3.23 ppb**	
			Ice cream (5 pooled)	Total coplanar 0.0001 ng/g; mono- ortho and di-ortho ND **Total 0.0001 ppb**	
			Milk (5 pooled)	Total coplanar 0.0004 ng/g; mono and di-ortho ND **Total 0.0004 ppb**	
			Vegan diet (1 pooled)	Total coplanar 0.0002 ng/g; mono-ortho 0.015 ng/g; di-ortho 0.144 ng/g **Total 0.159 ppb**	
Across U.S. (1995)	Fast Food Restaurants	PCBs 105, 118,156,128,138,153, 180	Hamburger, McDonalds Big Mac (5)	mono & di-ortho total 0.957 ppb	Schecter and Li 1997
			Pizza, Pizza Hut Supreme (5)	mono & di-ortho total 1.180 ppb	
			Chicken, KFC Original Recipe (5)	mono & di-ortho total 1.170 ppb	
Binghamton, NY (1996)	Grocery Stores	Coplanar PCBs 77,126, 169	Hamburger, cooked (5)	Mean 1.401; range 1.204-1.601 ppb	Schecter et al. 1998
			Hamburger, uncooked (4)	Mean 1.270; range 1.028-1.736 ppb	
			Bacon, cooked (5)	Mean 2.734; range 1.722-5.370 ppb	
			Bacon, uncooked (4)	Mean 1.556; range 1.205-1.971 ppb	
			Catfish, cooked (4)	Mean 3.188; range 1.945- 3.963 ppb	
			Catfish, uncooked (4)	Mean 4.691; range 2.200-6.387 ppb	
Across U.S. (IL, NE, CA, GA, NY, PA, MD) (1998)	Baby Food Grocery Stores	PCBs 77, 126, 159	Chicken	Range 0.883-0.228 ppb	Schecter et al. 2002
			Turkey	Range 0.144-0.160 ppb	
			Beef	Range 0.150-0.225 ppb	
			Lamb	Range 0.579-0.844 ppb	
			Ham	0.771 ppb	
Dallas, TX (2009)	Supermarkets	PCBs, 52, 101, 118, 138, 153, 180	Hambuger	PCB-153 1.2 ppb; PCB-180 0.21 ppb	Schecter et al. 2010
			Salmon	PCB-52 0.28 ppb; PCB-101 0.51 ppb; PCB-118 0.43 ppb; PCB-138 0.93 ppb; PCB-153 1.21 ppb; PCB-180 0.44 ppb	
			Canned Sardines	PCB-52 0.28 ppb; PCB-101 0.67 ppb; PCB-118 0.80 ppb; PCB-138 1.80 ppb; PCB-153 1.83 ppb; PCB-180 0.49 ppb	
			Bacon	Non-detected	
			Turkey	Non-detected	
			Sausages	Non-detected	
			Ham	Non-detected	
			Chicken breast	Non-detected	
			Roast beef	Non-detected	
			Canned chili	Non-detected	
			Catfish fillet	Non-detected	
			Tilapia	Non-detected	
			Cod	Non-detected	
			Frozen fish sticks	Non-detected	
			Butter	Non-detected	
			American cheese	Non-detected	
			Other cheese	Non-detected	
			Whole milk	Non-detected	
			Yogurt	Non-detected	
			Cream cheese	Non-detected	
			Eggs	Non-detected	

PCB Contribution to Dioxin TEQ in Food

Coplanar PCB congeners act toxicologically like dioxins and contribute to the total dioxin TEQ in foods. The National Academies of Science provided a comprehensive compilation of data on dietary exposure to dioxin and dioxin-like compounds including PCBs (NAS 2003). Estimates of exposure are based on concentrations of dioxins and dioxins-like compounds measured in foods and dietary consumption habits of those foods. The dioxin and PCB food concentration data were based on the U.S. Food and Drug Administration's Total Diet Study (FDA 2006). The NAS report identifies that dietary sources of animal fat are by far the largest source of dioxin exposure to the general population, with 90% of total exposure being due to consumption of food – namely animal products and their associated animal fats (beef, pork chicken, fish, fats (butter), and dairy products. The NAS reported that estimates of the contribution of PCBs to dietary dioxin TEQ range from 37-57% and concluded that 50% was a reasonable estimate (NAS 2003).

EPA's 2000 Draft Dioxin Reassessment (EPA 2000a) summarized the available data on background concentrations in foods for the United States. Using that data and standard assumptions for intake, they developed an estimate of general background exposure to coplanar PCBs. The estimates assume concentrations in food reported as nondetected are present at ½ the detection limit. EPA estimated background exposure to adults in the general population to be 0.64 pg/kg/day for dioxins and furans and 0.34 pg/kg/day for dioxin-like PCBs. Based on EPA's analysis, coplanar PCBs account for approximately one-third of total dioxin-TEQs (Smith and Frohmberg, 2008). See Table 34.

Table 34. Estimates of Background Dietary Exposure to Dioxins and Coplanar PCBs on a Toxic Equivalents (TEQs) Basis.

Chemical	Estimated Exposure Assuming NDs = 0 (pg/kg/day)	Estimated Exposure Assuming NDs = 1/2 DL (pg/kg/day)
Dioxins/Furans TEQs	0.38	0.64
Coplanar PCB TEQs	0.34	0.34
Total TEQ	0.72	0.98
% Contribution of Coplanar PCBs to Total TEQs	47%	35%
EPA RfD for dioxin (non-cancer endpoints)	0.7 pg/kg/d	0.7 pg/kg/d

(Source: Smith and Frohmberg, 2008)

ND = non-detected, DL= Laboratory detection limit

In summary, food and especially fish appear to be major contributors to PCB exposure in the U.S. population. With few exceptions, freshwater fish species have the highest PCB levels. Mean PCB concentration of all Washington state freshwater fish for which Health has issued fish consumption advisories is over 150 times higher than the mean PCB concentration reported in other common food items. Freshwater fish species in Washington are over ten times higher than

PCB concentrations measured in commercially available fish in Washington State stores. Commercially purchased fish PCB levels are also greater than mean PCB concentrations of other non-fish foods tested by a factor of ten (Figure 29). Such comparisons illustrate the relative contribution of freshwater fish species to an individual's dietary PCB exposure, particularly for high fish consumers. Those individuals or groups that rely on freshwater fish species as an important component of their diet are potentially at greater exposure to PCBs and correspondingly at greater health risk.

Dioxin-like PCBs are significant contributors to dietary dioxin burden in the U.S. and the total dioxin burden appears to be at the EPA reference dose for dioxin TEQ.

The PCBs in fish are relatively well studied and this source of exposure has a robust health literature pointing to adverse health impacts. The most vulnerable lifestage to PCB exposure appears to be fetal development. Although U.S. exposures have dropped dramatically since the 1980s, Avid consumers of Puget Sound salmon, freshwater sports fish like bass, and other fish from contaminated waters are still at risk for elevated exposure. Reducing PCB exposure in people will require efforts to reduce PCB levels in freshwater and some marine fish and to mitigate the sources of PCB loading to the waters where they live.

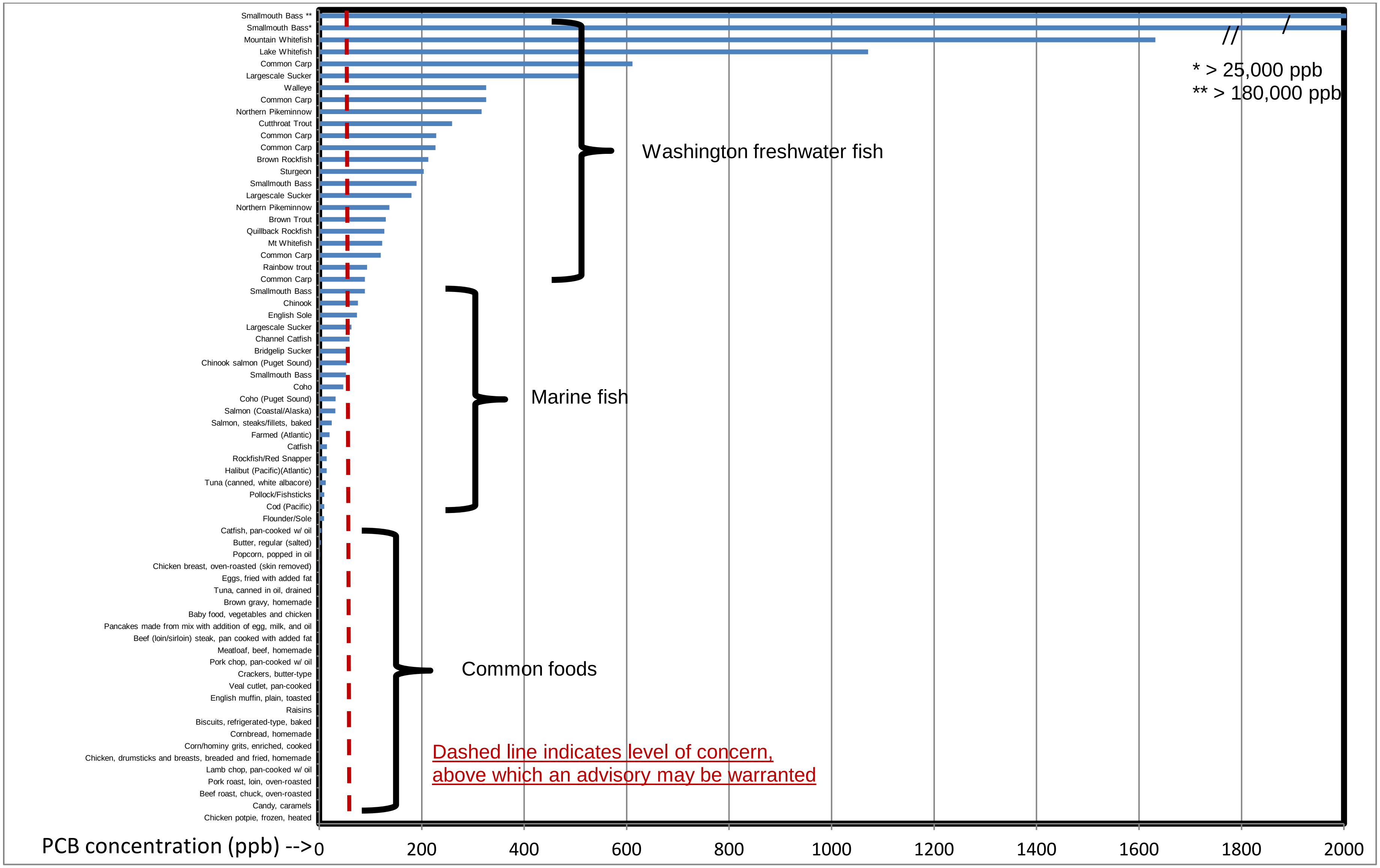


Figure 29. PCB Concentrations (ppb) in Sportcaught Fish Collected in Washington, Commercial Fish Purchased in Washington, & Common Foods

Data sources: WA freshwater fish data 2001-12 in Ecology EIM database, U.S. Dept. of Energy, WA Dept of Health commercial fish study (McBride et al.,2005), FDA Total Diet Study 1991-2006. Only a subset of the state's freshwater fish PCB data are shown to represent those species associated with Health fish advisories based on PCB levels.

PCBs in Ambient Air

PCBs with fewer chlorines can volatilize from water, soil, or contaminated materials and effect local air concentrations (ATSDR 2000, Carlson and Hites 2005, Du *et al*. 2009).They can also be transported long distances by global air currents and have been detected polar research stations where no local sources exist (Choi *et al*. 2008). Heavier PCBs are not volatile but can contaminate air when adhered to demolition dust, wind-blown dust, and airborne particulate. Inhalation was thought to be the primary pathway of occupational exposure to PCBs historically but was largely ignored for general population exposures until recently. PCB air monitoring started in Chicago in 1995 and found surprising PCB concentrations in urban air. Hu *et al*. 2008 analyzed for 209 PCB congeners in Chicago area air and reported an annual average of 0.000835 $\mu g/m^3$ (range 0.000075 to 0.0055 $\mu g/m^3$). Similar air concentrations results have been reported from the Philadelphia, PA (Du *et al*. 2009) and Cleveland, OH (Basu *et al*. 2009). Air levels at least an order of magnitude lower have been detected in polar regions (Choi *et al*. 2008) and in various remote locations around the Great lakes region (Basu *et al*. 2009).

Only limited air sampling for PCBs has been conducted in Washington State. Bulk air deposition samplers have been used to estimate the load to Puget Sound soils and water surfaces (Ecology 2010d). These results are not adequate for estimating inhalation exposure for health risk assessment because the lighter gas phase PCBs would be largely missed by these samplers.

One EPA pilot study analyzed air samples collected in year 2000 from rural areas of the U.S. for six PCBs that are considered dioxin-like. This study included one site on the Olympic Peninsula in Washington. PCB 118 was the most common dioxin-like PCB detected (0.337 pg/m^3). PCB 105 was detected at 0.115 pg/m^3, PCB 156/7 were detected at 19.7 fg/m^3, 77 was detected at 16 fg/m^3, and PCB 126 and 169 were detected at 1 fg/m^3 or less (EPA 2007).

The reported levels of PCBs in ambient air are generally well below EPA level of human health concern. The maximum PCB levels reported on hot summer days in Chicago was 0.0055 $\mu g/m^3$ which is still below the EPA “de minimus” cancer risk estimate for chronic inhalation of evaporated PCB congeners (0.01 $\mu g/m^3$).

Congener profiles of PCBs in ambient air differ both from profiles of commercial Aroclors and from congeners that partition to fish. Although the full spectrum of congeners has been detected in ambient air, lightly chlorinated congeners predominate (PCBs 1-52). Lighter congeners are more quickly eliminated from the body and their toxicological properties are less studied. This introduces uncertainty in the exposure and toxicity assessment of the mixture. Norström *et al.* 2010 conducted modelling to predict the contribution to PCB body burden from breathing urban air contaminated with a profile of PCBs similar to those detected in Chicago air. Their model suggests that urban air would not significantly impact the body burden of congeners associated with dietary uptake such as PCB 153, 180, and 183 but could contribute to human body burden of PCB 28, 33, and 52 by as much as 30% depending on the congener (Norström *et al.* 2010).

The toxicological relevance of inhaling congeners in urban air was investigated by Hu *et al.* 2012. They conducted a subchronic rodent inhalation assay with a cocktail of PCBs representing urban air and observed for immune responses, microsomal enzyme induction, cellular toxicity and histopathologic abnormalities. The minimal effects detected are suggestive of mild oxidative stress during the course of treatment.

The toxicological relevance of lighter chlorinated congeners must also consider the impact of metabolites produced once these lighter PCBs are absorbed into the body. For example, some PCB 3 metabolites are genotoxic and have been shown to cause point mutations in rodents (Xie *et al.* 2010, Robertson and Ludewig 2011). There is also limited evidence of tumor initiating activity of PCB 3, 15, 52 and 77 in a rodent model (Espandiari *et al.* 2004). Congeners like PCB 11 may be transformed into metabolizes that contribute to oxidative stress and cellular damage (Zhu *et al.* 2013). This is an active area of research and more study is needed to understand the potential toxicity of lower chlorinated PCBs and their metabolites.

PCBs in Indoor Air (Caulk, Joint Sealants, Lamp Ballasts)

PCBs were used as plasticizers and flame retardants in building materials such as some elastic caulks, joint sealing compounds for brick and masonry buildings, exterior paints, window glazing, ceiling tile coatings, and some floor finishes sold in the 1950-1970s. PCBs were also widely used in fluorescent lighting ballasts installed during this same period. In different investigations since 1980, these materials have been identified as sources of elevated PCB levels in air in schools, office buildings, large apartment complexes, and other buildings. A few examples are listed below.

EPA investigated PCB levels at six unoccupied schools in New York that were scheduled for major renovation or demolition and were suspected of containing PCB sources (EPA 2012c). EPA measured PCBs in air and surface wipes in the buildings and estimated student exposures before and after PCB remediation. EPA estimated doses for an average student were 0.022 µg/kg/day before remediation and 0.007 µg/kg/d after. Estimates of higher student exposure scenarios were 0.041 µg/kg/day before remediation and 0.012 µg/kg/day after. PCB light ballasts and caulk were considered the primary PCB sources in the schools and inhalation the primary exposure route. Remediation reduced estimated exposure by approximately two thirds. EPA conducted congener specific PCB analysis in one of the six schools. Average indoor air concentration of total PCBs in air was 0.50 µg/m^3, the average TEQ of dioxin-like congeners in air was 0.788 pg/m^3. (EPA 2012c). EPA research associated with this project confirmed that caulk with high levels of PCBs caused elevated PCB in the surrounding air, that light ballasts emit PCBs at normal operating temperatures even when there was no visible liquid leaking, that caulk with low levels of PCBs can be encapsulated to reduce emissions, and that a special treatment system can be effective in removing PCBs from thin surfaces such as wall paint (EPA 2013h). In December 2010, EPA released national guidance recommending that schools remove

all PCB-containing lighting ballasts.
http://www.epa.gov/epawaste/hazard/tsd/pcbs/pubs/ballasts.htm

Elastic joint sealants containing PCBs were found to be the source of elevated PCBs in indoor air at 29 sampling sites in various public buildings in Switzerland (Kohler *et al*. 2002). In most cases the indoor air levels of PCBs were below 1.50 µg/m^3 but levels up to 4.20 µg/m^3 were detected. As a comparison, this study reported air levels of 13.0 µg/m^3 PCB at an industrial building that formerly produced transformers. PCB congeners 28 and 52, used as indicators for the more volatile PCBs, predominated in all air samples. Dioxin-like PCBs were also measured (primarily PCB 118 and 105) and the TEQ was calculated using WHO 1998 TEFs. Emissions from joint sealants had a consistent ratio of dioxin-like PCBs to total PCBs. Air levels of 1.0 µg/m^3 total PCBs corresponded to a 1.2 pg/m^3 of dioxin-like PCBs. (Kohler *et al*. 2002).

Joint sealants were also the source of PCB contamination in a public building in Germany (Schettgen *et al*. 2012). Investigations included air measurements and biomonitoring of people who worked in the building. Workers from an uncontaminated building served as controls. Median air levels for total PCBs were reported as 1.74 µg/m^3 with a maximum of 4.28 µg/m^3. Exposed workers had significantly higher blood levels of the more volatile PCBs (28, 52, 101 and the dioxin-like congeners 105 and 118) but not heavier PCBs which constitute the bulk of human body burden and are taken up primarily in the diet (PCB 138, 153, or 180). The calculated TEQ for dioxin-like congeners did not differ statistically between the groups. Follow-up monitoring of three people who were removed from the building demonstrated that levels of PCB 28, 52, and 101 declined steadily after removal and that the biological half-lives were 4.5 ± 0.9 years for PCB 28, 1.3 ± 0.1 years for PCB 52, and 2.8 ± 0.7 years for PCB 101 (Schettgen *et al*. 2012). Longer retention of PCB 28 in the body may explain why PCB 52 predominated in air samples but PCB 28 predominated in serum samples.

Liebel *et al*. 2004 reported significantly higher median serum concentrations for PCBs 28, 52, and 101 in 377 children from the contaminated school in Germany compared to 218 students attending an uncontaminated school. There was a significant positive association between years spent at the contaminated school and serum levels of the combined lower chlorinated congeners. Air levels measured in multiple locations over two years in the school building ranged 0.004-0.600 µg/m^3 for PCB 28, 0.038-2.300 µg m^3 for PCB 52, and 0.003-1.100 µg/m^3 for PCB 101. Very little PCB 138, 153 or 180 were detected in air sampling. The authors estimated total PCB in air over the two-year period to be 0.690- 20.80 µg/m^3 (mean 2.044 µg/m^3) based on measurement of six indicator congeners. When PCB congeners associated with dietary intake were considered, there was no statistically significant difference between overall PCB body burden in students from the two schools. Nor was there a detectable difference in a survey of children's subjective symptoms.

Frederiksen *et al*. 2012 reported that mean PCB air levels in Danish multiunit housing was 1.03 µg/m^3 (range 0.168-3.843 µg/m^3) in apartments that contained PCB in elastic sealants verses a mean of 0.006 µg/m^3 in apartments sealed with PCB-free sealants. PCB sealants contained up 20% (221,680 ppm) PCB and were the primary determinant of indoor PCB levels. A survey of residents about their adherence to advice about minimizing their exposure showed that frequent ventilation, vacuuming, dusting, and floor washing were associated with lower indoor air levels.

In 2009, PCBs were discovered in chipping exterior paint on the former Rainier Brewery in Seattle at concentrations over 10,000 ppm PCB. This 4.5 acre site now houses mixed residential, restaurant, and business spaces. Paint chips were suspected to be the source of elevated PCBs in a nearby stormwater collection area. In 2010, sampling by EPA detected PCBs in indoor air in some office areas (0.010-0.028 µg/m^3) and in an outside stairwell that had been enclosed with the exterior paint intact (0.052 µg/m^3). Sampling also detected PCBs in vacuum dust at concentrations between 1.4-15.6 ppm in residential and office spaces, 3.4-36 ppm in storage and warehouse areas, and 470 ppm in stairwell dust. DOH concluded that there was a very low to insignificant increase in cancer risk associated with the levels detected. Recommendations for mitigation included removing all paint with more than 50 ppm PCB (as required by law), warning occupants to avoid regular use of the external stairwell until remediation could take place, and adopting cleaning techniques that would reduce potential for human exposure (WDOH 2013b).

PCBs can bind to indoor dust and present an inhalation or ingestion pathway for people. Dust intake is associated with higher molecular weight PCBs than air exposures. A 2006 survey of PCBs in residential settings reported median dust concentrations to be 0.200 µg/kg dust (ppb) in 20 Texas homes and 0.260 µg/kg in ten Toronto homes (Harrad *et al.* 2009). Maximum detected was 0.820 µg/kg PCBs in dust. While ingestion of house dust was a minor contributor to adult exposures, it contributed 1-20% of total PCB exposure to toddlers in exposure modelling (Harrad *et al.* 2009). Homes built before 1980 had higher PCB loading in house dust than more recently constructed homes in a large sampling of 415 homes in California (Whitehead *et al.* 2013). A recent study in China reported that house dust levels of PCBs were associated with subtle neurodevelopmental effects in pre-school aged children (Wang *et al.* 2015).

It appears that PCBs in older building materials can elevate PCBs in indoor air and dust and cause higher body burden of certain PCB congeners in the bodies of people living or working in these buildings. In some cases the detected air concentrations were high enough to exceed residential or occupational health guidelines. In the EPA investigation into older schools in New York, indoor air levels frequently exceeded EPA health guidance for schools of 0.70-0.60 µg/m^3 depending on the age of the children present. EPA requires caulk with more than 50 ppm of PCBs to be removed. PCB containing fluorescent light ballasts that remain in service are generally more than 30 years and have exceeded their expected life-span. They are at high risk for over-heating and failing and should be removed and properly disposed.

PCBs in Pigments and Dyes

Residual PCBs in dyes and pigments have been detected in ambient air, food, water, and consumer products. Air monitoring studies of urban air and remote polar air detect PCB 11, occasionally as a major component (Hu *et al.* 2008, Du *et al.* 2009, Basu *et al.* 2009, Choi *et al.* 2008). Consumer product testing has measured residual amounts of PCB 11 and other congener in a variety of colored paper, cardboard, and plastic packaging. (Hu and Hornbuckle 2010, Rodenberg *et al.* 2010). In Washington, PCBs associated with pigments and dyes have been identified in Columbia River water and clams and in a majority of fish sampled in a recent study in the Mid-Columbia. River (McCarthy 2007, Ecology 2005, U.S. Department of Energy 2010)

There is only limited investigation of absorption of PCB 11 into people. In rats, PCB 11 was rapidly absorbed via inhalation, distributed to tissue, and eliminated with half-lives in lung, liver and serum of approximately two hours. A hydroxylated metabolite of PCB 11 was also detected in the rodent liver (Hu et al 2013). Biomonitoring for PCB congeners unique to dyes and pigments (PCB 11) has detected PCB 11 in people. In a recent study, 65% of 85 women in a mid-West had traces of PCB 11 in their blood (Marek *et al* 2013). Three potentially toxic metabolites of PCB 11 have also been detected in human serum (Zhu *et al*. 2013).

There is only limited toxicity information specific to PCB 11. Zhu et al 2013, tested PCB 11 and the 4-hydroxymetabolite detected in human serum for toxicity *in vitro*. In this study, the PCB 11 metabolite suppressed cell growth, created oxidative stress, and resulted in cytotoxicity. Co-administration of antioxidants partially protected against the observed effects. PCB 11 had no effect in the test (Zhu et al 2013). Further study is needed to understand the magnitude of human exposure to PCB 11 and its metabolites as well as their toxicity.

Other Environmental Exposures

Small amounts of PCBs can be found in almost all soil surfaces and sediments. Most soil levels of PCBs are less than 0.010-0.040 µg/kg (ppb) but soil at hazardous waste sites may be much higher. In water, a small amount of PCB may remain dissolved but most tends to stick to organic particles and sediments or evaporate from the water surface (ATSDR 2000).

Direct contact with PCB in old paint, caulk or fluids leaking from capacitors can lead to skin absorption or ingestion of PCBs. It is important to wear protective clothing gloves and respiratory protection if repairing or handling equipment like light ballasts that may have PCBs in them.

Occupational Exposures

Current occupational exposures can come from exposure to leaking electrical equipment made before 1979, from PCB abatement programs, or during demolition or recycling of PCB-contaminated structures and equipment.

Herrick *et al*. (2007), conducted biomonitoring for 54 PCB congeners in a small group of construction workers with a history of removing PCB caulk from buildings in the greater Boston area. The workers had higher proportions of lighter PCB congeners (PCBs 6-74) in their serum than a reference population of men who sought health care in the Boston area. The construction workers as a group had more than five times more PCB 6, 16, 26, 33, 37, 41, 70, 97, and 136 than the referent population. One worker, who was actively involved with removal of PCB products at the time of blood collection, had 25% of his body burden comprised of the lighter PCBs compared to 7% of the PCB serum levels in the referent population.

Wingfors *et al*. 2006 collected blood samples from 36 workers directly involved in abating PCB sealants in Sweden. These were compared in a biomonitoring study to 33 age- and sex-matched construction workers who did not work in the abatement program. The exposed workers had PCB serum levels (sum of 19 congeners) that were twice as high as the controls (mean of exposed workers were 575 ng/g lipid compared to 267 ng/g for the controls). The PCB congener patterns also differed between the workers and the controls, with much higher levels of many less chlorinated PCBs in the exposed workers, compared to the controls. The authors concluded that PCBs 56/60 and 66, were good markers of general occupational exposure; PCB 44, 70, and 110 were good markers for recent occupational exposures; and PCB 153 and 180 reflected background (dietary) exposure. Follow-up samples taken 10 months later showed that serum concentrations of rapidly excreted congeners (PCB 52, 44, 70, and 110) declined after workers were given information about protecting themselves from exposure.

Adequate safeguards are also important during recycling and disposal of PCB containing materials. Electronic waste recycling and disposal practices in China have resulted in elevated exposures in workers, PCB release into nearby soils and rivers, and subsequent contamination of staple foods grown in surrounding areas. (Yang *et al.* 2013, Tue *et al.* 2013, Labunska *et al.* 2015).

Existing Washington State Health Advice

Fish consumption is the primary exposure pathway that most Washingtonians have to many Persistent, Bioaccumulative Toxics (PBTs). Many PBTs such as PCBs, DDT, and mercury are linked to a variety of adverse health effects (e.g. neurological, developmental, immunological, and cancer). The paradox of consuming fish is that it also known to be one of the healthiest forms of protein due in part to the high levels of omega-3 fatty acids that have been associated with a variety of positive health outcomes (e.g. prevention of heart disease, inflammation, arteriosclerosis, and cognitive development). Results from the most recent Behavioral Risk Factor Surveillance System (BRFSS) conducted by DOH indicate that nearly three quarters of the adult general population in Washington State consume fish. Washington State is also the home of numerous federally recognized tribes whose fish consumption rates are often well above that of the general population (Ecology 2013b). Additionally, there are other high fish consuming populations within the state including Asian and Pacific Islanders and sports fishers.

Because of potential exposure to PBTs to fish consuming populations, DOH collaborates with numerous state and federal agencies on the collection and analysis of contaminants in fish. DOH’s role is to evaluate fish contaminant levels in fish tissue for potential public health impacts and to convey information on risks and benefits to fish consumers by way of fish advisories. Currently, Washington State has thirteen waterbody specific fish advisories based on PCB levels in tissue. PCBs account for the greatest number of waterbody specific advisories in Washington State and across the country (WDOH 2013a, EPA 1999b).

Table 35 lists those waterbodies and fish species that currently have a fish advisory due to elevated PCB levels.

Table 35. Washington State PCB Fish Advisories

Water Body/Location	Fish Species	Advisory
Green Lake	Common Carp	1 meal per month
Lake Roosevelt	Largescale Suckers	2 meals per month
Lake Washington	Common Carp	Do not eat
	Northern Pikeminnow	Do not eat
	Cutthroat Trout	1 meal per month
	Yellow Perch	1 meal per week
Lower Duwamish River	Resident fish	Do not eat
	Shellfish	Do not eat
	Crab	Do not eat
Lower Columbia – (Bonneville Dam)	Resident fish	Do not eat
Bonneville Dam to McNary Dam	Resident fish	1 meal per week
Okanogan River	Common Carp	1 meal per month
Puget Sound	Chinook	1 meal per week
	Chinook (Blackmouth)	2 meal per month
	English Sole/Flatfish	Varies by location
	Rockfish	Varies by location
Spokane River		
Idaho Border to Upriver Dam	All species	Do not eat
UpRiver Dam to Nine Mile Dam	All species*	1 meal per month
	* Exception: Largescale Suckers	Do not eat
Long Lake (Lake Spokane)	Largescale Suckers, Brown Trout	1 meal per week
	Mountain Whitefish	1 meal per month
Walla Walla River – Lower	Carp	1 meal per month
Walla Walla River – Lower & Upper	Northern Pikeminnow	Do not eat
Wenatchee River	Mountain Whitefish	Do not eat
Yakima River	Common Carp	1 meal per week
Pending Advisories*		
Mid-Columbia	Lake Whitefish	1 meal per month
	Largescale Suckers	2 meals per month
	Sturgeon	2 meals per month
	Common Carp	1 meal per month
	Walleye	2 meals per month
	Bass	2 meals per month
Snake River	Channel Catfish	2 meals per month
	Common Carp	2 meals per month
* preliminary assessment, meal recommendations may change Fish advisories apply to all individuals. Women of childbearing age and young children should pay particular attention.		

Food Preparation and Cooking Advice to Reduce Exposure

Chemical contaminants are not distributed uniformly in fish. Fatty tissues typically concentrate organic chemicals such as PCBs and dioxins more readily than lean muscle tissue (ATSDR 2004). To reduce the level of PCBs in fish, remove the fish skin and visible fat before cooking. Do not use the fat for gravy or sauces. For further information on reducing contaminants such as PCBs in fish, visit http://www.doh.wa.gov/CommunityandEnvironment/Food/Fish/ReduceContaminantExposure.

Benefits of Fish Consumption

The primary health benefits of eating fish are well documented for children and adults. Dietary fish is associated with reduction of cardiovascular disease (Yuan *et al*. 2001, Rodriguez *et al*. 1996, Hu *et al*. 2002, Marckmann and Gronbaek 1999, Mozaffarian *et al*. 2003, Simon *et al*. 1995, Burr *et al*. 1989, 1994, Singh *et al*.1997, and Harrison and Abhyankar 2005) and positive pregnancy outcome (Jorgensen *et al*. 2001, Olsen *et al*. 1992, Olsen *et al*. 1995, Olsen and Secher 2002, Carlson *et al*. 1993, 1996, Fadella *et al*. 1996, San Giovanni *et al*. 2000, and Helland *et al*. 2003). Limited data also show a link between fish consumption and a decrease in development of some cancers (SACN 2004, IOM 2007). Additionally, eating fish has been associated with impacts on brain function, including protection against cognitive decline (SACN 2004, IOM 2007).

At present, we know that fish is an excellent protein source that is low in saturated fats, rich in vitamin D, omega-3 fatty acids, and other vitamins and minerals. The health benefits of eating fish are associated with low levels of saturated versus unsaturated fats. Saturated fats are linked with increased cholesterol levels and risk of heart disease while unsaturated fats (e.g., omega-3 polyunsaturated fatty acid) are an essential nutrient. Replacing fish in the diet with other sources of protein may reduce exposure to contaminants but could also result in increased risk for certain diseases (Pan *et al*. 2012). For example, replacing fish with red meat could increase the risk of cardiovascular disease due to the fact that red meat has higher levels of saturated fat and cholesterol (Law, 2000).

DOH fish advisories work to be protective of human health while acknowledging the benefits of eating fish. This is done by recommending decreased consumption of fish known to have high concentrations of contaminants in favor of fish that are lower in contaminants. DOH supports the American Heart Association and the U.S. Food and Drug Administration recommendation of consuming at least two servings (12 oz.) of fish per week as part of a healthy diet.

Health benefits of eating fish deserve particular consideration when dealing with groups that consume fish for subsistence. Removal of fish from the diet of subsistence consumers may have serious health, social, cultural, and economic consequences. In order to decrease the potential risks of fish consumption, these populations are encouraged to consume a variety of fish species, to fish from locations with low contamination, and to follow recommended preparation and cooking methods.

Recommendation for Breast-feeding

DOH recommends that babies be breast fed because breast feeding has many demonstrated health benefits for the developing child and the mother. (Washington State Department of Health website http://www.doh.wa.gov/YouandYourFamily/WIC/BreastfeedingSupport.aspx)

Many investigations have looked for adverse effects associated with PCBs in breast milk and duration of breast feeding. Most studies have shown that prenatal, not postnatal PCB exposure correlates with neurobehavioral effects (Michigan, NC, Patandin *et al*. 1999, Darvill *et al*. 2000). Breast feeding appears to have a net positive effect on neurobevavioral test performance regardless of PCB concentration of the milk (Jacobson *et al*. 1990b).

A Dutch study on PCB and dioxin exposures to children recently found that PCB body burden at 42 months is associated with possible immune deficits. However, when the researchers controlled for length of breast feeding, they found that the negative effect of higher postnatal PCB exposure was counteracted by the positive effect of longer duration of nursing in infancy (Weisglas-Kuperus *et al*. 2000). Using this same cohort, other researchers conducted neurological and cognitive assessments at 42 months and found that breast-fed children performed better than their formula fed counterparts despite higher prenatal and postnatal exposure to PCBs (Lanting *et al*. 1998, Patandin *et al*. 1997 and 1999). Follow-up with these children at 6.5 years showed that effects of prenatal exposure to PCBs on cognitive and motor abilities were still measureable in the formula fed group and not measurable in the breast-fed group (Vreugdenhil *et al*. 2002). Analysis of parental and home characteristics suggested that an advantaged home environment contributed significantly to the resilience of the breast-fed group (Vreugdenhil *et al*. 2002).

Current Regulatory Approaches for PCBs

This chapter describes the existing regulations relevant to PCBs and the activities that generate them at the federal, state, and international levels. It includes a brief summary of many laws and regulations directly related to management of processes that produce PCBs, the production, use, and disposal of products that contain PCBs, and exposure limits and cleanup levels for PCBs themselves. This chapter is not an exhaustive review of all of the regulations pertinent to PCBs.

In many instances, federal laws and regulations delegate the authority for implementing these laws and regulations to state or Tribal governments. In some cases, states adopt laws and promulgate regulations that are more stringent than their federal partners.

Federal Laws & Regulations

Toxic Substances Control Act

15 USC 2601 et seq., Toxic Substances Control Act

The Toxic Substances Control Act (TSCA) of 1976 (15 USC 2601 et seq.) gives EPA the authority to regulate new and existing substances. TSCA gives EPA the authority to require reporting, record-keeping and testing requirements, and restrictions relating to chemical substances and/or mixtures. Certain substances are generally excluded from TSCA, including, among others, food, drugs, cosmetics, and pesticides.

TSCA is the primary federal law pertinent to PCBs in the United States. PCBs are regulated by Title I Section 6 of the Act and by EPA implementing regulations, Title 40, Part 761 of the Code of Federal Regulations (CFR). Section 6(e)(2)(A) of TSCA states that "…effective one year after January 1, 1977, no person may manufacture, process, or distribute in commerce or use any polychlorinated biphenyl in any manner other than in a totally enclosed manner." Section 6 of TSCA further prohibited the manufacture of all PCBs by 1979, but allowed the EPA administrator to authorize certain processing, distribution in commerce, and use of PCBs manufactured before 1979 if the Administrator determined that such activity did not present an unreasonable risk of injury to health or the environment. Table 37 summarizes several subparts of TSCA and their contents

Table 36. Subparts of TSCA

Subpart A	General regulations governing PCBs including definitions
Subpart B	Manufacturing, processing, distribution in commerce and use of PCBs and PCB items Prohibitions and exceptions Authorizations including: • Totally enclosed uses (e.g. transformers, capacitors) • Non-totally enclosed uses (including requirements for servicing PCB-containing equipment) • Other uses (carbonless copy paper, research and development, scientific instruments, continued use of porous surfaces contaminated with PCBs)
Subpart C	Marking of PCBs and PCB items
Subpart D	Storage and disposal, including: • PCB disposal requirements • Remediation waste disposal and cleanup levels • Bulk product waste • PCB household waste storage and disposal • PCB decontamination standards and procedures • Storage for disposal
Subpart E	Manufacturing, processing, and distribution in commerce exemptions, including: • Research and development for disposal technologies • Analytical reference samples • 5 ml or less PCB fluids from electrical equipment for analysis •
Subpart F	Transboundary shipments of PCBs for disposal
Subpart G	PCB spill cleanup policy
Subpart J	General records and reports
Subpart K	PCB waste disposal records and reports
Subparts M-R	Sampling requirements for various media and disposal authorizations
Subpart S	Double wash/rinse method for decontaminating non-porous surfaces
Subpart T	Comparison study for validating a new performance-based decontamination solvent

Continued use and disposal of existing PCBs is governed by a framework of controls driven by the form the PCBs take (liquid form, non-liquid form, or multi-phasic, meaning a combination of liquid and non-liquid forms), the amount of PCBs in each form, and the original source of PCBs for media contaminated by a release.

While not a complete summary of all sections in TSCA that pertain to PCBs: below are some important requirements:

- Prohibits of manufacture, sale, and distribution, with exceptions.
- Mandates proper disposal for any PCBs unauthorized for use.
- Does not require testing to find PCB sources, but does require proper use and disposal of identified PCB contaminated items.
  - Many unauthorized uses are therefore not found until a release to the environment has occurred.
- Limits use of PCBs to certain “totally enclosed” uses, such as transformers and capacitors, or concentrations below 50 ppm in bulk product. Various other levels exist for remediation waste and other limited uses, typically with EPA approval.
- Requires that by December 1998, all known transformers containing PCBs >500 ppm be registered with EPA.
  - There is no requirement to determine if transformers contain >500 ppm PCBs, only to register it if it is known to be a PCB Transformer (>500 ppm PCBs).
- Allows many forms of PCB waste to be disposed of as municipal solid waste, which does not require PCBs to be listed on a manifest. Examples include:
  - Small non-leaking PCB capacitors.
  - Plastics (such as plastic insulation from wire or cable; radio, television and computer casings; vehicle parts; or furniture laminates); preformed or molded rubber parts and components; applied dried paints, varnishes, waxes or other similar coatings or sealants; caulking; Galbestos; non-liquid building demolition debris; or non-liquid PCB bulk product waste from the shredding of automobiles or household appliances from which PCB small capacitors have been removed (shredder fluff).
    - Any of these may also be disposed as landfill daily cover or as roadbed under asphalt.
  - Other PCB bulk product waste that leaches PCBs at <10 µg/L of water measured using a procedure used to simulate leachate generation.
  - PCB bulk product waste other than those materials listed above if:
    - The PCB bulk product waste is segregated from organic liquids disposed of in the landfill unit.
    - Leachate is collected from the landfill unit and monitored for PCBs.
- Requires labels identifying electrical equipment containing over 500 ppm PCBs.
- Requires quarterly inspections of PCB transformers containing more than 60,000 ppm PCBs. Transformers with less than 60,000 ppm PCBs and those with appropriate secondary containment must be inspected for leaks at least annually.

- Requires removal or reclassification of high-voltage network PCB-containing transformers to prevent fires. Requires enhanced electrical protection be added on many types of PCB transformers in, or within 30 meters of, commercial buildings.[12]
- Requires EPA authorization for commercial storage of PCBs. Non-Commercial storage does not always require EPA oversight.

TSCA *Rules*

Under TSCA EPA has promulgated 29 rules for the regulation of PCBs. A list of rules, with the associated notices, drafts, etc. can be found on the EPA website at http://www.epa.gov/wastes/hazard/tsd/pcbs/pubs/laws.htm. The current regulations can all be found in the Federal Code of Regulations (CFR) part 761. In general, each rule addresses a specific portion of managing PCBs, such as labeling and spills. Below is some information on three specific regulations.

1. 44 FR 31514 PCBs; Manufacturing, Processing, Distribution in Commerce and Use Bans.

This 1979 rule implemented the ban on PCBs and established 50 ppm PCBs as the general regulatory limit.

2. 49 FR 28172 Polychlorinated Biphenyls (PCBs) Manufacturing, Processing, Distribution in Commerce, and Use Prohibitions; Exclusions, Exemptions, and Use Authorizations

EPA promulgated a rule in 1984 for inadvertent generation of PCBs that are not in closed or controlled manufacturing processes (49 FR 28172). EPA found the societal benefit of these products and the cost of not producing PCBs outweighed the risks to human health and the environment from these sources of PCBs. The rule was based on a consensus proposal from the Environmental Defense Fund, Natural Resources Defense Council, and Chemical Manufacturers Association (now known as the American Chemistry Council). It requires that the concentration of inadvertently generated PCBs in products, including recycled paper, must have an annual average of < 25 ppm, with a maximum of 50 ppm. Detergent bars are treated differently as they are consumer products with a high potential for exposure, and are limited to 5ppm (soap and deodorant are regulated by the FDA).

There were several additional criteria in the rule:

- Releases to ambient air must be less than 10ppm.
- Discharges to water must be less than 0.1ppm, except from recyclable paper the limit is 3 ppb total Aroclors.

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[12] Panero, M., Boheme, S., and Muñoz, G. Pollution Prevention and Management Strategies for Polychlorinated Biphenyls in the New York/New Jersey Harbor. February 2005. New York Academy of Sciences, New York, NY. Available at: http://www.nyas.org/WhatWeDo/Harbor.aspx

- All wastes must be disposed of properly. Process wastes with PCB levels > 50 ppm must be disposed of in accordance with TSCA.
- The concentration of monochlorinated biphenyls is discounted by a factor of 50 and dichlorinated biphenyls are discounted by a factor of 5.
- Certification, reporting, and records maintenance.

The numerical limits in the law were set at the Limits of Quantification (LOQs) at the time.

The rule clarifies some overlap between TSCA with the Federal Insecticide, Fungicide and Rodenticide Act (FIFRA) and Federal Food Drug and Cosmetic Act (FFDCA). If a chemical is solely manufactured for a use that is regulated by FIFRA or FFDCA, then that substance is not regulated under TSCA. If only some uses are regulated under FIFRA, then the chemical is regulated under TSCA until it becomes part of an identified pesticide product. However, chemicals used in FDA-regulated products under FFDCA (like a food, food additive, drug, cosmetic, or medical device) are excluded from TSCA jurisdiction.

In 2013 EPA clarified the definition of “excluded PCB Products” to generally allow for the recycling of plastic separated from shredder residue containing < 50 ppm PCBs under specific conditions. The voluntary procedures to prevent the introduction of PCBs in shredder residue are (1) documented source control programs and (2) documented output control. The review was done at the request of the Institute of Scrap and Recycling Industries to clarify whether the plastic material should be managed as an Excluded PCB Product or as a PCB Remediation Waste. This interpretation reiterates EPA’s “generic exclusion for processing, distribution in commerce, and use, based on the Agency’s determination that the use, processing, and distribution in commerce of products with less than 50 ppm concentration will not generally present an unreasonable risk of injury to health or the environment” (FR Vol. 78, No. 66, April 5, 2013).

3. Several rules on transformers and other electrical equipment have been promulgated. The current regulations are in CFR part 761.

The regulations include several important definitions:

- PCB Transformer (≥500 ppm) *PCB Transformer* means any transformer that contains ≥500 ppm PCBs.
- PCB contaminated (50-500 ppm) *PCB-Contaminated* refers to liquid and non-liquid material containing PCBs at concentrations ≥50 ppm but <500 ppm, and non-porous surface having a surface concentration >10 µg/100 cm^2 but <100 µg/100 cm^2.

  The definitions specifically mention electrical equipment with a very similar definition. *PCB-Contaminated Electrical Equipment* means any electrical equipment including, but not limited to, transformers (including those used in railway locomotives and self-propelled cars), capacitors, circuit breakers, reclosers, voltage regulators, switches (including sectionalizers and motor starters), electromagnets, and cable, that contains PCBs at concentrations of ≥50 ppm and <500 ppm in the contaminated fluid. In the absence of liquids, electrical equipment is PCB-Contaminated if it has PCBs at >10 µg/100 cm^2 and <100 µg/100 cm^2.

- Non-PCB Transformer (<50 ppm) *Non-PCB Transformer* means any transformer that contains less than 50 ppm PCB.

Owners of PCB transformers (≥500 ppm) were required to register their transformers with the EPA by Dec. 28, 1998. Some important points about this requirement:

- There is no requirement to test a transformer to determine if it is a PCB Transformer.
- There is no requirement to register a transformer if the owner takes ownership after 1998.
- There is no requirement to register a PCB-contaminated transformer (50-500ppm PCBs)
- There is no requirement to request a registered transformer be removed from the database if it is physically removed from service.
- Other equipment, such as bushings with ≥ 500 ppm PCBs, are not required to be registered.

While testing for PCBs is not required, the regulations do include PCB concentration assumptions that are based on the age and size of the equipment. The assumptions include:

- Transformers with <3 pounds (1.36 kilograms (kgs)) of fluid, circuit breakers, reclosers, oil-filled cable, and rectifiers whose PCB concentration is not established contain PCBs at <50 ppm.
- Mineral oil-filled electrical equipment that was manufactured before July 2, 1979, and whose PCB concentration is not established is PCB-Contaminated Electrical Equipment (i.e., contains ≥50 ppm PCB, but <500 ppm PCB). All pole-top and pad-mounted distribution transformers manufactured before July 2, 1979, must be assumed to be mineral-oil filled.
- Electrical equipment manufactured after July 2, 1979, is non-PCB (<50 ppm PCBs).
- If the date of manufacture of mineral oil-filled electrical equipment is unknown, any person must assume it to be PCB-Contaminated.
- A transformer manufactured prior to July 2, 1979, that contains 1.36 kg (3 pounds) or more of fluid other than mineral oil and whose PCB concentration is not established, is a PCB Transformer (i.e., ≥500 ppm). If the date of manufacture and the type of dielectric fluid are unknown, any person must assume the transformer to be a PCB Transformer.
- A capacitor manufactured prior to July 2, 1979, whose PCB concentration is not established contains ≥500 ppm PCBs.
- A capacitor manufactured after July 2, 1979, is non-PCB (i.e., <50 ppm PCBs).
- If the date of manufacture is unknown, any person must assume the capacitor contains ≥500 ppm PCBs.

Water Regulations

33 USC 1251 et seq., The Clean Water Act (CWA)

EPA has established water quality criteria for certain compounds that define levels to protect human health and aquatic life. The Clean Water Act and its amendments prohibit discharging pollutants from a point source without a National Pollutant Discharge Elimination System (NPDES) permit. These permits include conditions to protect water quality. EPA authorizes states to issue and monitor compliance with these permits. The Clean Water Act also directs EPA to establish technology-based standards, known as Best Available Technology (BAT) requirements to prevent discharges of harmful amounts of pollutants.

Stormwater from certain industries and municipalities is also considered a point source of pollution that requires NPDES permitting[13]. PCBs from various sources that are deposited on land and washed into storm drains would be regulated under these stormwater permits. EPA's stormwater regulations establish two phases for the stormwater permit program:

- Phase I stormwater permits cover discharges from certain industries, construction sites involving five or more acres, and municipalities with a population of more than 100,000.
- Phase II stormwater permits cover all municipalities located in urbanized areas and construction sites between one and five acres. The EPA rule also requires an evaluation of cities outside of urbanized areas that have a population over 10,000, to determine if a permit is necessary for some or all of these cities.

National Recommended Water Quality Criteria[14]

PCBs are a Priority Pollutant under the CWA. EPA has national recommended water quality criteria for the protection of aquatic life and human health in surface water for about 150 pollutants. These criteria are published pursuant to Section 304(a) of the Clean Water Act and provide guidance to states. For aquatic health the chronic freshwater criterion is 0.014 ug/L and 0.03 ug/L for saltwater. For human health the criteria are 0.000064 ug/L both for the consumption of water and organism and for the consumption of organism only.

National Toxics Rule (40 CFR 131.36).[15]

The National Toxics Rule promulgated chemical-specific numerical criteria for priority toxic pollutants for 14 states to bring them into compliance with requirements of section 303(c)(2)(B)

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[13] Department of Ecology. How is Stormwater Regulated? Available at: http://www.ecy.wa.gov/programs/wq/stormwater/municipal/howregulated.html

[14] US EPA. National Recommended Water Quality Criteria. Available at: http://water.epa.gov/scitech/swguidance/standards/current/index.cfm (accessed 9 June 2011).

[15] US EPA. Water Quality Standards; Establishment of Numeric Criteria for Priority Toxic Pollutants; States' Compliances. 57 FR 60848. Available at: http://water.epa.gov/lawsregs/rulesregs/ntr/index.cfm (accessed 21 Oct 2013).

of the CWA. This rule became effective in 1993. The criterion for PCBs for both freshwater and marine water is 0.00017 ug/L, which has a fish tissue equivalent of 5.304 ug/kg.

42 USC 300f et seq., Safe Drinking Water Act (SDWA)

The Safe Drinking Water Act specifies water quality standards for drinking water. The National Primary Drinking Water regulations under the SDWA apply to public water systems with at least 15 service connections or more than 25 individuals for more than 60 days per year.

The SDWA sets two drinking water standards. The Maximum Contaminant Level Goal (MCLG) is a non-enforceable health goal. The Maximum Contaminant Level (MCL) is the legally enforceable standard. Water systems must reduce levels of the contaminant as close to the MCLG as feasible, considering technology, treatment techniques, and costs. For PCBs the MCLG is zero and the MCL is 0.0005 mg/L (ppm)[16]

Air Regulations

42 USC 7401, Clean Air Act and Amendments

PCBs are regulated under Section 112 of the Clean Air Act as Hazardous Air Pollutants (HAPs).

Regulation under Section 112 of the Clean Air Act requires major sources of HAPs to meet standards based on Maximum Achievable Control Technology (MACT). These standards must require the maximum degree of emission reduction that the EPA determines to be achievable by each particular source category. Different criteria for MACT apply for new and existing sources. For existing major sources, MACT is defined as the technology used to control emissions at the top 12% of facilities within the same source category. Eight to nine years after MACT is implemented, EPA is required to conduct a residual risk analysis. If the "residual risk" for a source category does not protect public health with "an ample margin of safety," the EPA must promulgate health-based standards for that source category to further reduce HAP emissions.

PCBs are one of several substances listed in Section 112(c)(6) of the Clean Air Act, which requires EPA to “list categories and subcategories of sources assuring that sources accounting for not less than 90 per centum of the aggregate emissions of each such pollutant are subject to standards.” EPA published this listing in a Federal Register notice in June 1997.[17] Various forms of waste incineration were identified as the primary industrial source categories emitting PCBs.

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[16] US EPA. List of Contaminants & their MCLs. Available at: http://water.epa.gov/drink/contaminants/index.cfm#List (accessed 9 June 2011).

[17] Notice of draft source category listing for section 112(d)(2) rulemaking pursuant section 112(c)(6) requirements. 62 FR 119 (20 June 1997). p. 33625 - 33638.

Waste, Hazardous Substance & Cleanup Regulations

42 USC 6901 et seq., Resource Conservation and Recovery Act (RCRA)

Under the authority of the Resource Conservation and Recovery Act of 1976, EPA implements regulations pertaining to solid waste, hazardous waste and underground storage tanks (40 CFR parts 239-299).

Hazardous wastes are managed under RCRA from their point of generation to their proper disposal or treatment. There are three means under RCRA of identifying if a waste is hazardous: (1) if the waste is specifically listed as hazardous, (2) if it exhibits hazardous characteristics, as determined by a Toxicity Characteristic Leaching Procedure (TCLP) test or 3) exhibits the characteristics of ignitability, corrosivity or reactivity.

Wastes are given waste codes based on their sources or specific properties. D codes are for characteristic wastes. P and U waste codes are assigned to discarded chemical products. F codes are for non-specific and K codes are for specific industrial sources.

Standards for the Management of Used Oil (40 CFR Part 279)[18] include management standards for generators, transporters, processors, burners, and marketers of used oil containing PCBs at less than 50 ppm. Used oil containing more than 50 ppm is regulated under TSCA (40 CFR part 761).

RCRA allows EPA to permit facilities to Treat, Store and Dispose of hazardous waste. Additionally, RCRA grants EPA the authority to require cleanup of any releases of hazardous waste to the environment from a permitted or interim status facility through the RCRA Corrective Action Program. RCRA cleanup sites regularly also have PCB contamination. Generally these sites come under both programs for approval of the waste cleanup.

42 USC Part 103, Comprehensive Environmental Response, Compensation and Liability Act (CERCLA)

CERCLA, passed in 1980, is the primary federal authority used to regulate and cleanup historic hazardous waste sites. The statute and implementing regulations establish procedures for the long-term remediation of such sites, but also provides authority to clean up hazardous waste sites in need of immediate action. The law has subsequently been amended, by the Superfund Amendments and Reauthorization Act of 1986 (SARA), and the Small Business Liability Relief and Brownfields Revitalization Act of 2002.

Under CERCLA Section 103, releases of hazardous substances are required to be reported to the National Response Center if they exceed the Reportable Quantity (RQ) for that substance, which

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[18] US EPA. Standards for the Management of Used Oil. 40 CFR Part 279. Available at: http://www.access.gpo.gov/nara/cfr/waisidx_07/40cfr279_07.html (accessed 10 June 2011).

is 1 pound for PCBs.[19] CERCLA implements TSCA as an Applicable or Relevant and Appropriate Requirements (ARAR), without need for separate approval under TSCA for PCB waste disposal.

42 USC Part 116, Emergency Planning and Community Right-to-Know Act (EPCRA)

EPCRA, or SARA Title III, is intended to protect public health and the environment from hazards posed by toxic chemicals by providing information about the presence of toxic chemicals in communities. The Act, passed in 1986, creates the annual hazardous chemical inventory as well as the toxics release inventory (TRI).

Under Section 302 of EPCRA, facilities that manufacture, process or use chemicals on the list of Extremely Hazardous Substances (EHSs) must report the presence of those chemicals above a certain quantity, known as the Threshold Planning Quantity (TPQ).

Section 313 of EPCRA establishes the Toxics Release Inventory (TRI). Under the TRI, the release or waste management of toxic chemicals by certain industries must be reported if the quantity of a chemical that is manufactured, processed, or otherwise used during the calendar year exceeds the reporting threshold. For most TRI chemicals, the thresholds are 25,000 pounds manufactured or 10,000 pounds otherwise used.[20] The reporting threshold for PBTs is lower and is 10 lbs for PCBs.

Worker & Product Safety Regulations

84 USC 1590 et seq., Occupational Safety and Health Act (OSHA)

The Occupational Safety and Health Act allows the Occupational Safety and Health Administration (OSHA) to set protective regulatory limits on the amount or concentration of a substance in the air in workplaces. These limits, called Permissible Exposure Limits (PELs) are based on an average exposure over an 8 hour workday, or a Time-Weighted Average (TWA).[21] OSHAs PEL is 1,000 µg/m^3 for PCBs containing 42% chlorine (CAS 53469-21-9) and 500 µg/m^3 for compounds containing 54% chlorine (CAS 11097-69-1). The PELs include “skin” to refer to the contribution to overall exposure through skin. These are based on the prevention of liver injury in exposed workers.

The National Institute for Occupational Safety and Health (NIOSH) recommends a 10-hour TWA of 1 µg/m^3 based on the minimum reliable detectable concentration and the potential

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[19] US EPA. List of Lists: Consolidated List of Chemicals Subject to the Emergency Planning and Community Right-To-Know Act (EPCRA) and Section 112(r) of the Clean Air Act. EPA 550-B-01-003. October 2012. Available at: http://www.epa.gov/emergencies/tools.htm#lol (accessed 21 October 2013).

[20] US EPA. List of Lists: Consolidated List of Chemicals Subject to the Emergency Planning and Community Right-To-Know Act (EPCRA) and Section 112(r) of the Clean Air Act. EPA 550-B-01-003. October 2012. Available at: http://www.epa.gov/emergencies/tools.htm#lol (accessed 21 October 2013).

[21] Occupational Safety and Health Administration. Permissible Exposure Limits (PELs). Available at: https://www.osha.gov/dsg/topics/pel/ (accessed 21 October 2013).

carcinogenicity of PCBs.[22] The NIOSH Recommended Exposure Limit (REL) was based on reproductive effects in animal models, carcinogenic effects, and prevention of liver injury. NIOSH also recommends that all workplace exposures be reduced to the lowest feasible level.

Washington State Laws and Regulations

Water Regulations

Chapter 90.48 RCW Water Pollution Control

Chapter 173-200 WAC Water quality standards for groundwaters of the state of Washington.

This regulation is intended to protect current and future beneficial uses of groundwater from deleterious effects, prevent degradation of waters of outstanding value, and actively maintain the higher quality of waters that exceed water quality criteria.

Chapter 173-201A WAC Water quality standards for surface waters of the state of Washington.

This regulation institutes narrative and numeric criteria for surface water quality, an anti-degradation policy, and use-based protection measures.

Chapter 70.142 RCW Chemical Contaminants and Water Quality

This law allows the State Board of Health to establish standards for allowable concentrations of chemical contaminants in public water supplies.

Chapter 246-290 WAC Water quality standards for groundwaters of the state of Washington

This regulation establishes regulatory requirements applicable to public drinking water supplies.

Multiple Statutes – Chapters 90.48, 70.105D, 90.70, 90.52, 90.54 and 43.21 RCW

Chapter 173-204 WAC, Sediment Management Standards

Enacted in 1991, this chapter establishes marine, low salinity and freshwater surface sediment management standards. The purpose of this chapter is to reduce health threats to humans and biological resources resulting from surface sediment contamination.[23]

Air Regulations

Chapter 70.94 RCW Washington Clean Air Act

The Washington Clean Air Act authorizes the Department of Ecology to develop and implement regulations that are needed to control air pollution.

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[22] Polychlorinated Biphenyls (PCBs). Current Intelligence Bulletin 45 (1986) http://www.cdc.gov/niosh/docs/86-111/ (accessed 21 October 2013)

[23] WAC 173-204-320. Table 1, Marine Sediment Quality Standards. Available at: http://apps.leg.wa.gov/wac/default.aspx?cite=173-204-320 (accessed 10 June 2011).

Chapter 173-460 WAC Controls for new sources of toxic air pollutants

Under this chapter, Ecology reviews new sources of toxic air pollutants and establishes emission control requirements that are needed to prevent air pollution that may impact human health and safety. This chapter, enacted in 1991, requires new sources to implement Best Available Control Technology for toxics (BACT). The owner or operator of a new toxic air pollutant source must also conduct an Acceptable Source Impact Level (ASIL) analysis for toxic air pollutants. When performing these assessments, the owner/operator must quantify the amount of toxic air pollutant likely to be emitted from the new source and estimate ambient air concentrations that might result from those emissions. Ambient air concentrations are estimated using air quality models. The model air concentrations are then compared to regulatory screening values (ASIL). If the modeled concentration exceeds the ASIL screening levels, the owner/operator must perform a comprehensive review using a more sophisticated model and, if necessary, apply additional emission controls. Violators may be subject to enforcement actions, civil penalties and/or criminal charges such as gross misdemeanor. Twelve PCB congeners and general PCBs (CAS 1336-36-3) are regulated as Toxic Air Pollutants (TAPs).[24]

Waste, Hazardous Substance & Cleanup Regulations

Multiple Statutes - Chapter 70.105 RCW and parts of chapters 70.105A, 70.105D and 15.54 RCW

Chapter *173-303 WAC, Dangerous Waste Regulations*

These regulations meet the requirements of the Federal Resource Conservation and Recovery Act (RCRA) and the Department of Ecology is authorized by the US EPA to implement RCRA within the state. Therefore, all the requirements identified under RCRA are also part of the state’s dangerous waste regulations. In addition, this chapter also contains specific state-only dangerous waste requirements for any waste generated or disposed of within the state. The dangerous waste regulations require a generator of dangerous waste to designate that waste according to the regulations and follow the associated requirements for waste of that designation.

Washington State has specific requirements that pertain to toxicity and persistent criteria. Halogenated organic compounds like PCBs are considered persistent in the dangerous waste regulations.

WAC 173-303-100(5) requires waste to be evaluated for mammalian and aquatic toxicity and WAC 173-303-100(5)(b)(i) provides a process to designate a specific waste stream based upon the toxicity of the individual components. In this evaluation, toxicity must be considered with other waste constituents to determine if the waste stream designates as a state-only toxic waste

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[24] WAC 173-460-450. Table of ASIL, SQER and de minimis emission values. Available at: http://apps.leg.wa.gov/WAC/default.aspx?cite=173-460-150 (accessed 10 June 2011).

and assigned the waste codes of WT02 as dangerous waste or WT01 as EHW (extremely hazardous waste).

In Washington State, PCB waste may be regulated as a state criteria dangerous waste or as a state listed dangerous waste. The Washington dangerous waste regulations separate wastes into four categories:

- Characteristic wastes.
- Criteria wastes.
- Discarded chemical products.
- Non-specific and specific industrial sources.

Wastes are given waste codes based on their sources or specific properties as discussed in the Federal Regulations section on RCRA. W codes are for state-only wastes.

PCBs as a state criteria dangerous waste

Since PCBs meet the definition of Halogenated Organic Compound (WAC 173-303-040), wastes containing PCBs (other than state listed PCB wastes discussed below) must be evaluated for state persistence. This requirement has been in place since early adoption of the State's Dangerous Waste Regulations in 1982. At 100ppm PCB, a waste would be considered a persistent dangerous waste (waste code WP02). A few examples of PCB persistent waste include: caulking, tar and rubber stripping at airport runways. If the PCB concentration exceeds 10,000 ppm (waste code WP01), the waste is recognized as an extremely hazardous waste pulling on additional requirements and/or prohibitions on the management of that waste.

PCBs as a state listed dangerous wastes

To address the management of the most problematic PCB wastes- liquid PCBs in transformers, bushings and capacitors- RCW 70.105.105 gives the authority to Ecology to regulate PCBs as a dangerous waste. In 1985, Ecology amended its Dangerous Waste Regulations to include certain PCB wastes (waste code WPCB). This is a source specific group of waste products that only applies to discarded transformers, capacitors or bushings containing 2 ppm PCB or greater (except when drained of all free flowing liquid) and to the following wastes generated from the salvaging, rebuilding, or discarding of transformers, capacitors or bushing at 2 ppm PCB or greater: cooling and insulation fluids, cores, and core papers.

Exclusions

1) -071(3)(k). PCB exclusion. One may manage a state only PCB waste under specific TSCA regulations instead of the state Dangerous Waste regulations. The waste would become excluded from the state Dangerous Waste regulations. Often, listed-WPCB dangerous wastes are managed under this exclusion.

2) -073 “Special waste exclusion”. If the waste meets the definition of special waste (WAC 173-303-040), then some listed WPCB wastes and some state only persistent criteria waste (due to PCB) can be managed this way.

Chapter 70.95I RCW Used oil recycling

Used oil is conditionally regulated under the dangerous waste regulations as long as 1) it is not contaminated with chlorinated solvents or PCBs and 2) it is managed appropriately. If used oil is not contaminated, it may be recycled or burned for energy recovery. Used oil with 2 ppm or greater PCBs is prohibited from being managed as used oil under the Dangerous Wastes used oil regulations when burned for energy recovery. WAC 173-303-515 contains management standards for used oil.

This statute requires local governments to include an element in their hazardous waste plans enumerating how they will collect used oil. It also requires used oil recycling containers and educational information about used oil to be provided at any business that sells above 1,000 gallons of lubricating oil to consumers (500 gallons in a city with an approved used oil recycling element in their hazardous waste plan).

Chapter 70.105D RCW Hazardous Waste Cleanup – Model Toxics Control Act

Chapter 173-340 WAC, Model Toxics Control Act – Cleanup

Chapter 70.105D RCW establishes the framework and authority for the development of a program dealing with the cleanup of sites contaminated with toxic chemicals. The MTCA Cleanup Regulation, issued in 1991, establishes procedures and standards for the identification, investigation and cleanup of facilities contaminated with hazardous wastes.

MTCA provides several methods for setting cleanup standards. Under MTCA Method A, pre-calculated protective cleanup levels are available in tables within the regulation for use at relatively simple sites.

Method B is the universal method for determining cleanup levels for all media at all sites. A target cancer risk level of one in one million (10^{-6}) is used when calculating cleanup levels under Method B. Toxicity equivalency factor (TEF) methodology (Van Den Berg *et al*. 2006) may also be used to evaluate the toxicity of PCBs, where the mixture is considered a single hazardous substance.[25]

Method C cleanup levels are established when cleanup levels established under Method A or B may be impossible to achieve or may cause greater environmental harm.

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[25] Department of Ecology. Evaluating the Toxicity and Assessing the Carcinogenic Risk of Environmental Mixtures Using Toxicity Equivalency Factors. Available at: https://fortress.wa.gov/ecy/clarc/FocusSheets/tef.pdf (accessed 16 June 2011).

Chapter 173-360 WAC, Underground Storage Tank Regulations

The Department of Ecology implements Chapter 90.76 RCW, Underground Storage Tanks, in order to protect human health and the environment from leaking underground storage tanks containing petroleum and other regulated substances. No underground storage tank systems, within the parameters of this chapter's scope, may operate without a valid permit. This chapter sets forth performance standards for underground storage tanks. Tanks must be monitored and owners and operators are required to comply fully with testing and inspection. Releases into the surrounding environment must be immediately reported to Ecology and appropriate cleanup and containment measures must be taken. Under most circumstances, MTCA cleanup standards apply to the remediation of releases from leaking underground storage tanks. This chapter was adopted in 1990 and violators face fines of up to $5,000 dollars per day per violation.

Worker & Product Safety Regulations

Chapter 49.17 RCW Washington Industrial Safety and Health Act

Chapter 296-841 WAC Airborne Contaminants

This chapter specifies Permissible Exposure Limits (PELs) of 1,000 µg/m^3 for PCBs containing 42% chlorine (CAS 53469-21-9) and 500 µg/m^3 for compounds containing 54% chlorine (CAS 11097-69-1) that mirror the federal OSHA requirements (see Federal Regulations).

They also specify Short-Term Exposure Limits (STEL) of 3,000 µg/m^3 for PCBs containing 42% chlorine (CAS 53469-21-9) and 1,500 µg/m^3 for compounds containing 54% chlorine (CAS 11097-69-1). STELs refer to 15 minute exposure periods.

Taxes

Chapter 82.21 RCW Hazardous substance tax – model toxics control act

Chapter 458-20-252 WAC Hazardous substance tax and petroleum product tax

PCBs are taxed under the Hazardous Substance Tax.

This law places a tax on the first possession of hazardous substances in Washington. The Department of Ecology determines which substances are subject to the tax. The tax applies to petroleum products, pesticides, and certain chemicals. There are currently over 8,000 different hazardous substances identified as being subject to the tax. The tax rate is .007 of the wholesale value of the product. Funds are distributed to the Department of Ecology to help clean up, manage and prevent solid and hazardous waste in the state of Washington.[26] The tax does not apply to components or contaminants, such as inadvertently generated PCBs in other products.

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[26] Washington State Department of Revenue. Hazardous substance tax. Available at: http://dor.wa.gov/content/findtaxesandrates/othertaxes/tax_hazard.aspx

The tax applies to

- Petroleum products.
- Substances designated as hazardous under the federal Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA – see Federal Regulations).
- Any pesticide product required to be registered under the Federal Insecticide, Fungicide and Rodenticide Act (FIFRA – see Federal Regulations).
- Other substances or categories of substances designated by Ecology.

Select Regulations in Other US Jurisdictions

Maine

http://www.mainelegislature.org/legis/statutes/38/title38sec419-B.html

This law requires public utilities to remove transformers with more than 50 ppm PCBs. There are earlier deadlines for transformers within 100 feet surface water, elementary school or secondary school.

Select International Regulations

Most countries have prohibited the commercial manufacturing of PCBs.

Stockholm Convention[27]

The Stockholm Convention on Persistent Organic Pollutants is a global treaty that aims to protect human health and the environment from the effects of persistent organic pollutants. The Convention has a range of control measures to reduce and, where feasible, eliminate the release of POPs. The Convention also aims to ensure the sound management of stockpiles and wastes that contain POPs.

The Convention was signed in 2001 and entered into force in 2004. The US is a signatory, but has not ratified the Convention, so is not a Party to it.

PCBs are one of the 12 initial POPs under the Stockholm Convention. The parties to the Convention are required to eliminate the use of PCBs in existing equipment by 2025 and ensure environmentally sound waste management of them by 2028. Each country is expected to develop inventories and identify contaminated sites. To help stakeholders achieve the goals in the Stockholm Convention they created the PCB Elimination Network (PEN) as a voluntary

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[27] http://chm.pops.int/Home/tabid/2121/mctl/ViewDetails/EventModID/871/EventID/407/xmid/6921/Default.aspx

collaborative arrangement to promote and facilitate information exchange to support the obligations of Stockholm Convention on environmentally sound management of PCBs. Canada

Sweden

In addition to banning the use of PCBs, Sweden has required inventorying buildings with PCB-contaminated materials, such as caulk, and removing them[28]. The initial program in 1980 was voluntary, with information to homeowners and voluntary work by the Ecocycle Council to inventory and remediate buildings. In 2007 an ordinance passed to make owners required to inventory and remediate structures built or renovated between 1956 and 1973 (when PCBs in open applications were banned in Sweden). Working at the local city level, the inventories have mostly been completed and the remediation is expected to be finished by 2016.

Canada

PCBs were never manufactured in Canada and most PCBs used in Canada were imported from the US. Like the US, Canada banned the import, manufacture, and sale of PCBs in 1977 and allowed PCB equipment to be used until the end of its service life in the original regulation. The release of PCBs to the environment was made illegal in 1985.

One significant difference between US and Canada regulations is that Canada does not regulate mono- and di-chlorinated biphenyls[29]. PCBs with more than 2 chlorines are on Schedule 1 of the Toxics Substances list in Canada.

The Canadian Environmental Protection Act (CEPA) covers PCB Regulations.[30] Many of the regulations are similar to TSCA, such as a general limit of 50 ppm. One major difference is the Canadian regulations set deadlines for the phase-out of PCBs in use:

- Dec. 31, 2009 for equipment containing more than 500 ppm PCBs
- Dec. 31, 2009 for equipment containing 50-500 ppm PCBs within 100 meters of a drinking water plant, food or feed processing plant, school, hospital, or care center
- Dec. 31, 2025 for other equipment containing 50-500 ppm PCB
- Dec. 31, 2025 for light ballasts and pole-top electrical transformers

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[28] Johansson, Niklas. PCBs in Schools: International Experience: Inventory, Remediation, and Outcomes. EPA webinar series PCBs in Schools Session II. April 28, 2014.

[29] http://www.ec.gc.ca/lcpe-cepa/default.asp?lang=En&n=0DA2924D-1&wsdoc=4ABEFFC8-5BEC-B57A-F4BF-11069545E434 (accessed 13 July 2014).

[30] SOR/2008-273 available at http://www.ec.gc.ca/bpc-pcb/default.asp?lang=En&n=663E7488-1 (accessed 21 October 2013).

Economic analysis

Cost Estimates of Various PCB Reduction Methods

Chapter 173-333 WAC, the Persistent Bioaccumulative Toxic Chemical rule, requires Ecology to follow a specific process while developing Chemical Action Plans (CAPs). Specifically, Chapter 173-333-420(1) (f) (iii) (B) requires Ecology to consider the potential economic and social impacts of implementing the recommendations within the CAPs. In this section, we present estimates for the likely costs associated with the recommendations within the polychlorinated biphenyls (PCBs) CAP. The majority of the initial costs would accrue to Ecology because of the amount of scoping and research necessary to grasp the extent of PCB contamination in the state of Washington.

Economic analysis, like all analytical exercises, depends on the quantity and quality of data. As described throughout the document, PCBs are ubiquitous in the environment, which dictates that the scope of activities taken to reduce PCBs involve the combined efforts of public and private actors across numerous sectors of the economy throughout Washington. To the greatest extent possible given data limitations, we estimated costs to Ecology and the entities directly impacted by the recommended actions.

As expected, we lack data on various processes involved in remediating PCB-contaminated buildings, equipment, and habitats. In some cases, representative data is not available. In other cases, we determined that systemic attributes of entities affected by the recommendations, especially public and quasi-public entities, hindered our ability to reasonably assume that past costs serve as a basis for future costs. We determined that a scarcity of data in some areas required us to consider certain figures as anecdotal that fail to describe what an average entity in Washington affected by the recommendations might face. When we faced questions concerning the availability or reliability of data, we proceeded with a qualitative analysis. That is, we focused more on identifying the variables that drive the costs to those affected by recommendations.

As evident from the recommendations below, we anticipate building databases concerning PCBs based upon data submitted from entities and collected by Ecology. This new data would enable us to revisit the qualitative cost estimates presented in this section. Accordingly, it is important to remember that qualitative analysis is suggestive as opposed to representative.

The economic analysis section proceeds with a statement of each recommendation, specific goals, related background, and analysis of likely costs.

Historic PCB-Containing Building Materials

1. Identify PCB-containing lamp ballasts in schools and other public buildings. Encourage replacement with more energy efficient PCB-free fixtures.

Goal: Remove remaining PCB lamp ballasts from schools and other publicly owned buildings.

Prior to 1979, PCBs were widely used in fluorescent lamp ballasts, including those in use at schools and other public buildings. The pre-1979 ballasts have likely outlived their useful lives and are at high risk for failing (dripping, smoking, and catching fire). Ballast failures can expose children and others that frequently use public school buildings to concentrated PCB oils and elevated PCBs in the air.

There is no easily accessible source of information on how many buildings are of the age and construction type likely to have PCB-containing light ballasts. Because children are more sensitive to PCBs and school buildings are typically publicly owned, Ecology recommends prioritizing public schools. The first step towards implementing this recommendation is to conduct a survey of the 295 school districts in Washington to identify how many of the approximately 9,000 school buildings are likely to have PCB-containing light ballasts. Other public buildings will be surveyed as time and resources allow. Ecology would use the survey results to construct a database with information on construction and renovation dates and activities of schools (and other public buildings if possible). Ecology would first use the database to identify schools where PCB-containing light ballasts are likely still in use. Lamp ballasts with PCBs can then be identified through visual inspection.

Ecology anticipates that this recommendation would require an additional FTE at Environmental Specialist 3 (ES3) level. One FTE at ES3 level would cost $90,931 annually. We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Ecology envisions this person would split time between working towards this recommendation (0.75 FTE) and working towards Recommendation 5 (0.25 FTE). Because the staff person would work on two goals, we estimate the cost based on the time spent (.75 FTE) on this recommendation as $68,198 annually. Ecology anticipates that work on this recommendation could span two years (FY2016- FY2017) for a total estimated cost of $136,396.

We do not have confident cost estimates for replacing light ballasts in schools or other public buildings. Estimating the cost of replacing the light ballasts to school districts is not possible without knowing how many schools might contain PCB-containing light ballasts, the condition of the light ballasts, and the extent of the problem within each building. However, replacing old, potentially dangerous light ballasts not only reduces the risk of exposure for children and others that use the school buildings frequently, it also reduces energy costs. Accordingly, it makes sense to combine PCB removal with initiatives to increase energy efficiency rather than create a new program just for removal of PCB-containing ballasts.[31]

Public money should be used to remove PCB-containing lamp ballasts from schools and other public buildings. Schools with PCB-containing lamp ballasts will be provided with information about the importance of removing these ballasts and referred to the Office of the Superintendent of Public Instruction (OSPI) (or Washington State Department of Health and other available resources) to replace these fixtures with more energy-efficient lighting. Ecology has requested $200,000 in public money to assist with some of this work.

2. Develop and promote best management practices to contain PCBs in building materials currently in use and those slated for remodel or demolition.

Goal: Reduce exposure to people from PCBs in historic building materials and prevent PCBs in building materials from getting into stormwater.

Historically, PCBs were used at high levels in some caulks and paints. Studies in other areas have shown the widespread occurrence of PCB-containing caulk in buildings from about 1950-1980, especially masonry buildings. Smaller sampling efforts in Washington support this conclusion.[32] Accordingly, developing best management practices and other materials to provide guidance for renovation and demolition of buildings that contain PCB materials would help to prevent the release of PCBs into the environment.

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[31] Since 2009, the Legislature has provided money to the Office of the Superintendent of Public Instruction (OSPI) and the Department of Commerce to support energy efficiency measures in schools and other public buildings. If the grant programs are not funded, the legislature could establish a fund to help offset the costs of replacing PCB-containing lamp ballasts.

[32] For example, studies suggest that buildings with PCB-containing materials exist in the Duwamish basin. In addition, source tracing from cleanup efforts in Tacoma revealed PCBs in sidewalk and building caulk.

The first step toward preventing PCBs in building materials from getting into the environment is to compile, compose, and distribute information concerning best management practices for containment of PCB-containing materials. Based on available data in Washington, other government programs, and scientific literature, Ecology would develop BMPs for containing PCBs to prevent exposure during the life of the building and during remodeling or demolition. Ecology should also provide education and outreach on BMPs to local governments and those in the building trades.

Ecology estimates that developing BMPs would require an additional FTE of an Environmental Specialist 3 (ES3) for a three-year period. We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). One FTE at ES3 would cost $90,931 annually. Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

While working on the BMPs, Ecology would also work to compile existing information into a PCB Source Control Guidance Manual to aid Local Source Control work. A number of urban waters programs around the northwest have performed PCB source identification work. However, to date, the lessons learned from each of these programs have not been synthesized and summarized for the benefit of future pollution prevention efforts at the state and local levels.

Ecology estimates that work on the best management practices and source control manual would last approximately three years (FY2016-FY2018) and result in total staff costs of $272,793.

3. Assess schools and other public buildings for the presence of PCB-containing building materials.

Goal: Reduce children's exposure to PCB-containing building materials.
Goal: Prevent PCBs in building materials from getting into stormwater.

Many buildings constructed prior to the ban of PCBs include materials, such as caulk, paint, and light ballasts that often contain high levels of PCBs. Industrial buildings, including schools, are more likely to contain PCB-contaminated materials than residential buildings. Other states have found high levels of PCB contamination in schools. Because children are more sensitive to PCBs and school buildings are typically publicly owned, Ecology recommends assessing public schools for possible PCB contamination first and expanding the effort to include other buildings, as appropriate.

To our knowledge, school districts in Washington have not systematically tested schools for PCBs. Schools built prior to 1980 are more likely to contain material with PCBs. The first step in assessing public school buildings that contain PCB material is to construct a centralized database based on information provided by school districts. The database would contain information on the date of construction and dates of renovation for each school building in Washington. The database would serve as a mechanism to identify schools, based on construction date, that require testing for PCBs. Initial testing would include visual inspections and then physical testing where appropriate. Ecology would use the database and test results to determine the scope of the problem in Washington and plan accordingly. A similar approach would be used to assess other public buildings once the assessment of schools is complete, and as resources allow.

Ecology estimates that the person retained to compile information on PCB light ballasts in schools would compile the database for building materials, as well. Ecology anticipates that two Environmental Specialist 3 (ES3) positions in other recommendations will merge tasks in FY2018:

- The 0.75 FTE at Environmental Specialist 3 (ES3) level at $68,198 annually would spend two years (FY2016-FY2017) focusing on light ballasts (Recommendation 1).
- The 0.25 FTE at the ES3 level at $22,733 annually would spend two years (FY2016-2017) focusing on electrical equipment (Recommendation 5).
- These positions would shift their database efforts to include other building materials at schools.

Ecology anticipates that work on this recommendation could span four years (FY2018- FY2021) for a total estimated cost of $363,724.

We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Ecology understands the time and budget constraints facing school districts across the state. However, this recommendation would not require school districts to generate new reports or information. We assume that school districts have information concerning construction and renovation of school buildings from routine recordkeeping, operations, and maintenance documents. Therefore, we do not expect a cost to school districts to submit documents to Ecology for the database beyond minimal expenditures of time and resources to submit records to Ecology.

After compiling the database and conducting initial testing, Ecology would work with school districts to plan and coordinate remediation efforts at schools that have PCB-contaminated materials. There is no one size fits all approach to remediation projects for buildings containing PCBs (Environmental Health & Engineering, 2012). Depending on the extent of contamination, schools decide whether to pursue abatement (reducing the amount of PCBs in building materials permanently) or mitigation (controlling exposure) procedures. Regardless of the remediation technique, schools would need to work with local health agencies, Ecology, and EPA to meet removal criteria and follow hazardous waste regulations.

Estimating the cost of remediating school buildings in Washington is not possible without knowing the scope (number of schools and extent of remediation needed) of the problem. The number of school buildings and extent of work necessary to bring a building in compliance would determine bids from contractors and others involved in remediation activities. In addition, remediation activities generally involve mandated testing procedures, extensive planning, feasibility studies, and permitting requests. School districts might also have to explore temporarily relocating students during the initial testing/cleanup stage (depending on age of building and likelihood of PCB contamination). Because of the extensive nature of remediation projects, we feel that a database is appropriate to enable Ecology and school districts to narrow the scope, identify economies of scale, and prioritize remediation projects.

As mentioned above, systemic attributes of public entities make some estimates less reliable. We consider the process school districts use to price construction projects such a structural constraint. Generally, available data suggests that the cost of remediating PCB-contaminated school buildings depends on the extent of contamination and approach used by schools (abatement or mitigation) to address the problem. To our knowledge, no state has addressed PCB contamination in schools in a comprehensive manner. It appears that most schools learn of PCB contamination by miscellaneous tests conducted prior to unrelated renovation work, and must react quickly to bring exposure levels below EPA guidelines. This creates immediate financial stress on local/state agencies responsible for public health, school facilities, etc. Further, school districts face unique budget constraints and absorb costs differently than owners of private buildings.

Schools generally face administrative procedures (feasibility studies, budget requests, and limited window for large remediation projects) that increase the overall cost of projects. However, it is difficult to compare how school districts determine costs for certain projects, especially when comparing school districts in different regions or states. School districts in Washington form cost estimates based on the needs of schools here in Washington. In sum, existing estimates of remediation projects based solely on PCB contamination are too limited to provide a meaningful basis for comparison, at this point.

Acknowledging the above limitations, though, illustrates the need for Ecology to identify the scope of the problem here in Washington. We found estimates for remediation work at five schools in New York and two schools in Massachusetts. Estimates from remediation projects at the five public schools in New York City ranged from $3.2 million to $3.6 million (2014$) per school depending on the techniques (abatement or mitigation) used to address the PCB-contaminated areas (TRC, 2011). In 2010, an elementary school in Lexington, MA found PCB-contaminated material. The school had to close for a week while workers performed testing required by the EPA and performed preliminary cleanup work. Feasibility studies suggested that officials faced temporary solutions ranging from $3.0 million to $4.6 million (2014$) to relocate students while remediating the school (Goddard, 2010). Ultimately, officials decided to replace the school with a new $40 million building (Parker, 2014). A different school in Westport, MA also found PCB material and encountered initial costs in excess of $3 million (Wagner, 2014). Currently, the school faces additional costs ranging from $1.8 million to $7.75 million (2014$) (CGKV Architects, 2013) to remediate the PCB-contaminated material. Again, we consider the estimates from New York and Massachusetts more suggestive than representative. That said, the expenses incurred by the school districts in New York City and Massachusetts, along with the extent of activity required to remediate the structures, indicate a need to determine the scope of the problem by compiling construction dates and preliminary testing of high risk schools here in Washington.

Current Manufacturing Processes

4. Learn more about what products contain PCBs and promote the use of processes that don’t inadvertently generate PCBs.

Goal: Reduce newly generated PCBs in manufacturing processes.

Unpermitted non-point releases, such as from consumer products, are becoming increasingly important to control in order to reduce total PCB delivery. In 1982, EPA identified 70 manufacturing processes that are likely to inadvertently generate PCBs, but little else is known about this potentially large source of uncontrolled PCBs. More information is known about PCBs in pigments and dyes, which are a known source of PCBs in the environment and a problem for paper recyclers in Washington. Ecology recommends adding additional staff and funding to determine the extent of inadvertently generated PCBs in consumer products.

To accomplish the goal of reducing newly generated PCBs in manufacturing processes, Ecology would work with EPA, manufacturers, and other partners to identify products that inadvertently contain PCBs and explore available alternatives. Ecology would test identified products, and provide relevant results to the Department of Enterprise Services (DES) to assist with implementing new purchasing policies that provide a preference for products that do not contain PCBs. Ecology would also work with the Department of Health and DES to develop a focus sheet to educate purchasers and vendors in the state supply chain about the prevalence and incidence of products that inadvertently contain PCBs and concisely outlines the problem and its potential impacts.

Learning more about processes that inadvertently generate PCBs would require 1 FTE of a Natural Resource Scientist 3 (NRS3) at $116,641. We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Ecology anticipates that the NRS3 would test approximately 100 products a year. Ecology anticipates that the employee retained to perform the product testing would continue to work on this issue on an ongoing basis, with costs from FY2016 through FY2021 totaling $699,846. Additionally, at $1,000 per test, Ecology estimates annual testing costs of $100,000.

In addition to testing for PCBs, we also estimated the cost of investigating alternate processes in chemical manufacturing, to prospectively replace processes that produce PCBs as a byproduct. Specifically, we estimated the cost of initiating an alternatives assessment process for pigments and dyes, based on the Interstate Chemicals Clearinghouse (IC2) Alternatives Assessment Guide.

Ecology anticipates that producing such an “Alternatives Assessment” would cost $470,000 in FY2016, which includes hiring a consultant. Ecology plans to engage business and other concerned stakeholders to participate in the alternatives assessment process. If there are currently no alternatives that do not generate PCBs, Ecology would engage Green Chemistry Northwest or a similar institution to develop alternative processes.

Historic Electrical Equipment

5. Survey owners of historic electrical equipment, including transformers and large capacitors.

Goal: Confirm estimates of EPA-regulated electrical equipment with more than 500 parts per million (ppm) PCBs, learn what is known about electrical equipment with the PCBs greater than 2 ppm, and find out when such electrical equipment is estimated for replacement.

Electrical equipment, such as transformers and capacitors, used by utility and large non-utility industrial sites are the primary historical use of PCBs. Concentrations of PCBs in many pieces of electrical equipment are high compared to other sources, so relatively rare leaks and spills can release a significant amount of PCBs into the environment. Ecology recommends a survey of the 61 utilities and other sites that provide electricity to construct a recent and relevant database of electrical equipment that contains PCBs in Washington.

We anticipate that this recommendation would most likely affect utilities and large non-utility entities that provide electrical transmission (operate transformers or large capacitors) on site. The nature of electricity and the regulatory status of utilities presents unique challenges regarding this recommendation. Electricity is difficult to store, impossible to substitute, and a necessity to most consumers and businesses. Accordingly, each utility in the state enjoys a natural monopoly in the transmission of electricity to end users within a defined area. In exchange for monopoly status, various regulatory agencies set prices administratively based on costs incurred by utilities. The process of setting prices administratively makes it difficult, if not impossible, for us to determine the cost of individual projects and to determine how the utilities absorb costs. Regulations also require utilities to provide electricity on demand that makes a recommendation to test all existing equipment difficult to undertake without potentially disrupting service. In sum, from a practical perspective, a recommendation for further testing could prove cost prohibitive to utilities and consumers.

Prior to the ban on PCBs, manufacturers of transformers, capacitors, and other electrical equipment used oil-containing PCBs as a coolant within the housing of utility equipment. Since the ban on PCBs took effect in the United States, utilities and owners of electrical equipment have worked to identify and replace equipment that contains PCBs. According to a report submitted by the Utility Solid Waste Activities Group (USWAG, 2010) to the EPA, utilities across the country have reduced the use of equipment containing PCBs through normal maintenance and replacement procedures. A similar statement by the Northwest Public Power

Association (2010), extensive discussions with representatives from various utilities, and local news reports (Kramer, 2014), suggest that utilities across Washington have also replaced many units that contain PCBs. Currently, however, no reporting requirement exists to verify the claims of utility and a non-utility users of electrical equipment concerning retirement of equipment that contains PCBs.

Ecology believes that surveying the state's utilities and compiling a centralized database based on past, present, and future efforts by owners of electrical equipment is within reason and at minimal cost to the owners of electrical equipment. Because electrical equipment that might contain PCBs are part of a complex transmission system that requires significant time and resources to maintain, Ecology assumes that owners of electrical equipment maintain records that form the basis of routine maintenance schedules. Over the past 15 years, increased demand for energy and the interconnectedness of the electrical grid has changed the regulatory environment in such a way that providers of electricity at all levels of the transmission process must maintain certain levels of service at all times. In addition, utilities upgrade old and inefficient transmission equipment (often the equipment that might also contain PCBs) to increase efficiency and output. The need to maintain a consistent supply of energy and increase profitability, where possible, necessitates that utilities maintain extensive records. Therefore, we assume that utilities and other large non-utility owners know which pieces of equipment have PCBs or may have PCBs based on the age of the equipment.

Because the utilities have taken action to address the use of equipment that contains PCBs, Ecology believes establishing a database of current equipment containing PCBs would require minimal effort and cost to Ecology and users and electrical equipment. The survey of electrical equipment would not include small capacitors. The survey would not require additional testing or disposal of equipment by a certain date. The survey would ask owners to report what they know about detectable levels of PCBs in their equipment, using the industry standard detection level that is currently 2 parts per million (ppm), and detail when they expect all the equipment to be replaced based on their current maintenance practices. A statewide inventory will allow the state to confirm current inventories and target efforts to prevent releases of PCBs from this equipment.

Ecology anticipates that the FTE assigned to collect data on PCBs in public schools (Recommendation 1) would also assemble and enter data on electrical equipment. Ecology envisions this person would split time between working towards this recommendation (.25 FTE on this goal) and working towards Recommendation 1.

One FTE at Environmental Specialist 3 (ES3) level would cost $90,931 annually. We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Because the staff person would work on two goals, we estimate the cost based on the time spent (.25 FTE) on this recommendation as $22,733 annually. Ecology anticipates that work on this recommendation could span two years (FY2016-FY2017) for a total estimated cost of $45,466.

Multi-Source

6. Expand environmental monitoring to identify any new areas requiring cleanup.

Goal: Find areas with highly concentrated PCBs and clean them up to prevent the wider release of PCBs.
Goal: Find out more about the distribution of PCBs in Washington to prioritize future actions.

Ecology should expand environmental monitoring of water, fish tissue, and sediment to identify PCB hot spots, such as the recently found historical landfill of electrical equipment on Bradford Island in the Columbia River. Historical disposal practices at Bradford Island near Bonneville Dam on the Columbia River contaminated resident fish with high levels of PCBs. Levels were high enough to prompt Oregon and Washington to issue a joint fish consumption advisory for select species from Bonneville Dam upstream to McNary Dam. A number of fish sampling efforts in the Columbia basin (CRITFC, Hanford Corridor Study and Ecology's Freshwater Fish Contaminant Monitoring Program) found other areas (mid-Columbia and Snake River) with elevated levels of PCBs and a number of hydroelectric facilities.

To our knowledge no work has been done to identify sources of contaminants in these areas. The proposed monitoring would conduct sampling to determine if contaminant sources exist in areas with a focus on hydroelectric facilities. Ecology may also use mapping and historical information on the location of potential sites, such as landfills, industrial sites, railroad switching yards, etc., to find potential sites. Newly identified sites would receive priority for clean up using existing procedures. In addition to identifying new hot spots, Ecology would continue its trend monitoring to show changes in PCBs in the environment and biota over time.

Air deposition is a potentially significant pathway for PCBs to move into the environment. Ecology proposes to investigate monitoring air deposition to assess the relative importance of this pathway.

Ecology estimates that the workload generated by Recommendation 6 would require the addition of three FTE. Ecology estimates that it would take 1 FTE Natural Resource Scientist 3 (NRS3) to identify new hot spots and implement necessary testing procedures. In addition, Ecology estimates that effectively monitoring hot spots would require an Environmental Specialist 2 (ES2) FTE to provide support for testing. The addition of one FTE at NRS3 level would cost $116,641 annually, and one FTE at the ES2 level would cost $79,513 annually. To monitor air deposition, Ecology would require an additional FTE at the Environmental Specialist 3 (ES3) level to conduct the air monitoring tests. The ES3 would cost $90,931 annually.

We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Ecology anticipates that the work of the NRS3 toward this recommendation would take 3 years, with assistance from the ES2 for 2 years. Ecology anticipates that testing to identify potential hot spots would result in lab costs of $87,192 annually for two years. Ecology anticipates that the work of the ES3 to monitor air disposition would take two years, and anticipates testing costs of $37,967 annually for two years to monitor air deposition.

Table 37 summarizes the costs associated with this recommendation.

Table 37. Summary of the costs associated with Recommendation 6.

	FY 2016	**FY 2017**	**FY 2018**
Hot spot monitoring			
1 FTE NRS3	$116,641	$116,641	$116,641
1 FTE ES2	$79,513	$79,513	
Hot spot testing	$87,192	$87,192	
Air monitoring			
1 FTE ES3	$90,931	$90,931	
Air testing	$37,967	$37,967	

7. Conduct a public educational campaign.

Goal: Provide information to residents about ways they can minimize exposure.
Goal: Raise awareness of the problems associated with current and past production of PCBs.
Goal: Educate residents to identify and address possible household sources of PCBs.

To reach the goal of raising public awareness for PCBs, Ecology and the Department of Health would work together to help residents, people who fish, schools, local governments and businesses understand the risks associated with PCB exposure and ways to reduce risks, including the availability of safer alternatives.

The public educational campaign will include perspectives on public health risks and information on where exposures to PCB are most likely. It will also include advice on how individuals can minimize their own exposures to PCBs. The campaign will include where to look for potential sources of PCBs in households, such as in old appliances, electrical equipment, and building materials. It will also teach the public how to safely remove and dispose of these materials to prevent PCB releases.

Ecology anticipates that this recommendation would require the addition of 1 FTE Communication Consultant 3 (CC3) to develop and disseminate information to businesses, purchasing agents, vendors, residents, fishers, schools, and local governments describing the hazards associated with exposure to PCBs and resources available to address the problem. This FTE would cost $92,957 each year. This work includes promotion of BMPs for containment and demolition of buildings containing PCB laden materials.

We employed Washington State employee pay grades at step H (DOP, 2014) and standard overhead cost assumptions used for legislative fiscal notes and related estimation (Ecology, 2013). Wages include the following adjustments for overhead expenses (per FTE):

- Benefits of 33.0 percent of salary
- Goods and services of $5,709 annually, or $2.74 per hour
- Travel costs of $1,394 annually, or $0.67 per hour
- Equipment costs of $1,131 annually, or $0.54 per hour
- Agency administrative overhead of 32.25 percent of salaries and benefits (Agency administrative overhead FTEs are included at 0.15 FTE per direct FTE, and are identified as Fiscal Analyst 2 and IT Specialist 2.)

Ecology anticipates that the community awareness work would continue on an ongoing basis. Estimated costs for this recommendation from FY2016 through FY2021 are $557,742.

8. Conduct a study on which PCB congeners are present in Washington residents

Goal: Learn more about PCB congeners to which Washington residents are exposed
Goal: Find out more about the distribution of PCBs in Washington to prioritize future actions

Within available resources, the Department of Health should conduct bio-monitoring of Washington residents for PCBs including PCB 11 and other inadvertently produced PCBs associated with dyes, pigments, and printing inks. Researchers would use the data to better understand 1) the extent of total human exposure from multiple potential pathways and 2) the relative contribution of these congeners to human body burden of PCBs. In addition, the data would enable researchers to estimate the statewide distribution of PCBs in Washington residents, which would provide a better baseline than national data given the elevated levels of PCBs in local fish populations and relatively high fish consumption in a number of Washington communities and regions.

Ecology believes that this sort of bio-monitoring falls within criteria already established under existing DOH programs. Accordingly, we do not anticipate new costs with this recommendation.

Summary of Economic Impacts

Table 38 presents estimated costs to Ecology to implement the recommendations above.

Table 38. Estimated costs to Ecology to implement recommendations

Recommendation		FY 2016	FY 2017	FY 2018	FY 2019	FY 2020	FY 2021
1	0.75 FTE (ES3) to survey and assess lamp ballasts in schools.	$68,198	$68,198				
2	1 FTE (ES3) to develop and promote BMPs.	$90,931	$90,931	$90,931			
3	1 FTE (ES3) to assess schools for PCB-containing building materials.			$90,931	$90,931	$90,931	$90,931
4	Conduct an alternatives assessment for pigments and dyes.	$470,000					
	1 FTE (NRS3)	$116,641	$116,641	$116,641	$116,641	$116,641	$116,641

	to look for sources of PCBs in products.						
	Lab costs for product testing.	$100,000	$100,000	$100,000	$100,000	$100,000	$100,000
5	0.25 FTE (ES3) to survey owners of electrical equipment.	$22,733	$22,733				
6	1 FTE (NRS3) to identify environmental hot spots.	$116,641	$116,641	$116,641			
	1 FTE (ES2) to assist with monitoring.	$79,513	$79,513				
	Environmental monitoring/ lab analysis.	$87,192	$87,192				
	1 FTE (ES3) for air monitoring	$90,931	$90,931				
	Sampling/lab analysis for air monitoring stations.	$37,967	$37,967				
7	1 FTE (CC3) to conduct an education campaign.	$92,957	$92,957	$92,957	$92,957	$92,957	$92,957
Total		**$1,373,704**	**$903,704**	**$608,101**	**$400,529**	**$400,529**	**$400,529**
All estimates in (2014 $)							
ES2 = Environmental Specialist 2							
ES3 = Environmental Specialist 3							
NRS3 = Natural Resource Scientist 3							
CC3 = Communications Consultant 3							

Ecology does not anticipate that these initial recommendations would increase compliance costs for affected firms or public entities.

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Appendices

Appendix A. List of 209 PCB Congeners (EPA, 2003)

CASRN	Congener Number	IUPAC Name
1336-36-3		Polychlorinated biphenyl (PCB)
2051-60-7	1	2-Chlorobiphenyl
2051-61-8	2	3-Chlorobiphenyl
2051-62-9	3	4-Chlorobiphenyl
13029-08-8	4	2,2'-Dichlorobiphenyl
16605-91-7	5	2,3-Dichlorobiphenyl
25569-80-6	6	2,3'-Dichlorobiphenyl
33284-50-3	7	2,4-Dichlorobiphenyl
34883-43-7	8	2,4'-Dichlorobiphenyl
34883-39-1	9	2,5-Dichlorobiphenyl
33146-45-1	10	2,6-Dichlorobiphenyl
2050-67-1	11	3,3'-Dichlorobiphenyl
2974-92-7	12	3,4-Dichlorobiphenyl
2974-90-5	13	3,4'-Dichlorobiphenyl
34883-41-5	14	3,5-Dichlorobiphenyl
2050-68-2	15	4,4'-Dichlorobiphenyl
38444-78-9	16	2,2',3-Trichlorobiphenyl
37680-66-3	17	2,2',4-Trichlorobiphenyl
37680-65-2	18	2,2',5-Trichlorobiphenyl
38444-73-4	19	2,2',6-Trichlorobiphenyl
38444-84-7	20	2,3,3'-Trichlorobiphenyl
55702-46-0	21	2,3,4-Trichlorobiphenyl
38444-85-8	22	2,3,4'-Trichlorobiphenyl
55720-44-0	23	2,3,5-Trichlorobiphenyl
55702-45-9	24	2,3,6-Trichlorobiphenyl
55712-37-3	25	2,3',4-Trichlorobiphenyl
38444-81-4	26	2,3',5-Trichlorobiphenyl
38444-76-7	27	2,3',6-Trichlorobiphenyl
7012-37-5	28	2,4,4'-Trichlorobiphenyl
15862-07-4	29	2,4,5-Trichlorobiphenyl
35693-92-6	30	2,4,6-Trichlorobiphenyl
16606-02-3	31	2,4',5-Trichlorobiphenyl

38444-77-8	32	2,4',6-Trichlorobiphenyl
38444-86-9	33	2,3',4'-Trichlorobiphenyl
37680-68-5	34	2,3',5'-Trichlorobiphenyl
37680-69-6	35	3,3',4-Trichlorobiphenyl
38444-87-0	36	3,3',5-Trichlorobiphenyl
38444-90-5	37	3,4,4'-Trichlorobiphenyl
53555-66-1	38	3,4,5-Trichlorobiphenyl
38444-88-1	39	3,4',5-Trichlorobiphenyl
38444-93-8	40	2,2',3,3'-Tetrachlorobiphenyl
52663-59-9	41	2,2',3,4-Tetrachlorobiphenyl
36559-22-5	42	2,2',3,4'-Tetrachlorobiphenyl
70362-46-8	43	2,2',3,5-Tetrachlorobiphenyl
41464-39-5	44	2,2',3,5'-Tetrachlorobiphenyl
70362-45-7	45	2,2',3,6-Tetrachlorobiphenyl
41464-47-5	46	2,2',3,6'-Tetrachlorobiphenyl
2437-79-8	47	2,2',4,4'-Tetrachlorobiphenyl
70362-47-9	48	2,2',4,5-Tetrachlorobiphenyl
41464-40-8	49	2,2',4,5'-Tetrachlorobiphenyl
62796-65-0	50	2,2',4,6-Tetrachlorobiphenyl
68194-04-7	51	2,2',4,6'-Tetrachlorobiphenyl
35693-99-3	52	2,2',5,5'-Tetrachlorobiphenyl
41464-41-9	53	2,2',5,6'-Tetrachlorobiphenyl
15968-05-5	54	2,2',6,6'-Tetrachlorobiphenyl
74338-24-2	55	2,3,3',4-Tetrachlorobiphenyl
41464-43-1	56	2,3,3',4'-Tetrachlorobiphenyl
70424-67-8	57	2,3,3',5-Tetrachlorobiphenyl
41464-49-7	58	2,3,3',5'-Tetrachlorobiphenyl
74472-33-6	59	2,3,3',6-Tetrachlorobiphenyl
33025-41-1	60	2,3,4,4'-Tetrachlorobiphenyl
33284-53-6	61	2,3,4,5-Tetrachlorobiphenyl
54230-22-7	62	2,3,4,6-Tetrachlorobiphenyl
74472-34-7	63	2,3,4',5-Tetrachlorobiphenyl
52663-58-8	64	2,3,4',6-Tetrachlorobiphenyl
32284-54-7	65	2,3,5,6-Tetrachlorobiphenyl
32598-10-0	66	2,3',4,4'-Tetrachlorobiphenyl
73575-53-8	67	2,3',4,5-Tetrachlorobiphenyl
73575-52-7	68	2,3',4,5'-Tetrachlorobiphenyl

60233-24-1	69	2,3',4,6-Tetrachlorobiphenyl
32598-11-1	70	2,3',4',5-Tetrachlorobiphenyl
41464-46-4	71	2,3',4',6-Tetrachlorobiphenyl
41464-42-0	72	2,3',5,5'-Tetrachlorobiphenyl
74338-23-1	73	2,3',5',6-Tetrachlorobiphenyl
32690-93-0	74	2,4,4',5-Tetrachlorobiphenyl
32598-12-2	75	2,4,4',6-Tetrachlorobiphenyl
70362-48-0	76	2,3',4',5'-Tetrachlorobiphenyl
32598-13-3	77	3,3',4,4'-Tetrachlorobiphenyl
70362-49-1	78	3,3',4,5-Tetrachlorobiphenyl
41464-48-6	79	3,3',4,5'-Tetrachlorobiphenyl
33284-52-5	80	3,3',5,5'-Tetrachlorobiphenyl
70362-50-4	81	3,4,4',5-Tetrachlorobiphenyl
52663-62-4	82	2,2',3,3',4-Pentachlorobiphenyl
60145-20-2	83	2,2',3,3',5-Pentachlorobiphenyl
52663-60-2	84	2,2',3,3',6-Pentachlorobiphenyl
65510-45-4	85	2,2',3,4,4'-Pentachlorobiphenyl
55312-69-1	86	2,2',3,4,5-Pentachlorobiphenyl
38380-02-8	87	2,2',3,4,5'-Pentachlorobiphenyl
55215-17-3	88	2,2',3,4,6-Pentachlorobiphenyl
73575-57-2	89	2,2',3,4,6'-Pentachlorobiphenyl
68194-07-0	90	2,2',3,4',5-Pentachlorobiphenyl
68194-05-8	91	2,2',3,4',6-Pentachlorobiphenyl
52663-61-3	92	2,2',3,5,5'-Pentachlorobiphenyl
73575-56-1	93	2,2',3,5,6-Pentachlorobiphenyl
73575-55-0	94	2,2',3,5,6'-Pentachlorobiphenyl
38379-99-6	95	2,2',3,5',6-Pentachlorobiphenyl
73575-54-9	96	2,2',3,6,6'-Pentachlorobiphenyl
41464-51-1	97	2,2',3,4',5'-Pentachlorobiphenyl
60233-25-2	98	2,2',3,4',6'-Pentachlorobiphenyl
38380-01-7	99	2,2',4,4',5-Pentachlorobiphenyl
39485-83-1	100	2,2',4,4',6-Pentachlorobiphenyl
37680-73-2	101	2,2',4,5,5'-Pentachlorobiphenyl
68194-06-9	102	2,2',4,5,6'-Pentachlorobiphenyl
60145-21-3	103	2,2',4,5',6-Pentachlorobiphenyl
56558-16-8	104	2,2',4,6,6'-Pentachlorobiphenyl
32598-14-4	105	2,3,3',4,4'-Pentachlorobiphenyl

70424-69-0	106	2,3,3',4,5-Pentachlorobiphenyl
70424-68-9	107	2,3,3',4',5-Pentachlorobiphenyl
70362-41-3	108	2,3,3',4,5'-Pentachlorobiphenyl
74472-35-8	109	2,3,3',4,6-Pentachlorobiphenyl
38380-03-9	110	2,3,3',4',6-Pentachlorobiphenyl
39635-32-0	111	2,3,3',5,5'-Pentachlorobiphenyl
74472-36-9	112	2,3,3',5,6-Pentachlorobiphenyl
68194-10-5	113	2,3,3',5',6-Pentachlorobiphenyl
74472-37-0	114	2,3,4,4',5-Pentachlorobiphenyl
74472-38-1	115	2,3,4,4',6-Pentachlorobiphenyl
18259-05-7	116	2,3,4,5,6-Pentachlorobiphenyl
68194-11-6	117	2,3,4',5,6-Pentachlorobiphenyl
31508-00-6	118	2,3',4,4',5-Pentachlorobiphenyl
56558-17-9	119	2,3',4,4',6-Pentachlorobiphenyl
68194-12-7	120	2,3',4,5,5'-Pentachlorobiphenyl
56558-18-0	121	2,3',4,5',6-Pentachlorobiphenyl
76842-07-4	122	2,3,3',4',5'-Pentachlorobiphenyl
65510-44-3	123	2,3',4,4',5'-Pentachlorobiphenyl
70424-70-3	124	2,3',4',5,5'-Pentachlorobiphenyl
74472-39-2	125	2,3',4',5',6-Pentachlorobiphenyl
57465-28-8	126	3,3',4,4',5-Pentachlorobiphenyl
39635-33-1	127	3,3',4,5,5'-Pentachlorobiphenyl
38380-07-3	128	2,2',3,3',4,4'-Hexachlorobiphenyl
55215-18-4	129	2,2',3,3',4,5-Hexachlorobiphenyl
52663-66-8	130	2,2',3,3',4,5'-Hexachlorobiphenyl
61798-70-7	131	2,2',3,3',4,6-Hexachlorobiphenyl
38380-05-1	132	2,2',3,3',4,6'-Hexachlorobiphenyl
35694-04-3	133	2,2',3,3',5,5'-Hexachlorobiphenyl
52704-70-8	134	2,2',3,3',5,6-Hexachlorobiphenyl
52744-13-5	135	2,2',3,3',5,6'-Hexachlorobiphenyl
38411-22-2	136	2,2',3,3',6,6'-Hexachlorobiphenyl
35694-06-5	137	2,2',3,4,4',5-Hexachlorobiphenyl
35065-28-2	138	2,2',3,4,4',5'-Hexachlorobiphenyl
56030-56-9	139	2,2',3,4,4',6-Hexachlorobiphenyl
59291-64-4	140	2,2',3,4,4',6'-Hexachlorobiphenyl
52712-04-6	141	2,2',3,4,5,5'-Hexachlorobiphenyl
41411-61-4	142	2,2',3,4,5,6-Hexachlorobiphenyl

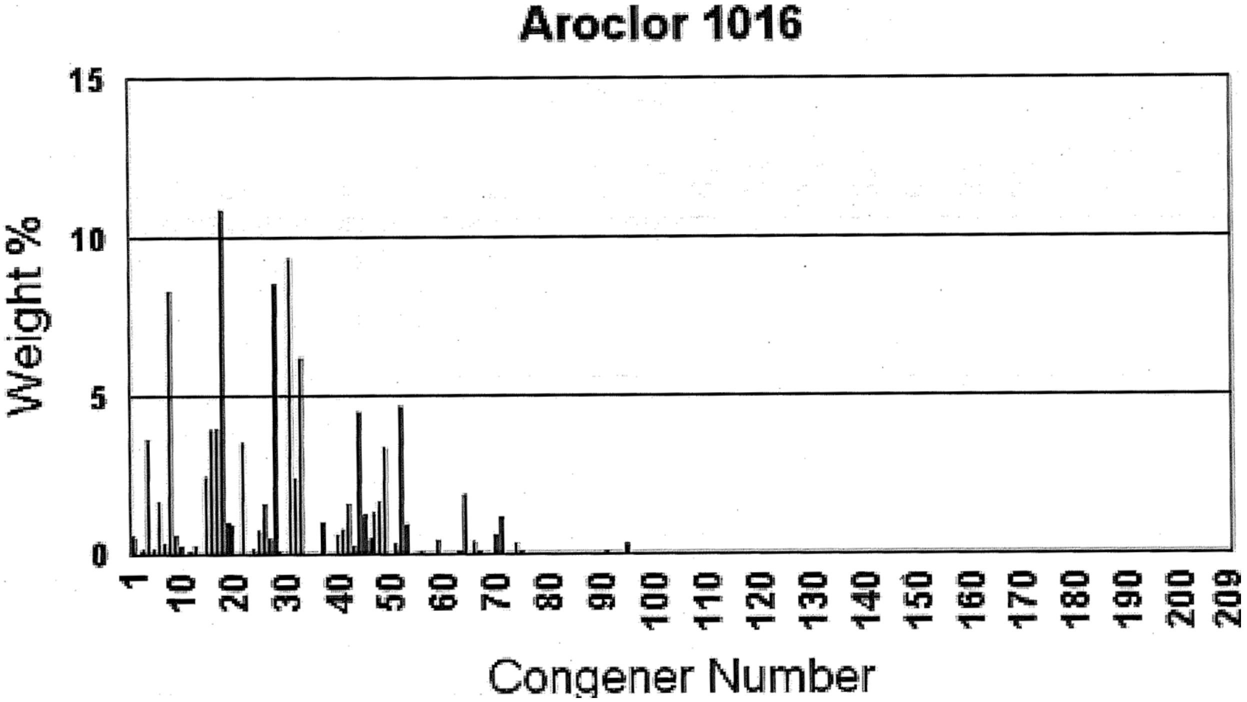
68194-15-0	143	2,2',3,4,5,6'-Hexachlorobiphenyl
68194-14-9	144	2,2',3,4,5',6-Hexachlorobiphenyl
74472-40-5	145	2,2',3,4,6,6'-Hexachlorobiphenyl
51908-16-8	146	2,2',3,4',5,5'-Hexachlorobiphenyl
68194-13-8	147	2,2',3,4',5,6-Hexachlorobiphenyl
74472-41-6	148	2,2',3,4',5,6'-Hexachlorobiphenyl
38380-04-0	149	2,2',3,4',5',6-Hexachlorobiphenyl
68194-08-1	150	2,2',3,4',6,6'-Hexachlorobiphenyl
52663-63-5	151	2,2',3,5,5',6-Hexachlorobiphenyl
68194-09-2	152	2,2',3,5,6,6'-Hexachlorobiphenyl
35065-27-1	153	2,2',4,4',5,5'-Hexachlorobiphenyl
60145-22-4	154	2,2',4,4',5,6'-Hexachlorobiphenyl
33979-03-2	155	2,2',4,4',6,6'-Hexachlorobiphenyl
38380-08-4	156	2,3,3',4,4',5-Hexachlorobiphenyl
69782-90-7	157	2,3,3',4,4',5'-Hexachlorobiphenyl
74472-42-7	158	2,3,3',4,4',6-Hexachlorobiphenyl
39635-35-3	159	2,3,3',4,5,5'-Hexachlorobiphenyl
41411-62-5	160	2,3,3',4,5,6-Hexachlorobiphenyl
74472-43-8	161	2,3,3',4,5',6-Hexachlorobiphenyl
39635-34-2	162	2,3,3',4',5,5'-Hexachlorobiphenyl
74472-44-9	163	2,3,3',4',5,6-Hexachlorobiphenyl
74472-45-0	164	2,3,3',4',5',6-Hexachlorobiphenyl
74472-46-1	165	2,3,3',5,5',6-Hexachlorobiphenyl
41411-63-6	166	2,3,4,4',5,6-Hexachlorobiphenyl
52663-72-6	167	2,3',4,4',5,5'-Hexachlorobiphenyl
59291-65-5	168	2,3',4,4',5',6-Hexachlorobiphenyl
32774-16-6	169	3,3',4,4',5,5'-Hexachlorobiphenyl
35065-30-6	170	2,2',3,3',4,4',5-Heptachlorobiphenyl
52663-71-5	171	2,2',3,3',4,4',6-Heptachlorobiphenyl
52663-74-8	172	2,2',3,3',4,5,5'-Heptachlorobiphenyl
68194-16-1	173	2,2',3,3',4,5,6-Heptachlorobiphenyl
38411-25-5	174	2,2',3,3',4,5,6'-Heptachlorobiphenyl
40186-70-7	175	2,2',3,3',4,5',6-Heptachlorobiphenyl
52663-65-7	176	2,2',3,3',4,6,6'-Heptachlorobiphenyl
52663-70-4	177	2,2',3,3',4,5',6'-Heptachlorobiphenyl
52663-67-9	178	2,2',3,3',5,5',6-Heptachlorobiphenyl
52663-64-6	179	2,2',3,3',5,6,6'-Heptachlorobiphenyl

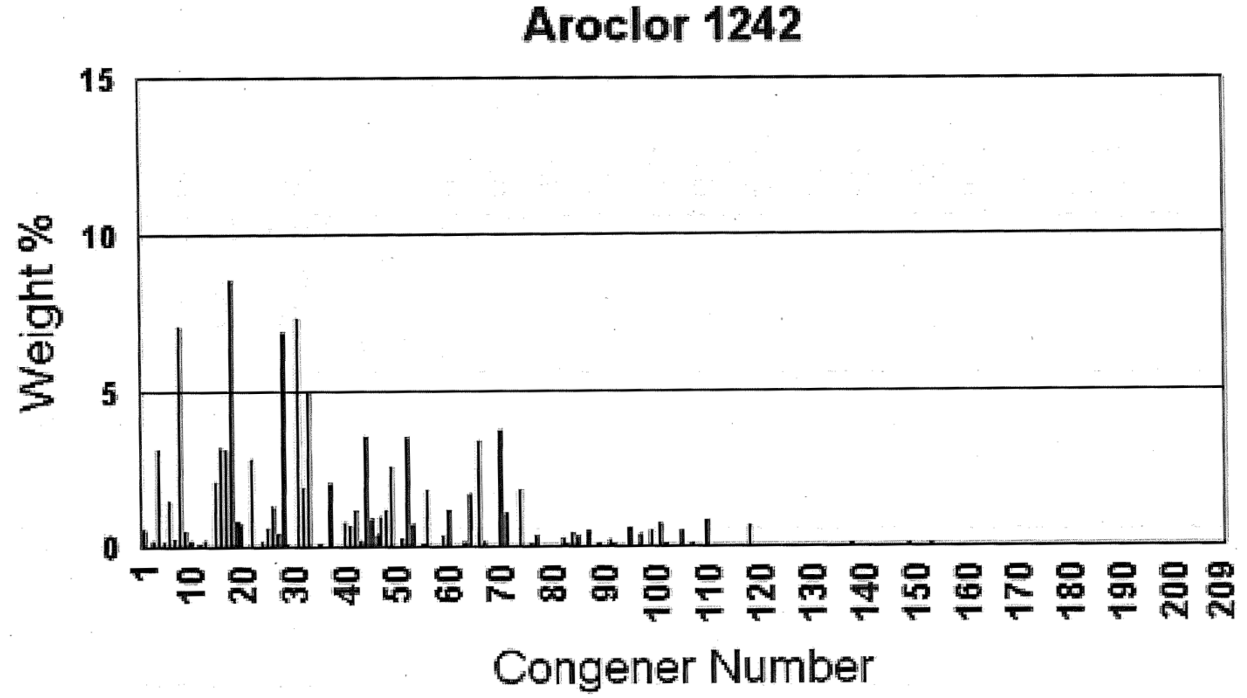
35065-29-3	180	2,2',3,4,4',5,5'-Heptachlorobiphenyl
74472-47-2	181	2,2',3,4,4',5,6-Heptachlorobiphenyl
60145-23-5	182	2,2',3,4,4',5,6'-Heptachlorobiphenyl
52663-69-1	183	2,2',3,4,4',5',6-Heptachlorobiphenyl
74472-48-3	184	2,2',3,4,4',6,6'-Heptachlorobiphenyl
52712-05-7	185	2,2',3,4,5,5',6-Heptachlorobiphenyl
74472-49-4	186	2,2',3,4,5,6,6'-Heptachlorobiphenyl
52663-68-0	187	2,2',3,4',5,5',6-Heptachlorobiphenyl
74487-85-7	188	2,2',3,4',5,6,6'-Heptachlorobiphenyl
39635-31-9	189	2,3,3',4,4',5,5'-Heptachlorobiphenyl
41411-64-7	190	2,3,3',4,4',5,6-Heptachlorobiphenyl
74472-50-7	191	2,3,3',4,4',5',6-Heptachlorobiphenyl
74472-51-8	192	2,3,3',4,5,5',6-Heptachlorobiphenyl
69782-91-8	193	2,3,3',4',5,5',6-Heptachlorobiphenyl
35694-08-7	194	2,2',3,3',4,4',5,5'-Octachlorobiphenyl
52663-78-2	195	2,2',3,3',4,4',5,6-Octachlorobiphenyl
42740-50-1	196	2,2',3,3',4,4',5,6'-Octachlorobiphenyl
33091-17-7	197	2,2',3,3',4,4',6,6'-Octachlorobiphenyl
68194-17-2	198	2,2',3,3',4,5,5',6-Octachlorobiphenyl
52663-75-9	199	2,2',3,3',4,5,5',6'-Octachlorobiphenyl
52663-73-7	200	2,2',3,3',4,5,6,6'-Octachlorobiphenyl
40186-71-8	201	2,2',3,3',4,5',6,6'-Octachlorobiphenyl
2136-99-4	202	2,2',3,3',5,5',6,6'-Octachlorobiphenyl
52663-76-0	203	2,2',3,4,4',5,5',6-Octachlorobiphenyl
74472-52-9	204	2,2',3,4,4',5,6,6'-Octachlorobiphenyl
74472-53-0	205	2,3,3',4,4',5,5',6-Octachlorobiphenyl
40186-72-9	206	2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl
52663-79-3	207	2,2',3,3',4,4',5,6,6'-Nonachlorobiphenyl
52663-77-1	208	2,2',3,3',4,5,5',6,6'-Nonachlorobiphenyl
2051-24-3	209	Decachlorobiphenyl

Appendix B. PCB Mixtures and Trade Names (EPA 2013d)

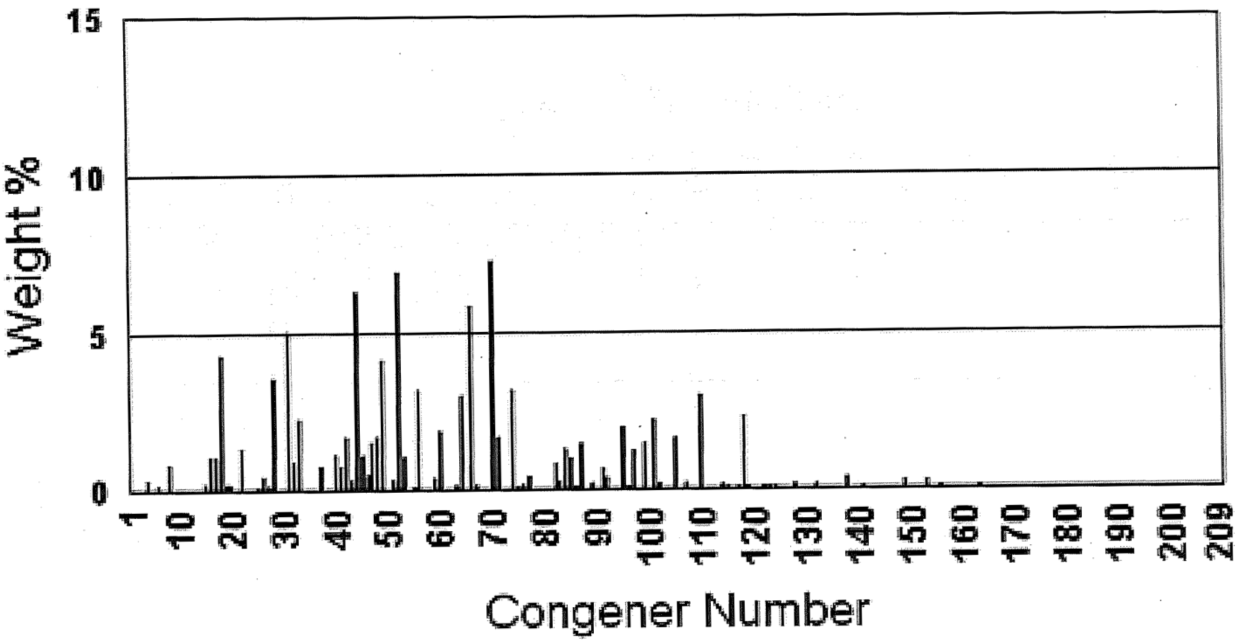
Acector
Adkarel
ALC
Apirolio
Apirorlio
Arochlor
Arochlors
Aroclor
Aroclors
Arubren
Asbestol
ASK
Askael
Askarel
Auxol
Bakola
Biphenyl, chlorinated
Chlophen
Chloretol
Chlorextol
Chlorinated biphenyl
Chlorinated diphenyl
Chlorinol
Chlorobiphenyl
Chlorodiphenyl
Chlorphen
Chorextol
Chorinol
Clophen
Clophenharz
Cloresil
Clorinal
Clorphen
Decachlorodiphenyl
Delor
Delorene
Diaclor
Dicolor
Diconal
Diphenyl, chlorinated
DK
Duconal
Dykanol
Educarel
EEC-18
Elaol
Electrophenyl
Elemex
Elinol
Eucarel
Fenchlor
Fenclor
Fenocloro
Gilotherm
Hydol
Hyrol
Hyvol
Inclor
Inerteen
Inertenn
Kanechlor
Kaneclor
Kennechlor
Kenneclor
Leromoll
Magvar
MCS 1489
Montar
Nepolin
No-Flamol
NoFlamol
Non-Flamol
Olex-sf-d
Orophene
PCB
PCB's
PCBs
Pheaoclor
Phenochlor
Phenoclor
Plastivar
Polychlorinated biphenyl
Polychlorinated biphenyls
Polychlorinated diphenyl
Polychlorinated diphenyls
Polychlorobiphenyl
Polychlorodiphenyl
Prodelec
Pydraul
Pyraclor
Pyralene
Pyranol
Pyroclor
Pyronol
Saf-T-Kuhl
Saf-T-Kohl
Santosol
Santotherm
Santothern
Santovac
Solvol
Sorol
Soval
Sovol
Soltol
Terphenychlore
Therminal
Therminol
Turbinol

Appendix C. Distribution of Aroclor mixtures (EPA, 2013d)

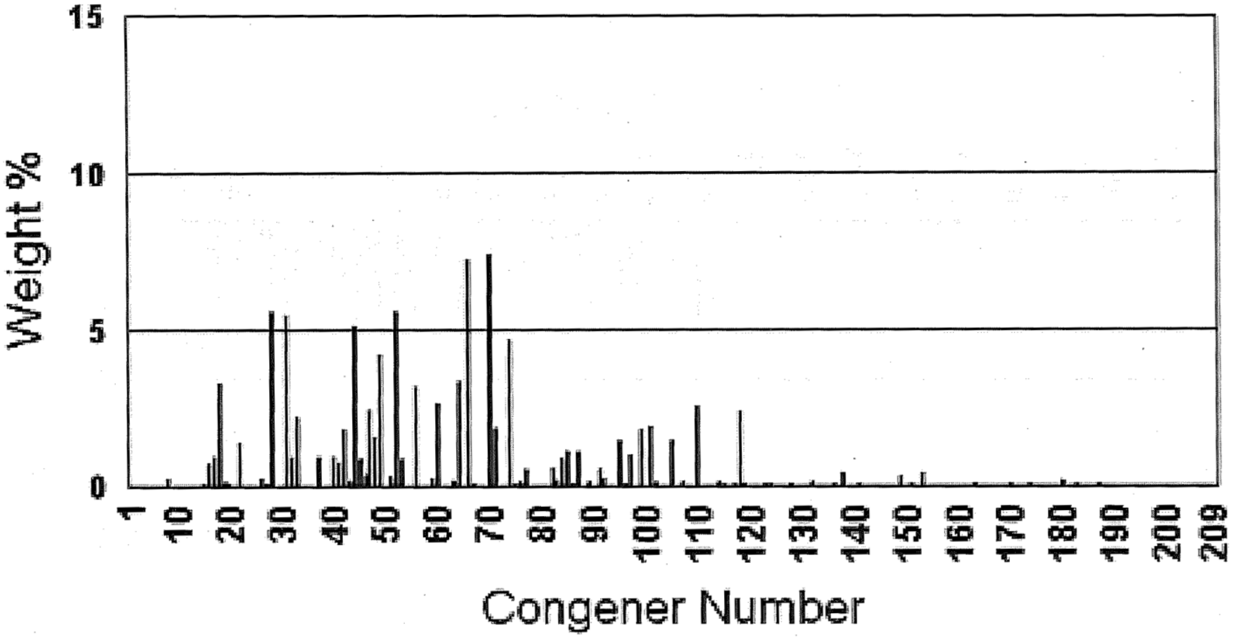


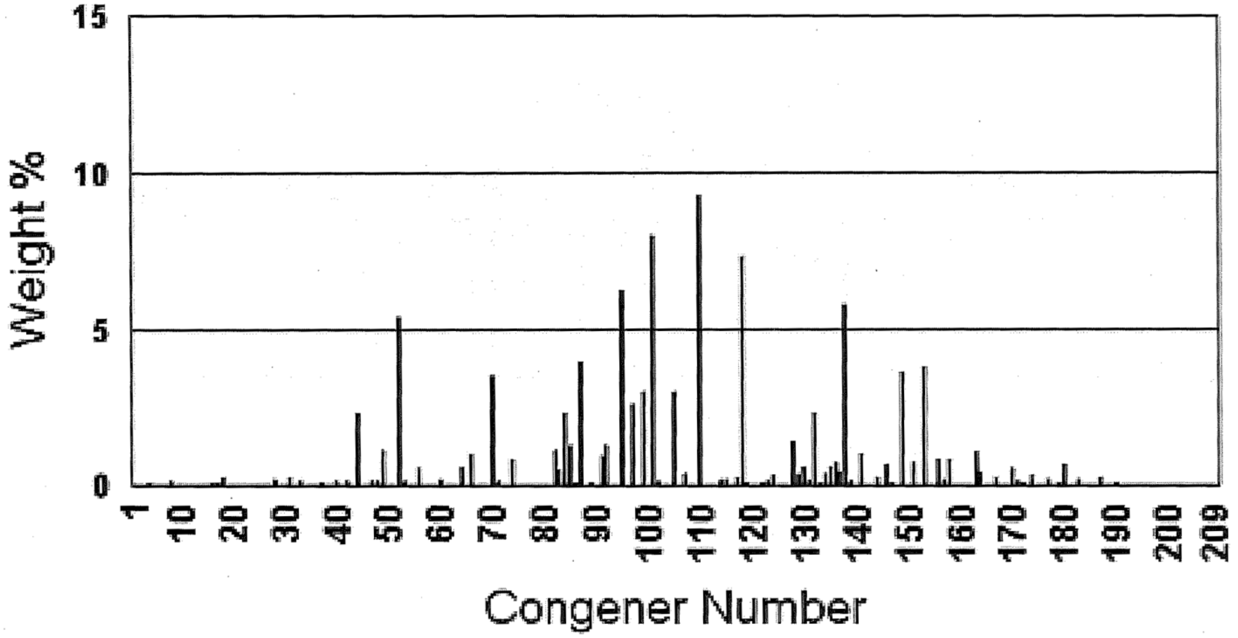


Aroclor 1248a

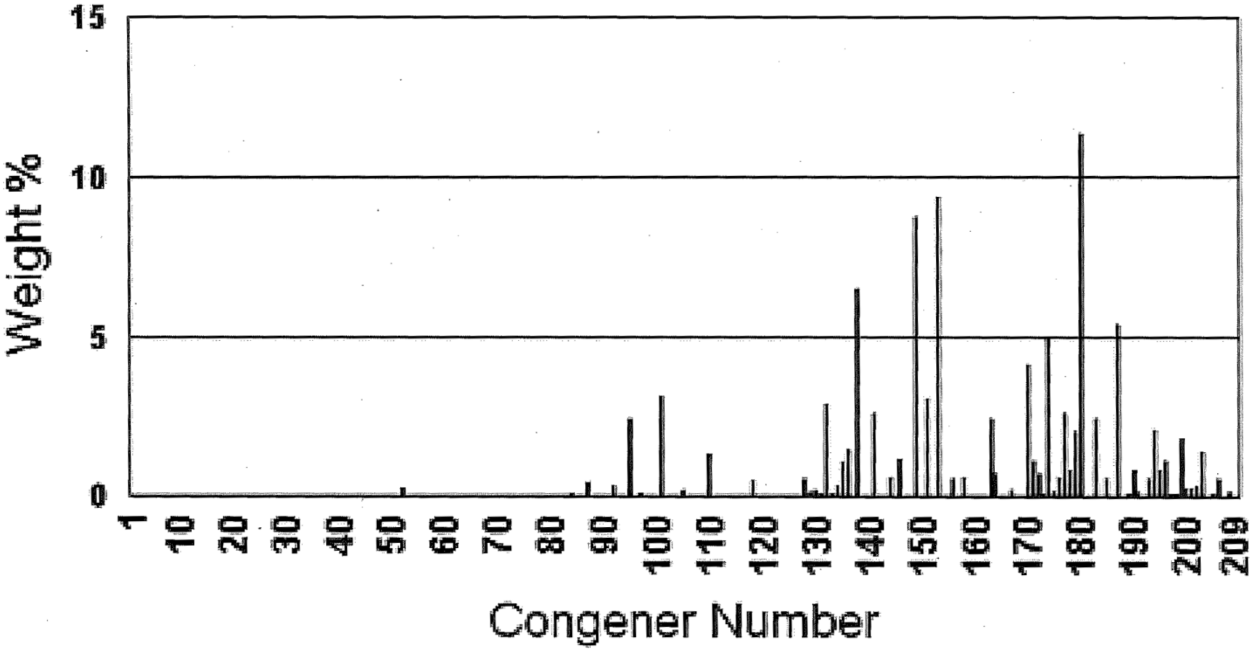


Aroclor 1248g





Aroclor 1254g



Aroclor 1260

Appendix D. Chemical Processes that have the Potential to Generate PCBs

The following was transcribed from EPA rulemaking records from “Polychlorinated Biphenyls (PCBs); Manufacturing, Processing, Distribution in Commerce, and Use Prohibitions: Exclusions, Exemptions and Use Authorizations” Doc No. OPTS-62032. This was transcribed for Panero *et al*. (2005).

Chlorinated Compounds Produced Using Direct Chlorination	Chlorinated Compounds Produced Using Chlorinated Feedstocks	Non-chlorinated Compounds Produced Using Chlorinated Feedstocks	IPPPE U No.
Petroleum Feedstock: BENZENE			
Chlorinated benzenes	Chloronitrobenzenes	Phenol	8
Chlorinated phenols	Dichloronitrobenzenes	Aniline	9
Hexachlorocyclohexane	Dichloroanilines	o-Phenylenediamine	28
Chloranilines	Chlorinated methyl phenols	o-,p-Nitroanilines	29
Trichloroanilines	Chlorophenyl phenylethers	Diphenylamine	34
	Chlorinated benzidines	Acetanilide	17
Petroleum Feedstock: ETHYLENE			
Mono, di-chloroacetic acid	Ethyl chloroacetate	Glycine	108
Sodium chloroacetate	Vinyl chloride	Cyanoacetic acid	111
Chlorinated ethanes	Vinylidene chloride	Sodium, carboxymethyl cellulose	112
Chlorinated ethylenes	Bis (2-chloroethyl) ether	Ethyl cellulose	118
Ethylene chlorohydrin	Chlorinated acetophenones	Ethylene diamine	134
Chlorinated, fluorinated ethanes	Choline chloride	Aminoethylethanolamine	135
Chlorinated, brominated ethylenes	Hexachlorobutadiene	Mono-, di-, and triethylene glycol ethers	150
Chlorinated, fluorinated ethylenes		Tetramethylethylene diamine	(3341)
Chlorinated acetaldehyde			
Chlorinated acetyl chloride			
Hexachlorobenzene			
Petroleum Feedstock: METHANE			
Chlorinated methanes	Chlorinated, fluorinated methanes	Carbon tetrabromide	162
Phosgene	Chlorinated, brominated methanes	Carbon tetrafluoride	(812)
Tetrachloroethane	Bis (chloromethyl) ether		
Chlorodifluoroethane (?)	Cyanuric chloride		
Perchloromethyl mercaptan (?)	Trichloroethylene		
Cyanogen chloride			

Chlorinated Compounds Produced Using Direct Chlorination	Chlorinated Compounds Produced Using Chlorinated Feedstocks	Non-chlorinated Compounds Produced Using Chlorinated Feedstocks	IPPPE U No.
Petroleum Feedstock: NAPHTHALENE			
Chloronaphthalenes			
Tetrachlorophthalic anhydride			
Petroleum Feedstock: PARAFFINS			
n-Propyl chloride		n-Propylamine	231
Carbon tetrachloride		Butyronitrile	232
Perchloroethylene		Amyl amines	243
Hexachloroethane		Amyl alcohols	244
Amyl chlorides		Amyl Mercaptans	245
Chloroprene		Benzophenone	249
Hexachlorocyclopentadiene		Linear alkylbenzenes	(2417)
Methallyl Chloride			
Petroleum Feedstock: PROPYLENE			
Dichlorohydrin	Epichlorohydrin	Isopropylphenols	272
Chloranil	Bis (2-chloroisopropyl) ether	Propylene oxide	280
Propylene chlorohydrin		Anisols	302
Chlorinated propanes		Allyl alcohol	317
Chlorinated propylenes		Glycerol	318/319
		Propyl amines	(1446)
Petroleum Feedstock: TOLUENE			
Benzyl chloride	Benzoyl chloride	Benzyl alcohol	334
Benzyl dichloride		Benzyl amine	335
Benzyl trichloride		Benzamide	337
Chlorotoluenes		Toluenesulfonamide	358
Chlorobenzaldehyde		Benzoyl peroxide	(495)
Chlorobenzoic acids & esters			
Chlorobenzoyl chlorides			
Toluenesulfonyl chloride			
Chlorobenzotrichlorides			

*The IPPEU No. refers to the process description in the 1977 EPA summary (EPA, 1977). Those numbers bracketed by parentheses refer to the OCPDB numbers in the 1980 EPA summary (EPA, 1980)

Appendix E. Reported Products with Inadvertently Generated PCBs

Manufacturers are required to report inadvertent generation of PCBs to EPA. Below is a summary table of the 77 reports received from 1994 to present. See the Regulations section for explanation of the requirement and "discounted."

Date	Reporter	Product	Concentration or amount	Category
4/13/1995	Sun Chem. Corp	2-Naphthalenecarboxylic acid, 4-[(2,5-dichlorophenyl) azo]-3-hydroxy, a dye precursor		pigments and dyes
2/11/2004	Clariant	imported dyes		pigments and dyes
6/13/2005	Clariant	imported dyes		pigments and dyes
5/19/2011	Clariant	imported dyes		pigments and dyes
7/29/1994	Ciba-Geigy Pigments Division	CBI		pigments and dyes
12/28/1994	Ciba-Geigy Pigments Division	CBI		pigments and dyes
12/29/1994	DIC Trading	3 pigments		pigments and dyes
6/22/1995	Ciba-Geigy Pigments Division	CBI		pigments and dyes
7/25/1995	Cappelle	4 pigments		pigments and dyes
7/2/1996	Uhlich Color Co	CI Pigment Orange 24		pigments and dyes
7/15/1996	Ciba-Geigy Pigments Division	CBI		pigments and dyes
8/16/1996	Engelhard	CI Pigment Violet 23	19.6 ppm	pigments and dyes
8/23/1996	Cappelle	CI Pigment Yellow 170		pigments and dyes
9/27/1996	UMC (United Mineral and Chem)	CI Pigment Green 7		pigments and dyes
1/13/1997	Zeneca	7 pigments		pigments and dyes
7/29/1996	CDR Pigments and Dispersions	6 pigments		pigments and dyes
6/18/1997	Fabricolor	12 pigments		pigments and dyes
7/1/1997	BASF	13 pigments		pigments and dyes
8/18/1997	Ciba Pigments Division	CBI for several pages		pigments and dyes
10/21/1997	Mil International	5 pigments		pigments and dyes
1/6/1998	Sun Chem. Corp	4 pigments		pigments and dyes
10/26/1997	Mil International	4 pigments		pigments and dyes
5/15/1998	Mil International	5 pigments		pigments and dyes
7/20/1998	Ciba Pigments Division	CBI		pigments and dyes
10/23/1998	Ciba Pigments Division	CBI		pigments and dyes
2/2/1999	Lansco Colors	7 pigments		pigments and dyes
7/15/1999	Ciba Colors	CBI		pigments and dyes

	Division			
7/31/1999	Sun Chem. Corp	CBI		pigments and dyes
2/2/2000	Ciba Colors Division	CBI		pigments and dyes
5/23/2000	Ciba Colors Division	CBI		pigments and dyes
8/31/2000	Ciba Colors Divison	CBI		pigments and dyes
9/8/2000	Avecia	7 pigments		pigments and dyes
11/22/2000	Mil International	7 pigments		pigments and dyes
12/13/2000	Ciba Colors Division	CBI		pigments and dyes
3/30/2001	Ciba Colors Division	CBI		pigments and dyes
5/4/2001	Magruder Color Co	3 pigments		pigments and dyes
6/1/2001	Sun Chem. Corp	9 pigments		pigments and dyes
7/18/2001	Ciba Colors Division	CBI		pigments and dyes
4/8/1994	PCL Group	Copper Phthalocyanine Blue		pigments and dyes
10/17/2001	Ciba Colors Division	CBI		pigments and dyes
1/25/2002	Ciba Colors Division	CBI		pigments and dyes
3/27/2002	Mil International	8 pigments		pigments and dyes
4/29/2002	Ciba Coating Effects	CBI		pigments and dyes
8/6/2002	Ciba Coating Effects	CBI		pigments and dyes
8/28/2002	Sun Chem. Corp	CBI		pigments and dyes
11/5/2002	Ciba Coating Effects	CBI		pigments and dyes
6/13/2003	Ciba Coating Effects	CBI		pigments and dyes
10/16/2003	Ciba Coating Effects	CBI		pigments and dyes
4/2/2004	Ciba Coating Effects	CBI		pigments and dyes
7/6/2004	Ciba Coating Effects	CBI		pigments and dyes
7/6/2004	Ciba Coating Effects	CBI		pigments and dyes
8/8/2005	Sun Chemical	CBI		pigments and dyes
5/25/2006	Cappelle	CI Pigment Yellow 17		pigments and dyes
1/30/1995	GE Silicones	CBI	<2.5 ppm discounted, total discounted quantity <1.1 lbs	silicones
1/30/1996	GE Silicones	CBI	<1.1 ppm discounted, total discounted quantity <0.9 lbs	silicones

1/24/1997	GE Silicones	CBI	<1.5 ppm discounted, total discounted quantity <0.6 lbs	silicones
1/24/1997	GE Silicones	CBI	<1.3 ppm discounted, total discounted quantity <0.53 lbs	silicones
2/25/1999	GE Silicones	CBI	<1.5 ppm discounted, total discounted quantity <0.8 lbs	silicones
2/7/2000	GE Silicones	CBI	<1.7 ppm discounted, total discounted quantity <0.5 lbs	silicones
3/13/2001	GE	CBI	<1.9 ppm discounted, total discounted quantity <0.7 lbs	silicones
5/28/2002	GE	CBI, adding hydrolyzed phenylchlorosilanes and phenylchlorosilanes	total discounted quantity < 0.83 lbs	silicones
4/30/1997	ABB	electrical capacitors	3.9 ppm, 134 liters	Unique
6/24/1994	Nagase America	2,4,6-TCPH (2,4,6-Trichlorophenylhydrazine)	9-12 ppm	Unique
11/30/1995	PHT International	2,6-Dichloro-4-Nitro Aniline		Unique
3/17/1998	ISK Biosciences	CBI, Chlorothalonil production		Unique
5/15/2001	PPG Industries	trichlorobenzene (TCB)		Unique
8/17/2012	Future Fuel	pesticide intermediate		Unique
4/7/1997	Elf Atochem		4 and 5 ppm	Unknown
2/18/2000	CBI	CBI		Unknown
6/13/2001	CBI	CBI		Unknown
2/4/2003	CBI	CBI		Unknown
CBI	CBI	CBI		Unknown
5/31/2011	CBI	CBI		Unknown
9/11/2012	CBI	CBI, 220 kg shipment		Unknown
8/23/2004	Formosa Plastics		up to 215-255 ppm, 143 lbs	vinyl chloride
6/24/1996	Geon		740 lbs PCB/ 62,676,000 lbs chemical feedstocks	vinyl chloride
11/13/1997	Dow			vinyl chloride

Appendix F. Washington PCB transformers in EPA database

We attempted to contact the registrants to find out about the current whereabouts of the registered transformers. While owners of PCB transformers (> 500 ppm PCBs) were required to register with the EPA, the EPA is not required to update the database.

Company	**City**	**Contact**	**Transformer street address**	**Trans. City**	**Trans. Zip code**	**No. of Trans.**	**Weight (kg)**	**Current whereabouts**
Puget Sound Energy	Bellevue	John Rork	Talcott Avenue & Columbia Street	Olympia	98501	3	7	In use
Puget Sound Energy	Bellevue	John Rork	14401 278th Avenue NE	Duval	98019	2	5	In use
Puget Sound Energy	Bellevue	Lea Boyle	14401 188th Avenue NE	Redmond	98052	2	4.52	In use
Puget Sound Energy	Bellevue	John Rork	S 173rd & 43rd Avenue S	Renton	98055	0	0	
Puget Sound Energy	Bellevue	John Rork	2211 Nevada Street	Bellingham	98225	0	0	
Puget Sound Energy	Bellevue	John Rork	24810 156th Avenue SE	Kent	98025	0	0	
Puget Sound Energy	Bellevue	John Rork	Hodgedon & Garfield Streets	Tenino	98589	0	0	
Puget Sound Energy	Bellevue	John Rork	70th Street E & Myers Road	Bonney Lk	98390	0	0	
Puget Sound Energy	Bellevue	John Rork	Dolarway Road	Ellensburg	98922	0	0	
Puget Sound Energy	Bellevue	John Rork	Jackson & Main Streets	Cle Elum	98922	0	0	
Puget Sound Energy	Bellevue	John Rork	19319 Electron Road	Orting	98360	0	0	
Puget Sound Energy	Bellevue	John Rork	W. side of Stottlemeyer Road	Poulsbo	98370	0	0	
Puget Sound Energy	Bellevue	John Rork	40801 268th Avenue SE	Enumclaw	98022	0	0	
Puget Sound Energy	Bellevue	John Rork	N. Tapps Highway & Vandermark Road	Auburn	98002	0	0	
Puget Sound Energy	Bellevue	John Rork	SE 80th Street & 246 Avenue SE	Issaquah	98027	0	0	
Puget Sound Energy	Bellevue	John Rork	13635 SE 26th	Bellevue	98004	0	0	
Puget Sound Energy	Bellevue	John Rork	3975 E. Highway 525	Langley	98260	0	0	
Puget Sound Energy	Bellevue	John Rork	1274 Thompson Road	Anacortes	98221	0	0	
Puget Sound Energy	Bellevue	John Rork	2857 S. 221st	Des Moines	98148	0	0	
Puget Sound Energy	Bellevue	John Rork	12251 Mt Baker Highway	Glacier	98244	0	0	

Puget Sound Energy	Bellevue	John Rork	7537 Portal Way	Ferndale	98248	0	0	
Puget Sound Energy	Bellevue	John Rork	13635 NE 80th	Redmond	98052	0	0	
Puget Sound Energy	Bellevue	John Rork	9512 Pacific Highway SE	Lacey	98503	0	0	
Puget Sound Energy	Bellevue	John Rork	9221 Wilows Road NE	Redmond	98502	0	0	
Puget Sound Energy	Bellevue	John Rork	34717 21st Avenue SW	Federal Way	98003	0	0	
Puget Sound Energy	Bellevue	John Rork	1035 Stevenson Avenue	Enumclaw	98022	0	0	
Puget Sound Energy	Bellevue	John Rork	Hanford Road & Centralia Steam Plt	Centralia	98531	0	0	
Puget Sound Energy	Bellevue	John Rork	South of I-90 between Exits 37 & 38	Snoqualmie	98065	0	0	
Puget Sound Energy	Bellevue	John Rork	Corner of Central Valley Road & Bucklin	Bremerton	98310	0	0	
Puget Sound Energy	Bellevue	John Rork	20th Street E & 169th Avenue E (2111)	Sumner	98340	0	0	
Western Washington University	Bellingham	Gayle Shipley	Commissary 781 25th St.	Bellingham	98225	0	0	
SDS Lumber Co	Bingen	Ronald Schultz	South Side BNSF RR	Bingen	98605	2	2138	Unknown
Kimberly-Clark Worldwide	Everett	Jim Ketchum	2600 Federal Ave.	Everett	98201	0	0	
Grays Harbor Paper L.P.	Hoquiam	Richard Johnston	801 23rd St.	Hoquiam	98550	5	50932	In use
Reynolds Metals Company	Longview	H.S. Hays	4029 Industrial Way	Longview	98632	0	0	
Washington Veneer	Omak	Joe Atwood	1100 Eighth Ave.E.	Omak	98841	7	12412	Unknown
PUD. No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 West 18th Street	Port Angeles	98362	4	505	Disposed of
City of Port Angeles	Port Angeles	Mark Shamp	321 E. Fifth Street	Port Angeles	98362	1		Disposed of
PUD. No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 West 18th Street	Port Angeles	98363	1	100	Disposed of
PUD No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 West 18th Street	Port Angeles	98363	1	68	Disposed of
Port Townsend Paper Corporation	Port Townsend	John M. Recht	100 Mill Hill Rd	Port Townsend	98368	0	0	
City of Richland	Richland	Wayne Collop	806 Thayer Drive	Richland	99352	2	45	Disposed of
US Dept of Energy Richland Oper. Office	Richland	B.J. Dixon	200 East Area	Richland	99352	1	137	Unknown
Energy Northwest	Richland	J.P. Chasse	HPCS Diesel Generator Rm, Nuclear Plant #2, N. Power Plant Loop	Blank	Blank	0	0	
Entercom Communications	Seattle	Martin Hadfield	910 Lone Oak Road	Longview	Blank	0	0	

Corp								
Total Reclaim, Inc	Seattle	Craig Lorch	2200 Sixth Avenue South	Seattle	98134	1	215	Disposed of
Seattle City Light	Seatle	Karen Dinehart	Laurelhurst Lane and 51st	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	4502 NE 41st Street	Seattle	98124	3	182	Disposed of
Seattle City Light	Seattle	Karen Dinehart	Bellevue Ave E & E. John	Seattle	98124	3	160	Disposed of
Seattle City Light	Seattle	Karen Dinehart	2826 NW Market Street	Seattle	98124	2	114	Disposed of
Seattle City Light	Seattle	Karen Dinehart	7710 35th Avenue, SW	Seattle	98124	1	68	Disposed of
Seattle City Light	Seattle	Karen Dinehart	6730 24th Avenue, NW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen. Dinehart	1414 NW Leary Way	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	7750 28th Ave NW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1405 NW 65th Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	8032 15th Avenue NW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	3209 NW 65th Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	2333 W Boston Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	2100 SW Andover Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	35th Ave SE & SW Genessee	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	5601 23rd Avenue SW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1605 SW Holden Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	3405 SW Graham Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	4118 SW Morgan Street	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	9370 52nd Avenue S	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1stE/Of Earl Ave NW, S/SI NW 90th	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1stN/Of S Holden,E/SI Rainier AveS	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1stS/Of W Bertona,E/SI 21st Ave W	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	48th NE & 47th NE	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	51st Ave NE & NE 41st Street	Seattle	98124	0	0	
Inland Power and Light	Spokane	Todd Hoffman	10110 W. Hallett Road	Spokane	99014		1,249.00	
Avista Utilities	Spokane	Clarice	various locations	Blank	Blank	157	16,434.	Disposed of

		Robertson					00	
Avista Corporation	Spokane	Clarice Robertson	Onion Creek Road	Colville	Blank	0	0	
Avista Corporation	Spokane	Clarice Robertson	SE corner of Rockwell and Monroe Streets	Spokane	Blank	0	0	
Tacoma Power	Tacoma	Russell Post	418 Gershick Rd	Silver Creek	98585	10	830	Disposed of
Tacoma School District #10	Tacoma	Margaret Ohlson	111 North E Street	Tacoma	98403	1	358	In use
Tacoma School District #10	Tacoma	Margaret Ohlson	2502 North Orchard	Tacoma	98406	1	358	In use
Pioneer Americas, Inc./Chlor Alkali Co. Inc.	Tacoma	Karl Iams	605 Alexander Ave.	Tacoma	98421	0	0	
TransAlta of Calgary	Alberta	Roger Carter	913 Big Hanaford Rd	Centralia	98531	42	34731	Disposed of

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
REGION 10
1200 Sixth Avenue, Suite 900
Seattle, WA 98101-3140

OFFICE OF THE REGIONAL
ADMINISTRATOR

February 24, 2015

Ms. Adriane Borgias
Spokane River Regional Toxics Task Force
Washington State Department of Ecology
4601 North Monroe Street
Spokane, Washington 99205-1295

Dear Ms. Borgias:

Thank you for your October 23, 2013, letter on behalf of the Spokane River Regional Toxic Task Force to Jim Jones, Assistant Administrator for the Environmental Protection Agency's Office of Chemical Safety and Pollution Prevention, and Cynthia Giles, Assistant Administrator for EPA's Office of Enforcement and Compliance Assurance, regarding the water quality challenges presented by polychlorinated biphenyls. I apologize for the delay in my response on behalf of the Agency, but your letter raises some particularly challenging issues for us. Specifically, your letter notes the potential problems from the release of inadvertently generated PCBs from products such as pigments, and requests that the EPA take two actions to address the problems. First, you ask that the EPA initiate enforcement of the existing prohibition on the imports of inadvertently generated PCBs at concentrations at or above 50 parts per million and, second, that EPA revise its regulations to eliminate all manufacture or import of inadvertently generated PCBs.

The EPA agrees with the Task Force on the importance of reducing PCBs in the environment and the need to look at all potential sources. The EPA participates on the Task Force and supports the collaborative approach being taken to reduce PCB sources in advance of completing a TMDL. Having approved the Spokane Tribe's water quality standards, the EPA understands the issues of concern associated with very low levels of PCBs and the challenges of meeting those standards.

Your request that the EPA initiate enforcement of the regulations regarding inadvertently generated PCBs raises very complex issues. Excluded Manufacturing Processes and the resulting products are excluded from the otherwise applicable statutory bans as long as certain requirements are met, including reporting those processes and products to the EPA and maintaining concentrations under specified limits. Given your request, we examined the potential for increased compliance and enforcement activity to address possible violations of these regulations and found a number of significant challenges. These challenges include the nature of the regulations, the EPA's ability to identify possible non-compliers, the resources necessary to implement an effective enforcement initiative, and the potential of any such initiative to effectively reduce PCB levels to meet water quality standards. Thus, an enforcement initiative targeted specifically at the regulations for inadvertently generated PCBs is not a promising approach.

Revising current regulations to reduce inadvertently generated PCBs presents both policy and scientific challenges. Before proposing more stringent regulations on the inadvertent generation of PCBs in pigments, the EPA would seek to further understand the complexities and contributions of not only PCB-11, but also other congeners that may be present in the Spokane River. At present, there are not sufficient data to assess such PCB congeners. However, in a step toward addressing this deficiency, the EPA has requested that toxicity testing on PCB-11, a congener identified to be incorporated into yellow pigments, be conducted through the National Toxicology Program at the National Institute of Environmental Health Sciences.

Excluded Manufacturing Processes and associated products may generate or contain a variety of inadvertently generated PCB congeners other than PCB-11. There are Toxicity Equivalence Factors (TEFs) established for the dioxin-like congeners, but, prior to revising TSCA regulations or the EPA's recommended water quality criteria for PCBs, the EPA would want to rely on additional toxicity information for many of the non-dioxin-like individual congeners. The aggregation of PCB congeners may in some instances be problematic for risk assessment because the toxicity of different PCB congeners varies and a fixed water quality concentration for total PCBs may not adequately represent the variable toxicity of the various congeners actually present in a particular water body. While the EPA is not proposing to undertake a comprehensive analysis of the remaining PCB congeners, we are examining the characterization of PCBs in water bodies. As stated above, characterizing individual PCB congeners' contribution to risk presents challenges. Therefore, the aggregation of all PCBs in the EPA's recommended water quality criteria for PCBs (i.e., expressed as total PCBs) is one topic we are discussing.

We note that states have taken the initiative to assess toxicity of specific chemicals in the past. One example is the toxicity criteria program managed by California's Office of Environmental Health Hazard Assessment. This process may be a reasonable approach that Washington can take to address the allowable amounts of specific PCB congeners generated inadvertently.

As you know, the EPA intends to propose to restrict and/or eliminate many of the remaining authorized uses of higher-concentration liquid PCBs. These remaining uses are the largest reservoir of commercial mixtures (Aroclors) that contain the dioxin-like PCBs for which there have been health concerns for decades. While these proposed changes will not address the inadvertently generated non-dioxin-like PCBs identified in your letter, the EPA believes this effort will help to reduce potential exposure and risk from remaining dioxin-like PCB uses.

One potentially promising strategy to address PCBs inadvertently produced in products is Green Chemistry. The EPA has provided funding to Ecology to establish a Green Chemistry Center and is a member of the Advisory Board for the Center. The Green Chemistry Center plans to host a workshop later this year on PCBs inadvertently produced in inks and pigments, perhaps leading to improvements in the production and use of PCB-free inks and pigments.

I understand that, having not heard back from the EPA in so long, you recently requested a meeting with the EPA senior managers to discuss these issues. If you still would like to meet after you and the other members of the Task Force have had a chance to review this response, I would be happy to assist in getting the meeting organized. Please feel free to contact me or have your staff contact Tom Eaton, Director of our Washington Operations Office at (360) 753-8086 or by email at eaton.thomas@epa.gov if you still wish to proceed with the meeting.

Thank you again for your letter, and again, I apologize for the delay. I look forward to continuing our work together and protecting human health and the environment.

Sincerely,

Dennis J. McLerran
Regional Administrator

cc: Wendy Cleland-Hamnett, Director, OPPT
Susan Shinkman, Director, OCE
Ken Kopocis, Deputy Assistant Administrator, OW
Ed Kowalski, Director, OCE, Region 10
Lauris Davies, Associate Director, OCE, Region 10
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OFFICE OF
WATER AND WATERSHEDS

September 5, 2014

Don Essig
Idaho Department of Environmental Quality
1410 N. Hilton
Boise, Idaho 83706

RE: EPA comments on Idaho's Discussion Paper #5 Anadromous Fish

Dear Don:

EPA appreciates the opportunity to provide comments on the discussion paper, which the Idaho Department of Environmental Quality (DEQ) provided at the July 23, 2014 negotiated rulemaking meeting. This discussion paper considers some of the issues regarding inclusion or exclusion of anadromous fish in the derivation of a fish consumption rate in Idaho. In our comments, we would like to note that we are discussing inclusion of fish that migrate in the consumption rate. EPA assumes that Idaho already intends to include species that ordinarily migrate but cannot because of physical barriers (e.g., dams) in their fish consumption rate. Whenever salmon are referred to in this comment letter, EPA is referring to salmon that can physically migrate.

EPA found Idaho's discussion paper to be quite comprehensive and well written. However, EPA believes the paper would have been strengthened by incorporating some of the issues analysis that Washington and Oregon did when considering the inclusion of anadromous species in the FCR. These issues are largely those associated with the uncertainties referred to later in these comments. The discussion paper also does not accurately describe the basis for EPA's subsistence default fish consumption rate of 142 g/day, implying it is solely derived from the 99th percentile of the general population (p. 4 under 'EPA's Position'). While EPA's recommendation corresponds to this percentile, the subsistence default rate was developed from a number of consumption estimates for subsistence fishers based on subsistence-fisher specific surveys. The subsistence default rate corresponds to a high-end (>90th percentile) consumption rate for a subsistence fisher population, as evidenced in particular by the 1994 CRITFC Survey. Although it also corresponds to the 99th percentile of the USDA's Continuing Survey of Food Intake by Individuals (CSFII) 1994-96, it was not derived from this data and the CSFII was not the basis for the selection of the 142.4 g/day value.

Although water quality criteria are developed by states and tribes on a local basis, the overall function of the water quality criteria is to support maintenance of appropriate water quality throughout the United States. Individuals should be able to safely consume the amount of fish they wish to and utilize water resources from any location within the U.S. Therefore, it is EPA's policy that consumption of freshwater and estuarine fish, regardless of source, should be used to develop water quality criteria for the protection of human health.

There are a number of uncertainties to consider in evaluating the inclusion of anadromous species, such as migrating salmon, in the FCR used to compute human health water quality criteria. Although EPA has, on a national basis, considered salmon to be largely a marine species, there is uncertainty as to the body burden of contaminants that salmon acquire in fresh water. Further, the marine food web that adult salmon rely upon is potentially affected by contaminant sources regulated under the Clean Water Act. The importance of considering these uncertainties is magnified by the fact that large amounts of salmon are consumed by tribal members and others in Idaho with associated contaminant exposures and consequent health risks. Yet another source of uncertainty is the market basket preferences of consumers. Individuals desiring to consume fish generally will vary in the species they consume. To protect the public in the face of these uncertainties, EPA believes that salmon should be included in the fish consumption rate used to derive human health water quality criteria in Idaho. Specifically, EPA believes that including salmon at a discounted rate (Option 2) or not including salmon at all (Option 3) would not be appropriate in Idaho.

There are additional arguments for including salmon in the fish consumption rate for chemicals regulated on the basis of their non-cancer toxicity. For these chemicals, EPA considers overall contaminant exposures in determining what allowable exposures are via fish consumption and drinking water ingestion. Specifically, the allowable exposure via fish consumption and drinking water should equal the total allowable exposure minus the exposure from other sources. EPA generally implements this consideration via the "relative source contribution" (RSC) term. The RSC is the fraction of the reference dose (e.g. the allowable daily intake) that can be allocated to fish consumption and water ingestion after considering the dose contributed by other routes of exposure. Though EPA guidance has stated that the primary approach for dealing with exposure to contaminants in salmon should be via reducing the RSC, it is difficult to do this using data because the necessary information is not always available. An alternate, acceptable approach to reducing the RSC is to fully include salmon consumption in the fish consumption rate. Assuming that Idaho would use the fish consumption rate to address exposure to contaminants found in anadromous fish, EPA believes that neither Option 2 nor Option 3 are appropriate.

EPA also has attempted to develop a cohesive regional perspective as it works with states and tribes to develop and/or update human health water quality criteria. Given that Washington and Oregon, which are downstream of Idaho, are considering including or have included salmon in their fish consumption rate, implementation of human health water quality criteria throughout the Pacific Northwest would be facilitated by uniformly including salmon in the fish consumption rate for Idaho. For all of the aforementioned reasons, EPA believes that salmon should be fully included in the fish consumption rate used to develop Idaho's human health water quality criteria.

We look forward to continued work with DEQ on this effort and are available if you would like to discuss our comments further. Please contact Lon Kissinger (206-553-2115) or myself (206-553-1834) if you have any questions.

Sincerely,

Lisa Macchio
Water Quality Standards Coordinator

ACWA Monitoring, Standards & Assessment Committee Call
Wednesday, April 17, 2013

Subject: Discussion on EPA's new FAQ: *Human Health Ambient water Quality Criteria and Fish Consumption Rates Frequently Asked Questions*: http://water.epa.gov/scitech/swguidance/standards/criteria/health/methodology/upload/hhfaqs.pdf

Comments from Washington & Idaho:

Cheryl Niemi, Washington Department of Ecology, cnie461@ecy.wa.gov
Don Essig, Idaho Department of Environmental Quality, don.essig@deq.idaho.gov

Overall comment:

Several states are dealing with development of human health criteria as they revise standards. Washington and Idaho are both starting the process and are dealing with particularly complex issues because of abundance of fisheries for anadromous fish, subpopulations that consume large amounts of anadromous and local fish and shellfish, a lack of state-specific data on the fish and shellfish consumption patterns of the general population, and a very motivated and concerned set of stakeholders who all have important interests to address. In addition, in Idaho and Washington there have been recent communications with EPA Region 10 that indicate that EPA is considering development of regional guidance or other decision-making processes on human health criteria development that could seriously affect the ability of the states to make the risk management decisions that have historically and appropriately been made by states – decisions on such issues as risk levels and fish consumption rates. This had led to an uncertain rule-making environment, and a real concern that EPA might develop guidance that could act as rule. Launching this FAQ into such a highly charged environment, without the benefit of state review and consideration of the issues being addressed in state rule-makings, is of significant concern.

Specific comments:

In the left column below is a copy of the EPA FAQ. The FAQ is divided below into a table format to facilitate discussion of individual Question/Response topics. State comments/concerns with the information in the FAQ are in the right column. Specific comments in each section are numbered across the columns to clarify the linkage between highlighted FAQ language and state comments/concerns.

Document Title:

Human Health Ambient water Quality Criteria and Fish Consumption Rates

Frequently Asked Questions

[Note: the answers below reflect existing EPA policy and guidance, as articulated in the 2000 Human Health Methodology]

Abbreviations:

HHC – Human health criteria, WQS = water quality standards, SDWA = Safe Drinking Water Act, CWA = Clean Water Act, RSC = Relative Source Contribution, MCLG = Maximum Contaminant level Goal, MCL = Maximum Contaminant Level

EPA FAQ Language	State Comment/concern
This guidance does not have a disclaimer.	EPA's new FAQ on multiple discharger variances (EPA-820-F-13-012, March 2013) contains some introductory language that clarifies the role of that FAQ – explaining that it is guidance and not rule. A similar disclaimer is desirable for this Fish Consumption Rate FAQ (EPA variance FAQ language below): ***DISCLAIMER*** *These Frequently Asked Questions (FAQs) do not impose legally binding requirements on the EPA, states, tribes or the regulated community, nor do they confer legal rights or impose legal obligations upon any member of the public. The Clean Water Act (CWA) provisions and the EPA regulations described in this document contain legally binding requirements. These FAQs do not constitute a regulation, nor do they change or substitute for any CWA provision or the EPA regulations.* *The general description provided here may not apply to a particular situation based upon the circumstances. Interested parties are free to raise questions and objections about the substance of these FAQs and the appropriateness of their application to a particular situation. The EPA retains the discretion to adopt approaches on a case-by-case basis that differ from those described in these FAQs where appropriate. These FAQs are a living document and may be revised periodically without public notice. The EPA welcomes public input on these FAQs at any time.*

Q1. What is the goal of the human health ambient water quality criteria? Clean Water Act (CWA) section 303(c)(2)(A) requires that water quality standards (WQS) protect “public health or welfare, enhance the quality of the water and serve the purposes of [the Act].” CWA section 101(a)(2) establishes as a national goal “water quality which provides for protection and propagation of fish, shellfish, and wildlife, and recreation in and on the water, wherever attainable.” EPA has interpreted the “fishable” language in section 101(a)(2) to refer not only to protecting water quality so the fish and shellfish thrive, but when caught they can also be safely eaten by humans. Thus, to be consistent with section 101(a)(2),the applicable criteria for such “fishable” designated uses must not only protect the aquatic organisms themselves, but also protect human health through consumption of fish and shellfish.1 1 See memorandum from Geoffrey H. Grubbs and Robert H. Wayland (October 2000) posted at http://water.epa.gov/scitech/swguidance/standards/upload/2000_10_31_standards_shellfish.pdf EPA’s recommended 304(a) water quality criteria to protect these “fishable” designated uses, and accompanying risk assessment methodologies, reflect the longstanding interpretation that a designated use consistent with the goals of the Act means that State and Tribal waters should support safe consumption of fish and shellfish. EPA has consistently implemented the Clean Water Act to ensure that the total rate of consumption of freshwater and estuarine fish and shellfish (including estuarine species harvested in near coastal waters) reflects consumption rates demonstrated by the population of concern. In other words, EPA expects that the standards will be set to enable residents to safely	**Comment 1.** Suppression effects are a very sensitive topic for many groups in the Pacific Northwest, and it is difficult to apportion the amount of suppression caused by different factors. Unfortunately the concepts of *availability* of fish and *contamination* of fish get mixed up. Some specific language here that speaks directly to the possible causes of suppression, and then directly pinpoints the suppression linked to contamination, would be useful for readers. It would also be helpful to acknowledge the difficulty in accurately quantifying suppression.

consume from local waters the amount of fish they would normally consume from all fresh and estuarine waters (including estuarine species harvested in near coastal waters). EPA does not necessarily expect all consumers to eat only fish from a single State, but individuals or groups should be able to do so without concern for their health. (see **comment 1** at right)It is also important to avoid any suppression effect that may occur when a fish consumption rate for a given subpopulation reflects an artificially diminished level of consumption from an appropriate baseline level of consumption for that subpopulation because of a perception that fish are contaminated with pollutants.

This approach is consistent with a principle that every State does its share to protect people who consume fish and shellfish that originate from multiple jurisdictions. In addition, the goal of water quality criteria for human health is to protect people from exposure to pollutants through fish and water over a lifetime, and the goal of a State's designated use should be that the waters are safe to fish in the context of the total consumption pattern of its residents. Likewise, because people are expected to continue consuming fish and shellfish throughout their lifetime regardless of where they live, and this consumption leads to similar exposure to pollutants, it is appropriate to derive protective human health criteria in State and Tribal water quality standards assuming a lifetime of exposure.

Although the human health ambient water quality criteria (AWQC) are based on chronic health effects data (both cancer and noncancer effects), the criteria are intended to also be protective against adverse effects that may reasonably be expected to occur as a result of elevated acute or short-term exposures.

Q2. What does the fish consumption rate (FCR) indicate in the calculation for human health ambient water quality criteria? The FCR indicates the amount of fish and shellfish in kilograms consumed by a person each day. For the purposes of human health ambient water quality criteria, the fish and shellfish to be reflected in the FCR include all of the fish and shellfish consumed that are species found in fresh and estuarine waters (including estuarine species harvested in near coastal waters). (see **comment 1** at right)Because the overall goal of the criteria is to allow for a consumer to safely consume from local waters the amount of fish they would normally consume from all fresh and estuarine waters, the FCR does include fish and shellfish from local, commercial, aquaculture, interstate, and international sources. It is not necessary for the FCR to include fish and shellfish species designated as marine species, as that exposure is addressed by relative source contribution (see question 4 for more detail). However, partitioning of fish and shellfish into the different habitats in order to develop a FCR can only be done where sufficient data are available for this to be done in a scientifically defensible manner. For example, if a State were to determine through scientifically collected data that its citizens consumed 25 grams of fish and shellfish per day where 5 grams came from marine fish, 5 grams came from a local fresh water stream (see comment 1 at right) 5 grams came from a neighboring state's fresh waters, 5 grams came from international imports of estuarine shellfish, and 5 grams came from aquaculture of a freshwater species, then the FCR would be 20 grams per day. Only the marine fish component would be excluded from the FCR (see discussion below on relative source contribution). (see **comment 2** at right) All of the other components represent the amount of fish and shellfish that could	**Comment 1.** If the overall goal is to allow consumers to safely consume freshwater and estuarine fish resources from local waters, then including all the fish and shellfish consumed from interstate and international sources does not make sense. The amount of consumption associated with the commercial availability of these sources does not necessarily reflect the amount of fish or shellfish that are, were, or might be attainable in local waters. For instance, a person from a state with no marine coastline might eat large amounts of prawns and bivalves harvested in a foreign country and purchased at the supermarket. This consumption does not reflect exposures from local waters or the fishery resources that would naturally be there. This consumption should be considered during the development of the RSC (if data are available to document contaminants in these new fishery sources (such as mercury in tuna)), but not in the overall FCR. Aquaculture resources are complex. Many types of aquaculture are practiced. Some types are almost completely dependent on the local waters for support (e.g. oyster industry), others use a mixture of in-situ exposure of local water and commercial or proprietary feed stock (e.g. net pens), and still others use upland facilities with waters piped to the facility in a manner analogous to industrial water use and combined with commercial or proprietary feed stock (upland facilities raising tilapia). The first type of aquaculture venture could closely fit the definition of locally harvested resources, the second is more ambiguous, and the third is more similar to an industrial operation and not a local waterbody harvest issue. Including all resources from aquaculture in the FCR does not take the complexity of these different types of exposure sources into account. The different sources merit further discussion to

be taken and consumed from local waters if the consumer chose to do so.	determine whether they should be included in the FCR and when they should be considered for development of the RSC. **Comment 2.** The last sentence states that all of the "*components represent the amount of fish and shellfish that could be taken and consumed from local waters if the consumer chose to do so*." This does not make sense. The international and national market for fisheries has created a market situation where people who previously would have little harvest available locally could (by eating commercially available non-local fish or shellfish) enhance their consumption to levels that would more closely mirror locally supported consumption patterns in areas with locally abundant fishery resources – but do not mirror the "*amount of fish and shellfish that could be taken and consumed from local waters if the consumer chose to do so.*"
Q3. How is the exposure to a pollutant due to marine fish consumption accounted for in the human health ambient water quality criteria? Human health ambient water quality criteria are to account for all sources of exposure to the pollutants for which they are developed. The exposure to pollutants from marine fish and shellfish species that are not included in the fish consumption rate should be accounted for in the relative source contribution (RSC) when setting criteria for threshold non-carcinogens and non-linear carcinogens.	**No comment.**

Q4. What does the relative source contribution (RSC) indicate in the calculation for the human health ambient water quality criteria? The relative source contribution component of the human health ambient water quality criteria (AWQC) calculation for threshold non-carcinogens and non-linear carcinogens allows a percentage of the reference dose's exposure to be attributed to ambient water and freshwater and estuarine fish consumption (including estuarine species harvested in near coastal waters) when there are other potential exposure sources. (see **comment 1** at right) The rationale for this approach is that for pollutants exhibiting threshold effects, the objective of the AWQC is to ensure that an individual's total exposure from all sources does not exceed that threshold level. The RSC includes, but is not limited to, exposure to a particular pollutant from marine fish consumption (not included in the fish consumption rate), non-fish food consumption (fruits, vegetables, and grains), dermal exposure, and respiratory exposure. In the absence of scientific data, the application of the EPA's default value of 20 percent RSC in calculating 304(a) criteria or establishing State or Tribal water quality standards under Section 303(c) will ensure that the designated use for a water body is protected. (see **comment 2** at right – boldface added) This 20 percent **default for RSC can only be replaced where sufficient data are available to develop a scientifically defensible alternative value**. If appropriate scientific data demonstrating that other sources and routes of exposure besides water and freshwater/estuarine fish are not anticipated for the pollutant in question, then (see **comment 3** at right – boldface added) the RSC **may be raised to the appropriate level, based on the data, but not to exceed 80 percent.** The 80 percent ceiling accounts for the	**Comment 1.** The 20%/80% RSC approach in the EPA 2000 guidance was developed as part of a process to "harmonize" the SDWA and the CWA. See EPA 2000 (bottom of page 1-5): *"Another reason for the 2000 Human Health Methodology is the need to bridge the gap between the differences in the risk assessment and risk management approaches used by EPA's Office of Water for the derivation of AWQC under the authority of the CWA and Maximum Contaminant Level Goals (MCLGs) under the Safe Drinking Water Act (SDWA). Three notable differences are the treatment of chemicals designated as Group C, possible human carcinogens under the 1996 proposed cancer guidelines, the consideration of non-water sources of exposure when setting an AWQC or MCLG for a noncarcinogen, and cancer risk ranges."* The SDWA MCLG derivation procedures use a 20%80% approach. Applying this RSC range to CWA HHC provides some harmonization between the two Acts, but does not take into account that the MCLG is not a regulatory level (it is a goal), and that the CWA human health criteria (HHC) are regulatory levels enforced both as ambient concentrations in the water body (303(d) listing process and through NPDES permit limits.) Under the SDWA the MCLG is modified to create an at-tap regulatory level (the maximum contaminant level - MCL) by taking into account factors such as available treatment and available analytical methods. Here is an example for nitrate taken from EPA's website (boldface added) at http://water.epa.gov/drink/contaminants/basicinformation/nitrate.cfm that gives some explanation of how MCLs are developed from MCLGs:

fact that some sources of exposure may be unknown. In cases where an 80 percent RSC is used, 20 percent of the exposure is reserved for unknown sources. Although the 20 percent RSC has not been consistently applied to national 304(a) criteria recommendations for non-carcinogenic pollutants, where there are inconsistencies between the 2000 Human Health Methodology recommendation and implementation in criteria, the Human health Methodology should prevail and the 20 percent RSC applied. EPA is moving to complete implementation of this guidance in existing 304(a) criteria.	*“The MCLG for nitrate is 10 mg/L or 10 ppm. EPA has set this level of protection based on the best available science to prevent potential health problems. EPA has set an enforceable regulation for nitrate, called a maximum contaminant level (MCL), at 10 mg/L or 10 ppm.* ***MCLs are set as close to the health goals as possible, considering cost, benefits and the ability of public water systems to detect and remove contaminants using suitable treatment technologies. In this case, the MCL equals the MCLG, because analytical methods or treatment technology do not pose any limitation.”*** NPDES permitting tools can sometimes accommodate some of these considerations during implementation, but final limits must always be based on the HHC. The larger reason why the MCLG does not mirror the HHC is that the MCLG is not in itself a level that must be attained, while the HHC is always a level that must be attained in ambient waters. The roll-out of this difference is apparent with the application of the 303(d) program, the requirement for allocation of loads, and subsequent permitting requirements found at 40 CFR 122.4(i) and the Pinto Creek decision (http://cdn.ca9.uscourts.gov/datastore/opinions/2007/10/03/0570785.pdf . In this context, the HHC themselves are strong regulatory numbers that drive resource intensive programs. This is important because the risk management/policy decision to use a RCS of 20% to 80% in the MCLG itself has no regulatory outcome – it simply provides a backdrop for development of the MCL. The risk management/policy decision to use a 20% to 80%

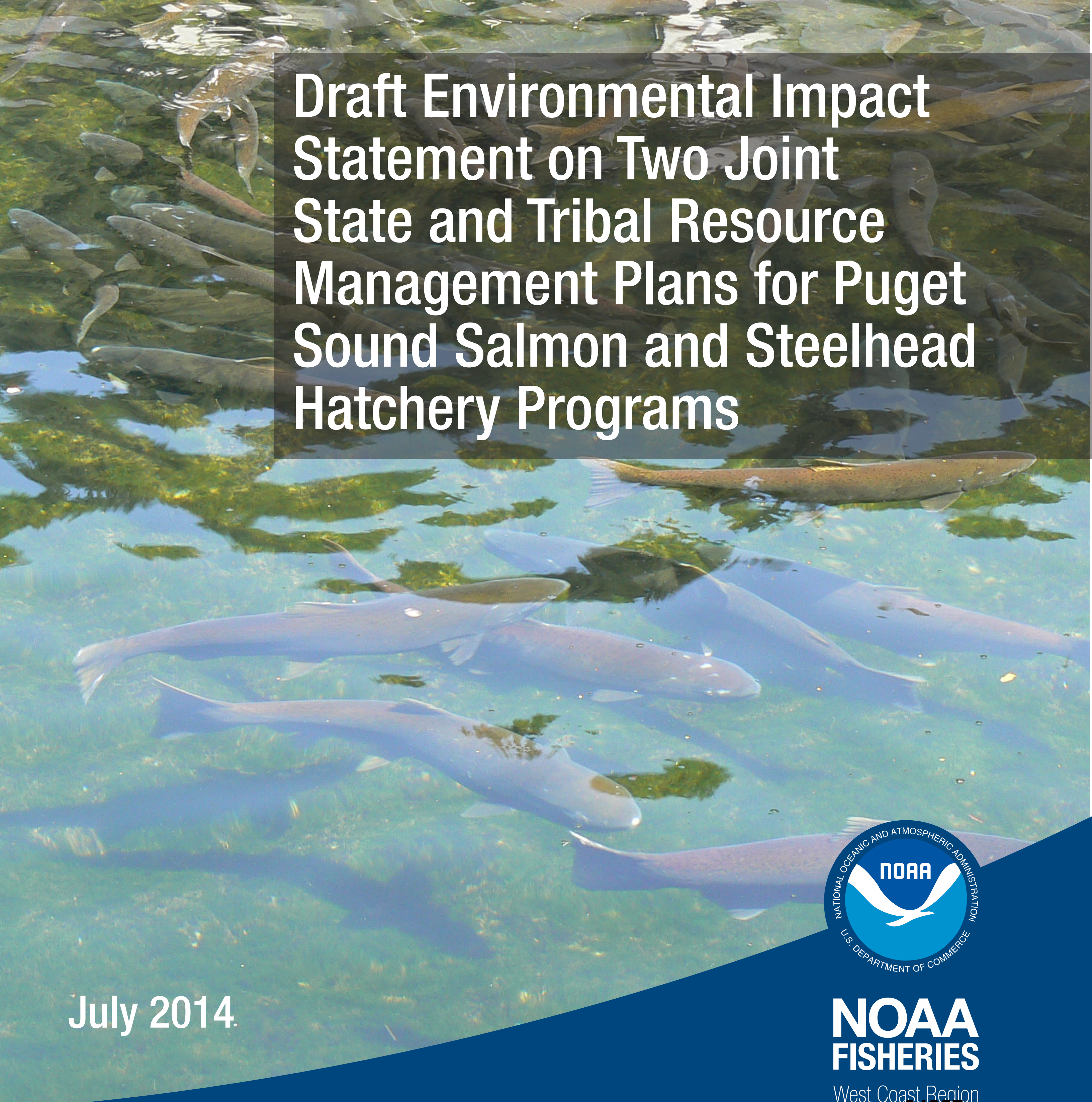
	RSC in HHC development directly affects a regulatory value with potentially large economic consequences (see Pinto Creek decision). We believe this background discussion is relevant because maybe the risk management/policy decision to use the SDWA RSCs to harmonize with the CWA HHC should be reconsidered now that states have had time to examine more fully the EPA 2000 guidance. The decision to try to harmonize the development of the MCLG and the CWA HHC may be like trying to harmonize apples and oranges: both are fruit – both are different from each other. Different regulatory programs address the same chemicals and effects in different ways in order to fulfill the requirements of enabling legislation, regulations, and local needs. Applying a default assumption (RSC = 20% to 80%) that might have no affect on a regulatory level (the MCL) from one program, to another program (NPDES) where the assumption can drive huge resource and compliance issues (through requirements to meet HHC in ambient waters) does not necessarily make sense. Trying to harmonize programs or regulatory levels seems like a good idea on the surface, but trying to harmonize programs or regulatory levels that are not completely analogous is not necessarily a good idea. An alternative to using the 20%-80% range would be to apply 100% as the RSC. 100% has been the RSC value traditionally incorporated into HH criteria development for the non-carcinogens, unless additional data to identify other exposure pathways are available (e.g., the new mercury HH criteria). Maybe this is the way to go until this issue has had more discussion. An inherent assumption in how the RSC for HHC is developed is that all other sources of the contaminant are

	required to be considered in the exposure scenario, and the HHC get the “left over” part of the reference dose. This results in the odd situation where, as the contribution of a contaminant from water becomes less and less important (a smaller part of the RfD allowed in water), the HHC get more and more stringent – in effect becomes a bigger and bigger driver for more restrictive limits. Because other regulatory programs (e.g., FDA action levels and food tolerances, SDWA MCLs, Superfund clean-ups) target lower levels of protection, the CWA program is at the mercy of the regulatory levels set in other programs, and is expected to “clean-up” the waters that are allowed higher levels of pollution than these other sources (even when these other sources may be ongoing sources even after their regulatory requirements have been fulfilled). It would be interesting to have a broader national discussion on how the RfD for any individual chemical is allocated among different regulatory programs. Maybe it would make more economic sense, and more opportunities might be available, to try to cut down the levels of contaminants allowed in other regulatory programs (that are based on cost, feasibility, etc.) so that the CWA criteria could focus only on the designated uses and CWA-regulated pollution sources within the geographic jurisdiction of each state. **Comment 2.** This reads like rule language instead of guidance. **Comment 3.** This reads like rule language instead of guidance.
Q5. Should an RSC also be applied to carcinogens? In the case of carcinogens based on linear low-dose extrapolation, the AWQC is determined with respect to the *incremental* lifetime risk posed by a substance’s presence in water, and is not being set	**Comment 1.** This statement in the FAQ causes confusion about who has the responsibility for making risk management decisions with regard to both risk level and FCR: EPA regions or the states? It would be useful for EPA to include a statement in

with regard to an individual's total risk from all sources of exposure. Thus, the AWQC represents the water concentration that would be expected to increase an individual's lifetime risk of carcinogenicity from exposure to the particular pollutant by no more than one chance in one million, regardless of the additional lifetime cancer risk due to exposure, if any, to that particular substance from other sources. For human health criteria, this exposure pathway considers consumption of freshwater and estuarine fish and shellfish (as described in the responses to Q1 and Q2) and drinking water ingestion. (see **comment 1** at right) EPA recommends that the incremental cancer risk from these exposure pathways not exceed more than 1 in 1,000,000 or 1 in 100,000 for the general population, nor exceed more than 1 in 10,000 for any sensitive sub-population (such as those who may consume a great deal more fish because of a subsistence lifestyle). States and tribes may consider adjusting the risk level according to guidance in the 2000 Human Health Methodology (and mentioned above), particularly if exposure to "other" sources besides water and fish is determined to be significant.	this FAQ similar to its statement in the 2000 Human Health methodology that: *"EPA believes that ambient water quality criteria inherently require several risk management decisions that are, in many cases, better made at the State, Tribal, or regional level."* This issue is particularly important, in an immediate sense, for Washington and Idaho. Both states have been told by EPA Region 10 that the Region is considering developing "region-specific" guidance (or some other framework to look at approvable criteria) on HHC, including risk levels and FCRs. The states have also been told that Region 10 thinks "the Oregon outcome was the right outcome." The Oregon outcome included risk management decisions, appropriately made by that state, for a FCR that included salmon consumption and application of that rate to a state-determined risk level. Washington and Oregon are concerned that development of regional guidance will usurp the risk management decisions appropriately and historically made by states, and instead have them made by EPA. If this is the approach then the issue of "rule-by-guidance" becomes important.
Q6. Could a state include a component of marine fish consumption in their FCR for deriving human health criteria? Yes, a state may include consumption of marine species in the FCR. (see **comment 1** at right) Coastal States and authorized Tribes that believe accounting for total fish consumption (i.e., freshwater/estuarine and marine species) is more appropriate for protecting the population of concern may do so. In the instance	**Comment 1.** As discussed above in the comments on Q2, commercial markets make marine fishery resources available to consumers in all states. Inland states may have just as much, or even maybe more, fish of marine origin sold in their markets than coastal states. This seems to be analogous to the inclusion of consumption of imported fish/shellfish from waters outside the US in the FCR used to calculate criteria. However, as

where the FCR includes freshwater, estuarine and all marine fish consumption, EPA recommends that states adjust the RSC estimate to reflect a greater proportion of the reference dose being attributed to water intake and the marine-inclusive FCR exposures. Including marine fish in the fish consumption rate may be particularly appropriate if a large proportion of fish consumption for the population to be protected consists of marine fish (such as salmon) and this exposure is clearly documented. Including marine fish in the fish consumption rate for criteria calculations would provide some calculations that are more stringent than those that don't include marine fish consumption, particularly for chemicals that are highly bioaccumulative.	discussed above, it seems that there is still much to discuss around the use, in HHC development, of commercially acquired fishery resources and resources from marine waters that are outside the jurisdictional boundaries of states. A serious national public policy discussion needs to take place about what we are trying to achieve by including non-local fish in the basis for water quality criteria that are going to be used to regulate local waters.
Q7. When fish consumption exposure is represented by a distribution of values, what are the appropriate percentiles to choose? In general, EPA considers protection of the general population to be represented by the 90th percentile of a total exposure distribution utilizing a "per capita" fish consumption distribution. If present in the state, subsistence fishers should be considered on a site specific basis. EPA has recommended the 99th percentile of a per capita fish consumption distribution as a surrogate for subsistence fishers, which corresponded to a range of average consumption estimates from actual surveys for subsistence fishers. (see **comments 1** at right) An analysis of protectiveness of the criteria for the general population, recreational fishers and subsistence fishers should be included in the criteria documentation.	**Comment 1.** We think it is clear from the EPA 2000 guidance, as reiterated in this FAQ, that final criteria development should be underlain by clear statements on risk management decisions made by the states and on the levels of risk/protection that are provided by new HHC. As stated at left, that clarity should apply to "the general population, recreational fishers and subsistence fishers." Note: Idaho's request for assistance in planning and/or conducting a survey of the general population of Idaho was recently refused by EPA. Given this FAQ direction, we would like greater clarification from EPA on why they were not supportive given their statement highlighted at left.

Draft Environmental Impact Statement on Two Joint State and Tribal Resource Management Plans for Puget Sound Salmon and Steelhead Hatchery Programs



July 2014

NOAA FISHERIES
West Coast Region

U.S. Department of Commerce | National Oceanic and Atmospheric Administration | National Marine Fisheries Service

Puget Sound Hatcheries Draft EIS

Appendix K

Chemicals Used in Hatchery Operations

Table of Contents

Hatchery operations routinely use a variety of chemicals to maintain a clean environment for the production of disease-free fish. These chemicals and safe handling requirements for the chemicals are described in this appendix. A brief description of commonly used chemicals in hatchery facilities and operations is provided below. In addition, a literature review is provided describing the potential for toxic contaminants in salmon and steelhead. This appendix provides information in support of Environmental Impact Statement (EIS) Subsection 3.7, Human Health, and EIS Subsection 4.7, Human Health.

1.0 Commonly Used Hatchery Chemicals

Common chemicals used in hatchery operations are disinfectants, therapeutics, anesthetics, pesticides and herbicides, and feed additives.

1.1 Disinfectants

Disinfectants are primarily used to clean equipment throughout hatchery facilities and may also be used to treat fish diseases. Hatchery facility workers are typically exposed to these chemicals through skin contact or inhalation during cleaning activities. However, Federal and state occupational safety and health programs (e.g., Occupational Safety and Health Act [OSHA], Washington State Industrial Safety and Health Act [WISHA]) ensure safe workplaces and require personal protective equipment and procedures (e.g., gloves, use of proper ventilation procedures, and/or respiratory protection in enclosed spaces). Following directions on product labels and using other hatchery-specific safety measures reduces chemical exposure to safe levels. Some common disinfectants used in hatchery operations are described below.

- **Chlorine (sodium hypochlorite).** Sodium hypochlorite is used for cleaning tanks and equipment and is the active component in chlorine. This compound may also be used to destroy fish fry that are infected with a disease.
- **Chloramine T.** Chloramine T is used for disinfecting tanks and equipment, and the treatment of bacterial gill diseases in salmon and steelhead. The active component is chlorine.
- **Formalin.** Formalin is a saturated aqueous solution of formaldehyde. It is used as a general disinfectant and is effective against fungal or parasitic infections.
- **Hydrogen peroxide.** Hydrogen peroxide is used as a general disinfectant and is effective against fish parasites (e.g., sea lice).
- **Iodophor.** Iodophor is a form of stabilized iodine employed as a general disinfectant. It is used to disinfect fish eggs and is effective against some bacteria and viruses.

- **Quaternary ammonium compounds (Hyamine).** Ammonium compounds or topical disinfectants are used to remove parasites from fish and have detergent and antibacterial properties.

1.2 Therapeutics

Therapeutics, which include antibiotics, are chemicals or veterinary medicines designed to be effective against parasitic, bacterial, or viral infections in fish. The most commonly used therapeutics in salmon and steelhead hatchery operations are:

- **Amoxicillin.** Amoxicillin is generally used as a veterinary antibiotic.
- **Erythromycin.** Erythromycin is generally used as a veterinary antibiotic.
- **Florfenicol.** Florfenicol is generally used as a veterinary antibiotic.
- **Oxytetracycline (terramycin).** Terramycin is widely used as an antibiotic. Oxytetracycline may be applied orally in fish feed or as a bath and is effective against a wide range of bacteria.
- **Potassium permanganate.** Potassium permanganate is primarily used as a bath treatment for fungal infections of finfish. It may also be used to alleviate acute oxygen shortage and to remove organic contaminants in fish ponds.
- **Penicillin.** Penicillin is generally used as a veterinary antibiotic.
- **ROMET®.** ROMET® is typically applied in fish feed and used to control a variety of bacterial infections.
- **Sulfamethazole trimethoprim.** Sulfamethazole trimethoprim is generally used as a veterinary antibiotic.
- **Vaccines.** Vaccines are generally used to treat viral diseases. There are a variety of vaccines available to treat animals in aquaculture. Salmon may be given vaccines to treat furunculosis, vibriosis, or yersiniosis. These vaccines are generally not considered a potential risk for human health since viral diseases of fish are typically not pathogenic to humans (World Health Organization [WHO] 1999), and the potential for exposure is minimal. The primary exposure pathway tends to be through accidental needle-stick injury (Douglas 1995; Leira and Baalsrud 1997).

Therapeutics typically are only applied when fish health specialists have determined that a disease is present in fish rearing in hatcheries. Human exposure to these chemicals typically would occur through skin contact by hatchery workers during application of the compound or through accidental needle pricks during vaccinations. However, Federal and state occupational safety regulations (e.g., Occupational Safety and Health Act of 1970 [29 United States Code [USC] 651 et seq.]) are in place to prevent these types of accidents.

Outside of the use of therapeutic chemicals in the workplace, there are two primary environmental concerns with the use of therapeutics in hatchery facility operations:

1. Therapeutic substances are not 100 percent absorbed by the fish and may be excreted into the holding water (Texas Agricultural Extension Service 1994; Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection [GESAMP] 1997; Milewski 2001). Government agencies typically do not regulate disposal of chemicals in fish waste products; therefore, there is a potential for these chemicals to enter the environment surrounding the hatcheries (Texas Agricultural Extension Service 1994; GESAMP 1997; Milewski 2001). Federal Clean Water Act and state surface water regulations prevent the discharge of chemicals at concentrations that may pose a threat to human health. However, water quality regulations currently do not exist for all veterinary products, medicines, or their by-products when incompletely metabolized. The environmental persistence of therapeutic substances varies, and some may degrade in a few hours to a few months (GESAMP 1997). Antibiotics used at hatcheries have been detected in receiving waters downstream of aquaculture operations (Boxall et al. 2004; Pouliquen et al. 2009; Martinez-Bueno et al. 2009). Moreover, studies suggest these compounds may persist in sediments (Pouliquen et al. 2009; Martinez-Bueno et al. 2009).

   Therapeutics are typically applied infrequently and at low doses (GESAMP 1997). The use of therapeutics is governed by the Federal Drug Administration (FDA) through the Animal Medicinal Drug Use Clarification Act of 1994 (21 Code of Federal Regulations [CFR] 530), which does not permit extra-label use of drugs that are administered through feed (MacMillan et al. 2006). Currently, the volume of therapeutics released from hatcheries and the potential risks associated with these releases are unknown. Concentrations that have been reported in receiving waters near fish farms and hatcheries in other parts of the United States and in Europe are usually well below those toxic to fish and invertebrates (Boxall et al. 2004). It is expected that limited use of veterinary medicines following label instructions in U.S. fish hatcheries poses minimal risk to human health and the environment (GESAMP 1997; MacMillan et al. 2006), although locally high concentrations could occur depending on the nature of the receiving environment.

2. The use of antibiotics may increase the potential for the development of resistance in certain strains of bacteria (Burka et al. 1997; GESAMP 1997; WHO 1999). Therefore, overuse of antibiotics could render them ineffective for control of some bacteria. Resistant bacteria that infect fish have the potential to transfer resistant genetic material to bacteria that infect non-fish organisms (e.g., humans). Genetic bacterial resistance may occur by the movement of plasmids (i.e., genetic elements independent of the chromosome) between bacteria. This type of transfer has been demonstrated in a number of microorganisms (Burka et al. 1997; GESAMP 1997; WHO 1999; Cabello 2006). Therefore, the improper use of antibacterial antibiotics may cause resistance in bacterial pathogens that can infect humans (Burka et al. 1997; GESAMP 1997; WHO 1999; Cabello 2006). The use of therapeutics is governed by the FDA through the Animal Medicinal Drug Use Clarification Act of 1994 (21 CFR 530), which does not permit therapeutics for uses not specified in the drug’s label (MacMillan et al. 2006). Adhering to this regulation and drug label recommendations minimizes the potential for the development of antibiotic resistance.

1.3 Anesthetics

Anesthetics are commonly used to immobilize fish during egg or milt collection, to calm fish during transportation, or during treatment with other therapeutics. They are typically applied or used at low concentrations and, thus, represent a low risk to human health (GESAMP 1997) when handled using general safety precautions (i.e., Federal OSHA or state WISHA regulations) and following label requirements. Some common anesthetics used in hatchery operations are:

- **Benzocaine.** Benzocaine is used during egg or milt stripping or during preparation for transport.
- **Tricaine methanesulfonate (MS-222).** MS-222 is used as a general sedative and applied as a bath in the holding tanks.

1.4 Pesticides and Herbicides

A wide variety of aquatic pesticides and herbicides is used in hatchery facility operations to protect fish from parasites and remove nuisance organisms, weeds, or algae. Due to their toxicity, a number of these chemicals are not approved for use in the United States. For hatcheries, pesticides and herbicides are typically highly toxic and are used in small concentrations to control algae growth or aquatic weed growth. Commonly used algaecides approved for use in the United States may contain various forms of copper. Some common aquatic herbicides include dichlobenil, diquat, endothall, fluridone, glyphosate, 2,4-dichlorophenoxyacetic acid, and 2-butoxyethyl ester. These products may be hazardous to human health if prolonged or accidental exposure (i.e., inhalation, ingestion, or dermal contact) occurs because these compounds may be toxic at certain concentrations. Some of these products have bacteria as the active ingredient (e.g., Microbe Lift and Liquid Live Micro-organism) rather than a chemical ingredient to reduce the growth of pests. These products are typically less toxic to human health than synthetic chemicals. Safety measures on the product label and the material safety data sheet (MSDS) provide directions for proper use and applications. These safety measures, along with Federal OSHA and state WISHA regulations, serve to limit human exposure to potentially hazardous concentrations.

1.5 Feed Additives

While in hatchery facilities, hatchery-origin fish are fed with commercial diets containing fish oil and fish meal that can be from sources anywhere in the world. These feeds are known sources of toxic contaminants (Jacobs et al. 2002a; Carlson and Hites 2005; Maule et al. 2007; Johnson et al. 2010). The potential risk to human health from these contaminants is discussed further in Subsection 3.7.2, Toxic Contaminants in Hatchery-origin Fish.

Hatcheries may also use fish food that is supplemented with a variety of dietary additives. Fish raised and released from hatcheries are only fed (including dietary additives) while they are juveniles, which differs from fish raised in aquaculture farms that consume feeds and additives throughout their life. These additives may consist of artificial or natural pigments, fish oils, and/or vitamins. For example, astaxanthin and canthaxanthin are carotenoids commonly used in aquaculture to artificially color the flesh of salmon during the later stages of growth, since farm-raised fish tend to be less colorful than hatchery- or natural-origin fish. Vitamin C and Vitamin E are widely used to enhance the disease resistance of fish stocks. Exposure to feed additives from hatchery-origin fish is considered to be of low risk to human health because the concentrations used in hatcheries are typically below levels that would result in adverse health effects (GESAMP 1997).

1.6 Miscellaneous Chemicals

A variety of other chemicals are typically used at salmon and steelhead hatcheries. These chemicals are considered nonhazardous and, when used within the product label requirements and following OSHA regulations, are not expected to pose a risk to human health.

- **Anhydrous (3thyl) alcohol.** Ethyl alcohol is one of two chemicals used in a solution used to check the fertilization of eggs.
- **Lime (Type S).** Lime is widely used to neutralize acidity and increase total alkalinity of grow-out ponds.

- **Salt (sodium chloride).** Salt can be used to remove parasites or prevent stress during transport of fish.
- **Sodium thiosulfate.** Sodium thiosulfate is used to neutralize chlorine and iodophor prior to discharging wastewater.

2.0 Toxic Contaminants in Hatchery-origin Fish

Seafood consumption by humans is generally promoted due to the nutritional value of fish products. For example, fish contain elevated levels of omega-3 fatty acids, which are considered beneficial to the cardiovascular system (Mayo Clinic 2014). However, concerns have been raised that farm-raised and hatchery-origin fish may contain toxic contaminants (WHO 1999; Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004) that pose a health risk to consumers. Sources of contaminants in fish include chemicals or therapeutics, contamination of the nutritional supplements or feeds, and/or contamination of the environment where the fish are reared or released (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Carlson and Hites 2005; Johnson et al. 2007; Maule et al. 2007; Kelly et al. 2008; Johnson et al. 2010). The contaminants of primary concern are those that are persistent in the environment and are known to accumulate in the tissues of fish (e.g., methylmercury, dioxins, dichlorodiphenyltrichloroethane [DDT] and its metabolites, or polychlorinated biphenyls [PCBs]) (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Johnson et al. 2007; Maule et al. 2007; Kelly et al. 2008; Johnson et al. 2010).

Commercial diets fed to farm-raised and to hatchery-origin fish are known sources of toxic contaminants. Contaminant concentrations (e.g., pesticides, PCBs) measured in farm-raised fish are higher than in natural-origin fish (Hites et al. 2004; Hamilton et al. 2005), and the use of commercial feed in hatchery facilities may also contribute to higher concentrations of organic pollutants in hatchery-reared fish compared to their natural-origin counterparts (Johnson et al. 2007).

Hites et al. (2004) found that farm-raised salmon contained substantially more chemical pollutants than fish caught in the wild. This study suggested that these pollutants were originating from fish pellets that contain the dried and compressed body parts and toxicants from several whole fish, which they compared to a natural-origin salmon that eats a few bites of a single fish. In recent studies completed by Johnson et al. (2007), high concentrations of both PCBs and DDTs, comparable to those observed in farmed salmon, were found in juvenile hatchery-origin Chinook salmon. The authors attributed this effect in part to high body fat levels in hatchery-reared juveniles, which facilitates the uptake of lipid soluble contaminants, but concluded that there was too little information on contaminant concentrations in different lots of feed and in fish from different hatcheries, and concentrations were potentially too variable to determine how fish feed affects contaminant levels in hatchery-origin fish. The authors stated that more comprehensive sampling of fish and feed from hatcheries would be needed to determine the extent of the problem in the Pacific Northwest (which includes the project area). In a more recent study (Johnson et al. 2010), subyearling Chinook salmon were sampled from eight hatcheries that release juvenile salmon into the Columbia River. Concentrations of PCBs and DDTs were found to be lower than those reported from previous studies (i.e., in Johnson et al. 2007), and were generally comparable to levels observed in juvenile salmon from minimally contaminated rural estuaries. Contaminant concentrations were higher in the earlier study, in part, because the fish sampled were older and larger than those sampled in the more recent study, but the differences could also be related to differences in contaminant concentrations in feed or in the hatchery environment.

Various investigations have examined the amount of organic contaminants in commercial fish feeds, and found elevated levels of PCBs, polycyclic aromatic hydrocarbons [PAHs], and pesticides (Easton et al. 2002; Jacobs et al. 2002a; Jacobs et al. 2002b; Hites et al. 2004; Neergaard 2004; Carlson and Hites 2005). In a study of contaminants in fish feeds used at National Fish Hatcheries, Maule et al. (2007) found contaminants present, although generally at lower concentrations than those reported by the investigators cited above. The U.S. Geological Survey (USGS) and U.S. Fish and Wildlife Service (USFWS) have continued studying contaminants in feeds and fish (USGS 2012) at several Federal hatcheries in the USFWS Pacific Region to 1) evaluate and compare overall contaminant levels, 2) identify temporal differences in contaminant levels found in various feed forms, 3) evaluate contaminant levels and bioaccumulation rates of different commercial diets in various life-stage history classes, 4) assess the re-distribution of contaminants during smoltification, and 5) simulate the release of fish from a hatchery by fasting fish and monitoring the mobilization and re-distribution of contaminants.

Another potential source of contaminants for hatchery-origin fish includes construction materials found within hatcheries. For example, PCBs identified in fish from the Leavenworth National Fish Hatchery in the Columbia River basin were found to be related to the paint lining fish tanks (Cornwall 2005). Some hatchery facilities in Puget Sound were constructed in the early to mid-1900s and may contain chemicals in historical building materials (e.g., paint) that are banned in current materials. Other sampling for toxic substances is ongoing at national fish hatcheries (Cornwall 2005), and the U.S. Environmental Protection Agency’s (EPA’s) National Pollutant Discharge Elimination System (NPDES) general permit for Federal and tribal facilities requires hatcheries to include information on painted and caulked surfaces that regularly contact process water when they apply for general permit coverage (EPA 2009). While the potential for exposure of hatchery-raised fish to contaminants in building materials exists, further incidents have not been reported.

While hatchery-origin fish may contain chemicals of concern, the risks to humans from consumption of contaminants in hatchery-origin fish remain uncertain. The potential for human exposure to contaminants in fish is directly tied to the frequency of consuming fish (EPA 1999). Thus, consumer groups that eat large amounts of fish may have a higher potential for exposure to contaminants. Current information on consumption patterns suggests that some groups of people may consume greater quantities of fish than the general population (often termed subsistence consumers) (EPA 1999; ODEQ 2008; Ecology 2013). However, information is not available to determine what proportion of the diet of subsistence consumers comes from hatchery-origin or farm-raised fish. In addition, not all the contaminants in hatchery-origin fish are derived from hatchery facilities and their operation.

Migrating and rearing salmon and steelhead encounter and accumulate additional contaminants in the rivers, estuaries, and oceans that they inhabit (Missildine et al. 2005; Johnson et al. 2007). It is unknown what proportion of contaminants present in hatchery-origin fish originates from hatcheries and what proportion originates after release. It is also unknown whether those contaminant levels pose a risk to human health. Johnson et al. (2010) suggested that the greatest accumulation of contaminants in the bodies of hatchery-origin juvenile salmon that feed and rear in urban areas occurs after the fish are released from hatcheries. In contrast, for juvenile hatchery-origin fish that are released into relatively uncontaminated rural areas, hatcheries can be a primary source of contaminants. Contaminants accumulated during hatchery rearing would probably contribute very little to concentrations of contaminants in returning adult salmon, since concentrations acquired only during the relatively short juvenile rearing period would be diluted as the fish grew larger to adulthood. Studies suggest that, for returning adult salmon, most of the contaminants present in their bodies are acquired during their time at sea (Kelly et al. 2007; Cullon et al. 2009; O’Neill and West 2009). An exception would be resident Chinook salmon that rear in Puget Sound (about 4 percent of Chinook salmon releases), and may carry a heavier load of contaminants than other salmon that spend more time at sea. Outside of resident Chinook salmon, there is no available information that demonstrates hatchery-origin fish have a greater proportion of contaminates than natural-origin fish, and thus, it is assumed that hatchery-origin salmon and steelhead do not present a greater threat of contamination than natural-origin salmon and steelhead. The Washington Department of Fish and Wildlife currently monitors toxic contaminants in fish and other organisms, as a member of the Puget Sound Ecosystem Monitoring Program.

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CAAP
General Permit
Permit No.: MTG130000

MONTANA DEPARTMENT OF ENVIRONMENTAL QUALITY

CONCENTRATED AQUATIC ANIMAL PRODUCTION GENERAL PERMIT

AUTHORIZATION TO DISCHARGE UNDER THE

MONTANA POLLUTANT DISCHARGE ELIMINATION SYSTEM

In compliance with Montana Water Quality Act, Title 75, Chapter 5, Montana Code Annotated (MCA), and the Federal Water Pollution Control Act (the "Clean Water Act"), 33 U.S.C. 1251 et. seq., applicants issued an authorization letter for this Fish Farm General Permit, are permitted to discharge wastewater effluent from fish farms and hatcheries to state waters in accordance with effluent limitations, monitoring requirements and other conditions set forth herein.

A written authorization letter from the Department is required before an applicant is authorized to discharge under the Fish Farm General Permit.

This permit shall become effective: **July 1, 2011**.

This permit and the authorization to discharge shall expire at midnight: **June 30, 2016**.

COPY

FOR THE MONTANA DEPARTMENT
OF ENVIRONMENTAL QUALITY

Jenny Chambers, Chief
Water Protection Bureau
Permitting and Compliance Division

Issuance Date: July 1, 2011

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I. EFFLUENT LIMITATIONS, MONITORING REQUIREMENTS, & OTHER CONDITIONS

A. Specific Effluent Limitations

Effectively immediately upon issuance of an authorization under this general permit and lasting for the duration of the permit, the following effluent limitations apply to all fish farm facilities covered by this general permit.

1. All facilities must develop and implement a Best Management Practices (BMP) plan to minimize the discharge of hatchery wastes to state waters. The plan shall be developed and submitted to the Department for review and approval, postmarked within 90 days of the date on the authorization letter. Thereafter the plan shall be updated annually and a copy, dated and signed by the facility manager, shall be kept onsite and be available for inspection. The plan must include the minimum requirements described in Part I.C.

2. There shall be no discharge of polychlorinated byphenyls (PCBs) in excess of 0.00065 µg/L in any sample. Analytical results less than the required reporting value (RRV) of 1 µg/L shall be reported as zero on the DMR and will be considered in compliance with this limit.

3. Drug and chemical use shall be limited to those approved by the Food and Drug Administration for use in aquaculture in accordance with label requirements. Pesticides must be registered for use in Montana by the Montana Department of Agriculture. Any extra-label use of approved drugs and chemicals or use of unapproved drugs and chemicals will require case-by-case approval by the Department prior to the discharge to state waters. All drug and chemical use shall be documented in the annual BMP plan.

4. Any additional requirements specified in the authorization letter.

B. Self-Monitoring Requirements

Upon the effective date of this permit, the following constituents shall be monitored at the frequency and with the type of measurement indicated; samples or measurements shall be representative of the volume and nature of the monitored discharge.

Discharge monitoring shall be conducted at the end of pipe, prior to discharge to the receiving water. Samples from multiple discharge pipes shall be flow proportioned and composited prior to analysis.

Parameter	Frequency	Type[1]	RRV
Flow Rate (gpm)	Monthly [2]	Instantaneous	NA
PCBs µg/L	Semi-Annual	Grab	1 µg/L
Fish Food Fed (lbs/day)	Daily [3]	Measured	NA
Total Suspended Solids[4] (mg/L)	Semi-Annual	Grab	1 mg/L

(1) See the definitions in Part I.A. of the permit.

(2) Both the average flow during the monitoring period and the highest average monthly flow shall be reported. Flow rate may be established via either influent or effluent flow.

(3) Both the average daily feeding rate during the monitoring period and the maximum daily feeding rate shall be reported

(4) TSS monitoring is only required at facilities with production greater than or equal to 20,000 pounds per year

All monitoring shall be reported semi-annually on Discharge Monitoring Report (DMR) forms (EPA No. 3320-1).

All monitoring shall be conducted during the month of maximum feeding within the monitoring period.

C. Best Management Practices Plan

The BMP plan shall be developed and implemented as described in Part I.A. The plan shall include, at a minimum, the following:

1. A written plan for the efficient feeding of fish in the facility that will maximize feed conversion and minimize the amount of metabolic wastes and uneaten food produced, and still allow the achievement of production goals. This plan could include, but is not limited to, the following: projected annual production, feeding methods that will be used, appropriate record-keeping of feed consumption, feed storage and handling methods, and any other means employed to minimize waste solids.

2. A description and schedule of cleaning and maintenance activities that will minimize the amount of waste discharged at any one time. This must include, at a minimum,

the weekly cleaning of raceways, unless otherwise approved for a specific fish species' rearing requirements in an approved BMP plan. Records of raceway cleaning must be maintained on site.

3. A description, including dosage rates, total quantity used, and calculated concentrations, of all drugs and chemicals that will be used routinely in hatchery operations.

Facilities that produce 20,000 pounds or more of fish per year shall be subject to the following requirements and prohibitions:

4. As part of the BMP plan each facility shall record and maintain on site records of the total pounds of food fed for the previous calendar year, the total weight gain of all fish in the facility the previous calendar year, and the corresponding feed conversion ratio (FCR). FCRs may be calculated for individual lots of fish, providing all fish produced are accounted for.

5. Sweeping accumulated solids from raceways or ponds to state waters without treatment is prohibited.

6. Practices such as the removal of dam boards or standpipes in raceways or ponds, which allow accumulated solids to discharge to state waters without treatment, are prohibited.

7. The BMP plan must include a description of the methods for cleaning accumulated wastes from settling basins of other treatment units. The plan must also address the disposal of the wastes in such a manner that they will not reach state waters.

Facilities that produce 100,000 pounds, or more, of fish per year are also subject to the specific federal ELG limits of 40 CFR 451 as follows:

(a) Solids control. The permittee must:

1. Employ efficient feed management and feeding strategies that limit feed input to the minimum amount reasonably necessary to achieve production goals and sustain targeted rates of aquatic animal growth in order to minimize potential discharges of uneaten feed and waste products to water of the U.S.
2. In order to minimize the discharge of accumulated solids from settling ponds and basins and production systems, identify and implement procedures for routine cleaning of rearing units and off-line settling basins, and procedures to minimize any discharge of accumulated solids during the inventorying, grading, and harvesting of aquatic animals in the production system.
3. Remove and dispose of aquatic animal mortalities properly on a regular basis to prevent discharge to state waters, except in cases where the permitting authority authorizes such discharge in order to benefit the aquatic environment.

(b) Materials storage. The permittee must:

1. Ensure proper storage of drugs, pesticides, and feed in a manner designed to prevent spills that may result in the discharge of drugs, pesticides, or feed to state waters.
2. Implement procedures for properly containing, cleaning and disposing of any spilled material.

(c) Structural maintenance. The permittee must:

1. Inspect the production system and the wastewater treatment system on a routine basis in order to identify and promptly repair any damage.
2. Conduct regular maintenance of the production system and the wastewater treatment system in order to ensure that they are properly functioning.

(d) Recordkeeping. The permittee must:

1. In order to calculate representative feed conversion ratios, maintain records for aquatic animal rearing units documenting the feed amounts and estimates of the numbers and weight of aquatic animals.
2. Keep records documenting the frequency of cleaning, inspections, maintenance and repairs.

(e) Training

1. In order to ensure the proper clean-up and disposal of spilled material adequately train all relevant facility personnel in spill prevention and how to respond in the event of a spill.
2. Train staff on the proper operation and cleaning of production and wastewater treatment systems including training in feeding procedures and proper use of equipment.

II. MONITORING, RECORDING AND REPORTING REQUIREMENTS

A. Reporting Requirements

All required monitoring shall be reported on a Discharge Monitoring Report (DMR), postmarked no later than the 28th day of the month following the monitoring period. Submit completed DMR forms and all reports to the following address:

Montana Department of Environmental Quality
Water Protection Bureau
P.O. Box 200901
Helena, Montana 59620-0901
Phone: (406) 444-3080

B. Monitoring Procedures

Monitoring must be conducted according to test procedures approved under Part 136 Title 40 of the Code of Federal Regulations, unless other test procedures have been specified in this permit.

C. Additional Monitoring by Permittee

If the permittee monitors any pollutant at the location(s) designated herein more frequently than required by this permit the results of such monitoring shall be included in the DMR form. Such increased frequency shall be indicated.

D. Record Retention

All records and information resulting from the monitoring activities required by this permit shall be retained for a minimum of three (3) years, or longer if requested by the Department.

E. Noncompliance Notification

If for any reason, the permittee does not comply with or will be unable to comply with any effluent limitation specified in this permit, the permittee shall notify as soon as possible by phone and provide the Department with the following information, in writing, within five (5) days of becoming aware of such condition:

1. A description of the discharge and cause of noncompliance; and

2. The period of noncompliance including exact dates and times, or if not corrected, the anticipated time the noncompliance is expected to continue, and steps being taken to reduce, eliminate and prevent recurrence of the non-complying discharge.

F. Inspection and Entry

The permittee shall allow the head of the Department or the Regional Administrator, or an authorized representative, upon the presentation of credentials and other documents as may be required by law, to:

1. Enter upon the permittee's premises where a regulated facility or activity is located or conducted, or where records must be kept under the conditions of this permit;

2. Have access to and copy, at reasonable times, any records that must be kept under the conditions of this permit;

3. Inspect at reasonable times any facilities, equipment (including monitoring and control equipment), practices, or operations regulated or required under this permit; and,

4. Sample, or monitor at reasonable times for the purpose of assuring permit compliance, any substances or parameters at any location.

III. COMPLIANCE RESPONSIBILITIES

A. Duty to Comply

The permittee must comply with all conditions of this permit. Any permit noncompliance constitutes a violation of the Act and is grounds for enforcement action; for permit termination, revocation and reissuance, or modification; or for denial of a permit renewal application. The permittee shall give the Department advance notice of any planned changes at the permitted facility or of an activity, which may result in permit noncompliance.

B. Penalties for Violations of Permit Conditions

The Montana Water Quality Act provides that any person who violates a permit condition of the Act is subject to civil or criminal penalties not to exceed $25,000 per day of such violation. Any person who willfully or negligently violates permit conditions of the Act is subject to a fine of not more than $50,000 per day of violation, or by imprisonment for not more than 2 years, or both. Except as provided in permit conditions on Part III.G of this permit, "Bypass of Treatment Facilities" and Part III.H of this permit, "Upset Conditions", nothing in this permit shall be construed to relieve the permittee of the civil or criminal penalties for noncompliance.

C. Need to Halt or Reduce Activity not a Defense

It shall not be a defense for a permittee in an enforcement action that it would have been necessary to halt or reduce the permitted activity in order to maintain compliance with the conditions of this permit.

D. Duty to Mitigate

The permittee shall take all reasonable steps to minimize or prevent any discharge in violation of this permit, which has a reasonable likelihood of adversely affecting human health or the environment.

E. Toxic Pollutants

The permittee shall comply with effluent standards or prohibitions established under Section 307(a) of the Act for toxic pollutants within the time provided in the regulations that establish those standards or prohibitions, even if the permit has not yet been modified to incorporate the requirement.

F. Changes in Discharge of Toxic Substances

Notification shall be provided to the Department as soon as the permittee knows of, or has reason to believe:

1. That any activity has occurred or will occur which would result in the discharge, on a routine or frequent basis, of any toxic pollutant which is not limited in the permit, if that discharge will exceed the highest of the following "notification levels":

   a. One hundred micrograms per liter (100 µg/*l*);

b. Two hundred micrograms per liter (200 μg/*l*) for acrolein and acrylonitrile; five hundred micrograms per liter (500 μg/*l*) for 2,4-dinitrophenol and for 2-methyl-4, 6-dinitrophenol; and one milligram per liter (1 mg/*l*) for antimony;

c. Five (5) times the maximum concentration value reported for that pollutant in the permit application in accordance with 40 CFR 122.21(g)(7); or,

d. The level established by the Department in accordance with 40 CFR 122.44(f).

2. That any activity has occurred or will occur which would result in any discharge, on a non-routine or infrequent basis, of a toxic pollutant which is not limited in the permit, if that discharge will exceed the highest of the following "notification levels":

   a. Five hundred micrograms per liter (500 μg/*l*);

   b. One milligram per liter (1 mg/*l*) for antimony:

   c. Ten (10) times the maximum concentration value reported for that pollutant in the permit application in accordance with 40 CFR 122.21(g)(7); or,

   d. The level established by the Department in accordance with 40 CFR 122.44(f).

IV. GENERAL REQUIREMENTS

A. Planned Changes

The permittee shall give notice to the Department as soon as possible of any planned physical alterations or additions to the permitted facility. Notice is required only when the alteration or addition could significantly change the nature or increase the quantity of pollutant discharged. This notification applies to pollutants, which are not subject to effluent limitations in the permit.

B. Anticipated Noncompliance

The permittee shall give advance notice to the Department of any planned changes in the permitted facility or activity, which may result in noncompliance with permit requirements.

C. Permit Actions

This permit may be modified, revoked and reissued, or terminated for cause. The filing of a request by the permittee for a permit modification, revocation and reissuance, or termination, or a notification of planned changes or anticipated noncompliance, does not stay any permit condition.

D. Duty to Reapply

If the permittee wishes to continue an activity regulated by this permit after the expiration date of the authorization to discharge, the permittee must apply for and obtain a new authorization. The request must be submitted at least 180 days before the anticipated operation date. A new application must be submitted with the correct application fee after the fifth year of operation and shall be submitted within 180 days before the anticipated operation date.

E. Duty to Provide Information

The permittee shall furnish to the Department, within a reasonable time, any information which the Department may request to determine whether cause exists for modifying, revoking and reissuing, or terminating this permit, or to determine compliance with this permit. The permittee shall also furnish to the Department, upon request, copies of records required to be kept by this permit.

F. Other Information

When the permittee becomes aware that it failed to submit any relevant facts in a permit application, or submitted incorrect information in a permit application or any report to the Department, it shall promptly submit such facts or information.

G. Signatory Requirements

All applications, reports or information submitted to the Department shall be signed and certified.

1. All permit applications shall be signed as follows:

a. For a corporation: by a responsible corporate officer;

b. For a partnership or sole proprietorship: by a general partner or the proprietor, respectively;

c. For a municipality, State, Federal, or other public agency: by either a principal executive officer or ranking elected official.

2. All reports required by the permit and other information requested by the Department shall be signed by a person described above or by a duly authorized representative of that person. A person is considered a duly authorized representative only if:

   a. The authorization is made in writing by a person described above and submitted to the Department, and,

   b. The authorization specified either an individual or a position having responsibility for the overall operation of the regulated facility or activity, such as the position of plant manager, operator of a well or a well field, superintendent, position of equivalent responsibility, or an individual or position having overall responsibility for environmental matters for the company. (A duly authorized representative may thus be either a named individual or any individual occupying a named position.)

3. Changes to authorization. If an authorization under Part IV.G.2 of this permit is no longer accurate because a different individual or position has responsibility for the overall operation of the facility, a new authorization satisfying the requirements of Part IV.G.2 of this permit must be submitted to the Department prior to or together with any reports, information, or applications to be signed by an authorized representative.

4. Certification. Any person signing a document under this section shall make the following certification:

> "I certify under penalty of law that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified personnel properly gather and evaluate the information submitted. Based on my inquiry of the person or persons who manage the system, or those persons directly responsible for gathering the information, the information submitted is, to the best of my knowledge and belief, true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment for knowing violations."

H. Penalties for Falsification of Reports

The Montana Water Quality Act provides that any person who knowingly makes any false statement, representation, or certification in any record or other document submitted or required to be maintained under this permit, including monitoring reports or reports of compliance or noncompliance shall, upon conviction be punished by a fine of not more than $25,000 per violation, or by imprisonment for not more than six months per violation, or by both.

I. Availability of Reports

Except for data determined to be confidential under 40 CFR Part 2, all reports prepared in accordance with the terms of this permit shall be available for public inspection at the offices of the Department. As required by the Clean Water Act, permit applications, permits and effluent data shall not be considered confidential.

J. Oil and Hazardous Substance Liability

Nothing in this permit shall be construed to preclude the institution of any legal action or relieve the permittee from any responsibilities, liabilities, or penalties to which the permittee is or may be subject under Section 311 of the Clean Water Act.

K. Property or Water Rights

The issuance of this permit does not convey any property or water rights of any sort, or any exclusive privileges, nor does it authorize any injury to private property or any invasion of personal rights, nor any infringement of federal, state or local laws or regulations.

L. Severability

The provisions of this permit are severable, and if any provision of this permit, or the application of any provision of this permit to any circumstance, is held invalid, the application of such provision to other circumstances, and the remainder of this permit, shall not be affected thereby.

M. Transfers

This permit cannot be transferred to a new permittee. A new owner or operator of a facility must apply according to the application procedures in Part IV.D of this permit 30 days prior to taking responsibility for the facility.

N. Fees

The permittee is required to submit payment of an annual fee as set forth in ARM 17.30.201. If the permittee fails to pay the annual fee within 90 days after the due date for payment, the Department may:

1. Impose an additional assessment consisting of 15% of the fee plus interest on the required fee computed at the rate established under 15-31-510(3), MCA; or

2. Suspend the processing of the application for a permit or authorization or, if the nonpayment involves an annual permit fee, suspend the permit, certificate, license

or other authorization for which the fee is required. The Department may lift the suspension at any time up to one year after the suspension occurs if the holder has paid all outstanding fees, including all penalties, assessments and interest imposed under this subsection.

O. Reopener Provisions

This permit may be reopened and modified (following proper administrative procedures) to include the appropriate effluent limitations (and compliance schedule, if necessary), or other appropriate requirements if one or more of the following events occurs:

1. Water Quality Standards: The water quality standards of the receiving water(s) to which the permittee discharges are modified in such a manner as to require different effluent limits than contained in this permit.

2. TMDL or Wasteload Allocation: TMDL requirements or a wasteload allocation is developed and approved by the Department and/or EPA for incorporation in this permit.

3. Water Quality Management Plan: A revision to the current water quality management plan is approved and adopted which calls for different effluent limitations than contained in this permit.

4. Toxic Pollutants: A toxic standard or prohibition is established under Section 307(a) of the Act for a toxic pollutant which is present in the discharge and such standard or prohibition is more stringent than any limitation for such pollutant in this permit.

V. DEFINITIONS

1. "**Department**" means the Montana Department of Environmental Quality (MDEQ).

2. A "**grab**" sample, for monitoring requirements, is defined as a single "dip and take" sample collected at a representative point in the discharge stream.

3. An "**instantaneous**" measurement, for monitoring requirements, is defined as a single reading, observation, or measurement.

4. A "**mixing zone**" means a limited area of a surface water body or aquifer where initial dilution of a discharge takes place and where water quality changes may occur. Also recognized as an area where certain water quality standards may be exceeded.

5. "**Non-degradation**" means the prevention of a significant change in water quality that lowers the quality of high-quality water for one or more parameters. Also, the prohibition of any increase in discharge that exceeds the limits established under or determined from a permit or approval issued by the Department prior to April 29, 1993.

6. The term "**TMDL**" means the total maximum daily load limitation of a parameter, representing the estimated assimilative capacity for a water body before other designated uses are adversely affected. Mathematically, it is the sum of wasteload allocations for point sources, load allocations for non-point and natural background sources, and a margin of safety.

7. The **"receiving stream"** means the river, stream, or creek, which receives the wastewater discharge from the facility.

FRESHWATER

DRAFT Comparison: Washington National Toxic Rule (NTR) criteria versus Oregon Human Health Criteria

Grouped by: ➡	More protective criteria could result in new permit limit where detected	More protective criteria non-detect in effluent sample	New criteria	Equal or less protective than current NTR criteria

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NTR Chem #	Chemical Name Non-Carcinogens Carcinogens	CAS #	NTR Criterion Water and Organisms (µg/L) based on 6.5 Grams/Day	Oregon Criterion Water and Organisms (µg/L) based on **175 grams/day** Includes updated RSCs, RfDs, Cancer Slope Factors, and other modifications specific to Hg & As	Is Oregon's revised criterion is more protective?	Percent decrease between WA & OR Criteria	Priority Pollutant Scan Data Detection From preliminary data as presented in Policy Forum #3. (Ecology staff are in the process of reviewing all available effluent datasets to determine if any other chemicals have been detected in discharge.)	EPA method number	Detection Level (DL) (µg/L)	Quantitation Level (QL) (µg/L)
77	1,4-Dichlorobenzene	106467	**397**	**16**	Yes	**96%**	Detected	624	4.4	17.6
1	Antimony	7440360	**14**	**5.1**	Yes	**63%**	Detected	200.8	0.3	1
68	Bis(2-Ethylhexyl) Phthalate	117817	**1.8**	**0.20**	Yes	**89%**	Detected and Quantified	625	0.1	0.5
14	Cyanide	57125	**698**	**130**	Yes	**81%**	Detected	335.4	5	10
79	Diethyl Phthalate	84662	**22,631**	**3,800**	Yes	**83%**	Detected	625	1.9	7.6
36	Methylene Chloride	75092	**4.7**	**4.3**	Yes	**8%**	Detected	624	5	10
9	Nickel	7440020	**607**	**140**	Yes	**77%**	Detected and Quantified	200.8	0.1	0.5
38	Tetrachloroethylene	127184	**0.80**	**0.24**	Yes	**70%**	Detected and Quantified	624	1	2
39	Toluene	108883	**6,765**	**720**	Yes	**89%**	Detected and Quantified	624	1	2
119	Total Polychlorinated Biphenyls (PCBs)	multiple CAS #	**0.00017**	**0.0000064**	Yes	**96%**	Detected w/ non- 40 CFR 136 methods	1668C	~ 30pg/L	~50 pg/L
			0.00017	**0.0000064**	Yes	**96%**	--	608	0.25	0.5
37	1,1,2,2-Tetrachloroethane	79345	**0.17**	**0.12**	Yes	**30%**	--	624	1.9	2
42	1,1,2-Trichloroethane	79005	**0.61**	**0.44**	Yes	**27%**	--	624	1	2
29	1,2-Dichloroethane	107062	**0.38**	**0.35**	Yes	**9%**	--	624	1	2
85	1,2-Diphenylhydrazine	122667	**0.040**	**0.014**	Yes	**65%**	--	1625B	5	20
76	1,3-Dichlorobenzene	541731	**397**	**80**	Yes	**80%**	--	624	1.9	7.6
32	1,3-Dichloropropene	542756	**10**	**0.30**	Yes	**97%**	--	624	1	2
16	2, 3, 7, 8-TCDD Dioxin	1746016	**0.000000013**	**0.00000000051**	Yes	**96%**	--	1613B	1.3E-06	0.000005

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55	2,4,6-Trichlorophenol	88062	**2.1**	**0.23**	Yes	**89%**	--	625	2	4
46	2,4-Dichlorophenol	120832	**93**	**23**	Yes	**75%**	--	625	0.5	1
49	2,4-Dinitrophenol	51285	**70**	**62.0**	Yes	**11%**	--	625	1	2
82	2,4-Dinitrotoluene	121142	**0.11**	**0.084**	Yes	**24%**	--	609/625	0.2	0.4
48	2-Methyl-4,6-Dinitrophenol	534521	**13**	**9.2**	Yes	**31%**	--	625/1625B	1	2
78	3,3'-Dichlorobenzidine	91941	**0.039**	**0.0027**	Yes	**93%**	--	605/625	0.5	1
110	4,4'-DDD	72548	**0.00083**	**0.000031**	Yes	**96%**	--	608	0.025	0.05
109	4,4'-DDE	72559	**0.00059**	**0.000022**	Yes	**96%**	--	608	0.025	0.05
108	4,4'-DDT	50293	**0.00059**	**0.000022**	Yes	**96%**	--	608	0.025	0.05
17	Acrolein	107028	**321**	**0.88**	Yes	**100%**	--	624	5	10
18	Acrylonitrile	107131	**0.059**	**0.018**	Yes	**70%**	--	624	1	2
102	Aldrin	309002	**0.00013**	**0.0000050**	Yes	**96%**	--	608	0.025	0.05
103	alpha-BHC	319846	**0.0039**	**0.00045**	Yes	**88%**	--	608	0.025	0.05
58	Anthracene	120127	**9,567**	**2,900**	Yes	**70%**	--	625	0.3	0.6
19	Benzene	71432	**1.2**	**0.44**	Yes	**63%**	--	624	1	2
59	Benzidine	92875	**0.00012**	**0.000018**	Yes	**85%**	--	625	12	24
60	Benzo(a)Anthracene	56553	**0.0028**	**0.0013**	Yes	**54%**	--	625	0.3	0.6
61	Benzo(a)Pyrene	50328	**0.0028**	**0.0013**	Yes	**54%**	--	610/625	0.5	1

FRESHWATER

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62	Benzo(b)Fluoranthene	205992	**0.0028**	**0.0013**	Yes	**54%**	--	610/625	0.8	1.6
64	Benzo(k)Fluoranthene	207089	**0.0028**	**0.0013**	Yes	**54%**	--	610/625	0.8	1.6
104	beta-BHC	319857	**0.014**	**0.0016**	Yes	**88%**	--	608	0.025	0.05
66	Bis(2-Chloroethyl)Ether	111444	**0.031**	**0.020**	Yes	**36%**	--	611/625	0.3	1
67	Bis(2-Chloroisopropyl) Ether	108601	**1,389**	**1,200**	Yes	**14%**	--	625	0.3	0.6
20	Bromoform	75252	**4.3**	**3.3**	Yes	**23%**	--	624	1	2
21	Carbon Tetrachloride	56235	**0.25**	**0.10**	Yes	**61%**	--	624/601 or SM6230B	1	2
107	Chlordane	57749	**0.00057**	**0.000081**	Yes	**86%**	--	608	0.025	0.05
22	Chlorobenzene	108907	**677**	**74**	Yes	**89%**	--	624	1	2
23	Chlorodibromomethane	124481	**0.41**	**0.31**	Yes	**25%**	--	624	2	2
73	Chrysene	218019	**0.0028**	**0.0013**	Yes	**54%**	--	610/625	0.3	0.6
80	Dimethyl Phthalate	131113	**313,000**	**84,000**	Yes	**73%**	--	625	1.6	6.4
74	Dibenzo (a,h) Anthracene	53703	**0.0028**	**0.0013**	Yes	**54%**	--	625	0.8	1.6
111	Dieldrin	60571	**0.00014**	**0.0000053**	Yes	**96%**	--	608	0.025	0.05
81	Di-n-Butyl Phthalate	84742	**2,715**	**400**	Yes	**85%**	--	625	0.5	1
115	Endrin	72208	**0.76**	**0.024**	Yes	**97%**	--	608	0.025	0.05
116	Endrin Aldehyde	7421934	**0.76**	**0.030**	Yes	**96%**	--	608	0.025	0.05
33	Ethylbenzene	100414	**3,120**	**160**	Yes	**95%**	--	624	1	2

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86	Fluoranthene	206440	**296**	**14**	Yes	**95%**	--	625	0.3	0.6
87	Fluorene	86737	**1,276**	**390**	Yes	**69%**	--	625	0.3	0.6
117	Heptachlor	76448	**0.00021**	**0.0000079**	Yes	**96%**	--	608	0.025	0.05
118	Heptachlor Epoxide	1024573	**0.00010**	**0.0000039**	Yes	**96%**	--	608	0.025	0.05
88	Hexachlorobenzene	118741	**0.00075**	**0.000029**	Yes	**96%**	--	612/625	0.3	0.6
89	Hexachlorobutadiene	87683	**0.44**	**0.36**	Yes	**19%**	--	625	0.5	1
90	Hexachloro-cyclopentadiene	77474	**240**	**30**	Yes	**88%**	--	1625B/625	0.5	1
91	Hexachloroethane	67721	**1.9**	**0.29**	Yes	**85%**	--	625	0.5	1
92	Indeno (1,2,3-cd) Pyrene	193395	**0.0028**	**0.00130**	Yes	**54%**	--	610/625	0.5	1
34	Methyl Bromide	74839	**48**	**37**	Yes	**24%**	--	624/601	5	10
95	Nitrobenzene	98953	**17**	**14**	Yes	**19%**	--	625	0.5	1
96	N-Nitrosodimethylamine	62759	**0.00069**	**0.00068**	Yes	**1%**	--	607/625	2	4
98	N-Nitrosodiphenylamine	86306	**5.0**	**0.55**	Yes	**89%**	--	625	0.5	1
53	Pentachlorophenol	87865	**0.28**	**0.15**	Yes	**47%**	--	625	0.5	1
54	Phenol	108952	**20,905**	**9,400**	Yes	**55%**	--	625	2	4
100	Pyrene	129000	**957**	**290**	Yes	**70%**	--	625	0.3	0.6
12	Thallium	7440280	**1.7**	**0.043**	Yes	**98%**	--	200.8	0.09	0.36
120	Toxaphene	8001352	**0.00073**	**0.000028**	Yes	**96%**	--	608	0.24	0.5

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Blue values = less protective

NTR Chem #	Chemical Name Non-Carcinogens Carcinogens	CAS #	NTR Criterion Water and Organisms (µg/L) based on 6.5 Grams/Day	Oregon Criterion Water and Organisms (µg/L) based on 175 grams/day Includes updated RSCs, RfDs, Cancer Slope Factors, and other modifications specific to Hg & As	Is Oregon's revised criterion is more protective?	Percent decrease between WA & OR Criteria	Priority Pollutant Scan Data Detection From preliminary data as presented in Policy Forum #3. (Ecology staff are in the process of reviewing all available effluent datasets to determine if any other chemicals have been detected in discharge.)	EPA method number	Detection Level (DL) (µg/L)	Quantitation Level (QL) (µg/L)
43	Trichloroethylene	79016	2.7	1.4	Yes	48%	--	624	1	2
44	Vinyl Chloride	75014	2.0	0.023	Yes	99%	--	624/SM6200B	1	2
	1,2,4,5-Tetrachlorobenzene	95943	NC	0.11	Yes	NA	--			
101	1,2,4-Trichlorobenzene	120821	NC	6.4	Yes	NA	--	625	0.3	0.6
31	1,2-Dichloropropane	78875	NC	0.38	Yes	NA	--	624	1	2
40	1,2-Trans-Dichloroethylene	156605	NC	120	Yes	NA	--	624	1	2
	2,4,5-TP	93721	NC	10	Yes	NA	--			
	2,4,5-Trichlorophenol	95954	NC	330	Yes	NA	--			
	2,4-D	94757	NC	100	Yes	NA	--	6640B		
47	2,4-Dimethylphenol	105679	NC	76	Yes	NA	--	625	0.5	1
71	2-Chloronaphthalene	91587	NC	150	Yes	NA	--	625	0.3	0.6
45	2-Chlorophenol	95578	NC	14	Yes	NA	--	625	1	2
56	Acenaphthene	83329	NC	95	Yes	NA	--	625	0.2	0.4
	Barium	7440393	NC	1,000	Yes	NA	--	200.8	0.5	2
70	Butylbenzyl Phthalate	85687	NC	190	Yes	NA	--	625	0.3	0.6
	Chloromethyl ether, bis	542881	NC	0.000024	Yes	NA	--			
6	Copper	7440508	NC	1,300	Yes	NA	--	200.8	0.4	2

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DRAFT Comparison: Washington National Toxic Rule (NTR) criteria versus Oregon Human Health Criteria

Grouped by: ➡

More protective criteria could result in new permit limit where detected	More protective criteria non-detect in effluent sample	New criteria	Equal or less protective than current NTR criteria

NA = Not Applicable
NC = Not Calculated
Red values = more protective
Blue values = less protective

NTR Chem #	Chemical Name Non-Carcinogens Carcinogens	CAS #	NTR Criterion Water and Organisms (µg/L) based on **6.5** Grams/Day	Oregon Criterion Water and Organisms (µg/L) based on **175 grams/day** Includes updated RSCs, RfDs, Cancer Slope Factors, and other modifications specific to Hg & As	Is Oregon's revised criterion is more protective?	Percent decrease between WA & OR Criteria	Priority Pollutant Scan Data Detection From preliminary data as presented in Policy Forum #3. (Ecology staff are in the process of reviewing all available effluent datasets to determine if any other chemicals have been detected in discharge.)	EPA method number	Detection Level (DL) (µg/L)	Quantitation Level (QL) (µg/L)
	Dinitrophenols	25550587	NC	62	Yes	NA	--			
	Hexachlorocyclo-hexane,	608731	NC	0.0014	Yes	NA	--			
	Methoxychlor	72435	NC	100	Yes	NA	--	6630B&C and D3086-90		
8b	Methylmercury	22967926	NC	0.040 mg/kg	Yes	NA	--			
	Nitrates	14797558	NC	10000	Yes	NA	--			
	Nitrosamines	35576911	NC	0.00079	Yes	NA	--			
	N-Nitrosodibutylamine	924163	NC	0.005	Yes	NA	--			
	N-Nitrosodiethylamine	55185	NC	0.00079	Yes	NA	--			
97	N-Nitrosodi-n-Propylamine	621647	NC	0.0046	Yes	NA	--	607/625	0.5	1
	N-Nitrosopyrrolidine	930552	NC	0.016	Yes	NA	--			
	Pentachlorobenzene	608935	NC	0.15	Yes	NA	--			
10	Selenium	7782492	NC	120	Yes	NA	--	200.8	1	1
13	Zinc	7440666	NC	2,100	Yes	NA	--	200.8	0.5	2.5
41	1,1,1-Trichloroethane	71556	NC	NC	No	NA	--	624	1	2
28	1,1-Dichloroethane	75343	NC	NC	No	NA	--	624	1	2
30	1,1-Dichloroethylene	75354	**0.057**	230	No	NA	--	624	1	2
83	2,6-Dinitrotoluene	606202	NC	NC	No	NA	--	609/625	0.2	0.4

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25	2-Chloroethylvinyl Ether	110758	NC	NC	No	NA	--	624	1	2
50	2-Nitrophenol	88755	NC	NC	No	NA	--	625	0.5	1
52	3-Methyl-4-Chlorophenol	59507	NC	NC	No	NA	--	625	1	2
69	4-Bromophenyl Phenyl Ether	101553	NC	NC	No	NA	--	625	0.2	0.4
72	4-Chlorophenyl Phenyl Ether	7005723	NC	NC	No	NA	--	625	0.3	0.5
51	4-Nitrophenol	100027	NC	NC	No	NA	--	625	0.5	1
57	Acenaphthylene	208968	NC	NC	No	NA	--	625	0.3	0.6
15	Asbestos	1332214	**7,000,000**	**7,000,000 fibers/L**	No	NA	--			
112	alpha-Endosulfan	959988	**0.93**	**8.5**	No	NA	--	608	0.025	0.05
2	Arsenic (inorganic)	7440382	**0.017**	**2.1**	No	NA	--			
63	Benzo(ghi)Perylene	191242	NC	NC	No	NA	--	610/625	0.5	1
3	Beryllium	7440417	NC	NC	No	NA	--	200.8	0.1	0.5
113	beta-Endosulfan	33213659	**0.93**	**8.5**	No	NA	--	608	0.025	0.05
65	Bis(2-Chloroethoxy) Methane	111911	NC	NC	No	NA	--	625	5.3	21.2
4	Cadmium	7440439	NC	NC	No	NA	--	200.8	0.05	0.25
24	Chloroethane	75003	NC	NC	No	NA	--	624/601	1	2
26	Chloroform	67663	**5.7**	**260**	No	NA	Detected and Quantified	624 or SM6210B	1	2
5a	Chromium III	16065831	NC	NC	No	NA	--			

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5b	Chromium VI	18540299	NC	NC	No	NA	--	3500-Cr EC	0.3	1.2
106	delta-BHC	319868	NC	NC	No	NA	--	608	0.025	0.05
27	Dichlorobromomethane	75274	**0.27**	**0.42**	No	NA	--	624	1	2
84	Di-n-Octyl Phthalate	117840	NC	NC	No	NA	--	625	0.3	0.6
114	Endosulfan Sulfate	1031078	**0.93**	**8.5**	No	NA	--	608	0.025	0.05
105	gamma-BHC (Lindane)	58899	**0.019**	**0.17**	No	NA	--	608	0.025	0.05
93	Isophorone	78591	**8.4**	**27**	No	NA	--	625	0.5	1
7	Lead	7439921	NC	NC	No	NA	--	200.8	0.1	0.5
	Manganese	7439965	NC	NC	No	NA	--	200.8	0.1	0.5
8a	Mercury (freshwater)	7439976	**0.14**	NC	No	NA	Detected and Quantified			
35	Methyl Chloride	74873	NC	NC	No	NA	--			
94	Naphthalene	91203	NC	NC	No	NA	--	625	0.3	0.6
99	Phenanthrene	85018	NC	NC	No	NA	--	625	0.3	0.6
11	Silver	7440224	NC	NC	No	NA	--	200.8	0.04	0.2

Control of Toxic Chemicals in Puget Sound

Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011

Publication No. 11-03-055

DEPARTMENT OF
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State of Washington

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Control of Toxic Chemicals in Puget Sound

Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011

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November 2011

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Abstract

The Washington State Department of Ecology and other agencies initiated a multi-phase project in 2006, the Puget Sound Toxics Loading Analysis (PSTLA), to evaluate sources of toxic chemicals entering Puget Sound. The analysis focused on an abbreviated list of chemicals that were known to, or threaten to, harm the Puget Sound Ecosystem. The study included an assessment of major delivery pathways such as surface water runoff, groundwater, publicly-owned treatment works (POTWs), and direct air deposition. An assessment of the relative hazards posed by target chemicals was also performed.

The overall goal of PSTLA is to provide technical information to help develop toxic chemical control strategies for the Puget Sound basin. This report is a synthesis of information generated on (1) chemical releases from human-caused sources, (2) the rates of chemical loading through various pathways, and (3) a basin-wide hazard evaluation for chemicals of concern.

For most of the chemicals addressed, the rate of loading to Puget Sound was estimated to be one to three orders of magnitude lower than the rate of release from human-caused sources. In most cases, surface runoff was found to contribute the largest loads to Puget Sound, typically accounting for more than one-half of the total loads from all environmental pathways combined. Loads delivered through POTWs were generally the smallest among the pathways assessed, typically accounting for less than 10% of the total loading for each of the chemicals addressed.

Results of the hazard evaluation suggest that the following chemicals are most likely to be found at concentrations where effects are documented or at levels above criteria used to protect aquatic organisms and consumers of aquatic organisms:

- copper
- mercury
- polychlorinated biphenyls (PCBs)
- polychlorinated dioxins and furans (PCDD/Fs)
- the pesticide DDT (and its metabolites DDD and DDE)
- polycyclic aromatic hydrocarbons (PAHs)
- bis(2-ethylhexyl) phthalate (DEHP)

Results of the hazard evaluation were coupled with information on chemical sources and loading to suggest priorities for source control among the chemicals assessed. Recommendations are provided for source control strategies, and data needs are identified.

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Executive Summary

Background and Approach

The Washington State Department of Ecology (Ecology) and other agencies initiated the Puget Sound Toxics Loading Analysis (PSTLA) in late 2006 to provide scientific information that could be used to guide decisions about how best to direct and prioritize resources and strategies for controlling toxic chemicals in the Puget Sound basin. The primary focus of PSTLA was to estimate toxic chemical loading to Puget Sound through major pathways such as surface water runoff, publicly-owned treatment works (POTWs), and direct air deposition.

PSTLA used a phased approach to develop technical information on toxics chemicals in the Puget Sound basin.

- Phases 1 and 2 relied on existing data to estimate chemical loadings and identify the most important delivery pathways.
- Phase 3 studies included collection of new monitoring data to fill data gaps identified during earlier phases.

Other important components of the project included:

- Assessing pharmaceuticals and personal care products in wastewater treatment plants.
- Assessing persistent organic pollutants in three guilds of marine species.
- Developing numerical models for polychlorinated biphenyls (PCBs) in Puget Sound.
- Developing a framework for a toxicant-based biological monitoring system.
- Preparing a report on the estimated release of chemicals from human-caused (anthropogenic) sources.

The present *Assessment Report* aims to synthesize information from all phases of the PSTLA, focusing primarily on the Phase 3 loading studies. Since information on loading and delivery pathways, primary chemical sources, and other PSTLA studies may not by itself be sufficient to meet the overall goal of PSTLA, a screening-level hazard evaluation of selected chemicals was conducted and included in this report. The hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin. The information provides a scientific basis to develop a source control strategy for toxic chemicals in the Puget Sound Basin, and to prioritize actions.

The report is organized in a chemical-by-chemical fashion. For each of the selected chemicals addressed in the report, the major ongoing anthropogenic sources are discussed and release rates are estimated, an assessment of loading to Puget Sound and major pathways is presented, and the results of the hazard evaluation for the specific chemical are discussed. These elements are expressed by asking the following questions about toxic chemicals in the Puget Sound basin:

- Where do they come from?
- How much is being delivered?
- What delivery pathways contribute to the loading?
- What is the relative toxic hazard posed by these chemicals at observed concentrations?

In order to focus source control actions, a lines-of-evidence approach was developed to identify (1) chemicals with large ongoing anthropogenic releases from primary sources (does not include natural sources or legacy pollution, *and* (2) a relatively high potential to elicit effects based on the results of the hazard evaluation and regionally important biological-effects data. Combining information on chemical releases and loadings with the relative potential for effects provides for a more robust prioritization of possible future source control efforts. Recommendations for filling data gaps are also included.

The PSTLA focused on an abbreviated list of chemicals of concern (COCs). This list was developed during Phase 1 of the project based on observed harm or the threat of harm to the Puget Sound ecosystem. There is a wide range of chemicals in the Puget Sound basin for which we lack environmental information, and yet the chemicals may have the potential to cause biological or ecological harm. In addition, there is a large degree of uncertainty about the sources, pathways, and hazards for the chemicals that have been addressed. Therefore, this assessment should be viewed as the starting point for developing a larger toxic chemical control strategy in which a much broader spectrum of chemicals is considered.

Summary of Major Findings

Sources, Loads, and Pathways

COCs (listed in Table ES-1) were selected as the core group of chemicals analyzed in PSTLA studies (1) based on a documented history of their presence in Puget Sound and their capacity to harm or threaten the Puget Sound ecosystem and (2) to ensure that a broad variety of delivery pathways would be represented. While there is general consensus that a much larger number of potentially harmful chemicals are released to Puget Sound, the identification and evaluation of all of these chemicals were beyond the scope of the PSTLA projects.

To remain consistent with other PSTLA projects, the geographical scope of this *Assessment Report* includes Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

Chemical loads were calculated for most of the major pathways identified during the initial phase of the PSTLA effort. These include surface water runoff, POTWs, atmospheric deposition directly to marine waters, and direct groundwater discharge. Table ES-1 summarizes the estimated quantities of COCs released in the Puget Sound basin and the loads delivered to Puget Sound. Although these estimates are based on the best available information, releases and loads for some COCs remain incomplete or reflect high levels of uncertainty. In particular, air deposition and groundwater loading data are unavailable for many of the organic COCs.

In general, the load of metals to Puget Sound is approximately an order of magnitude lower than the total release from ongoing anthropogenic sources. Arsenic is an exception, apparently due to a high level of enrichment from natural sources.

For organic chemicals, loads are generally one to three orders of magnitude lower than releases from ongoing anthropogenic sources. The comparatively large differences between release and loading rates for organics may simply reflect the fewer number of pathways assessed for some organics. Closer agreement between anthropogenic releases and loads for metals may also be due in part to natural enrichment which contributes to the loads. Historic releases may also be a factor in loading of both metals and organic compounds.

Table ES-1. Toxic Chemical Releases and Loading in the Puget Sound Basin (metric tons/year).

COC	Total Release in the Puget Sound Basin[a]	Major Sources	Total Load to Puget Sound[b,c]	Major Pathway(s)
Arsenic	0.8	Industrial air emissions. CCA-treated wood leaching. Roofing material leaching.	14 – 25	Surface Runoff
Cadmium	1.0	Roofing material leaching.	0.05 – 0.53	Groundwater Atm. Deposition
Copper	180 - 250	Pesticides use on urban lawns and gardens.[d] Residential plumbing component leaching. Brake pad abrasion. Roofing material leaching. Vessel anti-fouling paint leaching.	33 – 80[e]	Surface Runoff
Lead	520	Ammunition and hunting shot use. Loss of fishing sinkers and wheel weights. Roofing material leaching. Aviation fuel combustion.	3.6 – 12	Surface Runoff
Mercury	0.5	Consumer product improper disposal. Crematoria and industrial air emissions.	0.11 – 0.37	Surface Runoff
Zinc	1,500	Roofing material leaching. Vehicle tire abrasion.	140 - 200	Surface Runoff
Total PCBs	2.2	Electrical equipment spills and leakage.[d] Residential trash burning. Building sealant (caulk) volatilization and abrasion.	0.003 – 0.02	Surface Runoff
Total PBDEs	0.7	Furniture, computer monitors, and other components of residential and commercial indoor environments.	0.028 – 0.054	Atm. Deposition POTWs
PCDD/Fs	0.000009[f]	Backyard burn barrels.	NA	NA
Total DDT	NA	NA	0.0025 – 0.032	Surface Runoff
Total PAHs	310	Woodstoves and fireplace combustion emissions. Vehicle combustion emissions. Creosote-treated piling, railroad ties, and utility poles.	0.19 – 1.0[e]	Groundwater Surface Runoff
DEHP	17	Polymer (primarily PVC) off-gassing. Industrial, commercial, and institutional air emissions. Roofing material leaching.	2.0 – 3.2	Surface Runoff
Triclopyr	150	Herbicide use on crops and golf courses.	0.64 – 0.69	Surface Runoff
Nonylphenol[g]	0.18	Industrial, commercial, and institutional air emissions.	0.023 – 0.024	Surface Runoff
Petroleum	9,300	Motor oil drips and leaks. Used oil improper disposal. Gasoline spillage during fueling.	330 – 500	Surface Runoff
Oil & Grease[h]	NA	NA	8,500 – 11,000	Surface Runoff

NA=Not analyzed

[a] Includes the Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

[b] Includes the Puget Sound and the U.S. portions of the Straits of Georgia and Juan de Fuca.

[c] Range of all pathways combined expressed as the sum of the 25th percentile values for each pathway – sum of the 75th percentile values for each pathway.

[d] Estimate is highly uncertain.

[e] Does not include estimated direct releases to marine waters (54 metric tons/yr for PAHs and 26 metric ton/yr for copper).

[f] Expressed as Toxic Equivalents (TEQs).

[g] Sources were not fully assessed.

[h] Category includes all hexane extractable material

The relationships between rates of initial release, rates of loading to Puget Sound, and the major delivery pathways are discussed for each COC. For most COCs, it is clear that a simple assessment of overall release from primary sources will not translate to levels measured in the environment. The type, mechanism, and setting of a chemical release are important factors governing their presence in environmental pathways. For instance, large quantities of polycyclic aromatic hydrocarbons (PAHs) are potentially released from creosote-treated railroad ties and utility poles throughout the basin, but the mechanisms and settings of release (volatilization and near-field leaching primarily to soil) do not translate to elevated concentrations of PAHs in surface runoff. In contrast, estimated zinc releases from major sources such as roof materials and vehicle tires occur in manners and settings (leaching from precipitation, abrasion to roadway surfaces) that result in substantial entrainment of zinc to surface waters.

For the majority of COCs, surface runoff contributed the largest loads to Puget Sound, typically accounting for more than one-half of the total loads from all pathways combined. Surface runoff from commercial/industrial land covers typically had the highest concentrations. However, agricultural areas produced the highest concentrations for several metals. Loading calculations were strongly influenced by the areas occupied by different land cover types. As a result, the largest loads were typically from forested areas (occupying 83% of all land cover), even though COC concentrations in forest areas were often the lowest among land covers or below reporting limits.

Loads delivered directly to the Puget Sound marine environment through groundwater were estimated using literature values for COC concentrations and discharge estimates. Groundwater loads were estimated to be an order of magnitude lower than surface runoff for most COCs. Exceptions were PAH loads, which were similar to surface water, as well as cadmium, gasoline, and diesel fuel, which had low to non-detectable surface water loads. Like surface runoff, load calculations for groundwater were strongly influenced by methods used to estimate concentrations from non-detected values.

Atmospheric deposition directly to marine waters was an important loading pathway for polybrominated diphenyl ethers (PBDEs) and high molecular weight PAHs. PBDEs was the only COC for which direct deposition from air appeared to be the largest delivery pathway to Puget Sound. COC flux from the atmosphere was comparatively high at a monitoring station located in a high-density urban area with nearby commercial, industrial, and major roadway contaminant sources.

POTWs generally accounted for less than one-tenth of the delivery to Puget Sound for each of the COCs assessed. Exceptions were diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate) and PBDEs, the latter of which had POTW loads larger than those in surface runoff. POTW loads for other chemicals not specifically addressed in this report, such as pharmaceuticals, are expected to be much higher in POTWs than in other delivery pathways.

Hazard Evaluation

To assess the relative hazards, observed environmental concentrations of COCs in various environmental media (surface water, sediment) were compared to available data on biological effects or to established criteria to protect aquatic life and consumers of aquatic organisms.

Evaluations were conducted for the following categories:

- Direct hazards to aquatic life through surface water exposure
- Direct hazards to benthic organisms through sediment exposure
- Direct hazards to aquatic life based on tissue residue levels
- Hazards to wildlife based on ingestion of prey, water, and sediment
- Hazards to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: Priority 1 level of concern, Priority 2 level of concern, or unknown (U) level of concern. Results were classified as Priority 1 when the upper end of a set of observed concentrations (e.g. 90th percentile values) exceeded the lower end of a set of effects concentrations (e.g. 10th percentile values), or exceeded selected threshold values such as water quality criteria. A Priority 2 level of concern was assigned in cases where the upper end of a set of observed concentrations was below the lower end of a set of effects concentrations or other threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U for a particular sub-category, this should not be interpreted to mean there are no hazards associated with that COC. All of the COCs evaluated pose some level of concern for Puget Sound. Locally, concentration hot spots may exist near major sources and may cause localized toxicity to aquatic organisms or lead to violations of standards that would not necessarily be emphasized in this broad regional assessment. To address this shortcoming in the hazard assessment, a limited review was conducted of regionally important biological effects information.

Results of the hazard evaluation showed that all of the COCs except lead are a Priority 1, where sufficient data were available for evaluation, for at least one of the categories evaluated. There were not sufficient data to conduct evaluations for PBDEs, triclopyr, nonylphenol, or petroleum. PCBs is the only COC assigned a Priority 1 for all five categories evaluated.

COC concentrations in surface waters and sediments – particularly freshwater – resulted in the most COCs assigned Priority 1. Tissue residue effects, wildlife, and human health evaluations generally resulted in fewer COCs receiving a Priority 1 assignment. However, only bioaccumulative chemicals were evaluated for these latter categories.

In addition to the hazard evaluation, reviews of regionally important biological-effects data showed that levels of the following chemicals found in the Puget Sound basin result in documented or potentially adverse effects to a variety of aquatic organisms:

- copper
- mercury
- PCBs
- PBDEs
- polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- PAHs

Priorities for Source Control Actions

A lines-of-evidence approach was used to move further toward a goal of deciding how best to prioritize actions and resources for controlling toxic chemicals in the Puget Sound basin. This lines-of-evidence approach considers information on the four major components of the assessment (sources, loading, pathways, and the relative hazards), but the approach mostly relies on COC sources and the relative hazards posed by COCs as determined by the hazard evaluation and review of other regional studies.

This approach adopts the rationale that chemicals with the greatest potential to elicit toxic effects at existing concentrations should be an important factor in determining the priority for source control efforts. Priority was also given to COCs for which we have large opportunities for source control. This reflects the extent to which there are existing regulatory actions to control releases, such as bans, management of materials, or other permanent actions which reduce releases to the environment. Given the uncertainty associated with individual estimates of releases or loadings, this lines-of-evidence approach provides a supportable rationale for establishing relative priorities for control actions.

Opportunities for source control are considered large where the major sources of a COC have not been addressed by control actions; where some of the major sources have been addressed, opportunities may be considered medium. In cases where actions have been implemented to control and reduce all or most of the major sources and this appears to have resulted in low rates of loading to Puget Sound, the opportunities for controlling a COC are considered small. This assessment relies principally on the *Sources Report* (Ecology, 2011) with limited input by Ecology staff and management to gauge the opportunities for source control; the assessment was not intended to be a detailed review of management initiatives. Table ES-2 summarizes major sources for each COC and possible opportunities for reducing those sources.

Based on the lines-of-evidence approach, copper, PAHs, DEHP, and petroleum sources were rated as have the highest priority for early actions. The reasoning for this determination is as follows:

- A substantial portion of the fresh and marine water copper data observed basin-wide falls within concentrations where effects have been documented (including reduced olfactory function in salmonids). Copper is released in large quantities from a variety of sources which appear to translate to substantial loads to the Puget Sound ecosystem. The use of copper in pesticide applications and the release of copper from roofing materials are sources which warrant further investigation. In addition, the effectiveness of recent legislation to limit copper in brake pads and vessel anti-fouling paint should be evaluated.
- A number of individual PAHs surpass (do not meet) freshwater sediment guidelines and human health criteria. In addition, a variety of studies have demonstrated links between PAH exposure and adverse effects to regionally relevant aquatic species. There appear to be numerous opportunities for control actions, primarily for combustion sources and for creosote-treated wood.

- Observed DEHP concentrations in both freshwater and marine environments exceed (do not meet) criteria for protection of benthic species and human health. Substantial amounts of DEHP are released in the Puget Sound basin, much of which occurs initially through releases to air from off-gassing of plasticized polymers and point-source air emissions. Several non-polymer uses of DEHP may also provide opportunities for source reduction.
- The relative hazard posed by petroleum in the Puget Sound basin was not able to be evaluated due primarily to the lack of biological-effects data and the absence of criteria to protect aquatic organisms, wildlife, or human health. However, some of the COCs addressed in this assessment are components of petroleum and may be released in substantial quantities along with the release of petroleum. In particular, substantial releases of PAHs are estimated to be released from petroleum. The major sources of petroleum are diffuse, such as motor oil drips and leaks and minor gasoline spillage during vehicle fueling, and therefore offer ample opportunities for reduction efforts.

Several COCs were found to be a Priority 1 level of concern based on the hazard evaluation but were not determined to be among the highest priorities for reduction actions since the major sources have been addressed through regulatory programs or other efforts. For instance, mercury poses a relatively high hazard to freshwater and marine aquatic organisms and wildlife based on doses calculated from observed data. However, many of the historical regional sources of mercury to the Puget Sound basin have been eliminated or are being addressed by the Mercury Chemical Action Plan (Ecology and WDOH, 2003). Similarly, PCBs are a Priority 1 level of concern for all hazard evaluation categories, but PCBs have been banned for decades, the major sources (use in electrical equipment) are highly regulated, and current loads to Puget Sound appear to be small.

Although the systematic prioritization approach identified four COCs for early actions, other factors should be considered to determine the need and feasibility for developing control and reduction strategies for other COCs. For instance, PBDEs are ubiquitous environmental contaminants, and although voluntary actions and bans have removed major PBDE formulations from new consumer products, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources. The hazard evaluation was not able to adequately assess the relative hazards associated with PBDEs due to a lack of environmental standards, although there is evidence in the available literature to suggest this COC may pose a hazard at observed concentrations.

Additional research is needed to assess the relative hazards posed by PBDEs and other COCs for which there are only limited environmental data. By the same token, COCs with limited source information should be further evaluated to assess additional opportunities for source control.
Of the COCs addressed in this report, PBDEs and nonylphenol were the COCs that should receive top attention for further research on potential hazard as well as possible opportunities for source control.

Table ES-2. Summary of Possible Actions to Reduce COCs in the Environment.

COC	Opportunities for Source Control	Major Ongoing Anthropogenic Sources	Possible Actions for Reductions
Arsenic	Medium	Industrial air emissions	Maintain existing permit controls.
		CCA-treated wood leaching	Continue ban for most non-structural uses.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Cadmium	Medium	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Copper	Large	Pesticides use on urban lawns and gardens	More data needed on actual pesticide use.
		Residential plumbing component leaching	Continue to implement Lead and Copper Rule.
		Brake pad abrasion	Continue to implement legislation enacted to reduce source.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vessel anti-fouling paint leaching	Continue to implement legislation enacted to reduce source.
Lead	Small	Ammunition and hunting shot use	Implement CAP and enforce existing regulations.
		Loss of fishing sinkers and wheel weights	Implement CAP and enforce existing regulations.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Aviation fuel combustion	Implement CAP and enforce existing regulations.
Mercury	Medium	Consumer product improper disposal	Continue to implement CAP and enforce existing regulations.
		Crematoria and industrial air emissions	Continue existing permit limits.
Zinc	Large	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vehicle tire abrasion	Investigate source where it poses local concern.
Total PCBs	Small	Electrical equipment spills and leakage	Continue programs for management and disposal.
		Residential trash burning	Continue enforcing existing ban.
		Building sealant (caulk) volatilization and abrasion	Investigate source where it poses local concern.
Total PBDEs	Medium	Furniture, computer monitors, and other components of residential and commercial indoor environments	Enforce ban on new products but consider control actions to reduce the release from existing products.
PCDD/Fs	Small	Backyard burn barrels	Continue enforcing existing ban.
Total DDT	Small	None apparent	Investigate source where it poses local concern.
Total PAHs	Large	Woodstoves and fireplace combustion emissions	Continue change out programs, investigate catalysts/capture devices, promote alternatives to wood heat.
		Vehicle combustion emissions	Anti-idling programs, continue/expand engine retrofits for private section engines, enforce existing vehicle controls.
		Creosote-treated piling, railroad ties, and utility poles	Control actions needed, gather information to identify highest priority areas.
DEHP	Large	Polymer (primarily PVC) off-gassing	Gather additional information on extent of releases.
		Industrial, commercial, and institutional air emissions	Maintain existing permit controls.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Triclopyr	Medium	Herbicide use on crops and golf courses	More data needed on pesticide use.
Nonylphenol	Unknown	Industrial, commercial, and institutional air emissions	More information needed on emissions from these sources and unidentified releases.
Petroleum	Large	Motor oil drips and leaks Used motor oil improper disposal	Expand existing education/workshop programs.
		Gasoline spillage (minor) during fueling	Possible opportunity for source control, but more data needed on extent of releases.

Bold=Recommended as priority for near-term actions based on lines-of-evidence approach.

Recommendations

Broad recommendations are provided below. These are intended to guide development of a long-term strategy to reduce toxic threats to Puget Sound. The reader is directed to the main body of the report for more detailed results of this assessment that should provide further direction towards specific chemical control actions and further source/pathway investigations.

While this report identifies sources of toxic chemicals entering Puget Sound and recommends ways to reduce this contamination, these recommendations should be prioritized and balanced alongside current efforts and regulatory programs that already keep millions of pounds of business-generated COCs safely managed.

In 2010 the U.S. Environmental Protection Agency selected Ecology to lead the development and implementation of a long-term toxic chemical control strategy for Puget Sound. Results from the PSTLA will be a key piece of information to help design and implement actions to reduce threats from the most important sources of toxic chemicals to the Puget Sound ecosystem. Ecology will use funding from a National Estuary Program grant to implement priority actions under this long-term toxics control strategy.

Major recommendations from this assessment can be summarized as follows:

- A variety of diffuse (nonpoint) sources appear to account for the majority of contaminant releases in the Puget Sound basin. In addition, surface water runoff during storms was identified as the major delivery pathway for most contaminants. High priority should be given to (1) implementing control strategies to prevent the initial release of contaminants and (2) reducing or treating stormwater inputs.

- Vehicles and vehicle-related activities represent an important source of a number of contaminants. Examples include: copper and zinc from brake and tire wear, PAHs from fuel combustion, and petroleum from motor oil drips and leaks as well as refueling operations. Source control strategies should be developed around reducing contaminant inputs from vehicles.

- Runoff and leaching from roofing materials were estimated to be a major source of several metals, particularly cadmium, copper, and zinc. Roof runoff may also be a substantial source of DEHP. Field investigations should be conducted to gauge the accuracy of this information, and if warranted, alternative assessments should be considered for this source category.

- Developed lands (commercial/industrial, agricultural, and residential) had higher concentrations of COCs compared to undeveloped forest land. Source control strategies should focus on identifying and controlling contaminant releases from existing and new developments.

- This assessment focused on a short list of contaminants that were known to, or threaten to, harm the Puget Sound ecosystem. Data are needed on the spatial distribution and impacts from a much wider range of potential contaminants (e.g. pharmaceuticals and personal care products, brominated flame retardants, nanomaterials) in the basin.

- Businesses in Washington that routinely handle large amounts of COCs should be inspected on a routine basis; once every three years appears to be a reasonable schedule.

Examples of other recommendations are provided below:

- One of the largest potential releases of copper is due to the urban lawn and garden use of products containing copper. Due to the lack of good pesticide-use information, there is a high degree of uncertainty surrounding this conclusion. Additional information is needed to determine release rates for this potentially important source of copper.

- Wood-burning stoves and fireplaces along with vehicle emissions were identified as some of the largest sources of PAHs in the Puget Sound watershed. Regional air programs should continue to pursue abatement programs to reduce wood smoke emissions and vehicle emissions.

- Creosote-treated wood represented approximately one-third of the PAHs released to the Puget Sound basin. In particular, direct release to Puget Sound occurs from treated pilings in marine (salt) water. Programs such as the Department of Natural Resources Marine Piling Removal program should be supported to reduce the release of PAHs from marine pilings and bulkheads.

- More information is needed to help distinguish natural and legacy sources of contaminants in environmental pathways such as surface water runoff. This will help gauge the feasibility and effectiveness of actions taken to reduce releases of chemicals from contemporary anthropogenic (human-caused) releases.

- Resources should be provided for local source control programs that identify and prevent the release of contaminants on a local scale.

Introduction

Background

The Puget Sound Basin covers more than 43,400 square kilometers (16,800 square miles) of land and water (Hart Crowser et al., 2007) and is home to 4.5 million people (U.S. Census Bureau, 2010) including large urban metropolitan centers such as Seattle and Tacoma (Figure 1). Although large urban and industrial areas have developed along the shores of Puget Sound and near the mouths of major rivers – particularly on the east side of the central Sound – much of the shoreline and the watershed remains undeveloped and the bulk of the upland basin is forest.

During the past 150 years, humans and their activities have released a wide variety of chemicals into Puget Sound and its surrounding watershed, many of which are toxic to humans and aquatic organisms. Due to its fjord-like structure and shallow sills, the entry of deep oceanic water into Puget Sound is restricted, which reduces flushing of the inland marine and estuarine waters (PSAT, 2007). As a result, toxic chemicals (toxicants) and other pollutants may accumulate in some inlets and embayments of Puget Sound, increasing their exposure to aquatic organisms.

While the marine waters of Puget Sound may be the ultimate sink for many of the toxic chemicals released in the basin, it is universally recognized that freshwater streams, rivers, and lakes in the basin may be at risk from contamination as toxic chemicals travel from their points of initial release to the Puget Sound. The health of Puget Sound ultimately depends on the health of its upland watershed, particularly since one of its most ecologically important and iconic organisms – pacific salmon – rely on the upland watersheds for some of their most vulnerable stages of life.

For several decades the loading of toxicants to Puget Sound has been recognized as a serious problem and has been documented in a number of reviews (e.g. Dexter et al., 1981; Romberg et al., 1984; PSWQA, 1986; PTI, 1991; PSAT, 2003; Redman et al., 2006). These reviews have primarily focused on identifying chemicals of concern (COCs), concentrations in marine sediments, and effects to aquatic organisms, but generally provided only conjecture about delivery pathways.

Puget Sound Toxics Loading Analysis

In December 2005, Governor Christine Gregoire and the Washington Legislature launched the Puget Sound Initiative, a comprehensive effort by local, state, federal, and tribal governments; business, agriculture and environmental communities; scientists; and the public to restore, protect, and preserve the Sound by 2020. Among the top recommendations put forth by the original Puget Sound Partnership (Partnership) in 2006 was to make the reduction of toxic chemicals entering Puget Sound waters a primary objective for the long-term agenda.

The Washington State Department of Ecology (Ecology) and other agencies responded to the original Partnership's "reducing toxics" recommendation by initiating the Puget Sound Toxics Loading Analysis (PSTLA). Phase 1 of PSTLA was an initial estimate of toxicant loading to Puget Sound through various pathways such as surface runoff and direct air deposition. The analysis relied on readily accessible data to estimate chemical loading to the marine basin, and the authors of the report acknowledged that there remained a number of significant gaps in determining an accurate toxics budget for the Sound (Hart Crowser et al., 2007). However, the Phase 1 study satisfied its primary goal of identifying data gaps and needs for additional studies, and informed the Puget Sound Action Agenda (PSP, 2008), the plan for restoring Puget Sound.

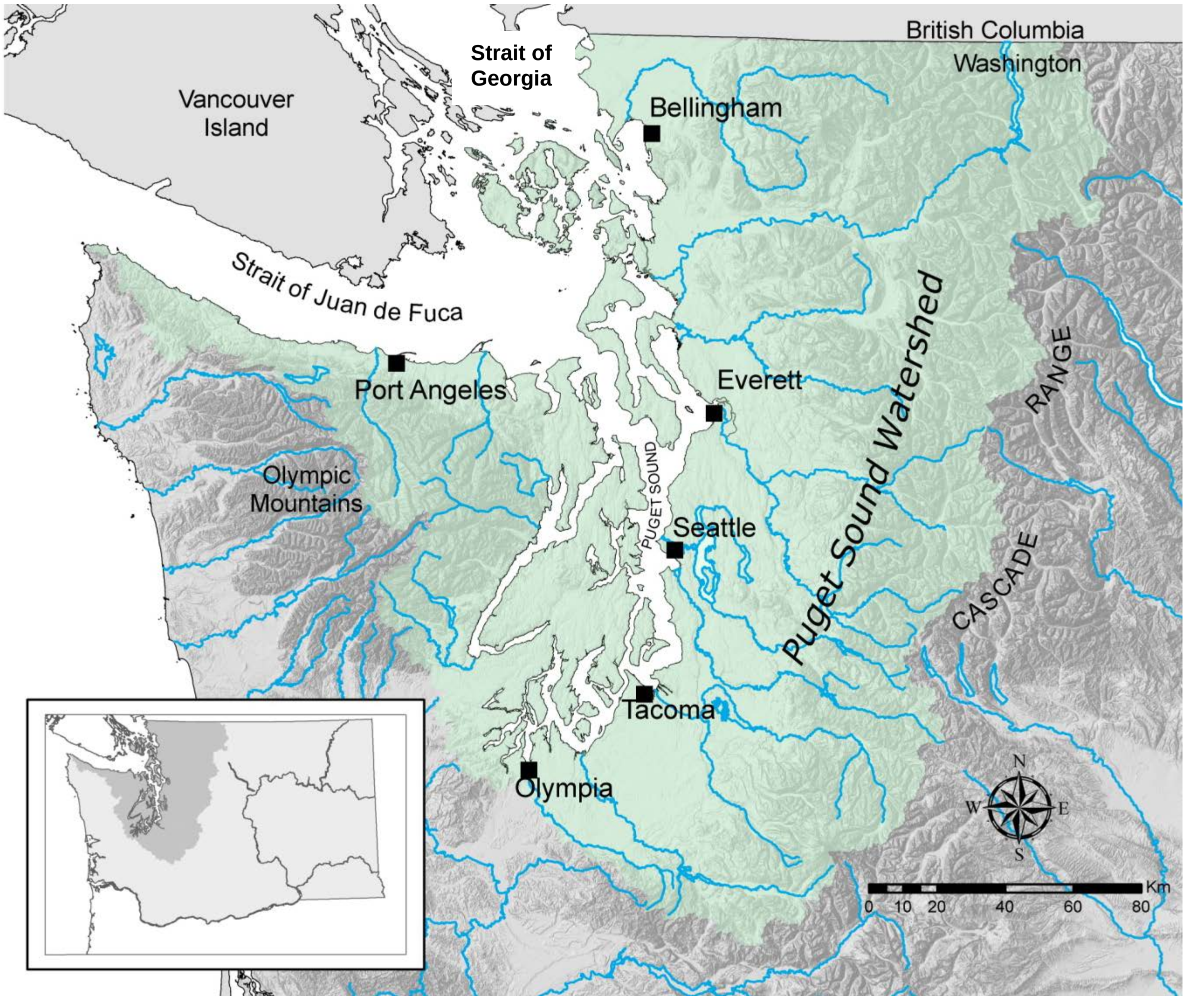


Figure 1. Watershed area for Puget Sound and the U.S. Portion of the Strait of Georgia and the Strait of Juan de Fuca.

The Partnership's Action Agenda re-iterated the conclusion that toxic chemical contamination is one of the largest problems facing Puget Sound, and a number of recommendations were developed to tackle various aspects of this complex problem. Although many of the recommendations were centered on implementation (e.g. programmatic, education, policy), the Partnership also recognized the need to gain a more thorough understanding of the problem as it currently exists. This need was expressed in the Action Agenda as Priority C.1.1.10: *Continue scientific work to better understand the sources of toxics, as well as transport and fate in the Puget Sound ecosystem, to better refine reduction strategies. This includes the toxic loadings assessments.*

In keeping with recommendations from the Action Agenda, two additional phases of PSTLA projects were mapped out:

- Phase 2 PSTLA studies were generally designed to refine loading estimates obtained during the Phase 1 effort and relied on readily available information much like the Phase 1 study.
- Phase 3 represented a departure from the earlier approaches in that nearly all of the Phase 3 projects included a sampling component so that refinements would include up-to-date and region-specific data.

In general, PSTLA projects were designed to assess the loading, sources, and to some degree, the impacts of toxic chemicals in Puget Sound. But the bulk of the PSTLA effort has been devoted to studying the delivery of toxicants through various pathways, such as direct atmospheric deposition, surface water runoff, and water exchange at the ocean boundary.

Purpose

The overall goal of PSTLA is to provide scientific information that will help guide decisions about how best to direct and prioritize resources and strategies for controlling toxic chemicals in the Puget Sound basin.

The purpose of this *Assessment Report* is to (1) distill the data and information generated by the PSTLA projects into a single document and (2) provide conclusions about toxic chemicals and their sources that can be used by those developing strategies to control toxic chemicals in Puget Sound.

The *Assessment Report* does not address all threats to Puget Sound. The issues regarding cleanup and restoration of Puget Sound are complicated and multi-faceted. For every pound of toxic chemicals released to Puget Sound by people, cars, households, etc., Washington businesses produce and handle hundreds to thousands times the amount of these chemicals, both in the form of chemical products and hazardous waste. While the *Assessment Report* identifies sources of pollution entering Puget Sound and recommends priorities to address this contamination, these recommendations should be balanced alongside current efforts that already keep millions of pounds of business-generated chemicals safely managed.

In 2010 the U.S. Environmental Protection Agency (EPA) selected Ecology to lead development and implementation of a long-term toxic chemical control strategy for Puget Sound. Results from the PSTLA will be a key component to (1) identify the most significant sources and delivery pathways of toxic chemicals and (2) prioritize reduction efforts to reduce threats from toxic chemical to the Puget Sound ecosystem. Ecology will use funding from a National Estuary Program grant to implement priority actions under the toxics control strategy.

Scope, Approach, and Organization of the Report

Many projects are included under the umbrella of the PSTLA, as shown in Figure 2. Appendix A provides additional information on PSTLA project subject matter, authorship, and completion status. All completed PSTLA reports are posted on the PSTLA internet homepage: www.ecy.wa.gov/programs/wq/pstoxics/index.html.

This *Assessment Report* focuses primarily on the PSTLA Phase 3 loading studies and draws on information provided in other PSTLA studies where appropriate. Since information on loading and delivery pathways, primary chemical sources, and other PSTLA-derived data may not by itself be sufficient to meet the overall goal of PSTLA, a screening-level hazard evaluation of selected chemicals was conducted and included in the *Assessment Report*. The hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin.

This *Assessment Report* is organized in a chemical-by-chemical fashion. For each of the selected chemicals addressed in the report, the major ongoing anthropogenic sources are discussed, an assessment of loading to Puget Sound and major pathways is presented, and the results of the hazard evaluation for the specific chemical is discussed. These elements may be re-phrased as the following questions for specific toxic chemicals in the Puget Sound basin:

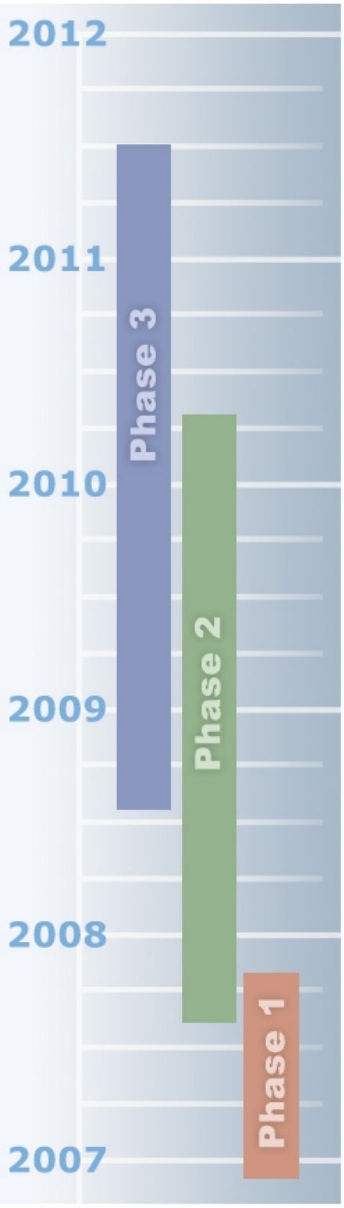
- Where do they come from?
- How much is being delivered to Puget Sound?
- What delivery pathways contribute to the loading?
- What is the relative toxic hazard posed by these chemicals at observed concentrations?

Figure 3 shows the major elements considered for this *Assessment Report*. As mentioned previously, loading estimates from different pathways come primarily from the PSTLA Phase 3 loading studies. Loading information from PSTLA Phases 1 and 2, as well as additional information on chemical transport and attenuation, may be used to supplement the PSTLA Phase 3 loading studies.

The PSTLA Phase 3 report on primary sources provides estimates of ongoing anthropogenic releases of selected chemicals. For each chemical addressed in this *Assessment Report*, the information on loading rates and pathways is discussed in context of their ongoing releases from primary sources (i.e. how these chemicals get in the environment in the first place).

As mentioned previously, the hazard evaluation provides information about the relative risk of toxic effects posed by selected chemicals at observed concentrations in the Puget Sound basin. The hazard evaluation was not produced as a separate project; it was conducted specifically for this Assessment Repot to enhance and supply additional context to the information on loading, pathways, and sources of toxic chemicals.

The major components used for this *Assessment Report* are described in more detail in subsequent sections. The reader should be aware that the *Assessment Report* does not consider these components exclusively. For instance, additional information on bioaccumulative chemicals in three guilds of marine organisms (West et al., 2011a and b; Noel et al., 2011) are used to provide additional context to the hazard evaluation for specific chemicals.



Phase 3: Targeting Priority Toxic Sources

Phase 3 Projects:

3A: Toxic Chemical Loadings via Surface Runoff
3B: Modeling Surface Runoff in Two Pilot Watersheds
3C: Evaluate Air Deposition
3D: Toxic Chemicals in Marine Waters and from Ocean Exchange
3E: Numerical Models and Scenarios
3F: Priority Pollutant Scans for POTWs
3G: Primary Sources of Toxic Chemicals
3H: Pharmaceuticals and Personal Care Products (PPCPs)
3J: Persistent Organic Pollutants in Three Guilds of Pelagic Marine Species from the Puget Sound
3K: Assessment Report
3L: Groundwater Discharge Directly to Puget Sound

Phase 2: Improved Loading Estimates

Phase 2 Projects:

2A: Loadings from Surface Runoff and Roadways
2B: Loadings from Dischargers of Municipal and Industrial Wastewater
2C: Bioaccumulation Model Estimates of Toxics from Sediments
2D: Water Column Data for Puget Sound and its Ocean Boundary
2E: Support for a Human Health Risk Assessment
2F: Numerical Models for Polychlorinated Biphenyls in Puget Sound
2G: Biological Observing System (TBiOS) for Toxics in Puget Sound

Phase 1: Initial Estimate of Toxic Chemical Loadings to Puget Sound

Phase 1 Project:

1A: Initial Toxics Loading Estimates

Figure 2. Timeline and Complete List of All PSTLA Projects.

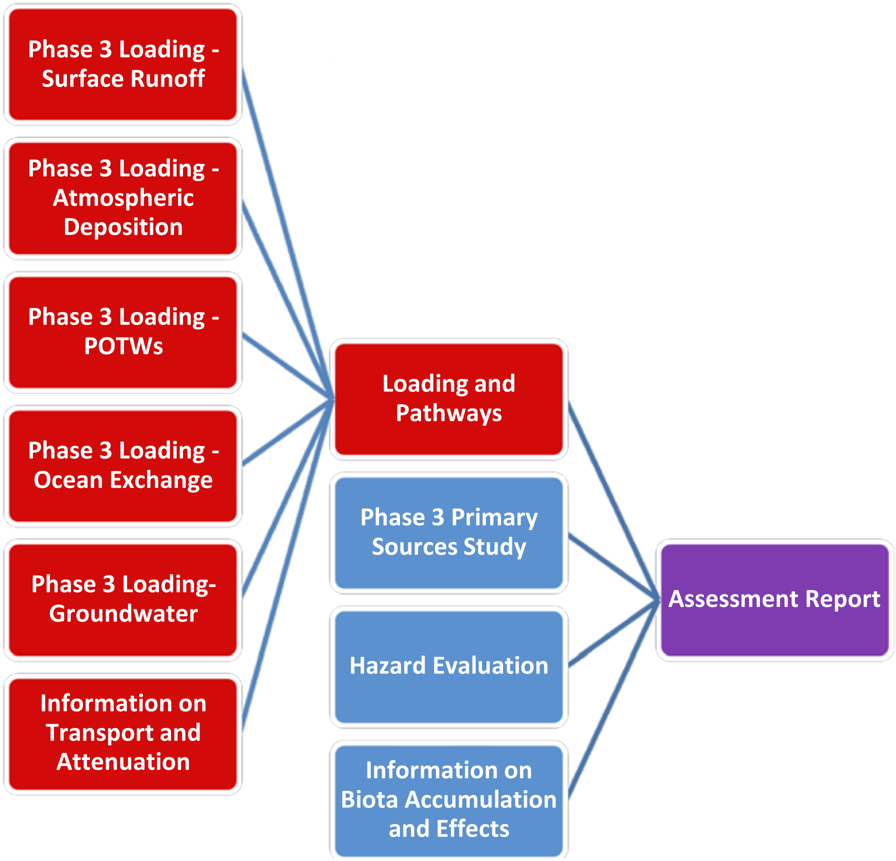


Figure 3. Elements Incorporated into the Assessment Report.

Selection of Chemicals

While there is general consensus that a large number of potentially harmful chemicals may be present in Puget Sound, the identification and evaluation of all chemicals was beyond the scope of the PSTLA projects. In order to focus on a group of chemicals that are known to be important and likely represent at least some of the most important ones in Puget Sound, the chemicals addressed in this *Assessment Report* are the chemicals of concern (COCs) first selected during the initial phase of the PSTLA.

The COCs were selected by a Chemicals of Concern Workgroup that had been convened to recommend a list of chemicals based on previous work and using best professional judgment. The workgroup sought to choose chemicals that had a documented history of presence in Puget Sound and "… that harm or threaten to harm the Puget Sound ecosystem and those that represent, or serve as an indicator for, a particular class of chemicals." The COCs list was developed to ensure that a broad variety of delivery pathways would be represented.

The workgroup settled on a list of chemicals that largely mirrored those identified by the then-lead agency for Puget Sound (Puget Sound Action Team; Redman et al., 2006). These chemicals became the core group of chemicals analyzed for the subsequent loading studies, although a few chemicals were excluded for particular studies while other studies included chemicals beyond the COCs.

The COCs are as follows:

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- Low molecular weight polycyclic aromatic hydrocarbons (LPAHs)
- High molecular weight polycyclic aromatic hydrocarbons (HPAHs)
- Carcinogenic polycyclic aromatic hydrocarbons (cPAHs)
- Diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate)
- Triclopyr
- Nonylphenol
- Oil and petroleum hydrocarbons

It is possible that this list of COCs represents only a small subset of those chemicals that may be impacting Puget Sound. Any conclusions drawn from the assessment of these COCs should not signify that other chemicals may not be of equal or greater concern.

Geographical Study Area

The geographical study area addressed in this *Assessment Report* is Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits (Figure 1). This is consistent with all of the land-based PSTLA loading projects (e.g. Hart Crowser et al., 2007; Envirovision et al., 2008a), except the groundwater loading analysis which excludes loads from the western Strait of Juan de Fuca.

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Puget Sound Toxics Loading Analysis (PSTLA) Projects

The bulk of COC data analysis in this report is contained in the Chemical-Specific Assessments section. The COC data analyzed in the Chemical-Specific Assessments section are primarily from the PSTLA studies. The following section summarizes the PSTLA studies used for these analyses in order to familiarize the reader with the subject and scope of the studies conducted under PSTLA. For more detailed information, the reader is advised to review the Chemical-Specific Assessments section or the original PSTLA studies which are referenced in the following section and are available online at www.ecy.wa.gov/programs/wq/pstoxics/index.html.

Loading Projects

PSTLA Phase 3 loading studies are described in the following sub-sections. PSTLA Phase 1 and 2 projects that were conducted to estimate COC loading are not included because their goal was generally to focus information for the development of the more refined Phase 3 loading studies. Aside from a few possible exceptions, the Phase 3 loading studies contain the best PSTLA loading data available.

Surface Water Runoff

Description

The Phase 3 study on COCs and other chemicals in surface water runoff (Herrera, 2011) was the primary off-shoot of the Phase 1 and 2 loading studies which identified surface runoff as the principal delivery pathway for most COCs. Like the other loading studies conducted under PSTLA, the surface runoff study was essentially a modeling effort which included the collection of sampling data to provide input data for the model used to estimate surface runoff loads of COCs.

The primary objective of the surface runoff study was to quantify the annual loading of COCs to Puget Sound through surface runoff, defined as the water flowing over the surface of the land at some point, including stream baseflow, stormwater, and groundwater discharging to surface waters. In addition to calculating COC mass loading to Puget Sound, the surface runoff study also provided data on chemical concentrations in surface runoff among different land cover types (commercial/industrial, residential, agricultural, and forests) and during different streamflow regimes (baseflow and storm flow).

Four sub-basins from the each of the representative land covers (commercial/industrial, residential, agricultural, and forests) were sampled twice during baseflow and six times during storm flow conditions between October 2009 and July 2010. Sampling was limited to stream sub-basins in the Snohomish River and Puyallup River watersheds for logistical reasons. Samples were analyzed for the entire list of COCs (except PCDD/Fs) as well as additional chemicals such as phthalates, semi-volatile organic compounds, pesticides, herbicides, and nutrients (Appendix B). Continuous stream discharges (flows) were recorded during the study period. The complete results of the chemistry and discharge data are included in the surface runoff report (Herrera, 2011).

The COC concentration data obtained from sampling were coupled with stream gauging data and converted to annual unit-area loads (e.g. kg COC/km^2) specific for a land cover and flow regime (baseflows or storm flows). The land area of the entire Puget Sound basin was assigned one of the four land covers, and the unit-area COC loads computed from study results were assigned to the corresponding land cover. Using this "scale-up" methodology, COC loads for the Puget Sound basin were calculated for both storm flows and baseflows.

Summary of Major Findings

The surface runoff project team found that PAHs, phthalates, semi-volatile organic compounds, pesticides, herbicides, and petroleum hydrocarbons were rarely detected or not detected at all. PCBs and PBDEs were detected in a majority of samples; however, only a few individual chemicals from each of these classes were commonly present. Most nutrients and six of the 15 metals evaluated in this study were detected in nearly all the samples. The frequency of detection and concentrations for most chemicals was generally higher for samples collected during storm flows than baseflow samples, a pattern generally consistent among all land cover types.

Since COC concentrations were typically higher during storm events, the increased discharge during storm events amplified the COC loads compared with baseflow loads. As a result, storm flow unit-area loads were much larger than baseflow unit-area loads for most chemicals. This suggests that environmental chemicals are mobilized during storm events; otherwise increased storm flows would simply dilute chemical concentrations and loads would remain the same regardless of discharge.

Although the surface runoff study demonstrated that chemicals are mobilized during storm events, it is unclear to what extent chemicals are transported to the stream corridors versus re-mobilization of chemicals residing in the stream corridors (e.g. sediments). It is likely that both circumstances occur to varying degrees, and may depend largely on the chemical in question.

COCs were generally detected more frequently and at higher concentrations in the commercial/industrial sub-basins compared to the other land cover types with a few exceptions; metals were occasionally detected more frequently and at higher concentrations in the agricultural areas. Metals aside, agricultural and residential areas had roughly the same level of chemical detection frequencies and concentration. Streams sampled in forested areas had the lowest overall concentrations as well as frequency of COC detection.

In general, COC unit-area loading rates for the four land cover types generally showed the same pattern as concentrations (commercial/industrial > agricultural ≥ residential > forest). However, since approximately 83.4% of the land base in the Puget Sound basin is forested and only 0.8% is commercial/industrial land cover, absolute loads from forested areas dominate the overall loading from surface runoff.

As with any study designed to extrapolate large values from a limited sample set, there are a number of uncertainties associated with the results. The low frequencies of detection for many organic COCs were particularly troubling since summary statistics for concentrations were required for calculation of loads. This required substitution methods for non-detected values within a concentration dataset, with one-half of the reporting limit typically used to substitute for non-detected results. Appendix B shows the method used to establish representative COC values for datasets with non-detected values. The result is that COC concentrations used to derive loads were determined largely by reporting limit values rather than measured concentrations.

The use of unit-area loads to compute basin-wide loading has the potential to introduce bias to the final load estimates. The unit-area load method assumes that all of a single land cover type (e.g. agricultural) in the Puget Sound basin delivers the same COC load regardless of the runoff volume (the runoff volume method was applied in the Phase 2 surface runoff estimates). This essentially has the effect of dampening the load signal from forest areas where the percentage of total runoff volume is even greater than the area. At the same time, loads from commercial/ industrial areas (and agricultural areas for some COCs) may be higher using the unit-area load method compared with loading calculations that use precipitation-driven runoff volumes. However, the overall effect of the unit-area load methodology results in lower absolute loads for the Puget Sound basin due to the diminished contribution of the forest loads.

Atmospheric Deposition

Description

The study of COCs atmospherically deposited directly to the surface of the Puget Sound marine waters (Brandenberger et al., 2010) was conducted based on recommendations from the Phase 1 report which concluded that PBDE and PAH loads delivered through this pathway may be comparable or greater than from surface runoff. The Phase 1 report also noted that there were few data to assess atmospheric deposition loads, and that what little data that existed was two decades old. There were no efforts among the Phase 2 projects to improve air depositions loads.

In order to obtain estimates of air deposition loads to Puget Sound, the project team designed a sampling network of seven locations geographically dispersed around Puget Sound that represented a variety of possible air pollution influences and precipitation patterns. One station was located in a high-density urban area of Tacoma with numerous potential industrial and roadway influences; a companion station was located in a nearby undeveloped area to assess the influence of highly localized air deposition.

Sampling was conducted at two-week intervals over the course of 14 months during 2008 – 2009. The funnel-type sampling devices used for the study collected bulk samples (dry + wet deposition) and were not designed to distinguish between dry and wet deposition. However, the large number (19) of discrete sampling events permitted a partial evaluation of differences between dry and wet deposition since sampling spanned wet and dry seasons.

Samples collected from the bulk deposition collectors were analyzed for the COC metals, PCBs, PBDEs, HPAHs, and cPAHs (Appendix B). Additional analyses were conducted for PAH markers and anhydrosugars in order to conduct analyses of PAH sources. Once COC concentrations from a sample were obtained, fluxes (e.g. ng COC/m^2/day) were calculated based on the sample volumes, funnel area, and elapsed collection time.

Using the COC fluxes, three scenarios were used to estimate total annual loads to Puget Sound:

1. The first scenario applied summary statistics derived from all of the pooled results to the entire marine surface.
2. The second scenario used location-specific deposition results and applied them to geographically associated marine compartments based on the Puget Sound Box Model (Pelletier and Mohamedali, 2009).
3. The third scenario used the same method as for the second scenario, but further divided the loading into dry and wet seasons.

The three scenarios for calculating annual loads from atmospheric deposition produced similar results, and the authors of the air deposition study did not explicitly state a preference for any particular scenario. Loading estimates from the third scenario are used for the present report because (1) values typically fell between estimated loads derived from the other scenarios, and (2) loads calculated for both dry and wet deposition could provide useful information for further analysis, much the same way baseflow and storm flow data from the surface runoff study are able to be evaluated independently.

Summary of Major Findings

All of the COCs analyzed were detected at all stations, and fluxes and loads were calculated based on summary statistics computed for each COC at each location. The high-density urban station in Tacoma had COC fluxes much higher than most other stations – an order of magnitude higher in most cases – including the nearby companion station. This finding supports the view that strong air deposition signals may be highly localized, particularly in high-density urban areas. Seasonal differences suggest a higher rate of wet deposition for metals, while the opposite appears to be the case for PBDEs. There does not appear to be any effect on seasonality for PAHs, although biomarker fluxes suggest a larger proportion of PAHs deposited during the winter originates from biofuel (e.g. firewood) combustion.

Users attempting to interpret results of the air deposition study should be aware of several limitations. The difficulties of attempting to extrapolate atmospheric deposition of chemicals across a large area using a limited sampling coverage are evident. As noted by the authors of the study (Brandenberger et al., 2010), the sampling coverage was able to capture only one location with elevated deposition rates due to highly localized conditions. Perhaps more important, mercury aside, there are few data on which to compare the results of the study. Most comparable data were collected 20 years prior to this study, and advances in analytical methods, increases in population, and emissions reduction efforts have all occurred during the intervening years. The current COC fluxes are much lower than those reported during the early 1990s.

Users of the data should also be aware that the method for handling non-detected PBDE congeners (14 were analyzed) was different from methods used in other loading studies. The method detection limit was used to replace each non-detect congener result during the summation of individual congeners to derive a total PBDE sum. The authors of the study acknowledged this resulted in a conservative (upward) bias of the results. This bias appears to result in fluxes (and resulting loads) on the order of 50% higher than those derived from using summing methods which substitute a zero for non-detected congeners when calculating a total PBDE sum.

In contrast to the non-detect substitution method described for PBDEs, non-detected PCB congeners were substituted with a zero, unless no congeners were detected for a sample, in which case one-half the highest method detection limit among congeners was used as the result. This likely resulted in a downward bias of the results. Appendix B shows the method used to establish representative COC values for datasets with non-detected values.

Publicly-Owned Treatment Works (POTWs)

Description

The Phase 3 project to assess COC loading through POTWs (Ecology and Herrera, 2010) was conducted based on recommendations from the Phase 1 report and results of a Phase 2 study of discharge data from POTWs and industrial wastewater discharges (Envirovision et al., 2008b). Both the Phase 1 and 2 studies found a limited amount of data which could be used to calculate reliable load estimates and recommended collection of additional data in order to refine an assessment of loading.

In order to estimate loads from POTWs, ten facilities were sampled during two events each. Sampling was conducted during February and July 2009 to represent wet and dry seasons, respectively. The POTWs were selected to represent varying types of treatment process, size, and source of wastewater, and were geographically distributed around the Puget Sound region. To obtain load estimates, the project team computed summary statistics for representative COC concentrations then multiplied concentrations by the average annual volume of treated wastewater discharged from all of the 96 POTWs in the Puget Sound basin.

Samples collected from POTWs were analyzed for the COCs (except arsenic, cadmium, mercury, and PCDD/Fs) as well as additional chemicals such as phthalates, semi-volatile organic compounds, pesticides, and herbicides (Appendix B). Few of these chemicals are routinely monitored by POTW operators.

Summary of Major Findings

Most classes of chemicals were detected in POTW effluent, and loads were calculated for all of the COCs analyzed except DDT, cPAHs, nonylphenol, triclopyr, and oil/petroleum. To obtain load estimates, the project team computed summary statistics for representative COC concentrations then multiplied concentrations by the average annual volume of treated wastewater discharged from all of the 96 POTWs in the Puget Sound basin. Due to the limited number of sampling events and atypical weather during the sampling period, the project team was not able to assess any seasonal variations in loadings.

Like the other loading studies, methods for handling non-detected results were required in order to obtain representative COC concentrations for datasets that contained a combination of detected and non-detected results. The POTW project team used the regression-on-order statistical analysis to calculate summary COC concentration statistics rather than simple substitution with one-half of the reporting limit. Perhaps more importantly, no loads were calculated for chemicals that had less than 50% overall frequency of detection. This appears to have introduced a downward bias in basin-wide loading estimates when compared with loading estimates that would have been obtained using the same non-detect substitution methodology that was applied in the surface runoff study. Appendix B shows the method used to establish representative COC values for datasets with non-detected values.

The absolute Puget Sound COC loads were calculated based on discharge volumes from 96 POTWs in the Puget Sound basin, yet not all of these POTWs discharge directly to the marine waters of Puget Sound. Efforts to distinguish POTW loads from surface runoff loads potentially run the risk of double-counting surface runoff loads if surface water sampling is conducted downstream of a POTW outfall. However, none of the surface runoff sampling stations were located downstream of POTW outfalls, and therefore distinct COC loads can be attributable to POTWs and surface runoff based on the methodologies used in the respective projects.

Ocean Exchange and Major Tributaries

Description

Ecology’s study of chemical load exchange at the ocean boundary (Gries and Osterberg, 2011) was originally conceived to provide chemical input data for the Puget Sound Box Model (Pelletier and Mohamedali, 2009) after a review of existing data (Serdar, 2008) found the available data were inadequate for the model. The ocean exchange project was designed with a number of marine water column stations to provide calibration data for the box model. The study design was later expanded to include measurements of chemical concentrations in the five rivers having the greatest annual discharges to Puget Sound in order to assess the relationships between river COC concentrations and those in corresponding inland marine waters.

The final sample design for the project included shallow and deep water sampling at three ocean boundary stations (eastern Strait of Juan de Fuca and Haro Strait), marine water at four locations representing the Main, Whidbey, South Sound, and Hood Canal basins, and freshwater and suspended particle sampling near the mouths of the Skagit, Snohomish, Nooksack, Stillaguamish, and Puyallup Rivers. Samples collected from freshwater and marine waters were analyzed for the COCs (except mercury, PCDD/Fs, and triclopyr; oil and petroleum was analyzed in freshwater only) as well as additional chemicals such as phthalates, semi-volatile organic compounds, and chlorinated pesticides (Appendix B).

The exchange of chemical loads at the ocean boundary was calculated by using concentrations in the deep water boundary stations to represent inflow to Puget Sound, and chemical concentrations from the Main, Whidbey, and Hood Canal basin sites to represent water flowing out of Puget Sound. The concentrations representing the inflow and outflow were then multiplied by the known volumes of water flowing in and out of Puget Sound, respectively.

Summary of Major Findings

The study found metals, PCBs, and PBDEs at detectable concentrations in the marine water column, but other COCs were either not detected or detected at such low frequencies that load estimates were not calculated.

Metals, PCBs, and PBDEs were found at detectable concentrations in the water column of rivers, but other COCs were either not detected or detected at such low frequencies that load estimates were not calculated. However, PAHs and DEHP were detected at high frequencies in suspended particles (sampled once in each river versus three water sampling events for each river).

Due to the inherent variability of chemical loads carried by rivers due to constantly shifting stage and conditions, the authors (project team) did not feel that annual loading estimates were supportable based on three sampling events. However, instantaneous daily loads were calculated and reported based on the river discharges recorded during the sampling events.

Uncertainties and limitations of the ocean exchange and major tributaries project were due more to assumptions about hydrology than difficulties associated with handling non-detected laboratory results. Perhaps the largest assumption used by the project team is that COC concentrations at deep water locations in the eastern Strait of Juan de Fuca and Haro Strait represent those flowing into Puget Sound, and COC concentrations from various Puget Sound locations represent concentrations in the outflow. Recommendations were made to improve the assessment of chemical exchange by sampling at more representative boundary locations, namely Admiralty Inlet (sill) and Deception Pass.

Aside from PBDEs, there was little need for the project team to adopt a method for calculating summary statistics for datasets containing non-detects. For PBDEs, the method of substituting a non-detect result with one-half of the estimated quantitation limit was adopted (Appendix B).

PBDE results also showed a very high degree of variability during the study, particularly for marine waters. The project team was unable to provide an explanation for the high degree of variability, but they did note that there was no evidence of sample contamination.

Groundwater

Description

The 2007 Phase 1 report included a specific recommendation for the development of loading estimates for *direct* groundwater discharge to Puget Sound. Although the authors of the Phase 1 report (Hart Crowser et al., 2007) acknowledged the potential for the groundwater pathway to contribute significant toxic chemical loads at the local scale, this pathway was not addressed at | a basin-wide scale. In light of the low mobility characteristics of many of the COCs and the assumed contaminant attenuation capacity of subsurface sediments, the transport of toxic chemicals to Puget Sound via direct groundwater discharge was generally considered to represent a comparatively minor component of the overall loading to Puget Sound marine waters, but this assumption had not been confirmed by a formal technical analysis.

The Phase 1 report recommendation to conduct a sampling-based groundwater loading assessment was not carried forward to a Phase 3 project. Instead, a loading study was conducted using readily available data to address the absence of data on COC loading through a direct groundwater pathway (Pitz, 2011). The study produced estimates of upper-bound (worst-case) mass loads of COCs delivered annually to Puget Sound through the groundwater pathway. The reader should note that i*ndirect* groundwater contributions of toxic chemicals to freshwater streams and rivers draining to Puget Sound are assumed to be represented in the load estimates developed for the surface runoff pathway (Herrera, 2011).

A substantial majority of the chemistry data used for the groundwater loading analysis was from industrial or commercial sites or from facilities that are known or suspected to have point-source-related toxic contamination, typically sampled in response to the Model Toxics Control Act (MTCA) or state Dangerous Waste regulations. Additional sources of data were also found to represent un-impacted (i.e. ambient) groundwater, but these datasets were small compared to the amount of data from impacted areas since most of the COCs are not typically analyzed during routine groundwater monitoring. Only data collected within 500 meters of the marine shoreline were used to derive representative COC concentrations for load calculations.

In an attempt to minimize potential bias from the high proportion of data from contaminated sites, groundwater data were divided into three categories: impacted areas, urban ambient areas, and non-urban ambient areas. Data from each category were pooled separately, and representative concentrations from each category were derived for loading estimates. Fluxes were then calculated by multiplying by the discharge (flow) for each shoreline segment by the COC concentration representing the groundwater data category for the associated 500-m buffer. This approach reduced the groundwater discharge associated with impacted areas to approximately 1% of the overall volume, while non-urban ambient areas discharged approximately 75% of the groundwater.

Sufficient groundwater data were generally available for metals but comparatively scarce for organic compounds. In particular, data on PCBs, PBDEs, PCDD/Fs, triclopyr, and nonylphenol were not sufficient to derive usable representative concentrations for groundwater.

Summary of Major Findings

The groundwater loading analysis found that COC loads from ambient areas generally exceed loads from impacted areas as a result of the comparatively high flow volume in ambient areas. This is similar to the dominant effect that forest land has on absolute COC loads as reported in the surface runoff study. For many of the PAHs, however, the highly elevated concentrations in groundwater from impacted areas more than compensated for lower flows, resulting in comparatively high PAH loads from impacted areas.

Users of the groundwater loading data should be aware of the many limitations and assumptions used in the report. It is an initial effort to calculate direct groundwater loads and has not benefited from the refinement process used for some of the other loading projects. The author (Pitz, 2011) found a low frequency of detection for nearly all of the COCs, and therefore the concentrations used for loading are largely driven by non-detected values (and the substitution methods used to handle non-detects). Appendix B shows the method used to establish representative COC values for datasets with non-detected values. Complex decision processes were developed to derive COC concentration for use in loading computations. Even the discharge volume estimates used to calculate loads, typically a consistent factor in loading studies for other pathways, ranged by an order of magnitude.

In addition to the limitations and assumptions discussed above, there are also uncertainties about the mobility and attenuation of COCs in groundwater. The groundwater loading study does not attempt to account for attenuation – an additional reason the results represent upper-bound estimates – but instead assumes that the COCs will migrate to the marine boundary in the same concentrations measured in upland groundwater. These and other uncertainties and limitations are thoroughly documented in the groundwater loading report (Pitz, 2011).

Other Projects

Inventory of COC Releases from Primary Sources

Description

In recognition that loading analyses by themselves may not provide adequate information to help Ecology, the Puget Sound Partnership, and others to develop and implement a toxics reduction and control strategy, an inventory of COC releases was undertaken by Ecology as part of the PSTLA Phase 3 effort. This sources inventory, referred hereto after as the *Sources Report* (Ecology, 2011), was conducted with the objectives of (1) identifying major ongoing anthropogenic sources of COCs in the Puget Sound basin and (2) estimating the quantities in which they are released into the environment.

The *Sources Report* focused specifically on the release of COCs from their primary sources. The term *source* was strictly defined as *the object or activity from which a COC is initially released to environmental media or released in a form which can be mobilized and transported in an environmental pathway* (Ecology, 2011). The term *primary source* was used to distinguish the initial release of a COC from a secondary release, such as mobilization of a chemical from a toxic cleanup site.

Examples of releases from primary sources include copper and zinc released from tire and brake pad wear, PAHs formed and released from combustion sources, and motor oil released from vehicle drips and leaks. The *Sources Report* did not estimate releases from secondary or natural sources.

All COCs except DDT were addressed in the *Sources Report*. The quantity of COCs released to the environment from approximately 110 primary sources was estimated from available information; no sampling was conducted for the project.

Summary of Major Findings

The study found that petroleum is released in the largest quantity among COCs, followed by zinc which is the only additional COC released at a rate greater than 1,000 metric tons (t) per year.

Lead, PAHs, and copper are released at rates greater than 200 t/yr, and triclopyr is released at a rate over 100 t/yr. Approximately 30 t/yr of phthalates are released, but the organic chemicals PCBs, PBDEs, PCDD/Fs, and nonylphenol, as well as the metals mercury, arsenic, and cadmium, are generally released at rates near 1 t/yr or less. A summary of release estimates for all COCs is included as Appendix C.

While the *Sources Report* provides only rough estimates for many of the COC release rates, useful information about the relative magnitude of releases from each source is contained in the report. The *Sources Report* also discusses how COCs are released from their sources, providing insight into their transport and fate in the environment following release.

The study of COC releases from primary sources was limited in its ability to provide refined estimates of COC releases, and the authors (project team) were not able to consider all sources of COCs to the Puget Sound environment. The release estimates were based on many assumptions and limited data in many cases. However, these assumptions and associated uncertainties are documented in the report.

Evaluation of Fate and Transport Mechanisms

Description

Understanding the behavior, transport, and fate of chemicals following their initial release from primary sources is a key element in developing strategies for controlling chemicals at their source as well as along their transport pathways following release. The transport and fate of chemicals in the environment is complex, particularly in the upland environment where various media and management practices can affect chemical concentrations, loads, sequestration, and removal. In order to better understand the transport and fate of chemicals in the environment, staff at the U.S. Geological Survey (USGS; Paulson et al., 2011-Draft) conducted an evaluation of transport and fate for copper released from several of the major sources identified in the *Sources Report* (Ecology, 2011). The evaluation of fate and transport mechanisms was not conducted as a discrete PSTLA project but instead was done specifically to enhance this *Assessment Report*. Upon its completion, it will be included as an amendment to this report.

The authors (Paulson et al., 2011-Draft) of the transport and fate evaluation used a mass-balance case study approach to examine factors affecting copper attenuation following release from roof runoff, brake pads, and vehicle tires. Using information from the *Sources Report*, the authors calculated the mass of copper released annually from these sources in two small urban watersheds in King County, Washington. The annual mass of copper discharged from these watersheds was estimated from historical sampling data for the streams comprising the respective watershed outlets. Finally, the authors incorporated information on the types of copper release, the likely locations of the releases, watershed characteristics including best management practices, and stream and water quality characteristics to provide possible explanations for copper attenuation.

The initial scope of this project included a proposal to conduct a similar evaluation for PCB and PBDE transport and fate. However, the lack of data on these chemicals in urban streams and stormwater precluded the authors' ability to conduct an evaluation for these chemicals using a case study mass-balance approach.

Summary of Findings

Findings of the copper transport and fate evaluation suggested that less than 10% of the copper mass estimated to be released in the watersheds was discharged at the respective watershed outlets. It appeared that the retention of copper in the watersheds was largely governed by the limited amount of water volume discharged from the watershed at the surface outlet; only about 15% of the estimated storm precipitation for each watershed was discharged at the surface outlets. The authors speculated that the retention of copper was due to infiltration into groundwater, trapping of particles by lawns, grassy road-side ditches, road shoulders and road right-of-ways, retention in the many structures installed in the watershed, and settling of particles in vegetated channels in low-gradient portions of the watersheds. Some of the copper released in the watersheds may have been removed by street sweeping, but this portion was likely minor.

The movement of copper by a variety of transport mechanisms through a variety of pathways was discussed by the authors of the fate and transport report (Paulson et al., 2011-Draft), although most of the information provided was speculative due to a lack of data for the multiple environmental media components required for an exhaustive evaluation. In cases where there were available data, the information did not always yield clear conclusions regarding copper transport and fate. For instance, data on copper analyzed from road surfaces resulted in divergent lines of evidence as to whether copper released from brake pads remains on the road surfaces or alternatively is suspended in air and blown away from the roads. This example underscores the complexity and difficulties in understanding the behavior, transport, and fate of chemicals at a small scale.

Hazard Evaluation for COCs in the Puget Sound Basin

The hazards posed by different COCs are not simply associated with the quantities released to the environment or loaded to Puget Sound, but are rather more appropriately evaluated by assessing their concentrations in various media. To assess the relative hazards posed by COCs, Ecology consulted with the ecological toxicology assessment team at King County Natural Resources and Parks to design and conduct a hazard screening of COCs in the Puget Sound basin. The hazard evaluation was not conducted as a discrete PSTLA project but instead was done specifically to enhance this *Assessment Report*.

To evaluate hazards potentially posed by COCs in the Puget Sound basin, King County used a methodology in which readily available observed environmental data for each COC were compiled then compared to concentrations where effects are documented, or to criteria established to protect aquatic life or consumers of aquatic organisms. These comparisons do not attempt to estimate absolute hazards but instead provide a rough discriminator of relative hazards among COCs using a specified methodology. A similar approach was used in a survey conducted several years earlier to evaluate endocrine disrupting chemicals in King County surface waters (King County, 2007).

The hazard evaluation was conducted for the following categories:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water, and sediment
- Hazard to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: a Priority 1 level of concern, a Priority 2 level of concern, or unknown (U) level of concern due to lack of sufficient data for an assessment. Results were classified as a Priority 1 when high observed concentrations (e.g. 90th percentile values) exceeded low effects concentrations (e.g. 10th percentile values), selected criteria, or other threshold values. A Priority 2 level of concern was assigned in cases where high observed concentrations were below threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U. Appendix D-1 details the thresholds used for comparisons and the minimum data required for the comparisons.

Environmental COC concentrations used for the hazard evaluation were obtained from a number of data sources including Ecology's Environmental Information Management (EIM) system, King County Laboratory Information Management System, USGS data obtained from their online database, the National Oceanic and Atmospheric Administration (NOAA) mussel watch program, and ENVVEST data from the U.S. Department of Defense. Data from the PSTLA Phase 3 Ocean Exchange, Pelagic Fish, and Surface Runoff studies were included among the observed data compilations. Due to the complexities associated with handling non-detects, only detected values were used to represent observed environmental COC concentrations for each of the media assessed.

Effects concentrations were obtained from a variety of sources. For surface waters, effects data were obtained from EPA's ECOTOX database for surface water. Freshwater and marine sediment data were compared to Washington State's Sediment Quality Standards (2003 Floating Percentile values for freshwater). Effects resulting from fish and invertebrate tissue burdens were evaluated using data from the Lower Duwamish Waterway Group and the Lower Willamette Group Remedial Investigation Baseline Ecological Risk Assessments. Wildlife hazards were evaluated by comparing daily COC doses through various exposure routes with daily doses where effects have been demonstrated. National Toxics Rule criteria were used as a basis for evaluating human health hazards.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U, this should not be interpreted to mean there are no hazards associated with that COC. Locally, concentration hot spots may exist near major sources, and may cause localized toxicity to aquatic organisms or lead to violations of standards. Finally, no attempt was made to evaluate hazards due to multiple COC exposures.

Details of the methodology and assumptions used for the hazard evaluation are in Appendix D-1. This includes rules for assigning COCs to Priority Levels for each media or receptor evaluated. Additional sections of Appendix D include box plots of the observed and effects concentrations and tabular summaries of the data and the results.

Puget Sound Box Model

Description

The Puget Sound Box Model study was developed as a tool to predict concentrations of PCBs in water, sediment, and biota of Puget Sound (Pelletier and Mohamedali, 2009). Three separate existing models were linked to achieve this task:

- A model to predict the circulation and transport of between regions of Puget Sound and between surface and deep layers of the water column.
- A contaminant fate and transport model to predict water and sediment concentrations of PCBs in response to external loading and internal processes.
- A food web bioaccumulation model to predict PCBs in Puget Sound biota in response to water and sediment concentrations.

Summary of Findings

Based on model outputs, concentrations of PCBs in sediments and biota were found to be very sensitive to external loading. However, the authors found that conclusions about increasing or decreasing trends in Puget Sound PCB mass could not be made due to the wide range in uncertainties regarding current external loading rates.

The median estimates of PCB loading used for the model showed slight increases in the total PCB mass for Puget Sound. However, the model was conducted using available information at the time and did not benefit from data collected during Phase 3 studies that may have aided in model calibration.

Persistent Organic Pollutants in Three Guilds of Marine Species

Description

Investigations of persistent organic pollutants (POPs) in three guilds of marine species were comprised of three separate studies carried out by Washington Department of Fish and Wildlife (WDFW), NOAA, and Fisheries and Oceans Canada. POPs were analyzed in plankton (West et al., 2011a), pelagic fish (West et al., 2011b), and harbor seals (Noël et al., 2010).

Previous studies have suggested that some pelagic species may accumulate higher levels of POPs from the water column and other pelagic components of the food web than from contaminated bottom sediment. In addition, pelagic fish are considered to be the primary source of POPs to southern resident killer whales. The overall goal of the studies on POPs in marine species was to assess where geographically the POPs enter the pelagic food web from stormwater and the atmosphere, the pathways of POPs within the pelagic food web, and the sources of POPs to species occupying the highest trophic levels (marine mammals, seabirds, and humans). The data from these studies were also intended to be used to refine the Puget Sound Box Model (Pelletier and Mohamedali, 2009).

For the plankton study, investigators analyzed phytoplankton (and other organisms and particles retained in a 20-micron net) and three species of krill which graze on phytoplankton (primarily *Euphausia pacifica*, but also *Thysanoessa spinifera* and *T. raschii).* Samples were obtained from numerous locations around Puget Sound during 2009. Phytoplankton and krill were analyzed for PCBs, PBDEs, PAHs, chlorinated pesticides, and ancillary parameters to assist with interpretation of the results.

In the study of pelagic fishes, researchers analyzed Pacific hake (*Merluccius productus*), which are an important prey item for harbor seals, and walleye pollock (*Theragra chalcogramma*). These species consume a variety of zooplankton such as the krill analyzed in the phytoplankton study, as well as small pelagic forage fishes. Hake or pollock were collected from a number of locations representing six hydrologically distinct waterbodies and one urbanized embayment during 2009. All fish were analyzed whole for PCBs, PBDEs, chlorinated pesticides, and ancillary parameters to assist with interpretation of the results.

The harbor seal (*Phoca vitulina*) study consisted of sampling 24 pups from four widely dispersed locations in Puget Sound. Investigators collected blood, fur, and skin/blubber biopsy samples from the pups. Skin/blubber samples were analyzed for PCBs, PCDD/Fs, PBDEs, chlorinated pesticides, and mercury. Hair and blood samples were analyzed for a variety of parameters to assess the feeding ecology, contaminant trends over space, and effects on their health.

Summary of Findings

For the plankton study, PCBs, PBDEs, DDTs, and PAHs in both particulate organic matter and krill exhibited a correlation with urban waters, and for PCBs and PBDEs in particular, concentrations were lower in less developed, more ocean-influenced basins. This suggests that urban waters represent areas where POPs enter the pelagic food chain.

Although PAHs are known to be metabolized and therefore do not accumulate in tissues of aquatic vertebrates (they were not analyzed for the companion fish and harbor seal studies), the authors of the plankton study found high levels of PAH accumulation in both phytoplankton and krill compared to other POPs. They also noted that a potentially significant implication of this finding was that pacific herring, a primary predator of krill in Puget Sound, exhibited significant exposure to PAHs possibly pointing to krill as a major contaminant transfer pathway. Another finding regarding PAHs was the relatively high concentrations in phytoplankton from non-urbanized basins, and in particular from samples collected near marinas, ferry terminals, or shoreline roadways. This suggests that shoreline development may play an important role in PAH transfer to the pelagic food web.

Patterns of PCB, PBDE, and chlorinated pesticide accumulation similar to plankton were found by authors of the pelagic fish study. Greater size- and lipid-specific accumulations of these chemicals were observed in Pacific hake from more developed basins compared to those with less development. As pointed out by the authors, the lipid-weighted PCB concentrations in Pacific hake were similar to Pacific herring and higher than in Chinook salmon, all species consumed by harbor seals. However, harbor seals may selectively prey on larger hake, which are typically females and have lower lipid and PCB concentrations, potentially making Pacific herring a greater overall contaminant transfer pathway than hake.

Results of the harbor seal study indicated that Hood Canal (south) seal pups were the least contaminated overall. Pups from the Main Basin of Puget Sound had the highest PCB and mercury levels. PBDE levels from all locations were similar, except Hood Canal which had the lowest concentrations. There was no discernible geographical trend in chlorinated pesticide levels, and PCDD/Fs were detected so infrequently that it was difficult to distinguish trends, although concentrations from the South Sound (east) basin were highest.

The authors of the harbor seal study note that several indicators suggest that the health of harbor seals may be impaired due to the contaminant exposure, particularly PCBs. These indicators include both threshold values based on effects and developed to protect marine mammals, as well as the health indicators measured during the study. They also noted that, while PCB exposure and accumulation may pose health risks to harbor seals, concentrations in seals have decreased appreciably during the 2000s.

There appear to be similarities in contaminant accumulation among the three studies conducted on POPs in marine organisms, and some inferences can be drawn from the results. For instance, there is a consistent geographical pattern seen in PCB concentrations across all species, which indicates the greatest exposure occurs in the Main Basin or embayments therein (e.g. Elliott Bay). However, at the time of this writing there has been no assessment of the results considered as a whole to: (1) Evaluate the consistencies in geographical patterns and (2) Assess the trophic transfer of contaminants between plankton and hake, and between hake and harbor seals. This may be best accomplished through updates and refinements to the Puget Sound Box Model.

A Toxics-Focused Biological Observing System for Puget Sound

Description

The concept of a toxics-focused biological observing system (TBiOS) was developed by authors from NOAA Fisheries, WDFW, and University of California (UC) Davis as a framework for assessing and monitoring toxic chemicals in Puget Sound through biological components (e.g. accumulation, responses, effects) rather than simply monitoring toxic chemicals through the more conventional analysis of water and sediments (Johnson et al., 2010a). Examples of recent research showing adverse effects to aquatic organisms from contaminant exposure are provided. The authors point out that biological monitoring would allow us to evaluate the impacts of toxic chemicals, the effectiveness of efforts to reduce toxic chemicals, effects to the ecosystem and the food web, and the effects of toxic chemical exposure coupled with other stressors.

As proposed, TBiOS would have three major components:

- Region-wide monitoring of toxic chemicals to assess large-scale geographical or temporal trends.
- Localized effectiveness monitoring to assess the impact of local source control or cleanup efforts.
- Diagnostic studies that would help uncover biological effects caused by toxic chemicals and develop monitoring tools to measure these effects.

The authors propose general ideas for the type and scale of monitoring and assessment programs that might be conducted under TBiOS. These programs would be a combination of new initiatives and building upon existing programs, such as the Puget Sound Assessment and Monitoring Program (PSAMP).

Benefits of TBiOS would include:

- Identifying toxic chemical-associated injury to the Puget Sound ecosystem, including the geographic extent and severity of the problem.
- Increasing our understanding of how toxic chemicals move through the Puget Sound ecosystem and accumulate in shellfish, fish, wildlife, and consumers of these organisms.
- Guiding our toxics reduction strategy efforts by helping to identify those watersheds where contaminants are the greatest problem and help us focus where detailed evaluations are most needed.
- Helping us evaluate the effectiveness of regional and localized toxics reductions strategies and actions.
- Establishing cause-and-effect linkages between toxicant exposure and biological impacts.
- Helping develop and establish more protective water quality and sediment guidelines.

Bioaccumulation from Sediments

Description

The study on bioaccumulation modeling was conducted to predict the concentrations of toxic chemicals in organisms resulting from specific concentrations in Puget Sound sediments (Ecology and Environment, 2009). In particular, the model was applied to the organic chemical criteria of Washington's Sediment Quality Standards (SQS). Since the model can also predict water column concentrations, surface water concentrations were predicted to provide comparisons with surface water criteria.

The model used for this effort was based on the Condon bioaccumulation model (Condon, 2007) which was developed to predict PCBs in biota from the Strait of Georgia, and therefore was deemed (with some modifications) adaptable for Puget Sound. This was the same model used as the food-web bioaccumulation component of the Puget Sound Box Model (Pelletier and Mohamedali, 2009; described above) to predict PCB concentrations in biota. The report documents modifications made to the Condon model to accommodate chemicals other than

PCBs, and in some cases, specimens in addition to those used by Condon (e.g. herring and salmon). PCBs, PAHs, and DEHP were the only COCs analyzed for the sediment bioaccumulation study.

Summary of Findings

Results of the modeling exercise showed that, at SQS levels in sediment, predicted water concentrations of PCBs and several PAHs may not be protective of human health, marine organisms, and piscivorous wildlife (PCBs only). Several non-COC organic chemicals were also predicted to be found at non-protective concentrations in water.

The authors of the report conclude that the model provides insights into the behavior and transfer of contaminants in the food web. However, they note the vast complexity of food-web modeling and caution users to consider the numerous assumptions and uncertainty before applying this or other generalized models, particularly if the model may be used for regulatory and management decisions.

Pharmaceuticals and Personal Care Products in POTWs

Description

The study of pharmaceuticals and personal care products (collectively referred to as PPCPs) was a screening-level effort carried out by Ecology and EPA Region 10 staff during 2008 (Lubliner et al., 2010). The study consisted of analyzing one-day composite samples of influent, effluent, and sludge from four POTWs in the Puget Sound region and one POTW in Hayden, Idaho. All of the plants had different processes for treatment of wastewater, with two plants employing secondary treatment and three plants employing tertiary treatment for nitrogen and phosphorus removal.

The screening-level of PPCPs was conducted due to concerns emerging from recent studies (cited in Lubliner et al., 2010) documenting PPCP presence in the aquatic environment and the possibility PPCPs may lead to effects which are not widely understood or have not been investigated. Since POTWs are a major step along the pathway from consumer use to release in the environment, the study sought to generate information about the effectiveness of POTWs in removing these chemicals. The study analyzed 72 PPCPs, 27 hormones and steroids, and 73 semi-volatile organic chemicals. PAHs, DEHP, and nonylphenol were the only COCs among the analytes selected for the PPCP study.

Summary of Findings

The authors of the study found that PPCPs are detected routinely in municipal wastewater and that their removal in POTWs varies by chemical and treatment process. Approximately one-fifth to one-half of the analytes were reduced to levels below reporting limits in the effluent. Overall, the combination of enhanced biological nutrient removal and filtration processes was found to provide the greatest PPCP removal effectiveness, although the authors note that this treatment process is employed by relatively few POTWs in the Puget Sound basin.

As for the COCs analyzed, DEHP was found in all influent samples, and concentrations were greatly reduced in all but one of the POTW effluents. Nonylphenol (4-nonylphenol) was rarely detected in POTW influent or effluent. None of the 16 PAHs analyzed were detected in either influent or effluent samples, although indeno(1,2,3-cd) pyrene was selected as one of the few target analytes for biosolids and was detected in three-quarters of the samples analyzed.

Chemical-Specific Assessments

Arsenic

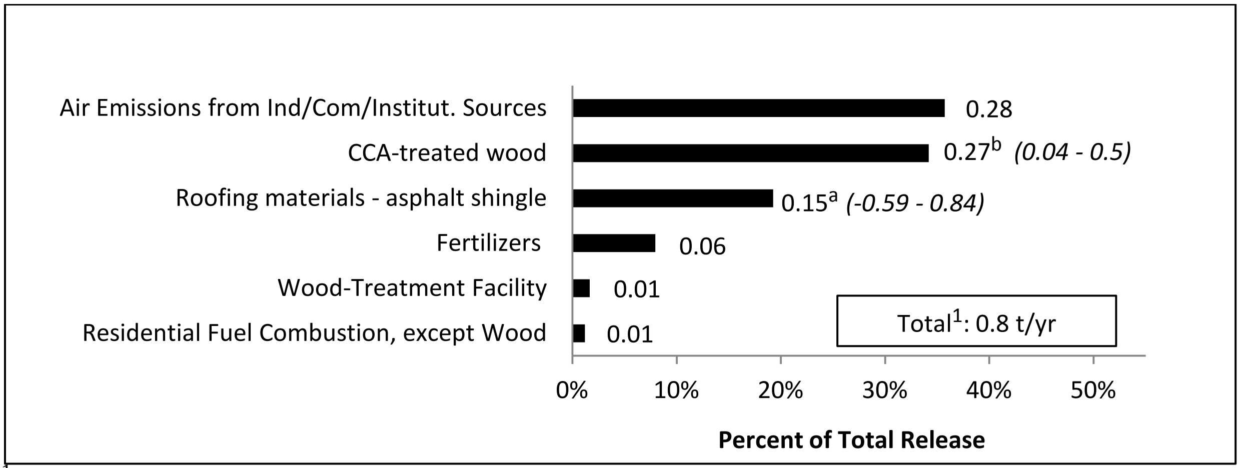
Arsenic is a naturally occurring element in the earth's crust and is nearly always detectable at concentrations >0.1 ug/l in water and >100 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Toxicity in water varies due to pH and redox potential (Eisler, 1988a).

Historically, approximately 70% of the global arsenic emissions are from anthropogenic sources, with the remaining 30% due to weathering of soils and rock (Moore and Ramamoorthy, 1984). In the Puget Sound region, historical releases may have occurred due to uses of arsenic compounds as pesticides as well as releases from large industrial sources. The Asarco Smelter in Tacoma emitted arsenic for decades and may have resulted in large swaths of the Puget Sound area with elevated arsenic (PTI, 1991; San Juan, 1994).

Major Releases from Primary Sources

An inventory of primary arsenic sources suggests that approximately 0.8 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin (Ecology, 2011). The largest current source of arsenic to the environment is point-source air emissions, accounting for approximately one-third of the total release (Figure 4).

There is a large amount of uncertainty around estimates of arsenic released from roof runoff and CCA-treated wood, sources which combined account for about one-half of anthropogenic arsenic released in the Puget Sound basin. For other sources – fertilizer application, a wood treatment facility that releases arsenic primarily to surface water, and residential fuel use (excluding wood) – arsenic releases are relatively minor.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mean

[b] Mid-point of range

Figure 4. Total Arsenic Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for arsenic loading were obtained from PSTLA loading studies and are included in Table 1. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 1. Total Arsenic Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.06	0.43	0.79
Air Deposition	0.25	0.35	0.54
Surface Runoff	13.5	16.9	23.4
POTWs	NA	NA	NA
Ocean Exchange (b)	-28	-23	-24

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary
NA=not analyzed

Estimated groundwater loads of arsenic range by an order of magnitude (0.06 – 0.8 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Arsenic was not measured in POTWs for the Phase 3 loading study. Phase 1 estimates suggest that arsenic loads from industrial wastewater are potentially substantial (0.2 – 14.6 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of arsenic reported in the Toxics Release Inventory (TRI) (0.01 t/yr total) (Ecology, 2011). In contrast, Phase 1 estimates for arsenic loading via municipal wastewater were based on a small set of non-detected sample results, and the load estimates appear to be unrealistically small (0 – 1 kg/yr). Due to the unreliability of the arsenic loads discharged through wastewater treatment, estimates from this pathway were not included in Table 1.

The deposition of atmospheric arsenic directly to the marine waters of Puget Sound is approximately 0.25 – 0.54 t/yr, based on fluxes ranging from 0.06 – 0.65 ug/m^2/d. Median arsenic fluxes are generally $\leq$0.2 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and in close proximity to major roadways including interstate highways. Fluxes at this location are consistently several times higher than at other locations.

Surface runoff loads for arsenic are estimated to be approximately 13.5 – 23.4 t/yr for the entire Puget Sound basin based on the surface runoff study. Nearly all (97%) of the total arsenic was in the dissolved form during baseflows; the fraction of dissolved arsenic decreased during storm flows to 74% of the total. Arsenic concentrations observed across base and storm flows were found to significantly correlate with total suspended solids (TSS) in residential and forest areas, partly due to a seasonal first-flush episode in the forested sub-basins.

Forested areas had much lower arsenic concentrations than other land covers. Commercial/ industrial and agricultural land covers had the highest arsenic concentrations, approximately double those in residential areas and four-fold higher than forests during baseflows. For all of the land covers, arsenic concentrations decreased during storm events, particularly in commercial basins.

Overall, there appears to be a net export of arsenic out of the Puget Sound due to exchange of marine waters, although total arsenic concentrations in the incoming marine waters (1.44 – 1.52 ug/l; 25th -75th percentile) are slightly higher than concentrations in the outgoing marine waters (1.36 – 1.49 ug/l). Total net export was 24 – 28 t/yr based on an inflow of 842 – 889 t/yr and an outflow of 870 – 913 t/yr.

The net sum of arsenic loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is -14, -5.3, and 0.7 t/yr, respectively. Under estimates at the 25th and median levels, there is a net export of arsenic out of Puget Sound due to the large mass exchanged at the ocean boundary. Estimates at the 75th percentile suggest that a net outflow at the ocean boundary is balanced by loads from the watershed and from air deposition.

Arsenic loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) is shown as ranges (25th -75th percentiles) and median for each pathway in Figure 5. Each pathway represented as a contribution to the total load is displayed in Figure 6.

Total arsenic loading from the major pathways assessed is 14 – 25 t/yr. Surface runoff accounts for the largest pathway (95% – 98%), followed by air deposition (2%). Groundwater potentially accounts for up to 3% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of arsenic in wells and variable estimates of groundwater flow. At the lower end of the load range, arsenic in groundwater is estimated to account for <1% of the load to Puget Sound. The loading studies failed to provide estimates of arsenic loading through POTWs, and earlier estimates (Envirovision et al., 2008b) provide little information on which to base reasonable load estimates.

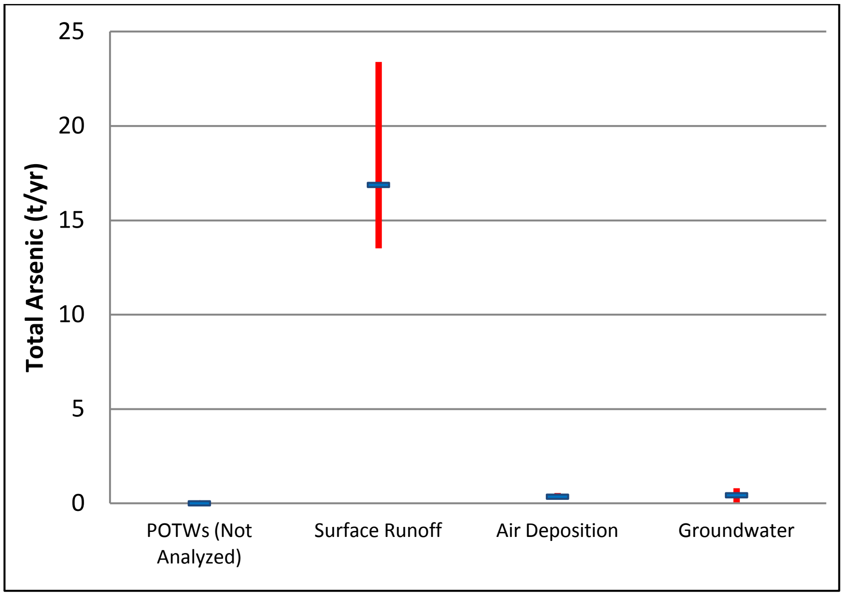


Figure 5. Range (25th - 75th percentiles) and Median Total Arsenic Loads to Puget Sound from Each Major Delivery Pathway.

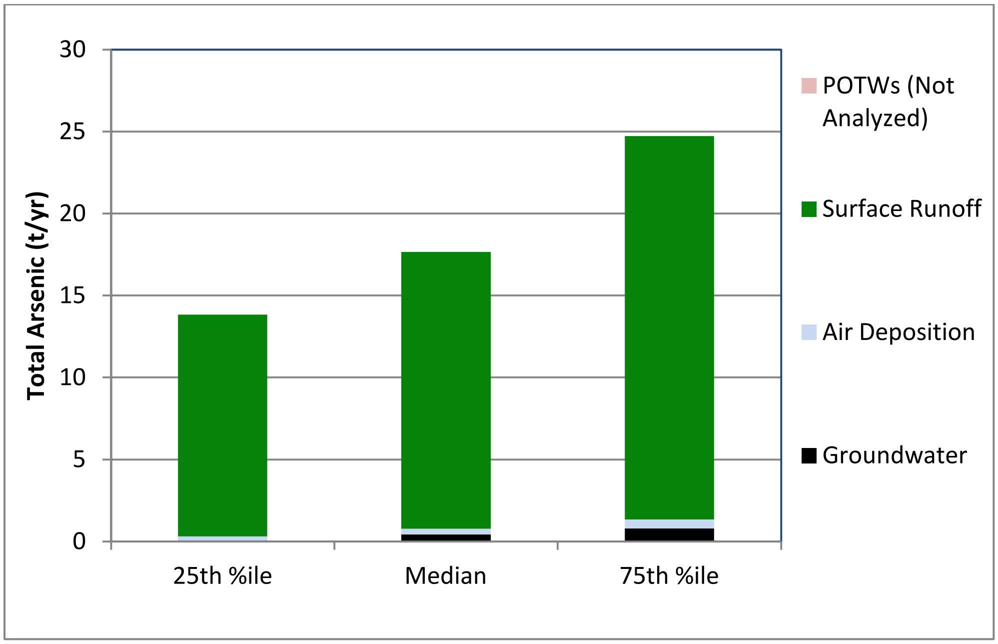


Figure 6. Total Arsenic Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of arsenic transport and fate following release suggests that much of the arsenic will be transported to Puget Sound through surface runoff. Direct deposition to marine waters from atmospheric deposition and loading through direct groundwater discharges appear to be only a small portion of overall loading.

Although not all anthropogenic sources of arsenic were assessed in estimates of primary releases, the total release of arsenic was only a small fraction (3 - 6%) of estimated loads delivered from major controllable pathways. Total arsenic initially emitted to the air (0.3 t/yr) is similar to air deposited on the marine waters (0.25 – 0.54 t/yr), although estimates for releases are for the entire Puget Sound basin and it is not known what portion of the air releases in the basin are transported out of the airshed and what portion of deposited arsenic is imported from outside airsheds.

The inventory of anthropogenic releases suggests that little arsenic is released directly to road surfaces or released through leaching by precipitation. Leaching of arsenic from CCA-treated wood, asphalt shingle roofs, and leaching or mobilization of arsenic-containing agricultural fertilizers only accounts for approximately 0.5 t/yr. In contrast, surface runoff loads range from 13.5 – 23.4 t/yr.

Arsenic loads in surface runoff during baseflow and storm flow conditions are roughly equal, and arsenic concentrations in streams from all land covers decline during storm events. This suggests little enrichment of streams by arsenic mobilized during storms. Instead, stormwater delivered to streams appears to simply dilute the baseflow levels of arsenic. This may indicate that arsenic originates primarily from groundwater.

Ongoing releases of anthropogenic sources of arsenic appear to be minimal, suggesting that a high proportion of the load is due to natural sources or historical releases which reside in the aquatic freshwater environment. Large historical sources of arsenic in the region include the Asarco Smelter in Tacoma which emitted arsenic for decades and may have resulted in large swaths of the Puget Sound area with elevated arsenic (PTI, 1991; San Juan, 1994).

Hazard Evaluation

The hazard evaluation indicates that arsenic is a Priority 2 or unknown level of concern for the categories assessed except for freshwater sediment (Table 2). There were few or no effects data available to adequately compare observed concentrations to surface water effects or human health criteria. Hazard due to tissue residue effects and effects to wildlife were not evaluated.

The limited set of freshwater effects for dissolved arsenic is three orders of magnitude above the 90th percentile value for dissolved arsenic in freshwater (approx. 2 ug/l) and two orders of magnitude below the chronic and acute water quality criteria.

Table 2. Summary of Hazard Evaluation for Arsenic.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,528	85%	No/INS	U
	Nearshore Marine	43	93%	INS	U
	Offshore Marine	58	91%	No/INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	623	82%	Yes	Priority 1
	Nearshore Marine	399	70%	No	Priority 2
	Offshore Marine	372	84%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	a	>99%	INS	U
	Nearshore Marine	a	100%	INS	U
	Offshore Marine	a	100%	INS	U

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

[a] N = 8 for freshwater fish, 11 for nearshore marine bivalves, 2 for nearshore marine invertebrates, 2 for offshore marine bivalves, and 0 for all other categories

Median arsenic concentrations in both freshwater and marine sediments are below the lowest guidelines or standards. However, in freshwater sediments the 90th percentile (approx. 40 mg/kg dw) of observed arsenic concentrations exceed the floating percentile SQS concentration. More than 25% of observed concentrations exceed the Canadian threshold effect level (TEL) and the consensus-based threshold effects concentration (TEC), more than 10% of values exceed the Canadian probable effects level (PEL) and the consensus-based probable effects concentration (PEC), and 5% of observed freshwater arsenic concentrations are at the floating percentile cleanup screening level (CSL).

In marine sediments, arsenic concentrations are nearly all (>95%) well below established guidelines and standards (including the SQS) except for the Canadian TEL which is exceeded by arsenic more than 25% of the observed concentrations in both nearshore and offshore sediments.

Arsenic hazards to human health were difficult to assess due to the paucity of data on inorganic arsenic, the form of arsenic for which the NTR has established criteria. No data were available for inorganic arsenic in edible freshwater fish or invertebrates. In marine waters, the few available data on inorganic arsenic in edible tissues (2 samples each for nearshore invertebrates and offshore fish, 11 samples for nearshore bivalves) all exceed (did not exceed) the NTR criteria. However, due to the small sample size available for assessment and lack of any data for one or more seafood categories, the overall human health level of concern for marine seafood was assigned an unknown level of concern.

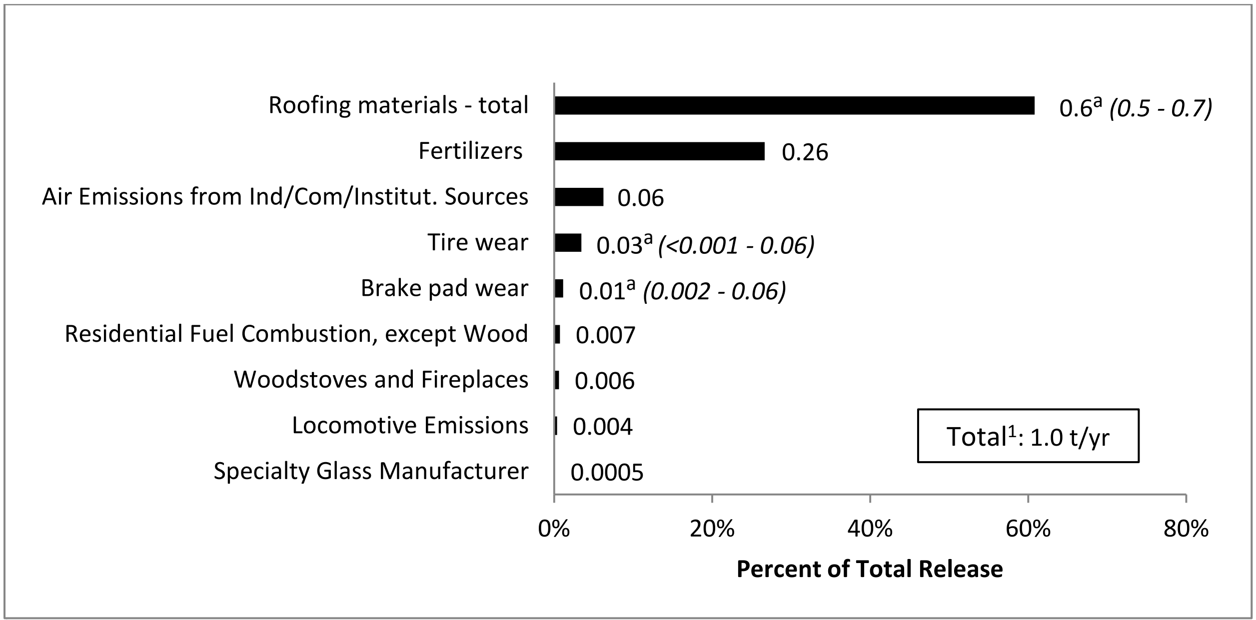
Cadmium

Cadmium is a heavy metal naturally occurring in the earth's crust and is nearly always detectable at concentrations >0.01 ug/l in water and >50 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Environmental levels of cadmium have been increased above natural levels due largely to manufacturing-related releases, combustion of fossil fuels, and the use of phosphorus fertilizers. Historically, the largest source of cadmium contamination was associated with waste from the now-defunct Asarco smelter in Tacoma (PTI, 1991).

Major Releases from Primary Sources

An inventory of primary cadmium sources suggests that approximately 1.0 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of cadmium to the environment appears to be leaching of cadmium from roofing material (Figure 7). Approximately 0.6 t/yr is released from rooftops, with 0.4 t/yr from asphalt composite shingles, 0.14 from built-up roofs, and a small fraction (0.03 t) from metal roofs.

Release from fertilizers accounts for most of the remainder of cadmium release (0.26 t/yr, 27% of total). Road-related sources such as tire and brake pad wear account for approximately 4% of the total release (0.04 t/yr). Comparatively little cadmium is released from industrial sources, with only a single facility reporting fugitive air releases during the previous ten years. Other air releases include locomotives and residential fuel use, but combined, these sources account for only about 6% of the total cadmium release.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mean

Figure 7. Total Cadmium Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for cadmium loading were obtained from PSTLA loading studies and are included in Table 3. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loading through other pathways was estimated from field studies specifically designed to estimate loads.

Table 3. Total Cadmium Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.012	0.22	0.43
Air Deposition	0.031	0.052	0.074
Surface Runoff (b)	0.01	0.01	0.02
POTWs	NA	NA	NA
Ocean Exchange	3.2	2.9	3.9

(a) Lowest, highest, and mid-point of estimated loads

(b) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows

NA=not analyzed

The estimated range of groundwater cadmium loads is large (0.012 – 0.43 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude. Cadmium was not measured in POTWs for the Phase 3 loading study. Phase 1 estimates suggest that cadmium loads through industrial wastewater may range from 0.02 to 0.9 t/yr (Hart Crowser et al., 2007), but this is based on a limited dataset and exceeds the industrial discharge of cadmium reported in the TRI by one to three orders of magnitude (Ecology, 2011). Phase 1 estimates for cadmium loading via municipal wastewater were based on a small set of non-detected sample results, and the load estimates appear to be small (1 – 4 kg/yr).

The deposition of atmospheric cadmium directly to the marine waters of Puget Sound is approximately 0.031 – 0.074 t/yr, based on fluxes ranging from 0.007 – 0.11 ug/m^2/d. Median cadmium fluxes were generally <0.02 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Cadmium fluxes at this location were elevated three- to four-fold above fluxes at other locations.

Surface runoff loads for cadmium are estimated to be approximately 0.01 – 0.02 t/yr for the entire Puget Sound basin based on the surface runoff study. Total cadmium was not detected in baseflow samples, although dissolved cadmium was detected in 13% of baseflow samples. Dissolved cadmium was also detected at a higher frequency than total cadmium in storm flow samples (34% and 8%, respectively). In either case, cadmium was detected at such low rates that estimates to calculate loads in surface waters were driven by the analytical reporting limits, and no estimate was derived for baseflow loads.

No cadmium was detected in surface runoff from residential or forested areas. Total cadmium was only detected in commercial/industrial areas – largely as a result of a seasonal first-flush episode – but the overall low frequency of detection was low (27%). However, dissolved cadmium was detected at a high rate in commercial/industrial areas (87%) and a more moderate rate in agricultural land covers (34%).

Overall, there appears to be a large net import of cadmium through exchange at the ocean boundary compared to other load pathways, although this estimate is based on very limited data. Total cadmium concentrations (25th -75th percentile) in the incoming marine waters are 0.091 – 0.097 ug/l, and total cadmium concentrations in the outgoing marine waters are 0.072 – 0.077 ug/l. Total net import was 3.2 – 3.9 t/yr based on an inflow of 53 – 57 t/yr and an outflow of 50 – 53 t/yr.

The net sum of cadmium loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 3.3, 3.0, and 4.4 t/yr, respectively. Under all of these estimates, there is a net cadmium load to Puget Sound.

Cadmium loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th -75th percentiles) and median for each pathway in Figure 8. Each pathway represented as a contribution to the total load is displayed in Figure 9.

Total cadmium loading from the major pathways assessed is estimated to be 0.05 – 0.53 t/yr. Groundwater potentially accounts for the largest loading pathway, at 24 – 82% of the total, but the groundwater numbers should be viewed with caution since they are driven largely by analytical reporting limits rather than measurable sample concentrations. The comparatively small contribution from surface runoff (4% – 15% of total) is also derived by non-detected values. Since the groundwater loading estimates are highly variable and load estimates at the low end of the range are small (0.012 t/yr), atmospheric deposition potentially represents the largest loading pathway (up to 62% of total load) if all estimates are assumed to be at the low end of the range.

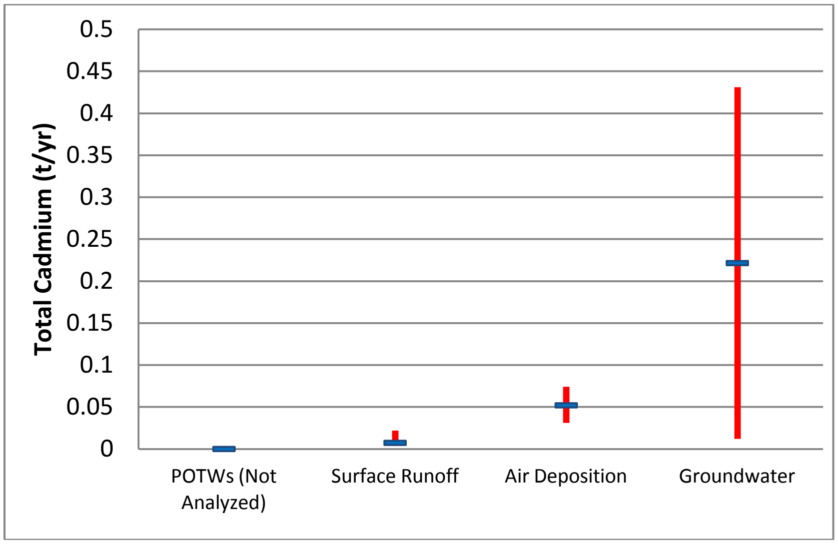


Figure 8. Range (25th - 75th percentiles) and Median Total Cadmium Loads to Puget Sound from Each Major Delivery Pathway.

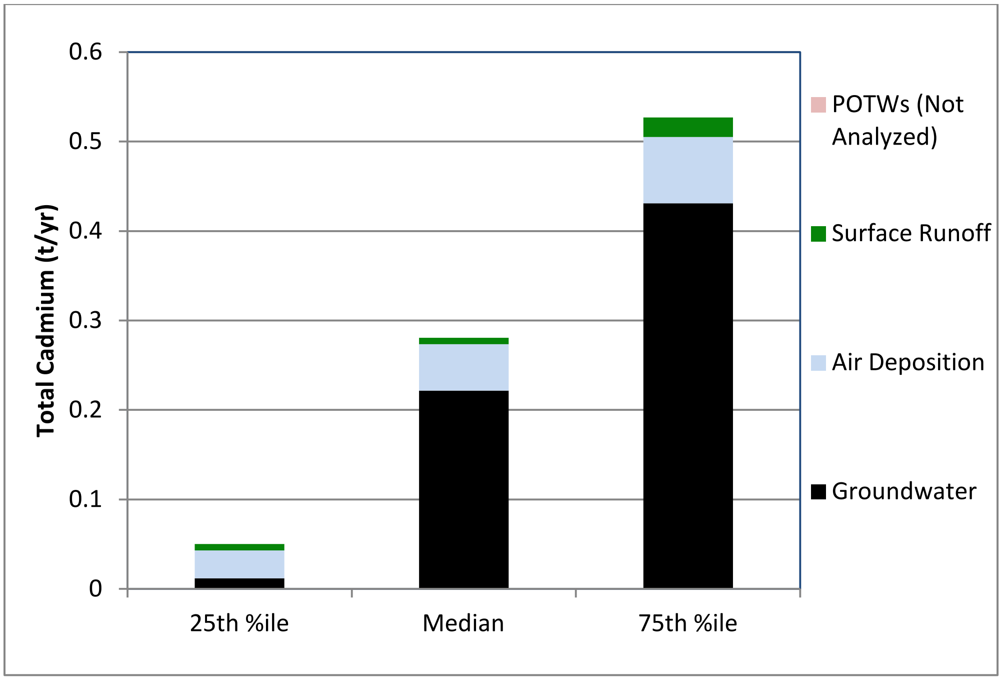


Figure 9. Total Cadmium Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of cadmium transport and fate following release is limited by a lack of data on cadmium in major loading pathways. The available information suggests that most of the anthropogenic cadmium released from primary sources – leaching from roofing material and fertilizers – would be initially mobilized in surface runoff. This may be reflected in the presence of cadmium in storm samples and a lack of cadmium in baseflow samples, but the data are too limited to establish any patterns with even moderate confidence.

The combined loading of cadmium from major controllable pathways (0.050 – 0.53 t/yr) is approximately 20 times lower than releases from all primary sources combined. Air releases of cadmium to the watershed (0.06 t/yr) are not substantially different than atmospheric deposition in marine waters (0.03 – 0.07 t/yr), although marine areas represent only about one-sixth of the watershed. It is not known what portion of the air releases in the basin is transported out of the airshed and what portion of deposited cadmium is imported from outside airsheds.

Hazard Evaluation

The hazard evaluation indicates that cadmium is a Priority 2 or unknown level of concern for the categories assessed except for freshwater sediment (Table 4). Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved cadmium in freshwater (approx. 0.3 ug/l) are one-half of the concentrations in freshwater where 10% of effects have been documented, and are below the chronic and acute water quality criteria. However, more than 5% of the observed cadmium concentrations in freshwater are above the chronic water quality criterion, and at least 10% of the observed values are above concentrations where 5% or more of the effects have been documented.

Despite enrichment from oceanic waters, the gap between observed cadmium concentrations and effects levels or criteria is much larger for marine waters than for freshwater. The 90th percentiles of dissolved cadmium concentrations in both nearshore and offshore marine waters (approx. 0.09 ug/l and 0.06 ug/l, respectively) are two orders of magnitude below the lowest 5-10% of effects as well as the acute and chronic water quality criteria. However, the limited dataset (n<50) for dissolved cadmium nearshore and offshore marine waters does not meet the criteria established for an adequate comparison, and this evaluation was assigned an unknown level of concern.

Median cadmium concentrations in both freshwater and marine sediments are below the lowest guidelines or standards. However, in freshwater sediments the 75th percentile (approx. 0.9 mg/kg dw) of observed cadmium concentrations exceed the floating percentile SQS concentration, and more than 10% of the observed values exceed the floating percentile CSL.

Cadmium concentrations in marine nearshore and offshore sediment have 90th percentile levels (approx. 1.0 mg/kg dw) one-fifth the SQS. However, the observed concentrations for both nearshore and offshore sediments exceed the Canadian TEL at the 75th percentile levels, and 5% of the nearshore concentrations are at the Lowest Apparent Effects Threshold (LAET).

Table 4. Summary of Hazard Evaluation for Cadmium.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,166	7%	No	Priority 2
	Nearshore Marine	32	100%	INS	U
	Offshore Marine	42	100%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	764	67%	Yes	Priority 1
	Nearshore Marine	462	62%	No	Priority 2
	Offshore Marine	471	70%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

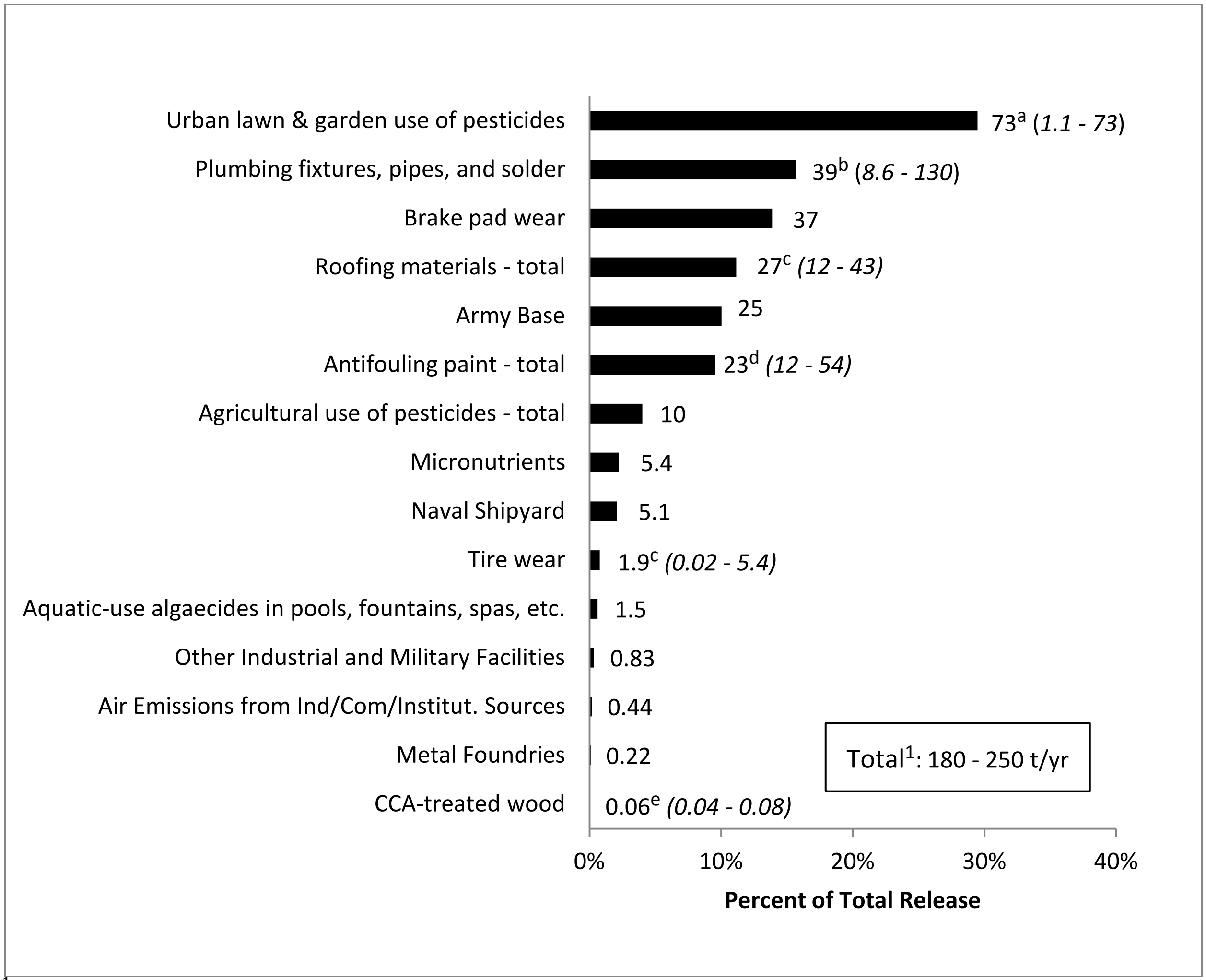
Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

Copper

Copper is a heavy metal naturally occurring in the earth's crust and is nearly always detectable at concentrations >0.5 ug/l in water and >3,000 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Copper is an essential element for all living organisms and is generally not toxic to humans and terrestrial wildlife at typical environmental concentrations. However, copper can be highly toxic to aquatic organisms at low concentrations.

Major Releases from Primary Sources

An inventory of primary copper sources suggests that approximately 180 – 250 metric tons (t) is released from anthropogenic sources annually in the Puget Sound basin. The largest current source of anthropogenic copper to the environment could potentially be due to urban lawn and garden use (Figure 10). Copper use in urban landscaping as an herbicide/fungicide or possibly as a micronutrient accounts for approximately 73 t/yr by some estimates, but may be as little as 1 t/yr based on other estimates. The authors of the *Sources Report* note the difficulty in obtaining accurate information on pesticide use and note that these data should be viewed with caution since they were not derived from market data, and use rates are difficult to estimate without this information (Ecology, 2011). Estimates of copper used as an agricultural pesticide and micronutrient in the Puget Sound basin are probably more accurate than estimates for urban use, but agricultural use of copper only represents 4% – 6% of the total annual release.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] High end of range

[b] Median

[c] Average

[d] Sum of means for recreational and commercial and mid-point for naval vessels

[e] Mid-point of range

Figure 10. Total Copper Release in the Puget Sound Basin (values shown are t/yr).

Other major releases of copper are leaching from plumbing components (39 t/yr), vehicle brake pad and tire wear (37 t/yr and 2 t/yr, respectively), and leaching from vessel anti-fouling paint (26 t/yr). Industrial, commercial, and institutional facilities are estimated to release a total of 31 t/yr of copper, with approximately four-fifths reported from activities at the Fort Lewis Army Base, possibly as solid copper from munitions use.

Smaller releases of copper also occur following the use of copper compounds in fountains and spas as an algaecide (1.3 t/yr), and from leaching of copper from CCA-treated wood (0.05 t/yr). There are no currently permitted uses of copper as an aquatic herbicide/algaecide in surface waters of the Puget Sound basin.

Major Delivery Pathways and Loading

Data on major delivery pathways for copper loading were obtained from PSTLA loading studies and are included in Table 5. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 5. Total Copper Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.099	2.19	4.27
Air Deposition	1.9	2.7	4.1
Surface Runoff	28.4	35.7	66.1
POTWs	2.5	4.33	5.5
Ocean Exchange (b)	-110	-100	-30

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary

Estimated groundwater loads of copper range by an order of magnitude (0.1 – 4.3 t/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Copper discharged from POTWs in the Puget Sound basin accounts for loads estimated to be 2.5 – 5.5 t/yr. Phase 1 estimates suggest that copper loads from industrial wastewater are potentially substantial (6 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of copper to surface waters or transferred to POTWs as reported in the TRI (<0.7 t/yr total) (Ecology, 2011).

The deposition of atmospheric copper directly to the marine waters of Puget Sound is approximately 1.9 – 4.1 t/yr, based on fluxes ranging from 0.5 - 19 ug/m^2/d. Copper fluxes were generally <2 ug/m^2/d, except for one location within a high-density urban area (Tacoma) including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Copper fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated five-fold above a nearby station that did not have the same air pollution influences in such close proximity.

Surface runoff loads for copper are estimated to be approximately 28.4 – 66.1 t/yr for the entire Puget Sound basin based on the surface runoff study. Overall, dissolved copper accounts for 63 – 76% of total copper concentrations in surface water, with the higher proportion of dissolved copper occurring during baseflows.

Total copper concentrations were highest in agricultural areas, whereas dissolved copper concentrations were similar in agricultural and commercial/industrial area. In agricultural, commercial/industrial, as well as forested areas, elevated copper concentrations appeared to correspond with elevations in TSS. The apparent links with TSS in forested sub-basins may be related to a substantial seasonal first-flush episode in forests. However, there appeared to be little overall increase in forest copper concentrations during storm events, whereas the median total copper concentrations increased by two- to three-fold during storms for all other land covers.

Based on sampling marine waters, there appears to be a large net export of copper at the ocean boundary, although this estimate is based on very limited data. Total copper concentrations (25th -75th percentile) in the incoming marine waters are 0.24 – 0.41 ug/l, and total copper concentrations in the outgoing marine waters are 0.38 – 0.46 ug/l. Total net export was 30 – 110 t/yr based on an inflow of 140 – 240 t/yr and an outflow of 250 – 270 t/yr.

The net sum of copper loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is -77, -55, and 50 t/yr, respectively. Under the 25th percentile and median estimates, there is a net export of copper out of Puget Sound due to the large mass exchanged at the ocean boundary. However, when the 75th percentile values are summed, the large surface water load outweighs the smaller export at the ocean boundary, leading to a net load of copper load to Puget Sound.

Copper loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th -75th percentiles) and median for each pathway in Figure 11. Each pathway represented as a contribution to the total load is displayed in Figure 12.

Total copper loading from the major pathways assessed is 33 – 80 t/yr. Surface water runoff accounts for the largest pathway (83 – 86%), followed by POTWs (7 – 8%) and air deposition (5 – 6%). Groundwater potentially accounts for up to 5% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of copper in wells and rough estimates of groundwater flow. At the lower end of the load range, copper in groundwater is estimated to account for <1% of the load to Puget Sound.

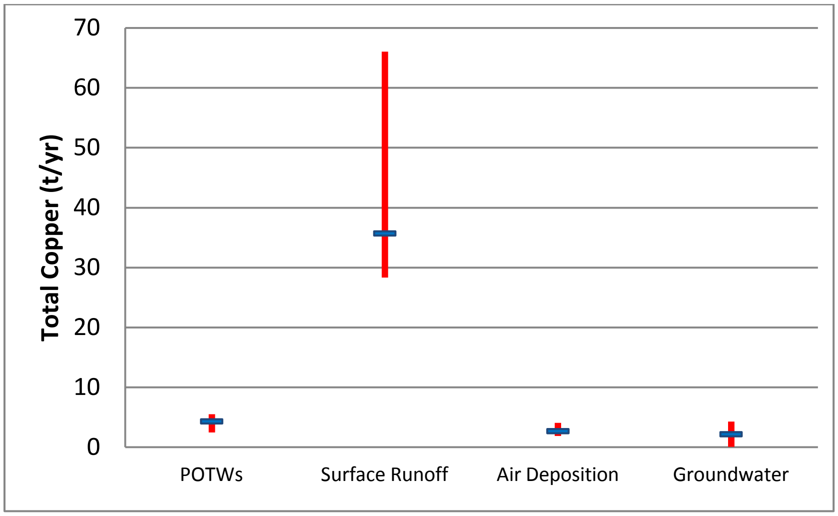


Figure 11. Range (25th - 75th percentiles) and Median Total Copper Loads to Puget Sound from Each Major Delivery Pathway.

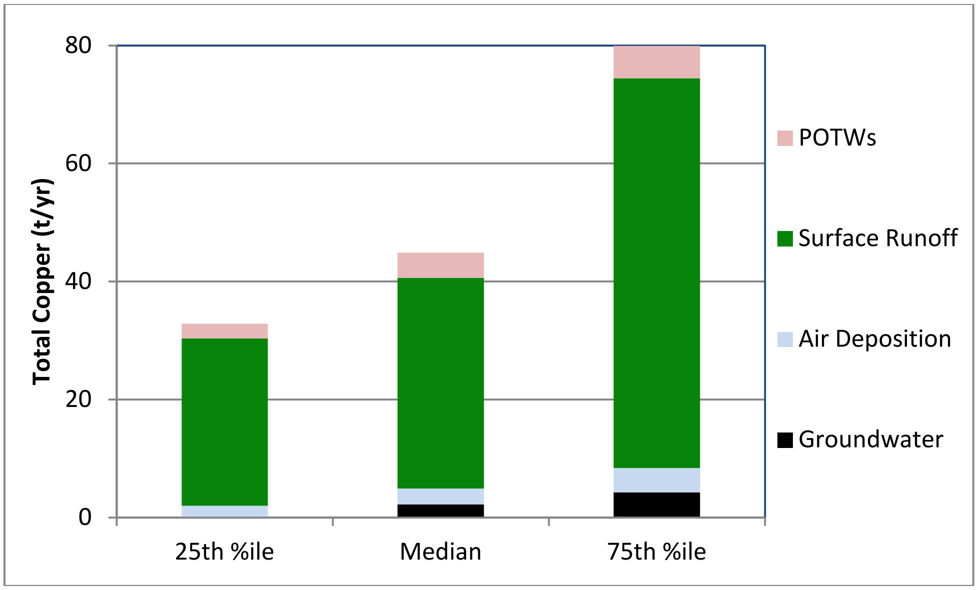


Figure 12. Total Copper Loads to Puget Sound as the Sum of Major Delivery Pathways

Relationship Between Sources and Pathways/Loading

The conceptual model of copper transport and fate following release suggests that much of the copper transported to Puget Sound will occur through surface runoff during storm events. Direct deposition to marine waters from atmospheric deposition and loading through direct groundwater discharges appear to be only a small portion (<15%) of overall loading. The high proportion of copper loading contributed by surface runoff is consistent with the types of sources accounting for the major copper releases.

Major unconstrained releases of copper are potentially from its use in urban landscaping and pesticides, leaching from rooftops, and from vehicle component wear. Together, these mostly urban sources account for as much as 140 t/yr of copper released to the environment in the Puget Sound basin.

It is difficult to draw conclusions regarding the fate of copper released from urban landscaping use and pesticide use. For instance, copper may be highly soluble depending on the formulation applied, and it may be released in dissolved form during storms or irrigation. However, since copper is likely to be used on recently disturbed soil in many instances, release through mobilization of particle-bound copper may also occur. This latter form of release is less likely to be infiltrated into groundwater, a seemingly distinct possible fate for copper solubilized in permeable soil. At the lower estimate of copper pesticide use in urban areas (1 t/yr), the comparatively low rate of use may be much more important locally than on a basin-wide scale.

Copper released from rooftops is likely to be in dissolved form since leaching is the likely release mechanism, but anticipating its pathway following initial release is difficult since it may run along roads and other impervious areas to surface waters, while some may be infiltrated into the soil or delivered to storm sewers and thereafter POTWs.

Brake pad and tire wear may result in a relatively high proportion of copper finding its way to surface runoff since approximately 50% is transported off-roadway as fugitive dust (Sinclair-Rosselot, 2006), although the fate of brake pad dust may vary greatly due to local conditions (Paulson et al., 2011-Draft). This and the possible attenuating factors discussed previously for copper initially released in water, mobilized by water, or released to an impervious surface such as a roadway likely account for much of the difference between amounts released from the unconstrained urban sources mentioned above (140 t/yr) and the amounts loaded to Puget Sound via surface runoff (28 – 66 t/yr).

The differences in copper releases from the unconstrained urban sources and the surface runoff on the regional scale are much smaller than the differences between releases and loads calculated during the mass-balance analysis of two small urban King County watersheds conducted by Paulson et al. (2011-Draft). They calculated that the annual copper mass discharged from these watersheds were less than 10% of the copper estimated to be released just from vehicle component (brake pad and tire) wear and roof runoff. At the much larger scale (entire Puget Sound basin), the high end (75th percentile) estimate of copper discharged to Puget Sound through surface water is equal to the mass of copper released just from vehicle wear and rooftops (66 t/yr). This suggests that while the conceptual relationships between copper sources and loading/pathways may be valid, these relationships may be much more complex and uncertain at finer scales.

Copper deposited atmospherically is difficult to link with its primary source(s). Copper released from stack air emissions is small compared to overall loads (<1%), and copper reported as fugitive air releases in the TRI accounts for only about 2% of the total annual release, for a total maximum release to air of approximately 6 t/yr to the Puget Sound basin. This is higher than the range of copper deposited directly to marine water from the atmosphere (1.9 – 4.1 t/yr), but the latter only accounts for one-sixth of the basin area. However, copper released as fugitive dust from brake pad wear is potentially substantial (>17 t/yr). This may account for the high levels of copper in atmospheric deposition samples at the high-density urban location, which is located near Interstates 5 and 705, associated interchanges, and other major roadways. Copper fluxes at this location were an order of magnitude higher than at most other locations, and the inventory of primary releases indicates that industrial sources may not be large contributors to airborne emissions. Copper fluxes at a location nearby but removed from close-proximity industrial and roadway influences were one-fifth those in the high-density urban area. This indicates that urban locations have strongly localized high copper flux signals, which are not reflected to the same degree in regional elevations of copper.

Disposal of copper at the Ft. Lewis Army Base (25 t/yr) presumably consists of solid copper material placed in waste piles or scattered throughout firing ranges. Solid copper disposal in the terrestrial environment will be retained in a soil reservoir and will only be transported to surface waters through gradual erosion or storm events with high energy flows.

Copper leached from vessel anti-fouling paint (26 t/yr) is presumed to be released entirely to marine waters with the possible exception of the Lake Washington/Lake Union system and freshwater marina areas near the mouths of the Snohomish and Duwamish Rivers.

Of the primary sources inventoried, copper released from residential plumbing components represents the only constrained source assessed. The annual load of copper discharged to POTWs from this source is estimated to be 28 t; discharges to septic systems were estimated to be an additional 11 t/yr (Ecology, 2011). The difference between the amount released and loading from POTWs (2.5 – 5.5 t/yr) is presumably due to the removal of solid material during the treatment process, an assumption that might be easily checked by conducting sampling of representative sludge material. Of course, copper from other constrained sources, naturally occurring copper in water, and copper in stormwater represent additional releases of copper to POTWs, but the exact extent of these contributions are not known.

Although patterns in copper loading pathways appear to be consistent with the types of primary sources, one confounding result is the high concentrations of copper in agricultural areas compared to other land covers. Overall copper releases as agricultural pesticides and micro-nutrients appear to be small (15.3 t/yr combined) compared to the other unconstrained releases previously mentioned, most of which are expected to occur in commercial/industrial and residential areas.

The high concentrations cannot simply be explained by excessive soil erosion in agricultural areas during storms, a scenario that would be expected to cause waters to become enriched with particle-bound copper. While copper in agricultural streams experienced a three-fold increase in concentration during storms, increases in TSS loads were lowest among all land covers (Herrera, 2011). One possible explanation for relatively high copper levels in agricultural streams may simply be that the form(s) of copper used and methods of application in agricultural settings are particularly prone to result in copper migration to surface waters.

Hazard Evaluation

The hazard evaluation indicates that copper is a Priority 1 level of concern for freshwaters, nearshore marine surface waters, and freshwater sediments (Table 6). Copper is a Priority 2 level of concern for offshore marine surface waters and marine sediments. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

Table 6. Summary of Hazard Evaluation for Copper.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	5,378	92%	Yes	Priority 1
	Nearshore Marine	107	100%	Yes	Priority 1
	Offshore Marine	71	100%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	826	>99%	Yes	Priority 1
	Nearshore Marine	519	>99%	No	Priority 2
	Offshore Marine	560	98%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

Dissolved copper concentrations in freshwater have a 90th percentile value of approximately 3.5 ug/l, higher than the level where more than 10% of effects documented for aquatic organisms occur and above the chronic water quality criterion (calculated at 25 mg/l calcium carbonate). At least 5% of the concentrations are above the acute water quality criterion (also calculated at 25 mg/l calcium carbonate).

In marine waters, nearshore concentrations of dissolved copper are distinctly higher than those observed offshore. At the 90th percentile level, the nearshore concentration (approx. 5 ug/l) is more than double the offshore concentration (approx. 2 ug/l) and exceeds both the chronic and acute water quality criteria. The upper levels (90th – 95th percentile values) of both the observed nearshore and offshore dissolved copper levels approach or exceed the 10th percentile values) of the ECOTOX dataset used for these comparisons, but only the observed nearshore data have 90th percentile values that exceed this threshold for assigning a Priority 1 level of concern.

Median copper concentrations in both freshwater and marine sediments are below all guidelines and standards except the marine Canadian TEL. However, in freshwater sediments the 90th percentile (approx. 90 mg/kg dw) of observed copper concentrations exceed the floating percentile SQS as well as the Canadian TEL and the consensus-based TEC. More than 5% of the observed freshwater sediment values exceed the Canadian PEL and the consensus-based PEC for copper.

In marine sediments, the 90th percentiles of the observed copper concentrations (approx. 90 mg/kg dw for nearshore and 70 mg/kg dw for offshore) are similar to those for freshwater sediments, but the marine SQS is much higher than the freshwater floating percentile SQS. As a result, all but possible outlier concentrations exceed the SQS in marine sediments.

Regionally Important Biological-Effects data

Copper is one of the most far-reaching potential priority toxicants in the Puget Sound region due largely to its ability to alter the sensory capacity and behavior of a wide variety of aquatic organisms. A number of local researchers have documented these effects in different organisms and in different ways.

Tierney et al. (2010) reviewed over 150 papers and found that avoidance behaviors were common in a variety of fresh and salt water fishes at less than 1 ug/l to concentrations ranging up to 20-30 ug/l. Tested species included coho and Chinook salmon as well as rainbow trout and golden shiner. Hecht et al. (2007) compiled a similar body of evidence for the disruptive effects of copper on juvenile salmonids. They used EPA methodologies to calculate benchmark concentrations predicted to represent 10% and 50% reductions in chemosensory response at 0.18 ug/l and 2.1 ug/l respectively. These values bracket a variety of other regional primary literature sources which confirm that the environmentally relevant range of <1.0 to 5.0 ug/l copper adversely impacts a variety of Puget Sound basin fish, particularly salmonids. Similar neurologic impacts were found by Linbo et al. (2006) on the mechanosensory lateral line of fish.

Sandahl et al. (2004) found copper concentrations of 4.4 ug/l produced sublethal neurotoxicity in coho salmon. In this laboratory study, copper reduced the ability of coho salmon to detect the natural odorants taurocholic acid and L-serine. Further study by Sandahl et al. (2007) confirmed that concentrations as low as 2 ug/l copper not only affect the neurologic systems of fish but also alter their behavioral responses to alarm pheromones. Other studies such as Baldwin et al. (2003) have also found olfactory inhibition at a comparable environmentally relevant concentration of 2.3 ug/l.

Hansen et al. (1999) produced a seminal work which was used by several of the subsequent reviews discussed above. In it they documented Chinook salmon avoidance behaviors at concentrations as low as 0.7 ug/l dissolved copper. However, Chinook also failed to avoid concentrations >44 ug/l due to the extensive neural saturation. This window of effect potentially contributes to mortality from prolonged copper exposure or impairment of olfactory dependent behaviors such as homing. Additional studies by McIntyre et al. (2008) found that water hardness had very little effect on copper's ability to alter olfactory function in coho salmon despite water hardness being a variable influencing the Washington State water quality criteria. All of these reviews and studies on regionally relevant species provide an additional line of evidence suggesting that copper is a very important toxicant at concentrations well within the range found it the Puget Sound regional environment.

Lead

Lead is a naturally occurring heavy metal that is a major constituent of more than 200 minerals and is nearly always detectable at concentrations > 0.01 ug/l in water and >100 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Meredith and Furl, 2009). Lead also accumulates in fish, particularly in bony material, but does not biomagnify to any meaningful extent (Eisler, 1988b).

Although lead occurs naturally in the earth's crust, human activity has increased environmental levels by one-thousand-fold during the past three centuries (ATSDR, 2007). Most recently, the use of tetra-ethyl lead in gasoline ("leaded gasoline") accounted for hundreds of millions of pounds of lead per year in the U.S. alone before being phased out from 1973 – 1996; by 1995, lead emissions from gasoline were estimated to be less than 1% of the level prior to initiation of the phase out (EPA, 1996).

Historically, concerns surrounding the effects of lead have been focused on toxicity to humans, particularly children. Ecology and WDOH (2009) have reviewed the history, exposure pathways, and effects of lead on humans in Washington. Environmental effects, particularly in the aquatic environment, are less apparent.

Major Releases from Primary Sources

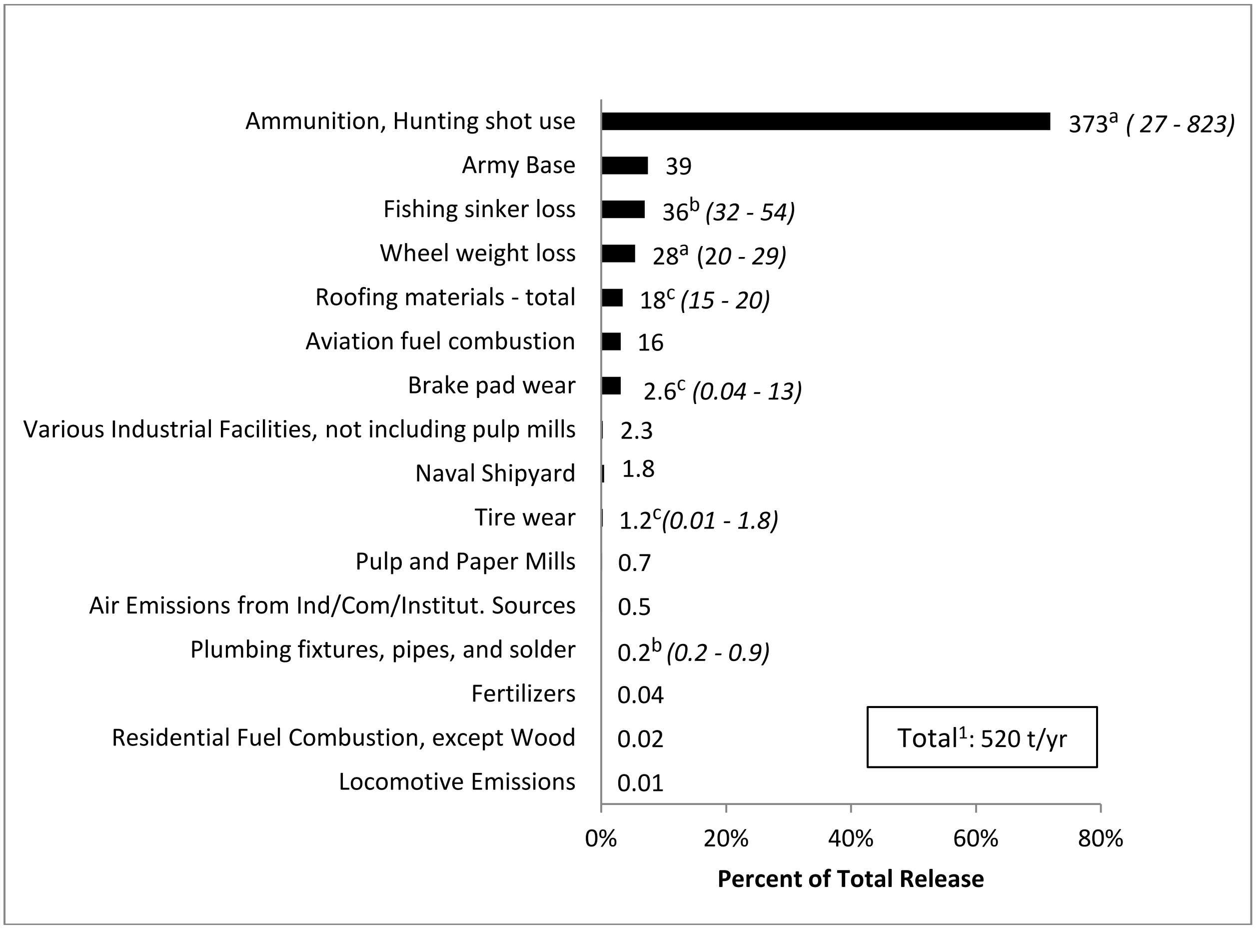
An inventory of primary lead sources suggests that approximately 520 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of lead to the environment is the use of ammunition and lead shot (Figure 13). Together with lost wheel weights, fishing sinkers, and a self-reported release at Fort Lewis presumed to be from ammunition use, approximately 90% (480 t) of the anthropogenic releases of lead in the basin may be as solid metallic lead in bulk form.

Other sources of lead include leaching from materials such as roof runoff (18 t/yr) and abrasion of vehicle brake pads and tires (4 t/yr combined). Although these releases are small compared to ammunition use, they represent the most likely sources of lead to be mobilized in stormwater following release. To be more precise, lead in roof runoff requires rain storms for its release, and approximately 50% of brake pad particles are released to the road surface (Garg et al., 2000;

Sinclair- Rosselot, 2006) and may be subsequently entrained in stormwater (depending on season and conditions). Tire particles are presumably released mainly to road surfaces, although some portion becomes transported away from the release point as fugitive dust.

Additional releases of lead include emissions from combustion of aviation fuel (16 t/yr) and approximately 0.5 t/yr released to the air from point sources; pulp mill emissions appear to make up the bulk of this latter category.

Other smaller releases include approximately 1 t/yr released from residential plumbing components, and 0.040 t/yr (40 kg/yr) released through fertilizer application.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Most reasonable estimate

[b] Median

[c] Mean

Figure 13. Total Lead Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for lead loading were obtained from PSTLA loading studies and are included in Table 7. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 7. Total Lead Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.044	1.07	2.10
Air Deposition	0.63	1.1	1.5
Surface Runoff	2.80	4.67	7.64
POTWs	0.14	0.18	0.25
Ocean Exchange	21	21	18

(a) Lowest, highest, and mid-point of estimated loads

Estimated groundwater loads of lead range by two orders of magnitude (0.044 – 2.1 t/yr). The range in estimates is due to differences in flows used to establish the possible range of loads as well as differences in methods used to estimate representative lead concentrations; most of the data were non-detected values.

Lead discharged from POTWs in the Puget Sound basin accounts for loads estimated to be 0.14 – 0.25 t/yr. Phase 1 estimates suggest that lead loads from industrial wastewater are potentially substantial (0.3 – 9 t/yr; Hart Crowser et al., 2007). This is based on a limited dataset yet brackets the estimate of the industrial discharge of lead to surface waters or transferred to POTWs as reported in the TRI (1.4 t/yr total) (Ecology, 2011).

The deposition of atmospheric lead directly to the marine waters of Puget Sound is approximately 0.63 – 1.5 t/yr, based on fluxes ranging from 0.15 - 8.6 ug/m^2/d. Lead fluxes were generally <1 ug/m^2/d, except for one location within a high-density urban area (Tacoma) including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Lead fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated five-fold above a nearby station that did not have the same air pollution influences in such close proximity

Surface runoff loads for lead are estimated to be approximately 2.8 - 7.6 t/yr for the entire Puget Sound basin based on the surface runoff study. Dissolved lead accounted for only a small portion of the total lead concentrations, and elevated concentrations of total lead appeared to correspond with elevated TSS (Herrera, 2011). Lead has a high affinity for particulate matter and is generally found at low proportions in the dissolved phase (Meredith and Furl, 2009; Hallock, 2010). Lead is therefore much more likely to be transported as particle-bound lead rather than in the dissolved phase.

The ratio of lead concentrations in storm flows-to-baseflows was consistently higher for lead than for other COCs analyzed. Lead in storm flows was higher compared to baseflows by factors of 3 to 6 depending on land cover (commercial/industrial was highest). This is consistent with particle-bound constituents which require storm flows to become mobilized.

Total lead was present at the highest concentrations in commercial/industrial land covers, generally by factors of 2 to 5. Overall median total lead concentrations in commercial/industrial areas increased by six-fold during storm events, while only increasing by factors of 2 to 3 in other land cover types. Although elevations in lead during storm flows were most pronounced in commercial/industrial areas, seasonal first-flush signals were only evident in residential (dissolved lead) and forest sub-basins (total lead).

Like cadmium, there appears to be a large import of lead through exchange at the ocean boundary compared to other load pathways, although this estimate is based on very limited data. Total lead concentrations (25th-75th percentile) in the incoming marine waters are 0.087 – 0.125 ug/l, and total lead concentrations in the outgoing marine waters are 0.047 – 0.087 ug/l. Total net import was 18 – 21 t/yr based on an inflow of 51 – 73 t/yr and an outflow of 30 – 55 t/yr.

The net sum of lead loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 25, 28, and 30 t/yr, respectively. Under all of these estimates, there is a net lead load to Puget Sound.

Lead loadings from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 14. Each pathway represented as a contribution to the total load is displayed in Figure 15.

Total lead loading from the major pathways assessed is 3.6 – 12 t/yr. Surface water runoff accounts for the largest pathway (66 – 77%), followed by air deposition (13 – 17%). Groundwater potentially accounts for up to 18% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of lead in wells and rough estimates of groundwater flow. At the lower end of the load range, lead in groundwater is estimated to account for 1% of the load to Puget Sound. Loading through POTWs represents only a small portion of total lead loads to Puget Sound (2 – 4%).

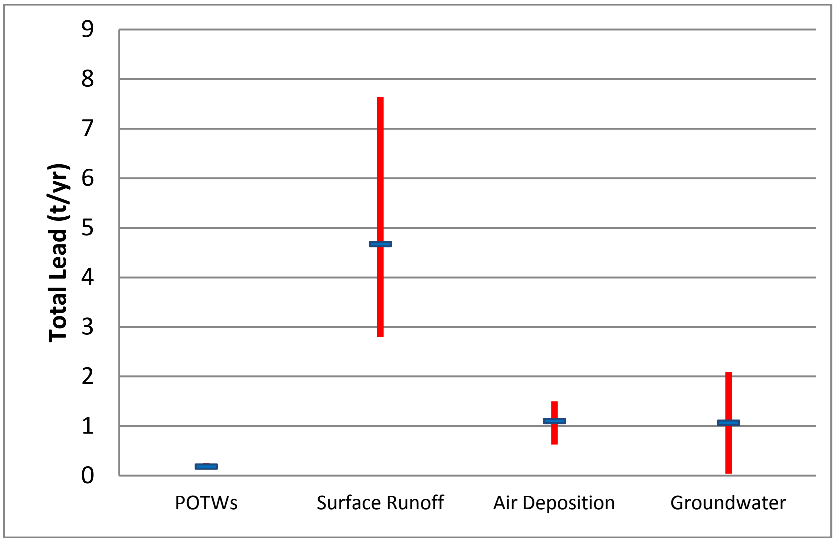


Figure 14. Range (25th - 75th percentiles) and Median Total Lead Loads to Puget Sound from Each Major Delivery Pathway.

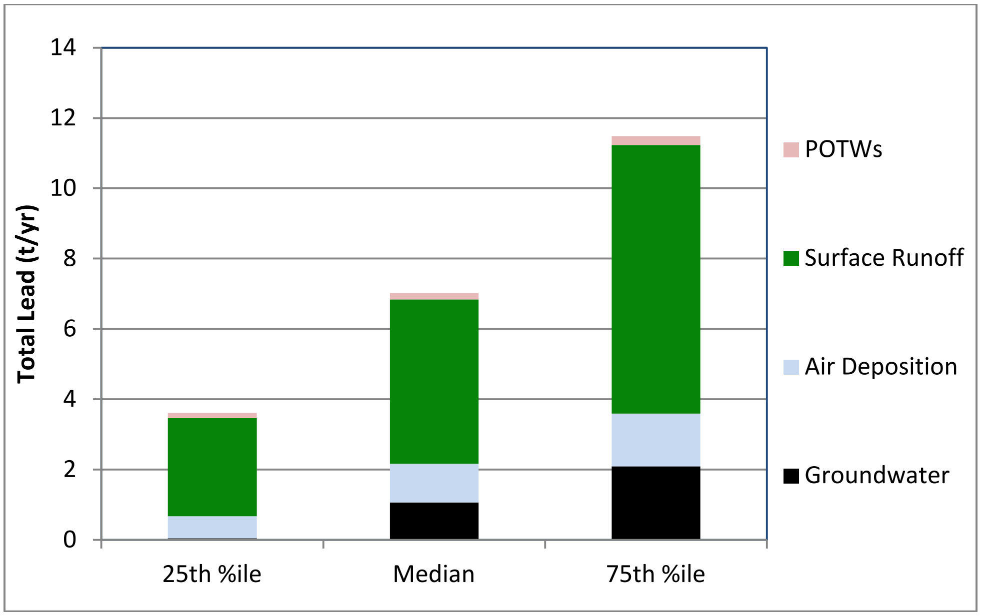


Figure 15. Total Lead Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of lead transport and fate following release suggests that much of the lead transported to Puget Sound will do so through surface runoff during storm events. Lead released to road surfaces through roof runoff, lost wheel weights, and tire and brake pad wear appears to represent the largest release to road surfaces. Combined, these account for an estimated 51 t/yr of lead released, or about 7-18 times the estimated lead load in surface runoff. These differences can be explained largely by the source for lead released to road surfaces. For instance, not all rooftop runoff will be delivered to roadways; some may be infiltrated or delivered to storm sewers and thereafter to POTWs. Lost wheel weights will largely remain in bulk metallic form and only gradually become pulverized and leached. Brake pad and tire wear may represent the lead source with the highest proportion of lead to find its way to surface runoff, even though a substantial fraction may be transported off-roadway as fugitive dust.

Lost fishing sinkers and lead shot and other ammunition landing in surface waters will gradual leach lead to the aquatic environment, although this likely results in only small increases in water column concentrations. Lead ammunition landing in the terrestrial environment will likely be retained in a soil reservoir and will only be transported to surface waters through gradual erosion or storm events with high energy flows. In general, lead from ammunition will be retained in the soil and not pose a problem to surface waters, with the exception of shooting ranges which may accumulate large enough masses of lead to impact nearby waterbodies. Two instances of high lead concentrations in streams and soils resulting from spent lead at shooting ranges have been documented recently in western Washington (Era-Miller, 2009; Ecology and WDOH, 2009).

The ultimate fate of lead emitted to air is less certain; only a small fraction of the estimated releases to air are deposited directly to the waters of Puget Sound, and it is not known what portion of the air releases in the basin are transported out of the airshed and the portion of deposited lead that is imported from outside airsheds. Approximately 19 t/yr are released to the air within the Puget Sound basin (not counting fugitive brake pad and tire dust) compared to 0.6 – 1.5 t/yr directly deposited to marine waters. Considering the area of the entire Puget Sound basin (six times the marine area) and assuming the same rate of deposition across the basin (an assumption that has not been verified), the ranges of air deposition fall within a factor of 2 compared with lead emissions to air. Atmospherically deposited lead would presumably be washed off surfaces in runoff – most likely in particulate form – and ultimately be transported to the Puget Sound through surface runoff, although infiltration to groundwater and paths to POTWs are other possible scenarios for secondary and tertiary pathways.

Although comparatively small amounts of lead are released in constrained sources, release of lead from residential plumbing fixtures (0.2 t/yr) almost certainly is received at POTWs. Annual loads of lead released to POTWs from this source appear to be nearly identical to the amount discharged from POTWs. Presumably, removal of lead would occur during the removal of solid material during the treatment process. Lead from other constrained sources, naturally occurring lead in water, and lead in stormwater represent additional releases of lead to POTWs, but the extent of these contributions are not known.

Hazard Evaluation

The hazard evaluation indicates that lead is a Priority 2 or unknown level of concern for all categories assessed (Table 8). Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved lead in freshwater (approx. 0.5 ug/l) are an order of magnitude below the 10th percentile of effects data and the acute water quality criterion, although the values are only slightly less than the chronic water quality criterion. In marine waters, the gap between observed concentrations and those where criteria are set and where effects occur is similar to that for freshwater. However, the dataset for observed nearshore marine water was too small (n<50) to assign a level of concern.

Table 8. Summary of Hazard Evaluation for Lead.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,427	33%	No	Priority 2
	Nearshore Marine	44	68%	INS	U
	Offshore Marine	77	88%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	838	96%	No	Priority 2
	Nearshore Marine	472	95%	No	Priority 2
	Offshore Marine	478	99%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

Unlike most other metals, lead appears to have a fairly distinct pattern of higher concentrations in offshore marine waters compared with concentrations observed nearshore. This may reflect lead enrichment of marine waters through ocean exchange. For cadmium, the only other metal assessed which is comparatively high in oceanic waters, the higher concentrations in offshore water may also be a reflection of this ocean enrichment process.

At least 10% of the observed lead concentrations in freshwater sediments exceed the Canadian TEL and PEL as well as the consensus-based TEC and PEC. However, the 90th percentile value (approx. 200 mg/kg dw) did not exceed the floating percentile SQS, making lead one of the few COCs that did not exceed this threshold.

Lead concentrations in marine nearshore and offshore sediment have 90th percentile values of approximately 100 mg/kg dw and 70 mg/kg dw, respectively. These concentrations are not above the SQS and exceed only the consensus-based TEC among the guidelines and standards used for comparison.

As mentioned previously, lead hazards to fish, wildlife, and human health were not assessed due to the lack of effects data. However, although lead shot has been prohibited in Washington for all waterfowl, coot, and snipe hunting since a nationwide phase-in of non-toxic shot was implemented during 1986-1991, lead poisoning of birds due to ingestion of lead shot remains a concern (Ecology and WDOH, 2009).

Mercury

Mercury is a naturally occurring heavy metal in the earth's crust and is nearly always detectable at concentrations >0.005 ug/l in water and >5 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). Unlike other metals, mercury is liquid at typical ambient temperatures, is volatile, and bioaccumulates to a high degree in edible tissues of fish (i.e. fillet), particularly those occupying high trophic positions.

Mercury naturally degasses from soils, the rate of which may be increased dramatically by disturbances such as logging and land development, and inundation caused by dam construction. Historically, a large source of mercury to Puget Sound was a now-defunct chloralkali plant in Bellingham (PTI, 1991), although mercury is released through numerous industrial and combustion sources as well.

Major Releases from Primary Sources

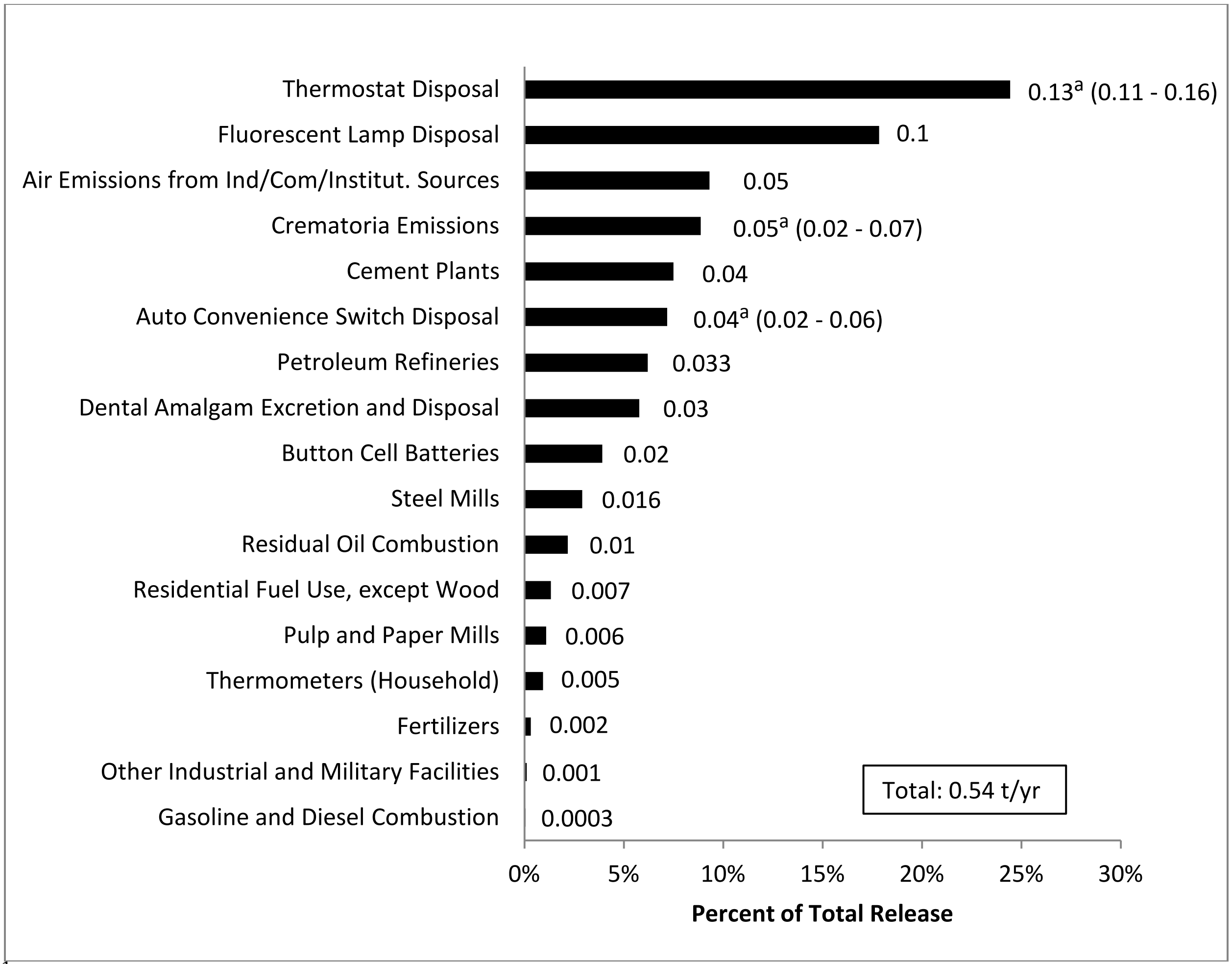
An inventory of primary mercury sources suggests that approximately 0.54 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of anthropogenic mercury to the environment may be due to thermostat and fluorescent lamp disposals (24% and 18%, respectively) (Figure 16).

Mercury releases from improper disposal of other materials may account for an additional 11% of the total release in the Puget Sound basin. These products contain mercury as liquid, or as in the case of fluorescent lamps, in vapor form. Since mercury has the potential to volatilize, there is likely some portion of liquid mercury that is released from landfills due to disposal of these products, even though collection and recycling efforts are underway to prevent these circumstances. Nearly all of the anthropogenic mercury sources assessed in the *Sources Report* have been addressed in the Mercury Chemical Action Plan (Ecology and WDOH, 2003), and efforts are underway to reduce or eliminate mercury releases from these sources.

Air emissions from major industrial facilities reporting under TRI represent approximately 18% of the total mercury release, and four-fifths of these are stack air emissions. As much as of one-quarter of the mercury release to air in the Puget Sound basin may be through combustions emissions. Much of this may be due to fossil fuel combustion, although mercury emissions from crematoria and cement plants may originate from the source material rather than the fuel.

Mercury is also released from the TransAlta Centralia Generating Plant at an annual rate of approximately 0.15 t (Ecology, 2011). Although the TransAlta plant was not included in the source inventory since its location falls outside of the Puget Sound basin, it is upwind of the basin and at least two nearby lakes in the Puget Sound basin have mercury levels in sediment that appear to reflect deposition from the plant (Furl and Meredith, 2010).

Other mercury releases which do not fall under air emission or landfill disposal categories include disposal and excretion of dental amalgam (0.03 t/yr, 6% of total) and mercury contained in fertilizer, particularly nitrogen and potassium material (0.002 t/yr, <1% of total).



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 16. Total Mercury Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for mercury loading were obtained from PSTLA loading studies and are included in Table 9. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads. Mercury was not measured in marine waters to assess exchange at the ocean boundary.

Table 9. Total Mercury Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.0047	0.049	0.094
Air Deposition	0.0094	0.02	0.033
Surface Runoff	0.091	0.136	0.238
POTWs (b)	NR	0.0024	NR
Ocean Exchange	NA	NA	NA

(a) Lowest, highest, and mid-point of estimated loads
(b) Estimate from Phase 2 (Envirovision et al., 2008b)
NR=not reported
NA=not analyzed

The estimated groundwater mercury load ranges by an order of magnitude (0.005 – 0.09 t/yr). The range in estimates is due primarily to the range in flows used to calculate loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Mercury was not measured in POTWs for the Phase 3 loading study. Phase 2 estimates for mercury loading through wastewater calculated an estimated release of 0.002 t/yr from municipal wastewater treatment plants and an additional 0.007 t/yr from industrial discharges, the latter of which is considered an underestimate due to incomplete sampling (Envirovision et al., 2008b).

The deposition of atmospheric mercury directly to the marine waters of Puget Sound is approximately 0.009 – 003 t/yr, based on fluxes ranging from 0.003 – 0.020 ug/m^2/d. Median mercury fluxes were generally ≤0.010 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Fluxes at this location were two- to three-fold higher than at other locations.

Surface water runoff loads for mercury are estimated to be approximately 0.09 – 0.24 t/yr for the Puget Sound basin based on the surface runoff study. Elevations in total mercury concentrations appeared to correspond with elevated TSS in all areas except agriculture land covers, although dissolved mercury accounted for one-half to two-thirds of the total mercury concentration.

During storm events, mercury concentrations in surface water increased by factors of 2 to 3 over baseflow concentrations. Agricultural areas have the highest mercury concentrations in surface water during both baseflow and storm flow conditions, followed by residential areas, commercial/industrial sub-basins, and forests. A substantial seasonal first-flush episode was evident for total mercury concentrations in forests but was not seen in other land covers.

The net sum of mercury loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 0.1, 0.2, and 0.3 t/yr, respectively. Under all of these estimates, there is a net mercury load to Puget Sound, although the possibility of a net export out of Puget Sound could not be explored since ocean boundary water was not sampled during the loading studies.

Mercury loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 17. Each pathway represented as a contribution to the total load is displayed in Figure 18.

Total mercury loading from the major pathways assessed is 0.11 – 0.36 t/yr. Surface runoff accounts for the largest pathway (65 – 87%), followed by air deposition (9%). Groundwater potentially accounts for up to 26% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of mercury in wells and rough estimates of groundwater flow. At the lower end of the load range, mercury in groundwater is estimated to account for 4% of the load to Puget Sound. The Phase 2 estimate of loading through POTWs – data obtained from the literature – is roughly 2% of the total mercury load to Puget Sound.

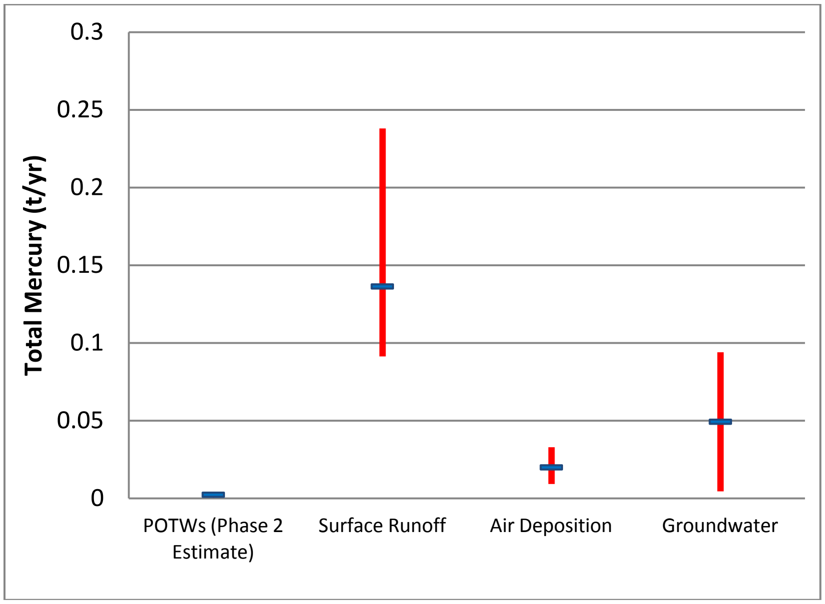


Figure 17. Range (25th - 75th percentiles) and Median Total Mercury Loads to Puget Sound from Each Major Delivery Pathway.

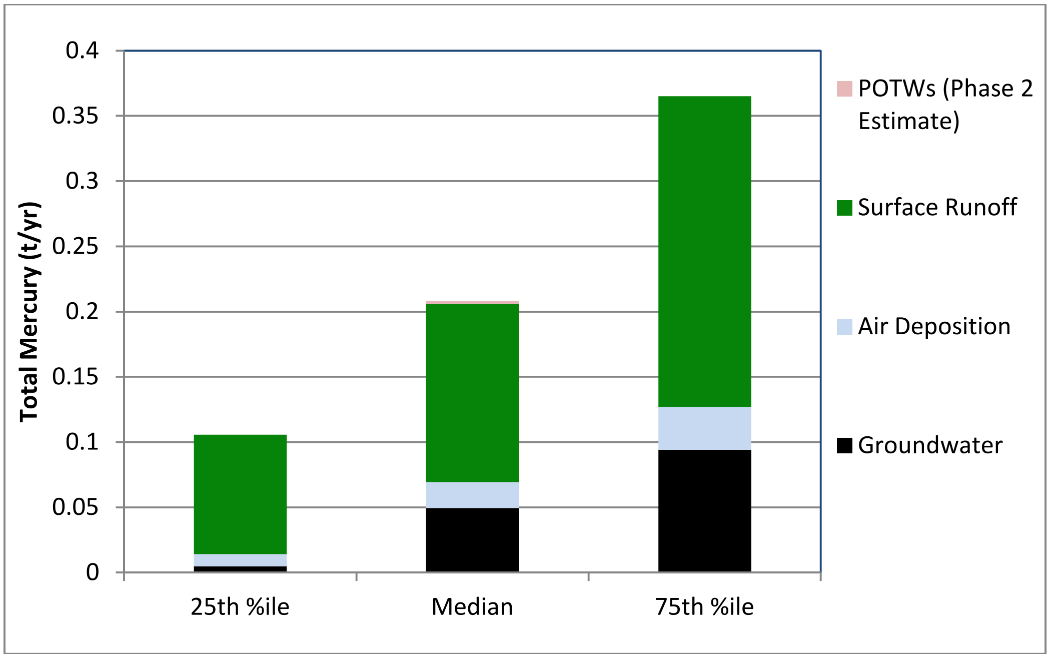


Figure 18. Total Mercury Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of mercury transport and fate following release suggests that much of the mercury transported to Puget Sound will occur through surface runoff, mostly during storm events, even as a high proportion of initial releases are emissions to air. Since most of the anthropogenic mercury releases occur as air emissions, the surface water pathway may be assumed to represent a secondary or tertiary pathway. Other possible explanations for the differences between the high proportion of air emission as a mercury source and the high proportion of surface runoff as a delivery pathway are natural sources of mercury or continuous mobilization of historic releases of mercury remaining in aquatic systems.

Total inventoried anthropogenic mercury releases to air in the Puget Sound basin are approximately 0.5 t/yr; closer to 0.7 t/yr if mercury released from the TransAlta plant is included. Since releases to air are at least double the loads from all major pathways combined, it seems reasonable that some of the mercury atmospherically deposited on land is entrained in surface runoff and transported to Puget Sound. Applying the known median atmospheric mercury deposition rates to marine waters to the entire Puget Sound watershed would result in an additional 0.1 t/yr of deposition. If all of the mercury assumed to be deposited to land surfaces under this scenario were mobilized in surface runoff, it would closely match the median load delivered to Puget Sound through the surface runoff pathway.

The air deposition study found that, unlike other trace elements measured, mercury did not vary significantly among stations, and much of the loading occurred as a result of washout during rain events. These findings appear to support widespread "dosing" of the watershed with aerially-transported mercury. However, the authors of the study (Brandenberger et al., 2010) note that while mercury deposition in the Puget Sound appears to be a result of regional transport, the bulk deposition methods used to estimate fluxes may underestimate the dry deposition of mercury. As a result, strong localized signals from industrial areas may be diluted out.

A consistent level of aerial mercury "dosing" across a watershed would likely result in similar mercury concentrations among land uses, particularly since much of the load is due to washout. The surface runoff results do not reflect a widespread dosing and washout scenario since forested areas have lower mercury concentrations than commercial/industrial and residential areas where combustion sources are likely to be located. However, higher mercury loads from commercial/ industrial and residential areas may simply be a result of enhanced mobilization from surfaces (due to the comparatively high proportion of impervious surfaces) rather than localized mercury sources.

The comparatively high concentration of mercury in agricultural area runoff samples remains a puzzle. Loss of mercury-enriched soil during storms may be one possible explanation, although baseflow mercury concentrations in agricultural areas are also higher than in other land covers, and TSS increases during storm flows are smaller in agricultural areas compared to other land types. It appears unlikely that major mercury sources would typically be located in close proximity to agricultural lands, and the one inventoried mercury release that is specific to agricultural practices is comparatively small (releases from fertilizer application, 0.002 t/yr).

The single inventoried source of mercury released in a constrained pathway is loss of dental amalgam through disposal (approximately 95% of the mercury used in dental offices is currently recovered) and excretion. Approximately 0.03 t/yr of mercury release in the Puget Sound basin is attributed to this source. This is approximately tenfold the estimated load from POTWs during the Phase 2 Study (0.002 t/yr) (Envirovision et al., 2008b). The differences are presumably due to the removal of solid material during the treatment process. Mercury from other constrained sources, naturally occurring mercury in water, and mercury in stormwater represent additional releases of mercury to POTWs, but the extent of these contributions are not known.

Hazard Evaluation

The hazard evaluation indicates that mercury is a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 10).

Most (>95%) of the observed mercury concentrations in surface waters are one to two orders of magnitude below concentrations where most (95%) of the effects have been documented. The 90th percentile of observed total mercury concentrations are slightly above the chronic water quality criterion (0.012 ug/l), but observed dissolved concentrations are well below the acute criterion (2.1 ug/l). Interestingly, the chronic water quality criterion is based on a value designed to avoid exceedance of the Food and Drug Administration Action Level for mercury in seafood (1.0 mg/kg; EPA, 1985).

Table 10. Summary of Hazard Evaluation for Mercury.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,313	63%	Yes	Priority 1
	Nearshore Marine	7	100%	INS[a]	U
	Offshore Marine	14	93%	INS[a]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	803	66%	Yes	Priority 1
	Nearshore Marine	459	70%	Yes	Priority 1
	Offshore Marine	367	79%	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Freshwater	b	100%	INS[a]	U
	Nearshore Marine	c	≥95%	INS[d]	U
	Offshore Marine	e	100%	INS[d]	U
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	Yes	Priority 1
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	f	>99%	No	Priority 2
	Nearshore Marine	g	>92%	No	Priority 2
	Offshore Marine	h	100%	No	Priority 2

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

INS[a] =Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

[b] N range is 11 – 16 and varies for organism type and tissue type

[c] N range is 42 – 169 and varies for organism type and tissue type

INS[d] =Insufficient effects data available for comparison to observed data

[e] N range is 5 – 190 and varies for organism type and tissue type

FW=Freshwater

SW=Saltwater

[f] N range is 34 – 776 and varies for organism type and tissue type

[g] N range is 107 – 197 and varies for organism type and tissue type

[h] N range is 37 – 346 and varies for organism type and tissue type

There are few observed data to compare mercury concentrations to effects data or criteria in marine waters. For instance, only seven usable values were found for nearshore marine waters and only 14 values for offshore waters. Ninety percent of the effects data were at least two orders of magnitude above the 90th percentile values for both the nearshore and offshore datasets. Although more than 25% of the offshore values exceed the chronic water quality criterion, the paucity of observed values precluded an adequate evaluation for any of these comparisons.

Mercury concentrations in freshwater sediments exceed the floating percentile at the 90th percentile concentration (approx. 0.6 mg/kg dw) of the observed dataset. Marine sediments also have observed mercury concentrations that exceed the SQS at 90th percentile levels for both the nearshore and offshore datasets (both approx. 0.5 mg/kg dw); these concentrations are also at or near the LAET.

Observed mercury concentrations in fish tissues are nearly identical to effects levels, but there were insufficient observed data for an adequate evaluation. Fewer effects values are available for marine waters. No marine effects concentrations were available for non-decapod invertebrates and fish, and only one marine decapod effect concentration was available. There are observed concentrations for all tissue types although only five for offshore decapods.

Evaluation of daily mercury doses based on fish and incidental sediment ingestion for the four species evaluated – great blue heron, osprey, river otter, and harbor seal – indicate that all species would be exposed to doses equal to or greater than the lowest effects dose. For the bird species, the lowest effects doses are based on reproductive effects, although heron also exceed a dose for growth effects. For the mammal species evaluated, both seal and otter exceed the lowest dose calculated for growth effects, while the otter also exceeds the lowest dose for mortality.

Edible tissues evaluated for comparison to the NTR criterion (based on the default consumption rate of 6.5 g/d) reveal that fish tissue generally has higher observed mercury concentrations than bivalves or other invertebrates, but for all organisms more than 95% of the observed values fell below the criterion (approx. 800 ug/kg wet weight).

Comparisons were also made using exposure assumptions outlined in the NTR but with varying daily consumption rates. More than one-half of the freshwater and marine fish tissue samples exceed the acceptable risk level using the EPA recommended subsistence rate of 142.4 g/d. In the nearshore marine areas, more than one-half of the mercury concentrations found in invertebrates other than bivalves exceed the acceptable risk at the 142.4 g/d rate, and more than one-half of the bivalves exceed the acceptable risk based on the Suquamish tribal rate (769 g/d). In the offshore marine areas, more than one-half of the mercury concentrations found in invertebrates other than bivalves exceed the acceptable risk at the 142.4 g/d rate, and more than one-half of the bivalves exceed the acceptable risk based on the Tulalip tribal/King Co. American Petroleum Institute (API) rate (242.5 g/d).

Regionally Important Biological-Effects Data

Grove and Henny (2008) analyzed mercury in river otter carcass livers obtained from trappers working in western Oregon and western Washington, including Puget Sound. Mercury liver concentrations were higher in Puget Sound adult river otters (mean of 7.89 mg/kg dw) than those from the northwest Washington area (mean of 5.85 mg/kg dw). Concentrations in Willamette River and coastal Oregon otter livers were slightly higher (mean of 9.2-9.3 mg/kg dw) but similar to Puget Sound levels. This study demonstrates that river otters living in the Puget Sound area bioaccumulate mercury.

In addition to the bioaccumulation of mercury by otters, accumulation in fish tissue has led to advisories for human consumption of fish in Puget Sound. The advisories, issued by Washington State Department of Health (WDOH), are based on data and consumption of particular species, and vary by region (Hardy and Palcisko, 2006). For instance, WDOH advises no consumption of yelloweye rockfish (*Sebastes ruberrimus*) caught anywhere in Puget Sound, and advises limited consumption of English sole (*Parophrys vetulus*) and other flatfish based on the marine area in which they are caught. In addition, there is a statewide mercury advisory for smallmouth bass (*Micropterus dolomieui*), largemouth bass (*M. salmoides*), and northern pikeminnow (*Ptychocheilus oregonensis*) with specific advice based on risks to population segments (e.g. children, nursing mothers) (McBride, 2003; WDOH, 2011).

Zinc

Zinc is a naturally occurring heavy metal abundant in the earth's crust and is nearly always detectable at concentrations >0.5 ug/l in water and >10,000 ug/kg (dw) in sediments from freshwater and marine environments (PTI, 1991; Serdar, 2008; Hallock, 2010; Appendix D). It occurs at comparatively high concentrations in natural waters, but zinc's wide use as a protective coating and alloy with other metals to reduce corrosion in outdoor environments may increase levels in the aquatic environment.

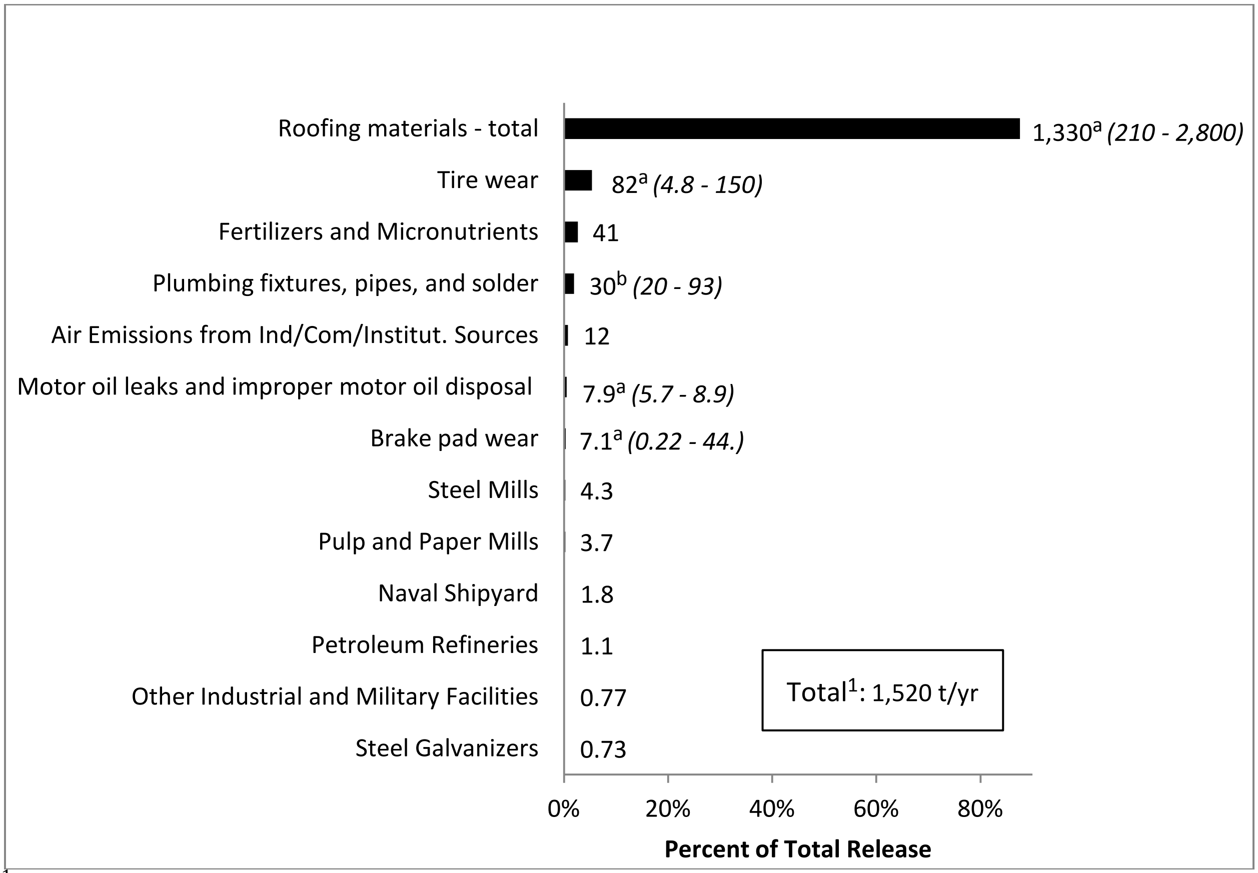
Major Releases from Primary Sources

An inventory of zinc released from primary sources suggests that approximately 1,500 metric tons (t) is released annually from anthropogenic sources in the Puget Sound basin. The largest current source of zinc to the environment is the leaching of zinc from rooftops, particularly those with galvanized components, accounting for approximately 1,300 t/yr of zinc released (Figure 19). The authors of the *Sources Report* note that total inventoried zinc releases to the Puget Sound basin probably underestimate the true extent of release since leaching from other galvanized items (e.g. culverts, light standards, guardrails) was not assessed (Ecology, 2011).

Although small in comparison to rooftop releases, zinc released from tire wear is substantial (82 t/yr), accounting for approximately 6% of the total. Brake pad wear accounts for approximately 5 t/yr of zinc released in the Puget Sound basin.

Zinc contained in fertilizers and micronutrients used in agricultural applications accounted for large zinc releases (41 t/yr). Similar to most other COC metals, concentrations were highest in phosphate fertilizers. However, zinc use in fertilizer applications (4 t/yr) is small compared to its use as an agricultural micronutrient (37 t/yr).

Other inventoried sources of anthropogenic release of zinc in the Puget Sound basin included leaching from residential plumbing components (21 t/yr) and industrial, commercial, and institutional emissions (approximately 24 t/yr). Of this latter category, approximately one-third of the zinc releases were from steel mills and pulp and paper mills.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mean

[b] Median

Figure 19. Total Zinc Release in the Puget Sound Basin (values shown are t/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for zinc loading were obtained from PSTLA loading studies and are included in Table 11. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 11. Total Zinc Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	1.97	10.8	19.7
Air Deposition	11	18	26
Surface Runoff	113	122	134
POTWs	16	19	24
Ocean Exchange (b)	-150	-80	10

(a) Lowest, highest, and mid-point of estimated loads
(b) Negative values indicate a net outflow at the ocean boundary

The estimated range of groundwater zinc loads is large (2.0 – 20 t/yr). The range in estimates is due almost entirely to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

Zinc discharged from POTWs in the Puget Sound basin has a much narrower range of loading, accounting for loads of 16 – 24 t/yr. Phase 1 estimates suggest that zinc loads from industrial wastewater are potentially substantial (16 t/yr; Hart Crowser et al., 2007), but this is based on a limited dataset and is far in excess of industrial discharge of zinc to surface waters or transferred to POTWs as reported in the TRI (<4 t/yr total) (Ecology, 2011).

The deposition of atmospheric zinc directly to the marine waters of Puget Sound is approximately 11 – 26 t/yr, based on fluxes ranging from 2.2 - 52 ug/m^2/d. Median zinc fluxes were generally <10 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. Zinc fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated five-fold above a nearby station that did not have the same air pollution influences in such close proximity. This pattern closely mirrored the deposition patterns of copper and lead among the air sampling locations.

Surface runoff loads for zinc are estimated to be approximately 113 – 134 t/yr for the entire Puget Sound basin based on the surface runoff study. Overall, dissolved zinc accounted for approximately one-half to three-quarters of the total zinc concentrations in surface water.

Absolute zinc loads were highest in forests, with little change in loading during storms compared to baseflows, although a seasonal first-flush episode was in forest, as well as agricultural areas. Zinc loads in other land covers increased dramatically during storms, particularly commercial/ industrial areas, where zinc loads increased by an order of magnitude. This leads to a disproportionately high loading of zinc in commercial/industrial areas relative to other land areas.

Based on sampling marine waters, there appears to be a large net export of zinc at the ocean boundary using estimates derived from the 25th and 50th percentiles of the data, although the dataset is very limited. However at the high end of the reported data range (75th percentiles), there is virtually no net flux across the ocean boundary. Total zinc concentrations (25th -75th percentile) in the incoming marine waters are 0.53 – 0.88 ug/l, and total zinc concentrations in the outgoing marine waters are 0.70 – 0.80 ug/l. Zinc inflow is estimated to be 310 – 520 t/yr, and zinc outflow is estimated to be 460 – 510 t/yr.

The net sum of zinc loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is -8, 90, and 210 t/yr, respectively. Under the 25th percentile estimate, there is a comparatively small net export of zinc out of Puget Sound due to the large mass exchanged at the ocean boundary. However, when the median values are summed, the large surface water load outweighs the smaller export at the ocean boundary, leading to a net load of zinc load to Puget Sound, and all loads estimated at the 75th percentile indicate net loads to Puget Sound.

Zinc loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 20. Each pathway represented as a contribution to the total load is displayed in Figure 21.

Total zinc loading from the major pathways assessed is 140 – 200 t/yr. Surface water runoff accounts for the largest pathway (66 – 83%), followed by POTWs (11 – 12%) and air deposition (8 – 13%). Groundwater potentially accounts for up to 10% at the upper end of the estimated range, but this value should be viewed with caution since it is based on literature values of zinc in wells and rough estimates of groundwater flow. At the lower end of the load range, zinc in groundwater is estimated to account for 1% of the load to Puget Sound.

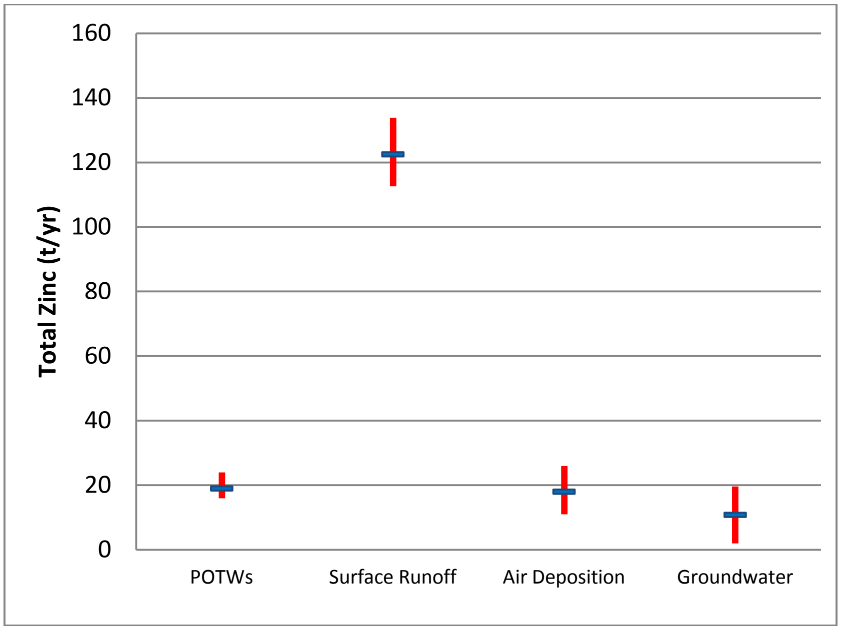


Figure 20. Range (25th - 75th percentiles) and Median Total Zinc Loads to Puget Sound from Each Major Delivery Pathway.

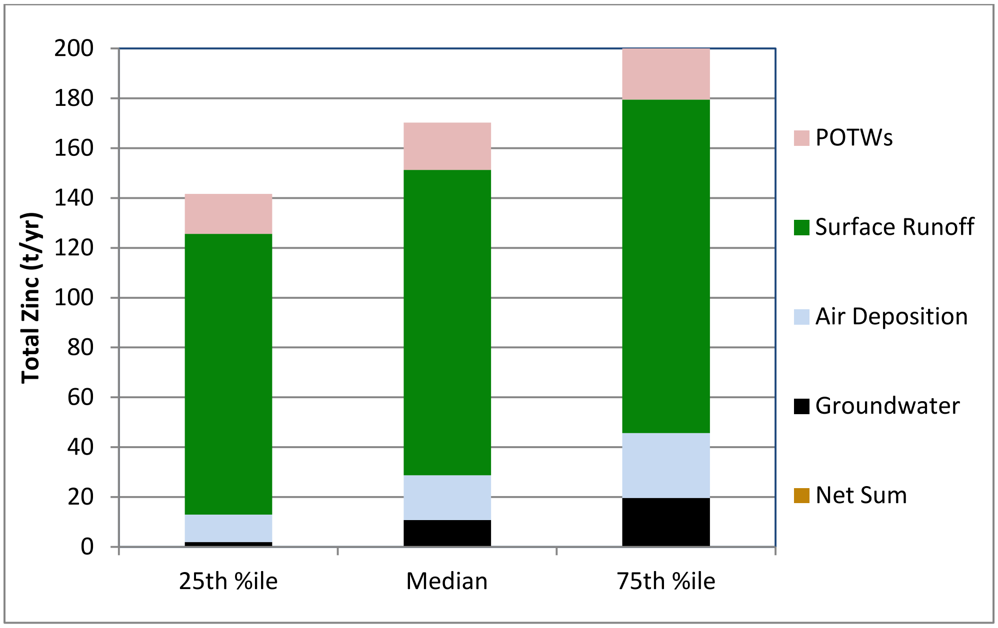


Figure 21. Total Zinc Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of zinc transport and fate following release suggests that much of the zinc transported to Puget Sound will occur through surface runoff during both baseflow conditions and storm events. The high proportion of zinc loading contributed by surface runoff is consistent with the types of sources accounting for the major zinc releases.

Major unconstrained releases of zinc are leaching from rooftops and from vehicle component wear. Together, these sources account for approximately 1,400 t/yr of zinc released to the environment in the Puget Sound basin.

Once zinc is released from rooftops it may be transported in runoff to any number of pathways. Zinc may continue to remain in runoff on impervious surfaces until it reaches surface waters or is diverted to wastewater treatment plants, or zinc may be initially or secondarily allowed to infiltrate into soils where it can migrate to groundwater or become retained in a soil reservoir. Once zinc is in soil, it can be slowly leached out in dissolved form, migrate to groundwater, or become released as soil particles during high-energy storms where it settles as aquatic sediments, including those found in catch basins.

The surface runoff results appear to be consistent with major zinc releases from rooftops. During storms, zinc is released through leaching, and in commercial/industrial areas, the high proportion of rooftop area and relative dearth of attenuating components would allow for the enriched runoff to reach surface waters. Most of the zinc released from rooftop and galvanized materials would presumably be in the dissolved form, consistent with elevations of dissolved zinc during storms.

Although this conceptual model appears to fit with commercial/industrial areas during storms, it does not explain the comparatively low concentrations of dissolved zinc from residential areas, nor does it explain the high zinc concentrations in commercial/industrial areas during baseflows. While there are few clues to explain the finding of low zinc in residential areas, the high levels in commercial/industrial area baseflows may be due to a high density of galvanized culverts or a reservoir of zinc-enriched sediments residing in aquatic systems.

Aside from zinc released from rooftops, zinc released from vehicle component wear would most likely occur in residential and commercial/industrial areas. Abraded particles released to roadways and mobilized in storm runoff would presumably result in increases in total zinc in excess of comparable increases in dissolved zinc. The increase in concentrations of total zinc in commercial/industrial and residential areas appears to be consistent with this supposition, rising two- to three-fold, with concentrations significantly correlated with TSS. There are virtually no elevations in total zinc in agricultural and forested land covers during storm events.

Overall zinc releases to air across the entire basin (not including fugitive dust from tire and brake pad wear) are estimated to be approximately 18 t/yr. Zinc released as fugitive dust from tire and brake pad wear is potentially substantial (48 t/yr), assuming 50% is emitted as airborne particles or becomes suspended in air following release to the road surface. This may account for the high levels of zinc in atmospheric deposition samples at the high-density urban location, which is located near Interstates 5 and 705, associated interchanges, and other major roadways. Zinc fluxes at this location were an order of magnitude higher than at other locations, and the inventory of primary releases indicates that industrial sources are not large contributors to airborne emissions. The total zinc air emissions across the basin (up to 66 t/yr) appears to match the deposition on marine waters (11 – 26 t/yr) when one considers that marine waters only constitute one-sixth of the basin area.

Of the primary sources inventoried, zinc released from residential plumbing components represents the only constrained source assessed. The annual load of zinc discharged to POTWs is estimated to be 21 t; an additional 8 t/yr is discharged to septic systems from this source. The estimate of zinc released from plumbing components is nearly identical to loads released from POTWs (16 – 24 t/yr), but it is unlikely that a mass balance has been achieved simply from these loading terms. It is much more likely that zinc from other constrained sources is delivered to POTWs, and naturally occurring zinc in water, as well as zinc in stormwater, represents additional releases of zinc to POTWs, but the exact extent is not known. At the treatment end, some quantity of zinc is presumably lost via solids removal prior to discharge.

Hazard Evaluation

The hazard evaluation indicates that zinc is a Priority 1 level of concern for offshore marine surface waters and for freshwater sediments (Table 12). A Priority 2 level of concern was assigned to fresh surface waters and marine sediments. The number of observed data in nearshore marine waters was insufficient for an adequate comparison with effects data or criteria. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

The 90th percentile values for dissolved zinc in freshwater (approx. 15 ug/l) are one-half the 10th percentile of effects levels and the chronic and acute water quality criteria. For marine waters, the differences between the observed and effects/criteria is even larger, with the difference between the 90th percentile of observed data (approx 2.1 ug/l for nearshore and 1.2 ug/l for offshore) are one to two orders of magnitude below the 10th percentile of the effects data and the chronic and acute water quality criteria.

Median zinc concentrations in both freshwater and marine sediments are about one-half of the lowest guidelines or standards. However, in freshwater sediments the 90th percentile (approx. 300 mg/kg dw) of observed zinc concentrations exceeds the floating percentile SQS concentration and floating percentile CSL.

Zinc concentrations in marine nearshore and offshore sediment have 90th percentile levels (approx. 180 mg/kg dw and 120 mg/kg dw, respectively) less than one-half the SQS concentration, although they exceed Canadian TEL.

Table 12. Summary of Hazard Evaluation for Zinc.

Surface Water		**Total N**	**FOD**	**90th %ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	4,844	88%	No	Priority 2
	Nearshore Marine	33	100%	INS	U
	Offshore Marine	57	95%	Yes	Priority 1
Sediment		**Total N**	**FOD**	**90th %ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	822	>99%	Yes	Priority 1
	Nearshore Marine	513	100%	No	Priority 2
	Offshore Marine	513	100%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th %ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th %ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

INS=Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

PCBs

Polychlorinated biphenyls (PCBs) were manufactured commercially in the U.S. from 1929 until their ban in 1979 after the negative health and environmental impacts associated with PCBs became apparent (Sittig, 1980; EPA, 1999). EPA (1997) estimated that as of 1977, U.S. manufacturers had produced a total of 635,000 metric tons (t) of PCBs. Prior to federally imposed use restrictions, the PCB market spanned a wide range of end products. While electrical equipment represents the majority of PCB use – 77% from 1929-1975 according to EPA (1997) – their chemical stability and plasticizing properties made them useful in a variety of applications. PCBs in open system applications such as plasticizers, hydraulics fluids and lubricants, and carbonless copy paper accounted for >20% of their historic use.

Although banned more than three decades ago, PCBs continue to be found in environmental media. Many of the same properties that made PCBs commercially desirable – their stability and resistance to degradation – make them extremely persistent in the environment, and they have become one of the most ubiquitous of all environmental contaminants.

There are 209 individual forms of PCBs, known as congeners, based on the degree of chlorine substitution and arrangement on the biphenyl molecule. The persistence of PCBs increases with the degree of chlorination. Mono-, di-, and tri-chlorinated biphenyls biodegrade relatively rapidly, tetrachlorinated biphenyls biodegrade slowly, and higher chlorinated biphenyls are resistant to biodegradation. PCBs accumulate in the lipids (fats) of fish and other animals, with lipid solubility typically increasing with the degree of chlorination (Mabey et al., 1982).

PCBs are typically present at very low concentrations in ambient waters, with water column concentrations typically in the 10 – 1,00 pg/l range for total PCBs (Dangerfield et al., 2007; Appendix D), although few data are available for marine waters (Serdar, 2008). In sediments, total dry weight PCB concentrations are typically found in the 1 – 100 ug/kg (dw) range (Appendix D).

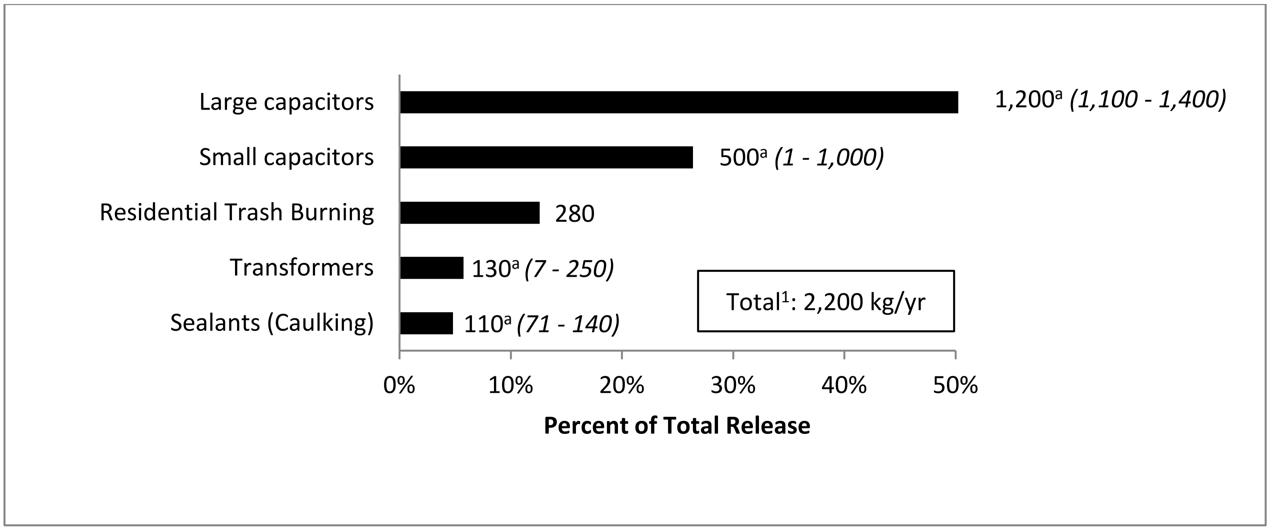
Major Releases from Primary Sources

An inventory of PCBs released from primary sources suggests that approximately 2,200 kg is released annually in the Puget Sound basin (Figure 22). PCB use can be placed in two categories: closed systems and opens systems. Closed systems include PCBs used in electrical transformers and capacitors, including those used in light ballasts. The release of PCBs due to leakage of closed systems was estimated at 1,800 kg/yr in the Puget Sound basin.

PCBs historically were used in a number of open-system products such as adhesives, carbonless copy paper, flame retardant coatings, pesticide extenders, lubricants, and caulking sealants. Of the products that incorporated PCBs, caulking sealants are among the most durable. Caulking used in commercial, industrial, and institutional buildings constructed during the 1940s through the 1970s may contain PCBs which may continue to be released through volatilization, leaching, or abrasion of the material. Based on the volume of commercial, industrial, and institutional buildings constructed in the Puget Sound basin during the era of PCB caulk use, an estimated 110 kg of PCBs are released from this source annually.

An additional source of PCB release is from residential trash burning. Based on modeling information generated by Ecology's Air Quality Program, PCBs are emitted from this source at a rate of 281 kg/yr in the Puget Sound basin.

The annual rate of PCB release, estimated to be 2,200 kg/yr, should be viewed with caution and likely overestimates actual releases. No regional sampling efforts to inventory PCB releases from primary sources have been conducted, and therefore PCB release estimates are based on literature values. There are also no regional field studies to estimate leakage rates, relative amounts released indoors and outdoors, and the proportion cleaned up and contained following spillage. The estimates displayed in Figure 22 assumed all leakage was unconfined.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 22. Total PCB Release in the Puget Sound Basin (values shown are kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PCB loading were obtained from PSTLA loading studies and are included in Table 13. No PCB loading data are available for groundwater.

The estimated discharge of PCB from POTWs is 0.13 – 1.75 kg/yr. The POTW load estimates were based on limited sampling conducted only during the wet season.

The deposition of atmospheric PCBs directly to the marine waters of Puget Sound is approximately 0.7 – 3.7 kg/yr, based on fluxes ranging from 0.16 – 2.8 ng/m^2/d. Median PCB fluxes were generally <0.7 ng/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. PCB fluxes at this location were elevated five-fold above most other locations around the Puget Sound region, including a nearby station that did not have the same air pollution influences in such close proximity.

Table 13. PCB Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	0.68	1.32	3.76
Surface Runoff	2.55	5.29	15.77
POTWs	0.126	0.342	1.75
Ocean Exchange (a)	-1.4	0.8	0.6
Returning Salmon (b)	NC	0.265	NC

NA=not analyzed
NC=not calculated
(a) Negative values indicate a net outflow at the ocean boundary
(b) Best estimate using available data

Surface runoff loads for PCBs are estimated to be approximately 2.6 – 15.8 kg/yr for the entire Puget Sound basin based on the surface runoff study. PCBs were detected in all land use types, and median concentrations among land covers were within a factor of 3 during baseflows. However, during storm flows, concentrations in commercial/industrial areas increased by an order of magnitude while concentrations in other areas remained virtually unchanged. PCB concentrations were particularly elevated during seasonal first-flush episodes in all land types except agricultural areas.

Overall PCB loads were generally proportional to land area when evaluated by land cover type, except commercial/industrial loads during storm events (percentage of the total PCB load was 15-fold higher than the percentage of commercial/industrial land cover in the Puget Sound basin). However, due to the large area and flows of forested areas, overall loads from forests accounted for 83% of the total PCB load.

Based on the sampling of marine waters, there appears to be an annual net export of 1.4 kg total PCBs at the ocean boundary using estimates derived from the 25th percentile of the data, although the dataset is very limited. At the median and higher range (75th percentile) of the estimates, there is less than 1 kg annual flux into Puget Sound at the ocean boundary. Total PCB concentrations (25th-75th percentile) in the incoming marine waters are 15 – 35 pg/l, and total PCB concentrations in the outgoing marine waters are 14 – 47 pg/l. PCB inflow is estimated to be 8.6 – 21 kg/yr, and PCB outflow is estimated to be 10 – 20 kg/yr.

In addition to major loading pathways, estimates of PCB influx to Puget Sound through returning adult salmon were based on typical whole-body PCB concentrations measured during 2004-2005 and escapement estimates from 2001. Rough estimates for five species of pacific salmon (*Oncorhynchus spp.*) suggest that the total influx of PCBs from this pathway is approximately 0.265 kg/yr based on whole-body PCB concentrations ranging from 4 ug/kg to 51 ug/kg (Sandie O’Neill, National Oceanic and Atmospheric Administration, written communication).

The net sum of PCB loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2.2, 8.0, and 22 kg/yr, respectively. Under all of these estimates, there is a net PCB load to Puget Sound.

PCB loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 23. Each pathway represented as a contribution to the total load is displayed in Figure 24.

Total PCB loading from the major pathways assessed is 3.3 – 21 kg/yr. Surface runoff accounts for the largest pathway (74 – 76%), followed by atmospheric deposition (18 – 20%) and POTWs (4 – 8%).

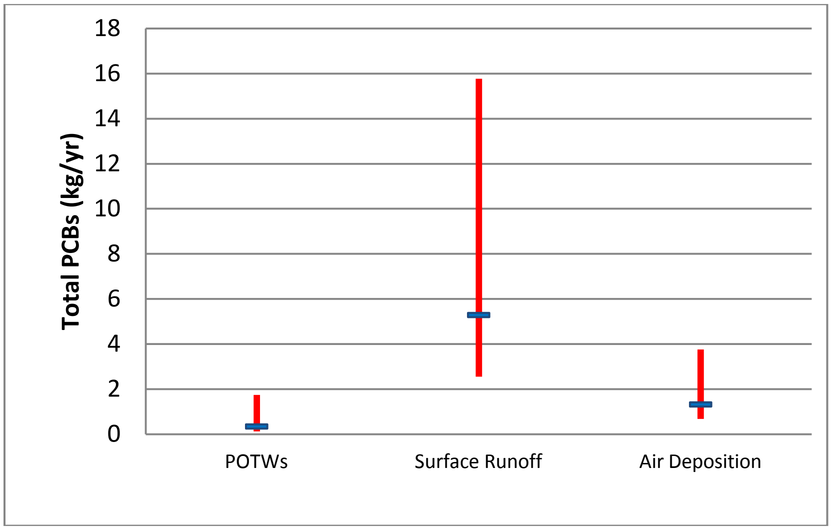


Figure 23. Range (25th - 75th percentiles) and Median Total PCB Loads to Puget Sound from Each Major Delivery Pathway.

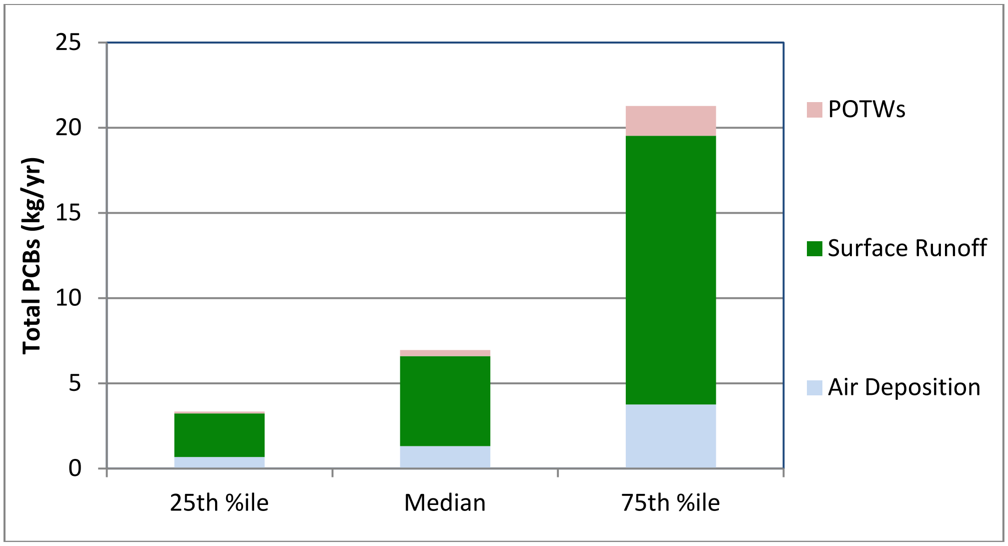


Figure 24. Total PCB Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PCB transport and fate following release suggests that much of the PCBs transported to Puget Sound will do so through surface runoff during both baseflow and storm flow conditions. Surface runoff data suggest that in residential, agricultural, and forested areas, a continuous and low level of PCB dosing from instream or upland sources occurs during baseflow. PCB concentrations remain the same or decrease slightly during storms, indicating that stormwater contains similar or lower PCB concentrations than the stream baseflows, and any mobilization of instream PCB reservoirs do not effectively increase concentrations.

In commercial/industrial areas, the dynamics of PCBs in surface runoff appear to be much different. PCB concentrations during baseflow conditions are 40% - 180% higher than in other land covers. During storm events, PCB concentrations in commercial/industrial areas increase six-fold. Based on the available information, it is impossible to ascertain whether the increase is due to mobilization of land surface PCBs, re-suspension of instream PCB reservoirs, or a combination of the two circumstances.

The major PCB sources identified in the *Sources Report* (Ecology, 2011) do not appear to have a direct link with the surface runoff pathway. Most of the PCBs releases are likely to occur in and around buildings and become bound to soil following release, volatilize and become transported off-site, or occur indoors. Residential trash burning presumably occurs in residential areas. However, commercial/industrial areas are the most likely to deliver PCBs to surface waters based on the primary sources since PCBs released from buildings and from transformers/ capacitors are more likely to occur in commercial/industrial areas than in the other land covers assessed. In addition, PCBs atmospherically deposited on land surfaces are more likely to become mobilized during storms if they are deposited on impervious surfaces which are more prevalent in commercial/industrial areas.

One other possible source of PCB enrichment of streams is marine-derived PCBs delivered upstream by salmon returning to spawn. PCB residues per whole-body fish range from approximately 7 ug for pink salmon (*O. gorbuscha*) to 336 ug for Chinook (*O. tshawytscha*) (Sandie O'Neill, National Oceanic and Atmospheric Administration, written communication). While the total annual PCB load entering Puget Sound from salmon is estimated to be approximately 0.3 kg/yr, the impact to streams is likely to be somewhat less. The load estimate does not include losses from commercial and recreational takes, live fish and carcasses removed from streams by wildlife, and maternal transfer to eggs (and subsequent flux from out-migrating smolts). In addition, approximately one-third of the PCB burden is carried by two species – pink salmon and chum salmon (*O. keta)* – which spawn much lower in the watershed than other species, thus diminishing upstream PCB transport.

PCB releases to air from inventoried sources are several orders of magnitude greater than air deposition to marine water (approx. 300 – 400 kg/yr versus 0.7 – 3.7 t/yr), and this discrepancy is large even when the deposition rates to marine water are scaled to the entire watershed area. The primary releases to air are combustion emissions originating from residential trash burning (280 kg/yr) and volatilization from PCB-containing building sealants (maximum of 140 kg/yr). There is a large degree of uncertainty surrounding release estimates from both of these sources;

neither has been sufficiently analyzed to determine if reported release rates are representative of the Puget Sound region.

PCB deposition patterns indicate similar fluxes occur throughout Puget Sound, suggesting a widespread atmospheric deposition pattern compounded with additional deposition from near-field sources in high-density urban areas. PCB emissions from building sealants are expected to be concentrated in cities, particularly older industrial cities, although it is not clear if they are likely to be deposited near their point of release.

Based on the inventory of sources, none of the PCB releases were exclusive to constrained systems such as sanitary sewers. Therefore it is difficult to quantitatively assess the relationship between releases and POTW loads (0.1 – 1.8 kg/yr). Some of the PCBs released from building sealants are likely to occur indoors and find their way to sanitary sewers after attaching to dust particles, although the quantity has not been determined. It is likely that some portion of the PCBs delivered to POTWs occurs through stormwater, but the extent of this contribution is not known. Since PCBs were historically used in a variety of commercial, industrial, and consumer applications (see *Sources Report* [Ecology, 2011] for a discussion of PCB uses), releases to constrained and unconstrained sources are likely to continue from a variety of sources.

Hazard Evaluation

Comparisons to effects levels, criteria, and guidelines are done separately for both PCB Aroclor and congener data. PCBs were historically marketed in the U.S. as Aroclors, mixtures of individual PCB compounds (a.k.a. congeners) based on average chlorine content. Environmental analysis of PCBs historically has focused on these Aroclor mixtures, although once in the environment Aroclors quickly alter their original composition due to unequal degradation, fugacity, and bioaccumulation rates of their individual components.

Beginning in the mid-1990s, the analysis of PCB congeners gained wider acceptance as the laboratory capacity for this method became more widely available and the utility of congener analysis became more evident. Although much more expensive than Aroclor analysis, congener analysis provides detection limits several orders of magnitude lower than Aroclors (e.g. mid parts per quadrillion levels versus mid parts per trillion levels in water). As a result, environmental sample datasets are generally a mix of Aroclor and congener data. Due to the lower congener detection limits, results of measurable PCBs are typically lower for congeners (as mentioned previously, the hazard evaluation results are shown for detectable concentrations only).

The hazard evaluation indicates that PCBs are a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 14).

Table 14. Summary of Hazard Evaluation for PCBs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	1,248[a]	4%[a]	Yes	Priority 1
	Nearshore Marine	11[a]	0%[a]	INS[b]	U
	Offshore Marine	84[c]	100%[c]	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	506[a]	43%[a]	Yes	Priority 1
	Nearshore Marine	612[a]	63%[a]	No	Priority 2
	Offshore Marine	387[a]	47%[a]	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Freshwater	d,e	88%	Yes/INS[b]	Priority 1/U
	Nearshore Marine	d,f	100%	No/INS[g]	Priority2/U
	Offshore Marine	d,h	≥99%	No/INS[i]	Priority 2/U
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	Yes	Priority 1
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	d,j	≥77%	Yes	Priority 1
	Nearshore Marine	d,k	>33%	Yes	Priority 1
	Offshore Marine	d,l	>66%	Yes	Priority 1

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

[a] Based on Aroclor data

INS[b]=Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

[c] Based on congener data

[d] Based on Aroclor and congener data

[e] N = 123 – 142 for non-decapod invertebrates. Observed data insufficient to evaluate fish tissue.

[f] N = 27 – 28 for decapods, N = 57 – 99 for non-decapod invertebrates, and N = 70 – 96 for fish tissue. Effects data insufficient to evaluate fish tissue.

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INS[g]=Insufficient effects data available for comparison to observed data

[h] N = 11 – 32 for non-decapod invertebrates and N = 26 – 324 for fish tissue. Observed data insufficient to evaluate decapods. Effects data insufficient to evaluate fish tissue.

INS[i]=Insufficient observed or effects data available for comparison

FW=Freshwater

SW=Saltwater

[j] N range is 51 – 918 and varies for organism type, tissue type, and PCB type.

[k] N range is 68 – 344 and varies for organism type, tissue type, and PCB type.

[l] N range is 10 – 477 and varies for organism type, tissue type, and PCB type.

PCB concentrations in surface waters vary depending on whether Aroclor or congener data are considered. The 90th percentile of observed freshwater Aroclor concentrations (approx. 0.05 ug/l) is several times higher than the chronic water quality criterion, but the 90th percentile of observed freshwater congener concentrations (approx. 0.002 ug/l) is several times lower than the criterion. Although there are more Aroclor data available, the detection frequency is much higher for congener data. For both sets of observed data, most (>95%) of the observed concentrations are one to two orders of magnitude below concentrations where most (95%) of the effects have been documented. It is notable that the numerical value for the chronic water quality criterion (0.014 ug/l) is driven by adverse reproductive effects to mink through consumption of fish, demonstrating the importance of PCBs’ high bioconcentration potential and manifestation of effects on higher trophic organisms.

There are no detectable PCB concentrations observed in nearshore marine waters to compare to effects data or criteria. For offshore marine waters, the 90th percentile of congener concentrations (approx. 0.00005 ug/l) is five orders of magnitude less than the 10th percentile of effects data and three orders of magnitude below the chronic water quality criterion.

In freshwater sediments, at least 25% of the observed PCB concentrations analyzed as congeners or Aroclors exceed the floating percentile SQS, although the number of congener samples is comparatively small. At least 5% of the Aroclor data exceed all of the guidelines and standards used for comparison.

PCB concentrations in marine sediments are difficult to characterize due to the vast differences between observed congener and Aroclor concentrations (Aroclors are two to four orders of magnitude higher), as well as large differences between concentrations in nearshore and offshore sediments (offshore Aroclors concentrations are two orders of magnitude higher than nearshore Aroclor concentrations). The congener concentration, assessed on either dry weight or organic-carbon normalized bases, are well below any guideline, whereas median nearshore Aroclor concentrations fall in the midst of guidelines, with the 75th percentile of the concentrations (approx. 20,000 ug/kg organic carbon) above the SQS.

Interestingly, although the nearshore Aroclor concentrations are two orders of magnitude higher than congeners, the concentrations are nearly identical on an organic-carbon normalized basis, suggesting that the differences can be partly attributed to high levels of organic carbon in the samples analyzed for Aroclor. The magnitude of difference between nearshore and offshore Aroclor concentrations remain after accounting for organic carbon. All level-of-concern assignments for sediments were based on Aroclor data since the congener datasets were not deemed sufficient for adequate comparisons (n ≥100; Appendix D-1).

Comparisons of observed PCB concentrations in tissues of fish, decapods, and other invertebrates to effects due to PCB burdens were difficult to evaluate due to a paucity of either observed or effects data. In freshwater where sufficient observed and effects data were available for non-decapod invertebrate, most of the observed concentrations measured as both Aroclor and congeners exceeded all of the effects concentrations.

For nearshore marine waters, both decapods and non-decapod invertebrates had sufficient (observed and effects) data to conduct an evaluation. For both organism types, the 90th percentile of observed data was at least five-fold lower than the 10th percentile of effects data. In offshore marine water, non-decapods invertebrate data were available to conduct comparisons; the 90th percentile value of observed concentrations was an order of magnitude below the 10th percentile of effects concentrations. There were not sufficient effects data for fish tissue to conduct an adequate evaluation in marine surface waters.

Evaluation of daily PCB doses based on fish and incidental sediment ingestion for the four species evaluated – great blue heron, osprey, river otter, and harbor seal – indicate that all species would be exposed to doses equal or greater than the lowest effects dose (assuming a 4% sediment ingestion rate for heron). For the bird species, the lowest effects doses are based on reproductive effects; osprey are exposed to doses three times the lowest of the effects level. For the mammal species evaluated, both seal and otter exceed the lowest dose calculated for reproductive effects by an order of magnitude.

Edible tissues evaluated for comparison to the NTR criterion (based on the default consumption rate of 6.5 g/d) indicate that PCB concentrations in nearly all of the fish and non-bivalves are higher than the NTR criterion. For freshwater, nearly all of the bivalve tissue had concentrations above the criterion as well. Bivalve PCB concentrations in nearshore areas are higher than those from offshore areas.

Regionally Important Biological-Effects data

PCBs have been detected in outmigrant juvenile salmon (Johnson et al., 2007) from multiple northwest estuaries and hatcheries, including three in the Puget Sound. Whole-body juvenile Chinook salmon from the Duwamish River contained the highest PCB concentration [103 ng/g wet weight (ww) or 3,100 ng/g lipid] of any of the locations tested. Johnson et al. (2007) note that this concentration is higher than the 2,400 ng/g lipid developed by Meador et al. (2002) as a tissue threshold for adverse health effects including reduced growth, altered enzyme and hormone activity, and increased mortality.

Separately, juvenile salmonid PCB exposures were documented as occurring via food source by an analysis of stomach content of outmigrants at three locations in Puget Sound (Stein et al., 1995). Meador et al. (2010) found that PCB tissue concentrations in outmigrant juvenile Chinook salmon from the Duwamish estuary varied by time and location within the estuary, suggesting that localized heterogeneity of sediment concentrations may substantially impact accumulation in fishes.

PCB concentrations in adult Puget Sound Chinook salmon tissues were found to be three to five times higher than those measured in six other populations of Chinook salmon on the West Coast of North America (O'Neill and West, 2009). Approximately 22% of maturing and sub-adult Puget Sound Chinook salmon had concentrations above the 2,400 ng/g lipid threshold mentioned previously. O'Neill and West (2009) note that these elevated tissue concentrations have resulted in consumption advisories, and have implications for the viability of these fish and southern resident killer whales. Cullon et al. (2009) found elevated PCBs in adult Chinook returning to the Duwamish River, as well as in Puget Sound Chinook smolts.

PCB concentrations in Puget Sound herring and Puget Sound flatfish have also been evaluated. Puget Sound herring were found to contain three to nine times higher concentrations of PCBs than herring from the Strait of Georgia, with Puget Sound whole-body concentrations ranging from about 120 to 160 ng/g wet weight (ww) (West et al., 2008).

The Puget Sound Partnership has adopted a target for PCB concentrations in fish as one of the first "dashboard indicators" for toxic chemicals in fish (PSP, 2011a). The dashboard indicators were developed to provide a broad range of measurements to assess the health of Puget Sound. The PCB target is based on the documented accumulation of PCBs in a variety of Puget Sound fish species and the availability of a PCB tissue threshold concentration (i.e., 2,400 ng/g lipid; Meador et al., 2002). Specifically, the target is to reduce PCB levels in Puget Sound so that 95% of the sampled species have tissue concentrations below the threshold. Currently, at least 15% and up too 100% of the Chinook salmon, Pacific herring, and English sole analyzed have PCB concentrations exceeding the threshold.

In addition to measurements of PCB accumulation in tissues, analyses of various biomarkers of pollution exposures in benthic flatfish were shown to successfully differentiate between sites with differing degrees of sediment contamination (Stein et al., 1992). Cullon et al. (2005) also found about seven times higher levels of PCBs in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia. Sol et al. (2008) found a statistically significant correlation between PCB concentrations in English sole livers and two biological effects parameters.

A number of studies have been conducted in the Puget Sound region investigating exposure and/or effects of PCBs and other persistent and bioaccumulative contaminants on wildlife, particularly marine mammals. Johnson et al. (2009) measured PCB concentrations in osprey eggs from the Lower Duwamish River and compared them to those sampled from the upper Willamette River. Total PCB residues were significantly higher in Lower Duwamish River osprey eggs (geometric mean = 897 ug/kg ww) compared to those from the Willamette River (geometric mean = 182 ug/kg ww). These results demonstrate that adult osprey bioaccumulation and maternal transfer of PCBs is occurring in osprey nesting in PCB contaminated areas of Puget Sound. This study also compared egg residues over time and determined that PCB concentrations in osprey eggs from the Lower Duwamish River had decreased 53% between 2003 and 2007.

Grove and Henny (2008) also demonstrated the bioaccumulation of PCBs in river otter livers from Puget Sound. The Puget Sound area river otters accumulated more PCBs (as total PCBs) than otters from other areas in western Washington. PCBs and other organochlorines have been shown to cause immunosuppression, thyroid disruption, and possibly cancer in harbor seals (Tabuchi et al., 2006; Ylitalo et al., 2005; Simms et al., 2000; Ross et al., 1996; Ross et al., 1995; de Swart et al., 1996; de Swart et al., 1995; Van Loveren et al., 1994). Vitamin A disruption has also been observed in harbor seal pups found on the Washington State coast whose mothers contained high PCB residues in their blubber (Simms et al., 2000). This effect on seal pups is suspected to result from exposure to contaminated milk.

There is substantial evidence that Puget Sound harbor seals and killer whales are bioaccumulating PCBs at very high concentrations in their blubber. The prey items of Puget Sound harbor seals were measured to have seven times higher concentrations of PCBs than prey from Strait of Georgia on a lipid basis (Cullon et al., 2005), which corresponds to PCB concentrations measured in harbor seal blubber.

Tissue concentrations of PCBs have often been reported as dioxin toxicity equivalents (i.e. TEQs) which are toxicities of dioxin-like PCBs, dioxins, and furans relative to the most toxic dioxin (2,3,7,8-TCDD). Puget Sound harbor seals have significantly higher TEQs in blubber (158 ng/kg lipid weight) compared to seals from the Strait of Georgia (33 ng/kg lipid weight) (Ross et al., 2004), and the TEQ contribution was greater from PCBs than dioxins and furans. Levin et al. (2005) also found that the majority of TEQs in harbor seal pups (from southern B.C.) were from PCBs, not dioxins and furans.

Ross et al. (2000) reported measured mean total PCBs in transient and Southern resident male killer whales were 251 and 146 mg/kg lipid, respectively; the authors concluded these marine mammals are among the most contaminated in the world.

Further research on the northern, southern, and transient killer whale communities have discovered that males bioaccumulate more PCBs than females due to maternal transfer (Krahn et al., 2007; Krahn et al., 2009). PCB concentrations in the blubber of mothers decrease temporarily during nursing and can reach concentrations below those in their calves. Mothers initiate bioaccumulation again after calves are weaned. Total PCB concentrations in southern resident killer whales were measured to range from about 5,000 to 180,000 ug/kg lipid. For all but three recent mothers, the measured concentrations exceed a marine mammal threshold for blubber concentrations (17,000 ug/kg lipid). Although environmental concentrations of PCBs are gradually declining, one modeled estimate of southern resident killer whale recovery projects that blubber concentrations will not reach the marine mammal threshold until 2063 (Hickie et al., 2007).

Accumulation of PCBs in fish tissue has led to advisories for human consumption of fish from Puget Sound marine waters, as well as limited freshwaters in the basin. The advisories, issued by WDOH, are based on data and consumption of particular species, and vary by region (Hardy and Palcisko, 2006). For instance, WDOH advises limited consumption of rockfish and flatfish based on the marine area in which they are caught. Consumption limits are also recommended for Chinook salmon due to elevated PCB levels, and vary depending on whether fish are migratory or the resident (blackmouth) species. WDOH also provides consumption advice for several Puget Sound region freshwater lakes and rivers due to PCBs in fish, including Lake Washington, Green Lake, and the Lower Duwamish River (WDOH, 2011).

PBDEs

Polybrominated diphenyl ethers (PBDEs) have been incorporated as flame retardants in numerous consumer products for decades and as a result, have gradually been released to the environment where they persist and accumulate in biota. Unlike most other persistent organic pollutants addressed in this report, environmental concentrations of PBDEs appear to be escalating in some cases, although available data on which to assess trends are also much more limited than for other chemicals.

Manufacturers of many different materials and products have used PBDEs as flame retardant additives in their products since the 1960s. These products include fabrics, television sets, computers, ABS resins, high impact polystyrene, textile coatings, carpet, polyurethane foams, cushions, mattresses, and insulation for wire and cables.

PBDEs are not chemically bonded to the matrices of those materials and products, and therefore they potentially escape from their matrix through volatilization to the air. Products and materials partially composed of or treated with PBDEs off-gas PBDEs to the environment during the useful lifetime of the product or material (i.e., while the product or material is still in use). Volatilization is one of the primary mechanisms of the release of PBDEs to the environment (Lorber and Cleverly, 2010).

Since PBDEs are a complex mixture of 209 congeners (varying by the number of bromine atoms and location on the molecule), their use and behavior in manufactured materials, as well as their behavior once released from materials, varies substantially. Major homolog groups (groups classified by the number of bromine atoms per molecule) include penta-, octa-, and deca-brominated diphenylethers (commonly referred to as Penta, Octa, and Deca, respectively). Penta was used widely in polyurethane foam and textiles, while the heavier homologs (Octa and Deca) were used primarily in polymers and electronics. The heavier homologs tend to be less volatile than the lighter BDEs, although once in the environment they may degrade to the lighter homologs. Heavier congeners such as Deca may also bind to dust more strongly than the more volatile congeners.

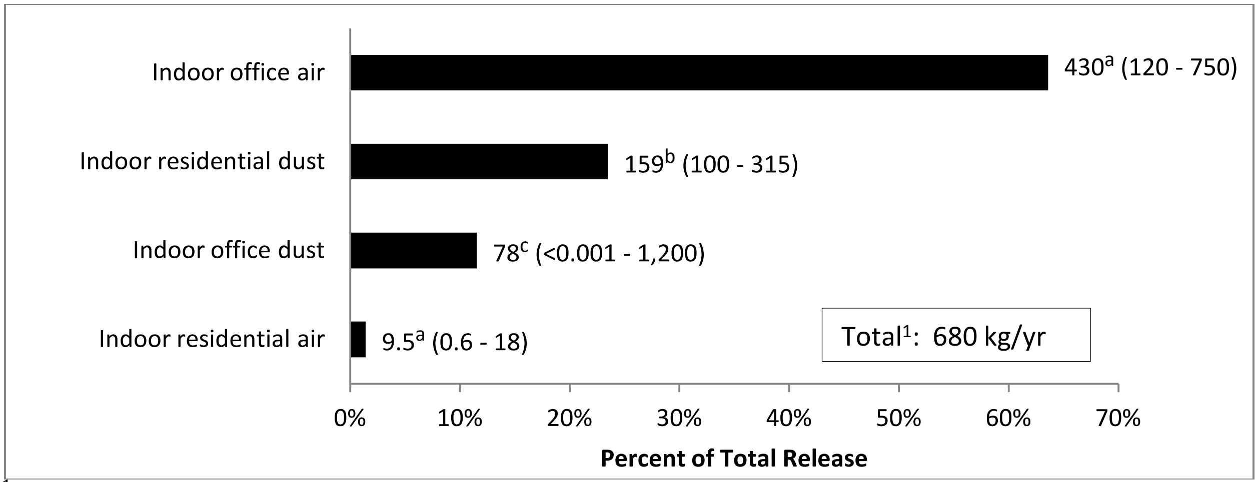
Beginning in the late-1990s, concerns began to emerge over the accumulation of PBDEs in animal tissues and the potential toxicity of PBDEs. Pressure to limit or ban PBDEs continued to mount until manufacturers of Penta and Octa voluntarily ceased production beginning in 2004. Deca manufacturers have agreed to discontinue the manufacture, import, and sales of Deca at the end of 2012, but in Washington State, Deca has been banned from mattresses since 2008 and was banned from televisions, computers, and residential upholstered furniture beginning January 1, 2011.

Major Releases from Primary Sources

Unlike most of the other COCs addressed in the present report, PBDEs are nearly all released by consumer products in constrained systems (i.e. indoors). For the inventory of primary sources (Ecology, 2011), no attempt was made to quantify environmental releases based on emissions from categories of PBDE-containing products such as computer monitors and mattress pads since most releases occur indoors and the attenuation between initial emissions and release to an environmental medium or pathway is uncertain. Therefore, release estimates were based on air exchange and dust generation in residential and commercial office spaces. This approach was taken to integrate individual component emissions and quantify PBDE releases in a simplified manner.

Total PBDE release from the four sources assessed totaled approximately 680 kg/yr (Figure 25). Indoor office space air accounted for 64% of the total release, while indoor residential air accounted for only 1%; indoor residential dust was the other major contributor at 23% while the indoor office dust contributed 12% to the overall releases. These release estimates should be viewed with caution since there is a high degree of uncertainty around all of the variables used to derive the values.

The PBDE Chemical Action Plan (Ecology and WDOH, 2006) largely addresses these sources and is consistent with the bans and restriction on PBDEs mentioned previously. However, many PBDE-containing consumer and office products are still in use and may represent diffuse sources of PBDEs to the environment during the remainder of their life cycle.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

[b] Median

[c] Geometric mean

Figure 25. Total PBDE Release in the Puget Sound Basin (values shown are kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PBDE loading were obtained from PSTLA loading studies and are included in Table 15. No loading data are available for groundwater.

Table 15. PBDE Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	15.7	20.3	23.8
Surface Runoff	5.14	5.67	9.95
POTWs	7.01	10.6	20.7
Ocean Exchange (a)	41	-11	240

NA=not analyzed
(a) Negative values indicate a net outflow at the ocean boundary

Loads from atmospheric deposition are estimated to be 16 – 24 kg/yr based on fluxes ranging from 4.5 to 27.3 ng/m^2/d (25th – 75th percentiles). Samples collected from the high-density urban station had higher PBDE fluxes than at other sites by factors of 3 to 4. PBDE flux patterns reflect some increased localized input, but regional sources may also play an important role in overall loading. The authors of the air deposition loading study point out that the estimates are likely to be conservative (high) due to the use of conservative assumptions to handle non-detected results.

PBDEs were frequently detected in the POTW loading study, with annual loads estimated at 7.0 – 21 kg. Although the authors of the POTW study caution against drawing conclusions about seasonal differences, they point out that PBDE concentrations are generally higher during the dry season.

Surface runoff loads for PBDEs are estimated to be approximately 5.1 – 10 kg/yr for the entire Puget Sound basin based on the surface runoff study. PBDEs were detected more frequently in storm runoff samples compared to baseflow. All samples from commercial/industrial areas contained detectable PBDE concentrations, but detection frequencies were close to 50% in other land covers (38% in forests).

Concentrations of PBDEs in commercial/industrial areas were higher than in other areas during both baseflows and storm flows. Like PCBs, concentrations in residential, agricultural, and forested areas were nearly identical among land types and were similar between baseflows and storm flows. PBDE concentrations in commercial/industrial areas increased by an order of magnitude during storms, although a seasonal first-flush episode was not evident in commercial/industrial areas whereas it was seen in other land covers. Normalized to land cover area, commercial/industrial area loads were 10- to 20-fold above other areas. However, absolute loads from forests were larger overall due to the large area of forested land cover.

Based on marine water sampling and subsequent calculation of loads at the ocean boundary, there appears to be an annual net export of 11 kg total PBDEs at the ocean boundary using estimates derived from the median concentrations, while loads calculated from the 25th percentile and the 75th percentile concentrations appear to show net PBDE imports of 41 and 240 kg/yr, respectively. Total PBDE concentrations (25th-75th percentile) in the incoming marine waters are 760 – 1,600 pg/l, and total PBDE concentrations in the outgoing marine waters are 603 – 1,071 pg/l. PBDE inflow is estimated to be 440 – 940 kg/yr, and PBDE outflow is estimated to be 400 – 700 kg/yr.

The net sum of PBDE loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 69, 26, and 290 kg/yr, respectively. Under all of these estimates, there is a net PBDE load to Puget Sound.

PBDE loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and median for each pathway in Figure 26. Each pathway represented as a contribution to the total load is displayed in Figure 27.

Total PBDE loading from the major pathways assessed is 28 – 54 kg/yr. Atmospheric deposition accounts for the largest pathway (44 – 56%), followed by POTWs (25 – 38%) and surface runoff (18%). This pattern of source contribution is notably different than other COCs which typically have the largest, and often the majority, load input from surface runoff.

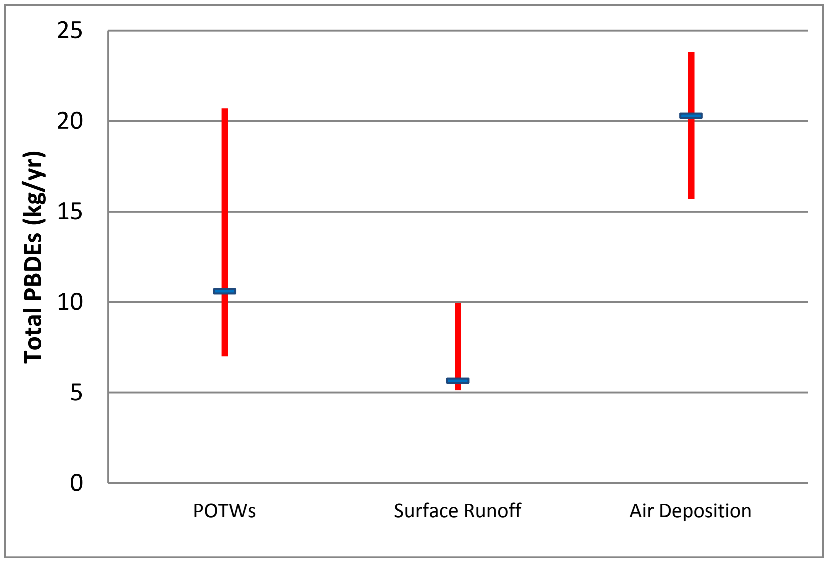


Figure 26. Range (25th - 75th percentiles) and Median Total PBDE Loads to Puget Sound from Each Major Delivery Pathway.

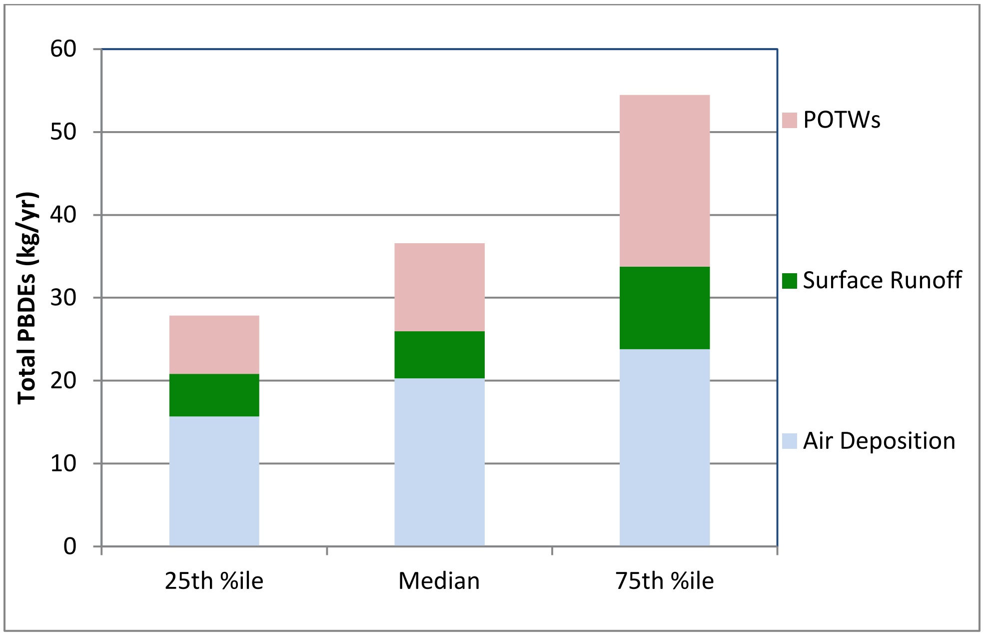


Figure 27. Total PBDE Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PBDE transport and fate following release suggests that much of the PBDEs will be initially released to air and atmospheric transport will deliver comparatively high loads directly to Puget Sound. PBDEs deposited on land will also be mobilized during storm events and delivered to surface waters, but in quantities lower than for direct atmospheric deposition. Some of the PBDEs deposited to land are also likely to be transported in storm sewers and delivered directly to Puget Sound or indirectly by way of POTWs.

The inventory of primary sources (Ecology, 2011) suggests that PBDEs are released in the highest quantities in commercial areas compared to other land covers; this notion is supported by results of the loading studies. The air deposition study found higher PBDE fluxes in the high-density urban (Tacoma) location relative to other sites. The surface runoff study found much higher PBDE concentrations in commercial/industrial areas compared to other land covers.
The potential for large releases from indoor air is also supported by seasonal PBDE fluxes in atmospheric deposition; the authors found higher flux rates during the warm season and surmised that they may have been due to increased indoor air release (i.e. windows left open and increased ventilation).

PBDEs are released from indoor consumer and office products, become attached to dust particles, and are subsequently delivered to the sanitary sewer through washing machine rinse water during the washing of fabrics with the attached PBDE-enriched dust, and rinsing other materials with attached dust particles. This appears to be a reasonable pathway for PBDE release and transport, although the estimated quantity delivered to POTWs via this route is highly uncertain. However, the high frequency of detection and relative large loads from POTWs lends support to this concept. Furthermore, due to the nature of these PBDE sources, washing machine rinse water concentrations would be expected to remain steady throughout the year. This appears to be consistent with PBDE loads from POTWs which did not vary appreciably between wet and dry seasons.

Hazard Evaluation

PBDE data are available for surface waters and sediment, yet the paucity of documented effects, standards, or guidelines for PBDEs consistent with those used for other COCs preclude the assignment of a Priority 1 or Priority 2 level of concern (Table 16).

Table 16. Summary of Hazard Evaluation for PBDEs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	255	59%	INS	U
	Nearshore Marine	0	--	INS	U
	Offshore Marine	126	20%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	77	97%	INS	U
	Nearshore Marine	1	100%	INS	U
	Offshore Marine	45	98%	INS	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS= Insufficient effects data available for comparison to observed data
U=Unknown level of concern due to lack of sufficient data for adequate comparison

Regionally Important Biological-Effects Data

PBDEs were detected in outmigrant Chinook salmon tissue and their stomach contents from four sites in Puget Sound (Sloan et al., 2010). Levels in wild outmigrant juveniles were higher than in hatchery fish, ranging from 67 to 13,000 ug/kg lipid, generally comparable to those measured in the Lower Columbia River and Estuary. Sloan et al. (2010) conclude that PBDEs may be contributing to reduced health and fitness in outmigrant juvenile Chinook salmon. PBDEs were detected in adult Chinook salmon returning to the Duwamish River and were not detected in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or Deschutes River (Cullon et al., 2009).

Lema et al. (2008) demonstrated that dietary exposures to certain PBDEs by adult fathead minnows can alter thyroid status and thyroid hormone-regulated gene transcription. Arkoosh et al. (2010) found that juvenile Chinook salmon exposed to moderate doses of PBDEs in their diet may be at increased risk of disease relative to those exposed to higher or lower doses of

PBDEs in their diet. PBDE levels were found to be about four to five times higher in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia (Cullon et al., 2005).

Very few studies have been conducted examining effects of PBDEs on birds. The studies reviewed indicate that PBDEs impact the reproduction and endocrine system similarly to PCBs. Exposure to BDE-71 for 75 days adversely impacted courtship and mating behavior of American kestrels (*Falco sparverius*)(Fernie et al., 2008). These birds also displayed significant delays in clutch initiation and smaller eggs (Fernie et al., 2009). Eggshell thinning and reduced hatching success also resulted. A study of species sensitivity to PBDEs (PBDE-71) observed that pentabrominated diphenyl ether (Penta BDE) exposure to eggs at 0.01 to 20 mg/kg caused decreased pipping and hatching success in American kestrels but not chickens (*Gallus gallus*) or Mallard ducks (*Anas platyrhynchus*)(McKernan et al., 2009). Species sensitivity was concluded to be Mallard ducks <chickens <American kestrels.

Total PBDE concentrations in osprey eggs and nestling plasma are significantly lower in the Lower Duwamish River (eggs: 321 ug/kg ww; plasma: 6 ug/kg ww) compared to those from the upper Willamette River (eggs: 897 pb ww; plasma: 22 ppb ww) (Johnson et al., 2009). Total PBDE concentrations in the osprey eggs did not change significantly between 2003 and 2007. Reproductive failure was observed in four of nine nests in the Lower Duwamish area. A small dataset from this study suggests that some nestlings may have experienced immunosuppression. However, the results were inconclusive due to the small sample size.

Compared to birds, a larger but still limited number of publications exist on the effects of PBDEs in mammals. Rodent exposure studies have demonstrated thyroid hormone disruption (Hallgren et al., 2001; Zhou et al., 2002) and developmental neurotoxic and behavioral effects (Ericksson et al., 2001; Viberg et al., 2003a; Viberg et al., 2003b). A study of grey seal pups and juveniles observed a relationship between circulating thyroid hormones, transport proteins, and PBDE uptake (Hall et al., 2003).

Similar to PCBs, there is evidence of bioaccumulation of PBDEs in marine mammals at high concentrations in blubber. However, absolute concentrations of total PBDEs appear to be lower than total PCBs. Cullon et al. (2005) measured PBDE concentrations five times higher in harbor seal prey from Puget Sound than the Strait of Georgia, but the mean PBDE concentration was five times lower than that measured for PCBs. Krahn et al. (2009) and Rayne et al. (2004) found the same pattern of killer whale blubber concentrations as found for PCBs in males, mothers, and calves. Krahn et al. (2009) measured total PBDE concentrations ranged from 680 to 15,000 ug/kg lipid. Mean PBDE concentrations in northern male killer whale blubber have been found to be significantly lower (203 ug/kg lipid) than those of southern resident (942 ug/kg lipid) and transient males (1,015 ug/kg lipid).

Although a quantitative effects assessment was not conducted for PBDE exposure to marine mammals, published research demonstrates that PBDEs are bioaccumulating to high concentrations in Puget Sound killer whales. This coupled with the growing evidence that PBDE exposure can cause thyroid and developmental effects in mammals strongly suggest that PBDEs are an important contaminant to monitor.

PCDD/Fs

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs; a.k.a. dioxins) are a class of persistent bioaccumulative compounds ubiquitous in the environment at low concentrations. There are 210 individual PCDD/F congeners (75 PCDDs and 135 PCDFs), but only the 17 congeners (seven PCDD and ten PCDF) with chlorines occupying the 2,3,7, and 8 positions on the molecules are considered toxic.

PCDD/Fs are generally found in mixtures, with the toxicity of the mixture translated to that of TCDD (and more recently, PeCDD) which is the most toxic congener. Each of the 17 PCDD/Fs are assigned a toxicity factor relative to that of TCDD and PeCDD, and the toxicity factor multiplied by the congener concentration is termed the toxic equivalent (TEQ) when the congeners are summed. The TEQ of an environmental sample is generally useful shorthand for assessing comparisons to regulatory thresholds and for assessing risks. Sampling and source data are commonly expressed as TEQs, and this convention has been adopted in the present document.

Unlike many other environmental contaminants, PCDD/Fs are not produced intentionally, but instead are formed and released as by-products of industrial production and combustion of certain chlorinated materials. As a result, industrial and combustion sources are responsible for the majority of PCDD/F releases. In the Puget Sound region, the major historical sources of PCDD/Fs – use of elemental chlorine in pulp bleaching, pentachlorophenol wood treatment operations, and combustion of saltwater-infused hog fuel – are all but gone (Yake et al., 1998). However, PCDD/Fs are extremely persistent in the environment, particularly at sites where these activities historically occurred.

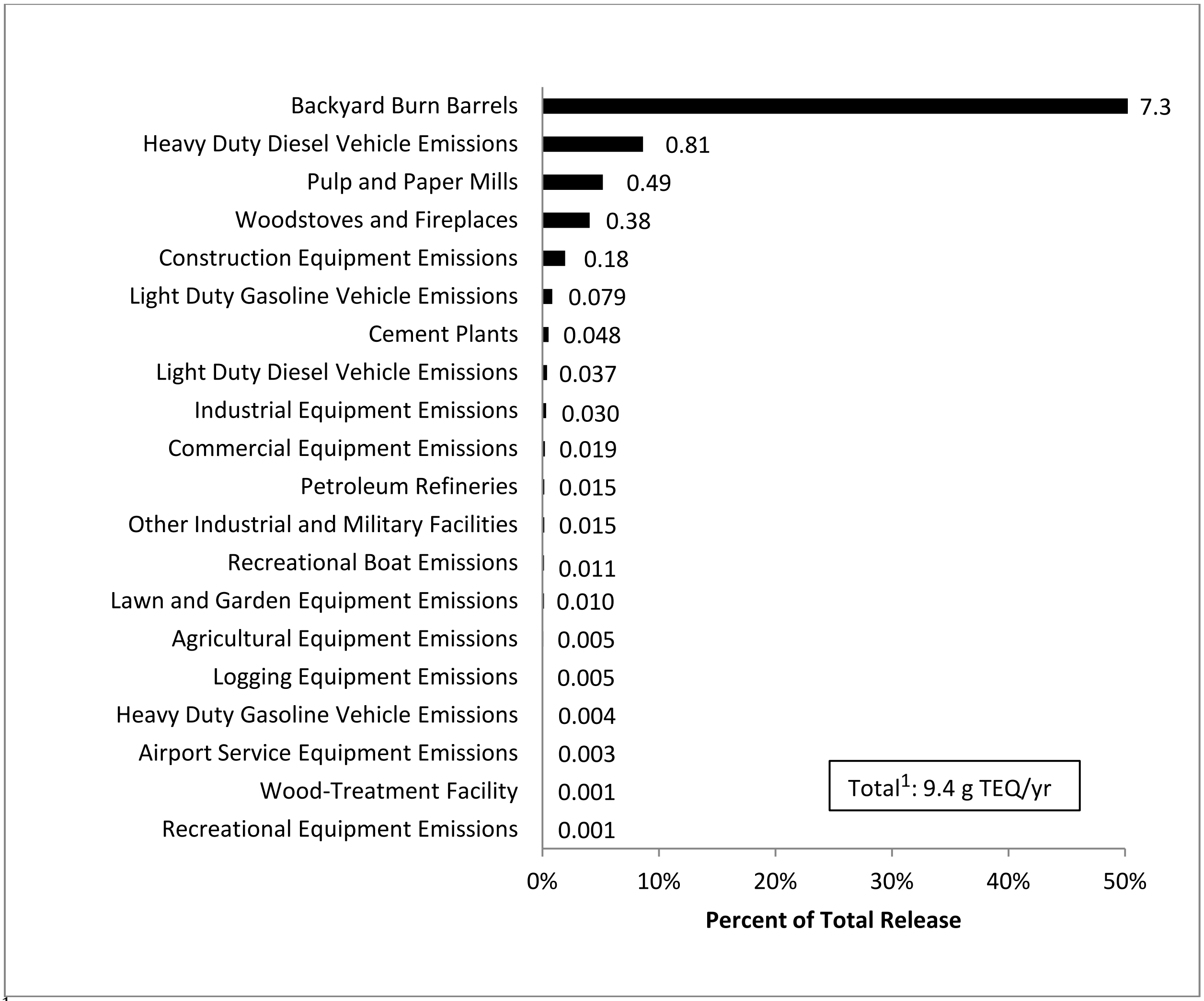
One other characteristic also distinguishes PCDD/Fs from most other environmental toxicants: their analysis requires expensive analytical techniques and is performed by only a limited number of commercial laboratories. As a result, they are commonly excluded from screening level investigations and are generally analyzed only if they are the focus of an environmental investigation. Since environmental PCDD/F data are not as prevalent as for other chemicals, there are fewer data on sources of their release to the environment.

PCDD/Fs are not typically detectable in ambient waters using conventional sampling and analytical techniques, even at detection limits in the low parts per quadrillion (pg/l) range. PCDD/Fs in sediments and biota are more typically found at low parts per trillion (ng/kg) levels, depending on the specific compound, organic carbon content of sediment, and lipid content in tissue.

Major Releases from Primary Sources

Annual PCDD/F release from the 20 sources assessed totaled approximately 9 g TEQ/yr (Figure 28). The largest single source is backyard burn barrels, accounting for nearly three-quarters of the PCDD/F release to the Puget Sound basin.

Combustion emissions to air account for 97% of the PCDD/F release. The remaining 3% of the release is to water, primarily from pulp and paper mills. This represents a substantial change from two to three decades previous when PCDD/F discharges to water from pulp mills represented the bulk of all releases to Washington State (EPA, 1991).



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 28. Total PCDD/F Release in the Puget Sound Basin (values shown are g TEQ/yr).

Major Delivery Pathways and Loading

None of the Phase 3 sampling studies included analysis of PCDD/Fs. The groundwater loading analysis included an estimate of PCDD/Fs from values reported in the literature. However, the groundwater data only included PCDD/Fs reported as total TCDD or total TCDF, with no indication of whether these are 2,3,7,8-substituted congeners.

Surface runoff load estimates were estimated during PSTLA Phase 2 based on a variety of U.S. and European runoff data, most of which were from urban areas (Envirovision et al., 2008a). The best estimates of the medians from these concentrations were 10 pg/l for commercial/ industrial areas, 5 pg/l for residential and agricultural areas, and 0.1 pg/l for forested area. When applied to the hydrologic model used in Phase 2, loads were estimated to be 6.1 – 103 g TEQ/yr (25th – 75th percentiles), with a median estimate of 25 g TEQ/yr. Most of the PCDD/F load (59%) was from residential areas, with the smallest percentage (6%) from commercial/industrial areas.

Estimates of PCDD/F loads to marine waters from atmospheric deposition were derived in Phase 1 from fluxes reported in Europe and adjusted to the Puget Sound region based on relative differences in air concentration (Hart Crowser at al., 2007). Fluxes of 0.1 – 10 pg/m^2/day (low to high end of range, 1 pg/m^2/day as medium value) were used to estimate the aerially deposited loads (0.31 – 31 g TEQ/yr, medium estimate of 3.1 g TEQ/yr).

Both the atmospheric deposition (Phase 1) and surface runoff (Phase 2) load estimates for PCDD/Fs have a large degree of associated uncertainty. The paucity of PCDD/F stream and air deposition data from the Puget Sound region makes gauging the representativeness of other data difficult. Therefore, these estimates should not be treated with the same level of confidence as those derived from Phase 3 sampling efforts.

Relationship Between Sources and Pathways/Loading

The conceptual model of PCDD/F transport and fate following release is complicated by the lack of sampling data. Based on the inventoried sources (Ecology, 2011), release to air is likely to be the primary initial pathway for PCDD/Fs. The dominance of combustion as a formation mechanism suggests that much of the PCDD/F release may be associated with airborne particles.

There are no reliable data to assess the degree of deposition for particle-bound PCDD/Fs in the Puget Sound basin, but near-field deposition may be expected for much of the emitted PCDD/Fs since the bulk of emission is from numerous non-point sources as opposed to high stacks designed to disperse emissions. Based on the primary sources in the basin, the release of PCDD/Fs is likely to occur in a mix of urban and rural locations, and may occur at a distance from the Puget Sound marine waters. Delivery to Puget Sound is therefore likely to occur through secondary pathways (e.g. surface runoff, POTWs) as well as direct deposition and would not be expected to be dominated by a single delivery mechanism.

Hazard Evaluation

The hazard evaluation indicates that PCDD/Fs are a Priority 1 level of concern for a wildlife mammalian species (river otter) and human health due to concentrations found in both the freshwater and marine aquatic environments (Table 17). However, there are insufficient observed or effects data to adequately evaluate PCDD/Fs in surface waters, sediments, or for tissue residue effects.

In fresh surface waters only five observed results are available to compare dioxin concentrations with effects data. Observed concentrations of 1,2,3,7,8-PeCDD and 1,2,3,4,7,8-HxCDD are one to two orders of magnitude below the lowest effects concentrations, and 2,3,7,8-TCDD is three orders of magnitude below levels where effects are documented. There are no observed environmental data available for PCDD/Fs in the marine water column.

In freshwater sediments, the median PCDD/F concentration (expressed as TEQ) is approximately equal to the lowest available guideline, the Canadian TEL. However, the interquartile range of concentrations spans four orders of magnitude, and at least 25% of the observed TEQs are well above the Canadian PEL where adverse effects are expected to frequently occur.

Median PCDD/F TEQ concentrations in marine sediments are similar to freshwater sediments, but the distribution of concentrations occupies a much narrower range. For both nearshore and offshore sediments, the median values exceed the Canadian TEL, but only 5 – 10% of the observed concentrations exceed the Canadian PEL.

There are few effects data to assess the potential concern of PCDD/Fs associated with tissue residue. For freshwater, there were sufficient effects data available for decapods, but no observed data. No tissue residue effects data were available for marine waters.

For the two bird species evaluated – great blue heron and osprey – only two daily effects doses of PCDD/Fs (as TEQ) were calculated: one for reproductive effects and one for mortality. Neither of the species are exposed to these doses, but heron are exposed to doses that are one-fifth to one-half of the lowest (reproductive) dose. Osprey are exposed to doses three orders of magnitude below the lowest effects dose. However, the low number of dose effects was not deemed sufficient for an adequate comparison with calculated doses (Appendix D-1).

Based on environmental concentrations of PCDD/Fs, river otter receive approximately five to ten times the daily doses (as TEQs) where reproductive effects have been documented, and within 10% of the lowest dose associated with mortality. Harbor seals are exposed to much lower daily PCDD/F doses; less than 10% of the lowest effect dose.

Concentrations of 2,3,7,8-TCDD in freshwater fish, bivalves, and other invertebrates were compared to the NTR criterion. Median concentrations for fish and tissues from other invertebrates were above the NTR criterion in both fresh and marine waters, although no detectable concentrations are available for 2,3,7,8-TCDD in marine nearshore fish. At least 90% of the 2,3,7,8-TCDD concentrations in freshwater bivalves are above the criterion, but in marine nearshore areas, less than 25% of the concentrations are above the criterion.

Table 17. Summary of Hazard Evaluation for PCDD/Fs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	7	>14%	INS[a]	U
	Nearshore Marine	0	--	INS[a]	U
	Offshore Marine	0	--	INS[a]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	36	89%	INS[b]	U
	Nearshore Marine	219	>99%	INS[b]	U
	Offshore Marine	106	>99%	INS[b]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Freshwater	c	57%	INS[b]	U
	Nearshore Marine	d	≥97%[e]	INS[b]	U
	Offshore Marine	f	≥86%	INS[b]	U
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Great Blue Heron (FW)	--	--	INS[b]	U
	Osprey (SW)	--	--	INS[b]	U
	River Otter (FW)	--	--	Yes	Priority 1
	Harbor Seal (SW)	--	--	No	Priority 2
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	g	≥25%	Yes	Priority 1
	Nearshore Marine	h	>18%[e]	Yes	Priority 1
	Offshore Marine	i	≥5%	Yes	Priority 1

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

INS[a]=Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

INS[b]=Insufficient effects data available for comparison to observed data

[c] N = 175 for non-decapod invertebrates. No observed data available for other organism types

[d] N = 76 for non-decapod invertebrates, N = 13 for decapods, and N = 1 for fish tissue.

[e] FOD=0% for fish tissue

[f] N = 28 for non-decapod invertebrates, N = 9 for decapods, and N = 10 for fish tissue.

[g] N range is 35 – 72 and varies for organism type and tissue type

[h] N range is 8 – 129 and varies for organism type and tissue type

[i] N range is 32 – 53 and varies for organism type and tissue type

Regionally Important Biological-Effects Data

Studies in Puget Sound of harbor seal and southern resident killer whale prey items have higher PCDD/Fs compared to the same prey items from the Strait of Georgia and British Columbia coast (Cullon et al., 2005, Cullon et al., 2009). Harbor seal prey were three to four times higher on a lipid basis than prey from the Strait of Georgia (Cullon et al., 2005). However, Ross et al. (2000) found that PCDD/F concentrations in killer whale blubber were much lower than PCBs, and there were no differences between whales from the northern and southern resident and transient communities. This was suspected to be due to metabolic removal of dioxins and furans.

DDT

The chlorinated insecticide dichlorodiphenyltrichloroethane (DDT) was banned in 1972 from general agricultural uses in the U.S. following concerns over its effects on wildlife and human health. However, potentially harmful levels are still found in the environment, together with its major breakdown and metabolic products dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD) due to their persistence and tendency to accumulate in fish and wildlife. In western Washington, DDT is only rarely detected in water, even at a typical analytical detection limit of about 1 ng/l (parts per quadrillion), but is nearly always detected in fish due to its extremely high bioaccumulation potential.

Major Releases from Primary Sources

DDT sources were not inventoried in the analysis of primary sources since it was banned nearly four decades ago and DDT had no uses other than as an agricultural insecticide and limited use as an urban pesticide. Due to its persistence, however, it continues to be found in environmental media, particularly soils and sediments, and is delivered to Puget Sound when these soil and sediment particles become entrained in surface water runoff.

Major Delivery Pathways and Loading

Data on major delivery pathways for DDT loading were obtained from PSTLA loading studies and are included in Table 18. No effort was made to assess DDT loads from direct deposition of atmospheric DDT to Puget Sound during Phase 3 sampling. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

The estimated range of groundwater DDT loads is large (0.2 – 7.3 kg/yr) due primarily to differences in flows used to establish the possible range of loads. DDT load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about DDT mobility in groundwater.

DDT compounds were analyzed in all ten POTWs during winter and summer sampling events, yet no concentrations were detectable. Reporting limits for DDT compounds in POTW samples were generally 2 – 3 ng/l.

Table 18. Total DDT Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	0.2	3.8	7.3
Air Deposition	NA	NA	NA
Surface Runoff (b)	2.2	23.9	25.1
POTWs	ND	ND	ND
Ocean Exchange	NC	NC	NC

(a) Lowest and highest estimated loads
(b) Detected in commercial/industrial areas only during baseflows and in commercial, agricultural, and forest areas only during storm flows
NA=not analyzed
ND=not detected
NC=not calculated due to insufficient data

Surface runoff loads for DDT are estimated to be approximately 2.2 – 25 kg/yr for the entire Puget Sound basin based on the surface runoff study. DDT compounds were detected in all land use types during storm events except residential covers, and >99% of the loads occurred during storm flows. Loads calculated during storms generally mirrored the amount of land area for each land use cover, a somewhat confounding result since agricultural areas would be expected to have larger loads relative to land area.

A single commercial/industrial area had the only detection of DDT compounds during baseflows, and only in very low concentrations. However, this particular location had DDT concentrations an order of magnitude above all other locations – regardless of land cover – during storm flows. For each land cover, the detection frequency for DDT compounds was low and never exceeded 50%, and therefore the calculated median loads were driven by non-detected values.

DDT loads were not calculated for the ocean exchange of marine waters due to the low frequency of detection. DDT compounds were detected in only three samples at locations in the northern boundary waters, all at low concentrations (<0.4 ng/l).

The net sum of DDT loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2.5, 28, and 32 kg/yr, respectively. Under all of these estimates, there is a net DDT load to Puget Sound.

DDT loading from pathways that may represent *partially controllable* sources of COCs is shown as ranges (25th - 75th percentiles) and medians for each pathway in Figure 29. Each pathway represented as a contribution to the total load is displayed in Figure 30.

Total DDT loading measured from the major pathways is 2.5 – 32 kg/yr, all contributed through surface runoff and groundwater. Surface runoff accounts for the largest portion of DDT loading (77 – 88%), with the remaining amount (12 – 23%) contributed by the groundwater pathway.

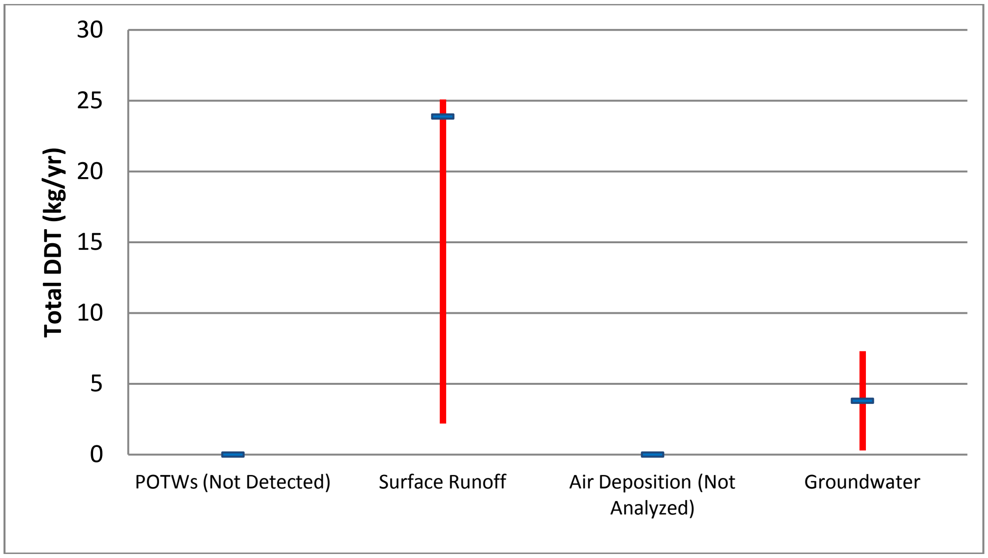


Figure 29. Range (25th - 75th percentiles) and Median Total DDT Loads to Puget Sound from Each Major Delivery Pathway.

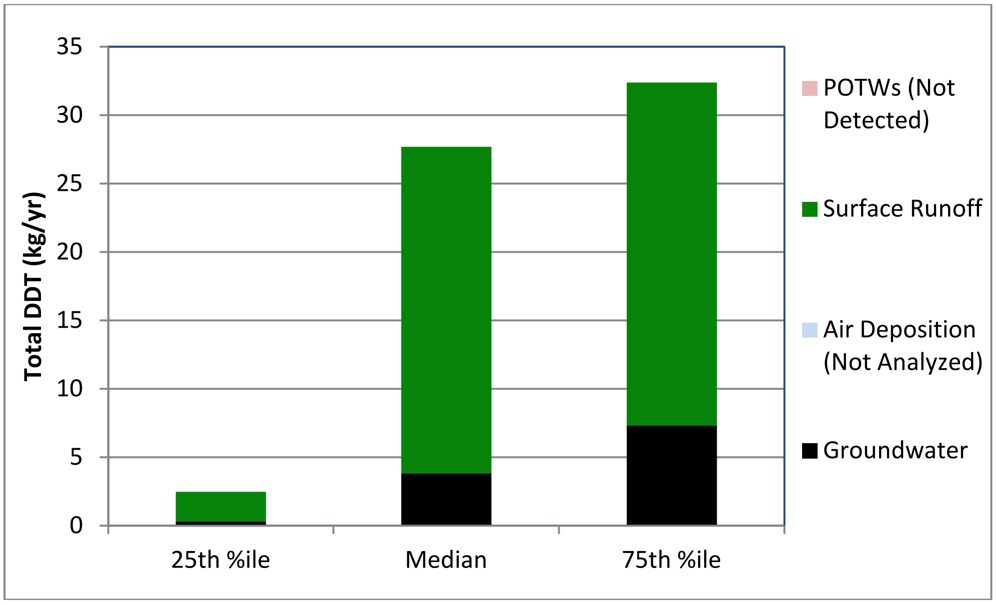


Figure 30. Total DDT Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of DDT transport and fate following release appears to be rather simple compared with other COCs. Releases from primary sources likely occurred during previous decades, and soils and sediments likely serve as the major reservoirs for DDT remaining in the environment. Soil and sediment-bound PCBs are mobilized during storm flows and delivered toward Puget Sound in surface runoff. Comparatively low DDT occurrence and concentrations in agricultural soils suggest that that the bulk of DDT mobilization from agricultural soils, where the majority of DDT was likely to have been originally released, has already occurred and the large DDT reservoirs may have advanced downstream and may largely reside in Puget Sound sediments. Conversion of agricultural lands to commercial or other land uses may explain high DDT concentrations in other land uses, particularly if soils are disturbed.

It is possible that DDT is also delivered to Puget Sound through direct atmospheric deposition to marine waters, or that some of the DDT in surface waters is due to aerially deposited DDT entrained in storm runoff. Loads of direct atmospheric deposition of DDT were calculated during the Phase 1 effort, and were estimated to be 1.2 – 31 kg/yr (low to high end of range, 6.2 kg/yr as medium value) based on fluxes reported in the eastern and midwestern U.S. (0.4 – 10 ng/m^2/day; 2 pg/m^2/day as medium value).

Hazard Evaluation

The hazard evaluation indicates that DDT compounds are a Priority 1 level of concern for a range of media and receptors in both the freshwater and marine aquatic environments (Table 19).

In fresh surface waters, the 90th percentile of total DDT (sum of DDT, DDE, and DDD) concentrations (approx. 1 ug/l) is slightly higher than the acute water quality criterion, and at least 95% of the observed concentrations are above the chronic water quality criterion. More than 90% of the values from the fairly extensive dataset on DDT effects are above the 90th percentile value for observed concentrations. There are not sufficient data to evaluate DDT in marine surface waters.

The chronic freshwater water quality criterion is three orders of magnitude lower than the acute water quality criterion. This difference stems from the high bioaccumulative potential of DDT, on which the chronic criterion is indirectly based (reproductive effects in the piscivorous brown pelican), whereas the acute criterion is based on LC_{50} concentrations for numerous fish and invertebrate species (EPA, 1980).

There are no SQS for freshwater or marine sediments to compare to DDT concentrations. However, median DDT concentrations exceed the Canadian TEL, and at least >75% of the concentrations exceed the consensus-based TEC, although concentrations are well below the Canadian PEL and consensus-based PEC.

In the marine environment, median DDT concentrations in both nearshore and offshore sediments are similar to the Canadian TEL. However, a number of the nearshore sediments have much higher concentrations than those from offshore locations, and at least 10% of the DDT concentrations exceed the Canadian PEL as well as the LAET and the 2LAET (2x the LAET).

Table 19. Summary of Hazard Evaluation for DDT.

		Total N	FOD	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	Level of Concern
Surface Water	Freshwater	2,179	4%	Yes	Priority 1
	Nearshore Marine	11	0	INS[a]	U
	Offshore Marine	0	--	INS[a]	U
		Total N	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
Sediment	Freshwater	365	30%	INS[b]	U
	Nearshore Marine	350	41%	INS[b]	U
	Offshore Marine	457	25%	INS[b]	U
		Total N	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
Tissue Residue Effects	Freshwater	c	≥92%	No	Priority 2
	Nearshore Marine	d	≥31%	INS[b]	U
	Offshore Marine	f	≥18%	INS[b]	U
		Total N	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
Wildlife	Great Blue Heron (FW)	--	--	Yes	Priority 1
	Osprey (SW)	--	--	Yes	Priority 1
	River Otter (FW)	--	--	No	Priority 2
	Harbor Seal (SW)	--	--	No	Priority 2
		Total N	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
Human Health	Freshwater	g	>68%	Yes	Priority 1
	Nearshore Marine	h	≥20%	Yes	Priority 1
	Offshore Marine	i	≥6%[j]	No	Priority 2

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

INS[a]=Insufficient observed data available for comparison to effects data

U=Unknown level of concern due to lack of sufficient data for adequate comparison

INS[b]=Insufficient effects data available for comparison to observed data

[c] N = 83 for non-decapod invertebrates and 139 for fish tissue

[e] N = 84 for non-decapod invertebrates, N = 20 for decapods, and N = 131 for fish tissue.

[f] N = 33 for non-decapod invertebrates, N = 5 for decapods, and N = 543 for fish tissue.

[g] N range is 56 – 634 and varies for organism type, tissue type, and DDT compound

[h] N range is 48 – 491 and varies for organism type, tissue type, and DDT compound

[1] N range is 33 – 1,036 and varies for organism type, tissue type, and DDT compound

[j] FOD=0% for 4,4’-DDD in bivalve tissue

Observed DDT concentrations in freshwater invertebrates and fish are well below concentrations where effects are documented. Insufficient effects data were available to adequately compare with observed concentrations from invertebrates or fish from the marine nearshore and offshore environments.

Calculation of DDT doses for the two bird and two mammalian species evaluated indicate that only great blue heron are exposed to daily DDT doses above a concentration where at least one effect (reproductive) has been documented. However, for osprey, the calculated daily DDT dose is only slightly (<50%) below this lowest effects dose. For both of the mammalian species evaluated – river otter for freshwaters and harbor seal for marine waters – calculated daily DDT doses are more than an order of magnitude below doses where effects have been documented.

NTR criteria for DDT compounds are based on individual DDT compounds (4,4'-DDD, 4,4'-DDE, and 4,4'-DDT) rather than total DDT. For freshwater tissues, DDD and DDE 90th percentile concentrations exceed the NTR criterion for invertebrates other than bivalves. In nearshore marine waters, the observed 90th percentile DDT concentration exceeds the criterion for fish and invertebrates other than bivalves. None of the tissues have observed 90th percentile values exceeding the NTR criterion in offshore marine waters.

In general, the interquartile range of concentrations for all three compounds falls between the NTR criteria at the 17.5 g/d EPA recreational consumption rate and the 769 g/d Suquamish Tribal rate. This pattern is consistent across the observed concentrations in freshwater and in nearshore and offshore marine waters.

Regionally Important Biological-Effects Data

Total DDT (sum of DDT, DDE, and DDD) concentrations in stomach contents of outmigrant juvenile Chinook salmon were found to be elevated in fish from the Duwamish Estuary and Commencement Bay relative to the stomach content concentrations of fish from the Nisqually Estuary (Stein et al., 1995). Whole-body total DDT concentrations of juvenile Chinook salmon were found to be relatively high (over 1,000 ng/g lipid or 25 ng/g ww) in fish from the Nisqually, Duwamish, and Columbia River estuaries (Johnson et al., 2007).

Johnson et al. (2007) also found detectable levels of DDTs in stomach contents, with stomach content concentrations substantially higher in Columbia River and Grays Harbor juvenile Chinook than in Duwamish and Nisqually Estuary. They suggested that at the levels measured, DDTs are unlikely to cause adverse effect by themselves, but that they may contribute via additive or synergistic effects with other contaminants. Substantially higher levels of DDTs were found in adult Chinook salmon returning to the Duwamish River than in adult Chinook returning to Johnstone Strait, the Lower Fraser River, or the Deschutes River (Cullon et al., 2009).

An analysis of DDT concentrations in Pacific herring tissues found that concentrations from Puget Sound herring were 1.5 to 2.5 times higher than those from the Strait of Georgia (West et al., 2008), with Puget Sound concentrations ranging from 19 to 27 ng/g ww (240 to 330 ng/g lipid). Cullon et al. (2005) found similar levels of DDTs in a mixture of fishes designed to represent the diets of Puget Sound and Strait of Georgia harbor seals.

Grove and Henny (2008) did not find detections of DDT and only low detections of DDE (mean of 0.004-0.28 mg/kg ww) in river otter livers from Puget Sound. These DDE levels are much lower than those found in river otters living along the Columbia River (mean of 0.12-1.65 mg/kg ww). The prey items of harbor seals in Puget Sound are 1.6 times higher in total DDT (lipid weight) than those from the Strait of Georgia (Cullon et al., 2005).

Puget Sound Chinook, the major prey of southern resident killer whales, have higher body residues of DDTs and lower lipids compared to Chinook from the British Columbia coast (Cullon et al., 2009). Krahn et al. (2009) found the same pattern of killer-whale blubber concentrations as found for PCBs in males, mothers and calves; total DDT concentrations ranged from 1,000 to 160,000 ug/kg lipid.

PAHs

Polycyclic aromatic hydrocarbons (PAHs) are a class of compounds characterized by two or more fused aromatic rings composed of carbon and hydrogen. There are hundreds of such compounds, but most studies have focused on 16 compounds that were designated as “priority pollutants” in the federal Clean Water Act. These 16 compounds are:

Low Molecular Weight PAHs (LPAHs)

- Acenaphthene
- Acenaphthylene
- Anthracene
- Fluorene
- Naphthalene
- Phenanthrene

High Molecular Weight PAHs (HPAHs)

- Benzo(a)anthracene*
- Benzo(a)pyrene*
- Benzo(b)fluoranthene*
- Benzo(k)fluoranthene*
- Benzo(g,h,i)perylene
- Chrysene*
- Dibenzo(a,h)anthracene*
- Fluoranthene
- Indeno(1,2,3-c,d)pyrene*
- Pyrene

*Designated as probable human carcinogens by EPA (cPAHs)

LPAHs tend to be found at elevated concentrations in uncombusted fossil fuels, while HPAHs are formed during incomplete combustion of fossil fuels and other organic materials such as wood (PTI, 1991). However, source identification of PAHs found in the environment cannot simply be distilled down to comparisons of LPAHs to HPAHs for a variety of reasons. LPAHs and HPAHs are not typically exclusive to particular sources; releases occur from diffuse sources, and degradation processes may alter PAH compositions following their release.

Most PAHs are found in mixtures, but biochemical processes and accumulation potentials, as well as toxicity of individual PAHs, may vary considerably. Creosote, a tarry substance formed as coke distillate, is used to preserve wood and has historically been a major source of PAHs in Puget Sound, particularly in areas with pole treating operations adjacent to marine waters.

In Puget Sound, PAHs are associated with liver disease and reproductive impairment of English sole, particularly in urban bays (PSAT, 2007). Concentrations in the water are difficult to measure, but limited data suggest that typical freshwater concentrations for total PAHs are in the 0.1 – 1.0 ug/l range (Appendix D-3), with marine water column concentrations slightly lower (Serdar, 2008). Total PAHs in freshwater and marine sediments are typically 100 – 1,000 ug/kg (dw) (Appendix D-4) although mean concentrations in urban bays may be up to ten-fold higher (Partridge et al., 2005).

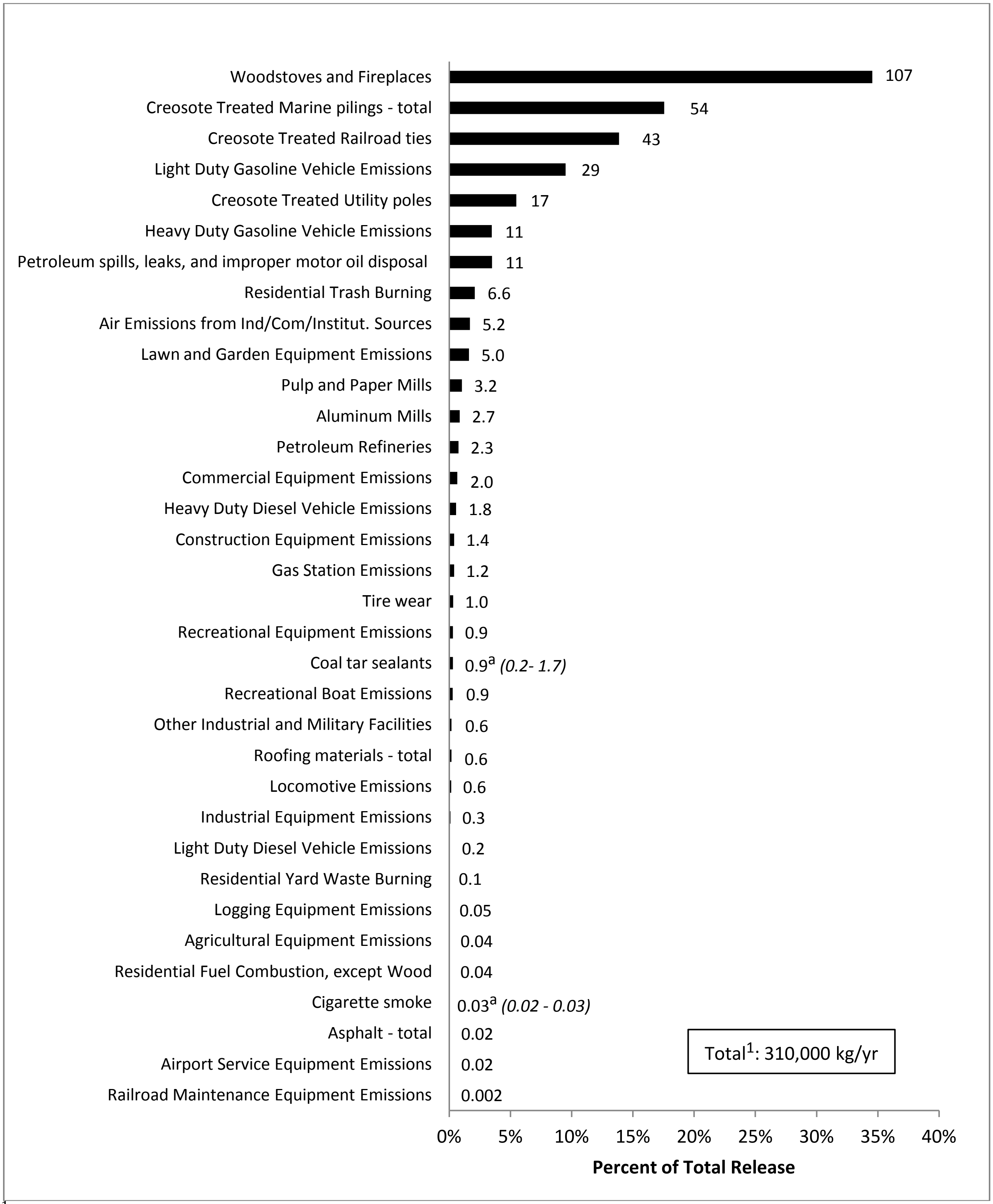
Major Releases from Primary Sources

Total annual PAH releases from sources inventoried total approximately 310,000 kg/yr in the Puget Sound basin (Figure 31). Generally speaking, the releases can be placed into two categories: combustion emissions and releases from creosote-treated wood.

Combustion releases account for most (55%) of the PAH release in the Puget Sound basin, with more than half of that amount due to woodstove and fireplace use, and smaller amounts due to residential trash burning and industrial emissions. The remainder of the combustion emissions is from petroleum fuel combustion, primarily due to gasoline use in vehicles.

Creosote-treated wood accounts for approximately one-third of the PAH release, with marine pilings (54 t/yr), railroad ties (43 t/yr), and utility poles (17 t/yr) representing the major sources.

Releases from large petroleum spills (≥ one gallon) and minor petroleum drips leaks, spillage, and improper disposal of motor oil account for PAH release of approximately 11 t/yr.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 31. Total PAH Release in the Puget Sound Basin (values shown are thousands kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for PAH loadings were obtained from PSTLA loading studies and are included in Table 20. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 20. PAH Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
		LPAH	
Groundwater (a)	7	159	311
Air Deposition	NA	NA	NA
Surface Runoff (b)	102	104	190
POTWs	3.27	8.05	34.9
Ocean Exchange	ND	ND	ND
		HPAH	
Groundwater (a)	6	124	244
Air Deposition	48.8	95.8	153
Surface Runoff (c)	25.2	36.2	50.7
POTWs	3.71	4.93	7.46
Ocean Exchange	ND	ND	ND
		cPAH	
Groundwater (a)	5	83	161
Air Deposition	20.8	43.2	69.8
Surface Runoff (d)	18.0	24.0	34.0
POTWs	NC	NC	NC
Ocean Exchange	ND	ND	ND
		Total PAH	
Groundwater (a)	13	284	555
Air Deposition (e)	48.8	95.8	153
Surface Runoff (b)	119	224	244
POTWs	7.55	18.5	45.8
Ocean Exchange	ND	ND	ND

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in commercial/industrial areas only during baseflows
(c) Detected in commercial areas only during baseflows and in commercial, residential, and agricultural areas only during storm flows
(d) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial, residential, and agricultural areas only during storm flows
(e) Total PAH based on HPAH only
NA=not analyzed
ND=not detected
NC=not calculated due to insufficient data

The estimated range of groundwater total PAH loads is large (13 – 555 kg/yr). The range in estimates is due to differences in flows used to establish the possible range of loads as well as differences in methods used to estimate representative PAH concentrations; most of the data were non-detected values. The groundwater loading calculations suggest that slightly over one-half of the groundwater loads of PAHs may be due to LPAH loads.

PAH load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about PAH mobility in groundwater. It is also notable that the “rule” used to sum individual PAHs to establish total PAHs (e.g. LPAH) resulted in higher values than results derived when applying summing rules used for other loading studies.

The deposition of atmospheric PAHs directly to the marine waters of Puget Sound is approximately 50 – 153 kg/yr, based on fluxes ranging from 0.012 – 0.69 ug/m^2/d. This estimate is based completely on HPAH; LPAHs were not analyzed. Median PAH fluxes were generally <0.05 ug/m^2/d, except for one location within a high-density urban (Tacoma) area including a nearby pulp mill, metal refiners, other industrial activities, and close to major roadways including interstate highways. PAH fluxes at this location were consistently an order of magnitude higher than other locations around the Puget Sound region, and were elevated six-fold above a nearby station that did not have the same air pollution influences in such close proximity. This pattern closely mirrored the deposition patterns of copper, lead, and zinc among the air sampling locations.

The total PAH load discharged from POTWs is estimated to be 6.6 – 46 kg/yr. Approximately one-half to three-quarters of the PAHs loaded from POTWs are LPAHs. Phase 1 estimates suggest that PAH loads from industrial wastewater are potentially substantial (2 – 87 kg/yr; Hart Crowser et al., 2007), although this is based on a limited dataset. The amount reported to be discharged to surface waters or transferred to POTWs from industries – as reported in the TRI – is also highly uncertain, with a high-end estimate of 90 kg PAH/yr (Ecology, 2011).

Surface water runoff loads for total PAH are estimated to be approximately 119 – 244 t/yr for the entire Puget Sound basin based on the surface runoff study, with LPAHs comprising approximately four-fifths of the load. The higher reported LPAH loads may be attributable in part to overall higher detection frequencies for LPAHs, although individual HPAH components were detected at generally higher frequencies than individual LPAH components.

In terms of land cover, PAHs are detected much more frequently in commercial/industrial areas (83%), about four times more frequently than in any other land cover. HPAH concentrations were highest in commercial areas, leading to loads calculated to be 16 – 32 kg/yr. Most of the remaining surface water PAH surface water loads were driven by a single LPAH constituent (phenanthrene) detected in 16% of the samples from forested lands, leading to a high calculated LPAH load for forests (102 – 190 kg/yr).

Most of the PAH detections occurred during storm events; PAHs were rarely detected during baseflows. Commercial/industrial areas were the only land covers where PAHs were detected during baseflow, and detection frequencies were so low (7% each for LPAH and HPAH) that baseflow loads for total PAH were ≤1 kg/yr under all scenarios used for load calculations.

PAHs were not detected in any of the marine water samples, and therefore no exchange could be calculated at the ocean boundary.

The net sum of total PAH loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 190, 620, and 1,000 kg/yr, respectively. Under all of these estimates, there is a net PAH load to Puget Sound. However, it was not possible to assess the possibility of a net export out of Puget Sound since no PAHs were detectable in marine waters.

PAH loading from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th - 75th percentiles) and medians for each pathway in Figures 32-35. Each pathway represented as a contribution to the total load is displayed in Figures 36-39.

The surface runoff and groundwater are the largest loading pathways, accounting for a combined total of 70 – 82% of total PAH loads. Estimates at the median and 75th percentile levels suggest that approximately one-half of the total PAH loading occurs through groundwater. Air deposition accounts for 15 – 26% of the total PAH loads, while POTW loads account for 3 – 5%.

As noted previously, estimates of PAH loads from surface runoff and groundwater are driven by concentrations derived from non-detected results. Since these are the two largest reported loading pathways for PAHs, there is a large degree of uncertainty surrounding these load estimates. The authors of the present report did not attempt to modify or otherwise recalculate loads reported in the individual loading studies. However, readers are encouraged to review the loading studies to gain an understanding of the methodologies used to estimate loads where many of the sample results were below reporting limits.

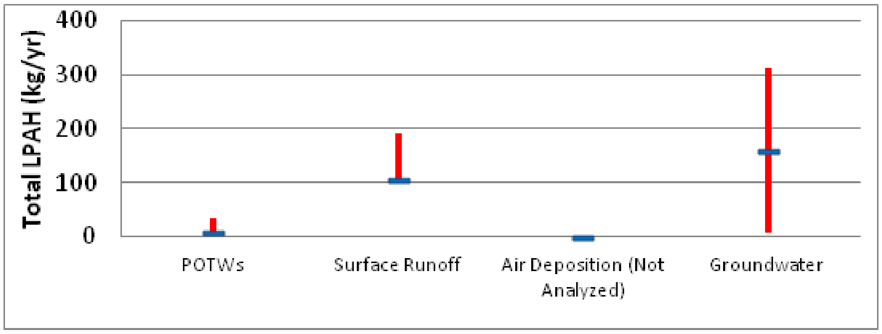


Figure 32. Range (25th - 75th percentiles) and Median LPAH Loads to Puget Sound from Each Major Delivery Pathway.

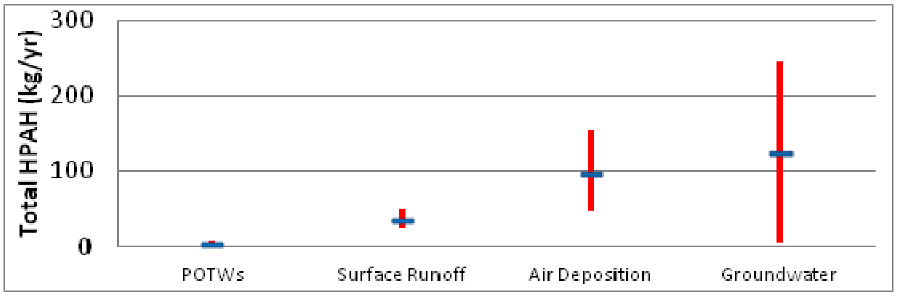


Figure 33. Range (25th - 75th percentiles) and Median HPAH Loads to Puget Sound from Each Major Delivery Pathway.

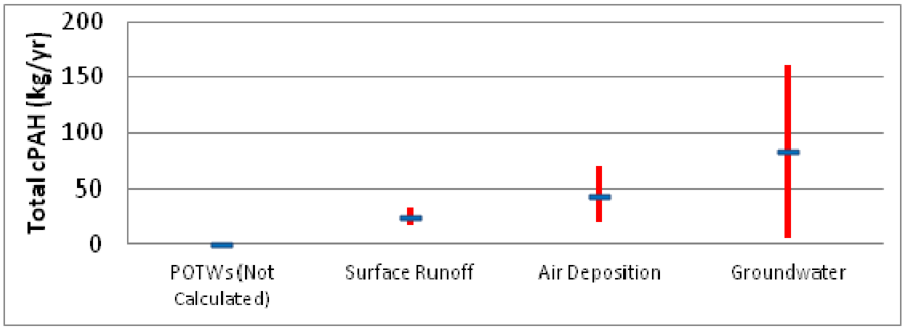


Figure 34. Range (25th - 75th percentiles) and Median cPAH Loads to Puget Sound from Each Major Delivery Pathway.

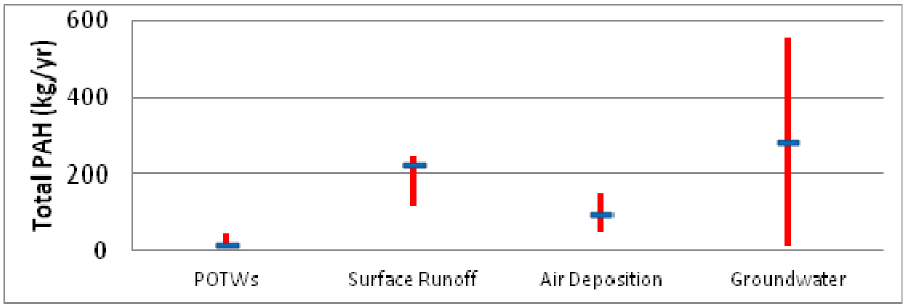


Figure 35. Range (25th - 75th percentiles) and Median Total PAH Loads to Puget Sound from Each Major Delivery Pathway.

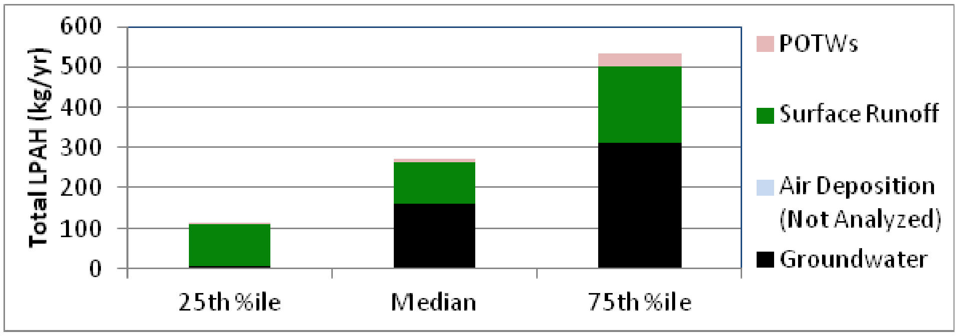


Figure 36. LPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

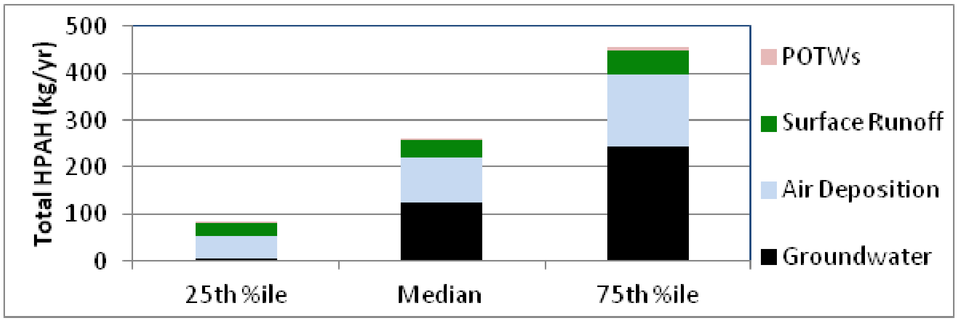


Figure 37. HPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

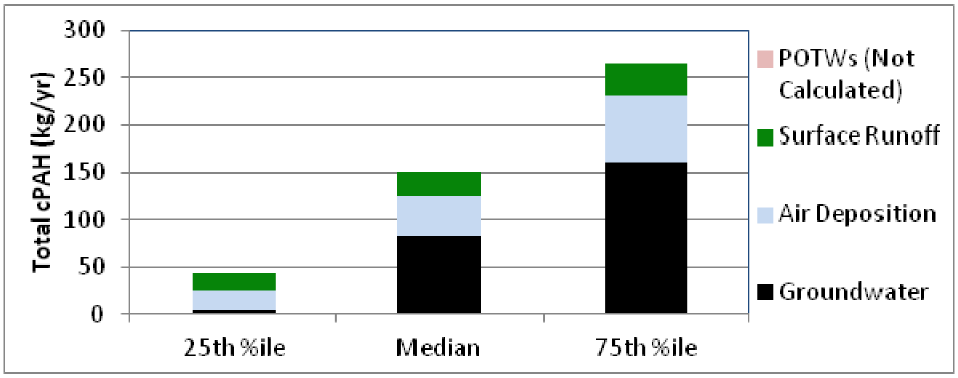


Figure 38. cPAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

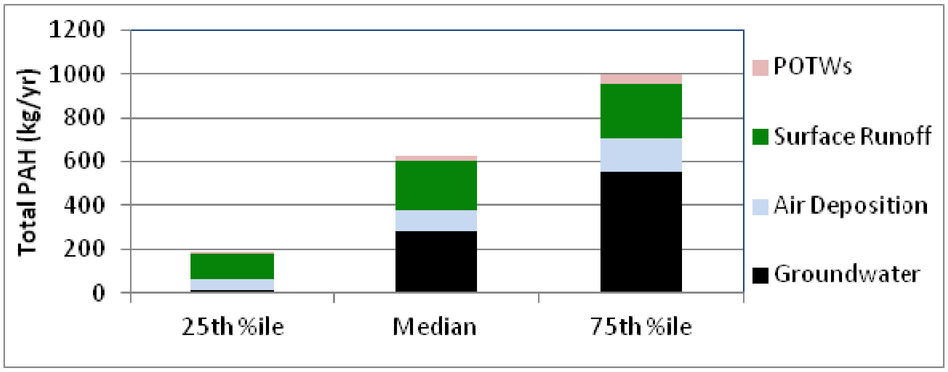


Figure 39. Total PAH Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of PAH transport and fate following release is complex, and the ability to draw conclusions is hampered by the limitations of PAH loading estimates derived for Puget Sound.

The inventory of sources (Ecology, 2011) suggests that releases to air – either in particle, aerosol, or vapor form – account for two-thirds of the PAH released in the Puget Sound basin (approximately 200,000 kg/yr). Releases of chemicals to air are by nature difficult to track through a watershed, but several generalizations may be made regarding PAH releases. For instance, nearly all of the PAHs released to air originate from combustion sources and would therefore be expected to be HPAHs, the major type of PAHs formed pyrogenically.

The Air Emissions Inventory (Ecology, 2007) indicates that woodstove use is the largest single combustion source (and overall PAH source) to the Puget Sound basin (Ecology, 2011). Through analysis of anhydrosugars (markers of wood combustion), the air deposition study appears to confirm a strong signal of woodstove use during the late winter months. Nearly all of the remaining PAH combustion sources are from petroleum, such as light-and heavy-duty vehicle emissions, and from the use of other machinery using internal combustion engines.

The air deposition study measured several LPAHs and related compounds to assess PAH source signatures. The authors concluded that all of the PAHs were derived from combustion sources, originating from a mixture of petroleum and biomass (Brandenberger et al., 2010). Geographically, the widespread and relatively homogeneous distribution of PAHs among many of the sampling stations – except those in urban areas – supports a notion of a widespread regional distribution of combustion products. Air deposition sites located in urban areas showed higher PAH concentrations apparently derived from fossil fuel combustion sources, a finding supported by correlation with metals (copper and lead) associated in part with vehicle traffic.

The air deposition study appears to be consistent with the types of PAH sources inventoried in the Puget Sound basin and with other contemporary studies of PAH sources conducted on a regional level (e.g. Stein et al., 2006). However, the amounts released are three orders of magnitude higher than the amounts delivered to Puget Sound from all major loading pathways combined. If the air deposition results were extended to the entire Puget Sound watershed (approximately 750 kg/yr at the 75th percentile), and all of the deposited PAHs were entrained and delivered through surface runoff, this load would still represent <0.5% of the combustion releases. One explanation for the difference is that PAHs loads reported for surface runoff may underestimate actual loads to Puget Sound, while at the same time releases from combustion sources may be largely overestimated. However, there are no clear lines of evidence to support either supposition.

Of the remaining non-combustion sources of PAHs (130,000 kg/yr), approximately 90% is released from creosote marine pilings, railroad ties, and utility poles (Ecology, 2011). Valle et al. (2007) estimated that in the New York/New Jersey Harbor area, approximately 13% of the PAHs released over the lifetime of these products is to air, resulting in a total loss to air of 14,000 kg/yr from all three product types combined. Remaining releases occur through leaching and washout, presumably directly to marine waters for pilings. PAH leaching from rail ties and utility poles is most likely to result in enrichment of adjacent soils (except in the case where these products are surrounded by impervious surfaces, or over-water in the case of railroad bridges). Once bound to soils, PAHs will generally remain immobile unless soils are removed or scoured through mechanical force. It is possible that some of the PAHs detected in commercial/ industrial area surface runoff during storm events were derived from utility poles or rail crossings, but the lack of major PAH components from creosote (naphthalene, acenaphthene) suggests that creosote leaching is not a major source in these instances.

Aside from PAHs released from creosote structures mounted on impervious surfaces, PAHs released from spillage of petroleum products is a potentially large source of LPAH to surface waters. An inventory of petroleum spills ≥ one gallon in the Puget Sound basin suggests that approximately 1,000 kg/yr of PAHs are released due to spillage of over 150,000 kg/yr of petroleum (Ecology, 2011). However, much larger quantities of PAHs released from small oil drips and leaks (8,500 kg/yr) and from improper disposal of used oil (1,300 kg/yr) account for the bulk of PAHs released as spills. Minor drips and leaks are likely to occur on impervious surfaces, which vastly increase the odds that PAH from this source will become entrained in runoff, some of which will find its way to surface waters. However, the results of the surface runoff study do not indicate motor oil as a major source of PAHs. Although the overall load of LPAH was higher than for HPAH, this did not appear to reflect widespread petroleum releases on impervious surfaces since the bulk of the LPAH load was from forested watersheds.

Of the POTW sources inventoried, only improper disposal of used oil is likely to be released in a constrained pathway. Most of the PAH loads discharged by POTWs is due to LPAHs, and the individual compounds frequently detected are those which are typically found at the highest concentrations in motor oil (fluoranthene, naphthalene, pyrene). This supports “down-the-drain” disposal of motor oil as a potential source of PAHs. Overall detection frequencies and concentrations of motor oil related PAHs in POTW effluent were similar between winter and summer season sampling, indicating a non-stormwater source of these PAHs and supporting the notion that improper disposal of motor oil may be a continuing PAH source to POTWs. However, there remain large differences in the quantity of PAHs released from this source (1,300 kg/yr) and the amount discharged from POTWs (8 – 46 kg/yr). At the treatment end, some quantity of PAH is presumably lost via solids removal prior to discharge, but the degree and extent of this removal is not known.

Hazard Evaluation

The hazard evaluation indicates that PAHs are a Priority 1 level of concern for freshwater sediments and for human health (Table 21). No effects data were available to adequately compare observed concentrations to tissue residue effects or effects to wildlife. The hazard evaluation indicates that levels of concern for PAHs may vary considerably depending on the media evaluated and whether PAHs are assessed individually or as groups (i.e. LPAH or HPAH).

In general, it appears that observed PAH concentrations in surface waters are much lower than effects data, although effects data are limited for freshwaters and observed data are limited for marine waters. Where both observed data and effects data were sufficient to conduct adequate comparisons, 90th percentile values of the observed concentrations were generally well below the 10th percentile values for effects concentrations.

Table 21. Summary of Hazard Evaluation for PAHs.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	a	≥5%	No[b]/INS[c]	Priority 2/U
	Nearshore Marine	11-12	>8%	INS[d]	U
	Offshore Marine	e	<4%	No[b]/INS[f]	Priority 2/U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	g	>25%	Yes/No[h]	Priority 1/2
	Nearshore Marine	i	≥44%	No	Priority 2
	Offshore Marine	j	>34%	No	Priority 2
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	k	>21%[l]	Yes/No[m]	Priority 1/2
	Nearshore Marine	n	>20%	Yes/No[o]	Priority 1/2
	Offshore Marine	p	>2%	No[b]/INS[f]	Priority 2/U

Yellow highlight indicates Priority 1 Level of Concern

FOD=Frequency of detection

[a] N range is 1,447 – 1,577 and varies for Individual PAH or PAH group

[b] No for several individual PAHs

INS[c]=Insufficient effects data available for comparison to observed data for several individual PAHs

U=Unknown level of concern due to lack of sufficient data for adequate comparison

INS[d]=Insufficient observed data available for comparison to effects data

[e] N range is 12 – 84 and varies for individual PAH or PAH group

INS[f]=Insufficient observed data available for comparison to effects data for several individual PAHs

[g] N range is 284 – 1,182 and varies for individual PAHs or PAH group

[h] No for benzo(a)anthracene; Yes for all other individual PAHs or PAH groups

[i] N range is 196 – 1,051 and varies for individual PAHs or PAH group

[j] N range is 217 – 906 and varies for individual PAHs or PAH group

[k] N range is 17 – 102 and varies for organism type, tissue type, and for individual PAHs

[l] FOD=0% for fish tissue

[m] Yes for 5 of 9 individual PAHs

[n] N range is 50 – 117 and varies for organism type, tissue type, and for individual PAHs

[o] Yes for 4 of 9 individual PAHs

[p] N range is 14 – 74 and varies for organism type, tissue type, and for individual PAHs

In freshwater sediments, for all individual PAHs (except benzo(a)anthracene), LPAH, and HPAH, the 90th percentile values of observed concentrations are above the floating percentile SQS. In many cases, more than 25% of the values exceed this threshold.

In marine sediments, none of the 90th percentiles of observed concentrations exceed the SQS. Median concentrations of LPAH in marine sediments are much closer to the lowest guideline, and the 75th percentile concentrations exceed this level (Canadian TEL). Sediments located in the nearshore environment had slightly higher LPAH concentrations than those located offshore. The 95th percentiles of nearshore LPAH concentrations exceed all of the guidelines. However, on an organic carbon-normalized basis, at least 95% of the nearshore and offshore concentrations are below the lowest guideline. HPAH concentrations in marine sediments mirror the patterns (relative to guidelines) of LPAHs, except median HPAH concentrations in both nearshore and offshore sediments exceed the lowest guideline.

Human health concerns were evaluated for nine of the 16 individual PAHs, but not for LPAH or HPAH since there are no NTR criteria for PAHs as groups. Several individual PAHs pose a Priority 1 level of concern for human health based on comparisons to the NTR criteria. For the nine PAHs evaluated for human health, benzo(a)pyrene, benzofluoranthenes (b and k), dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene exceed the NTR for at least one organism type in either freshwater or marine waters.

Regionally Important Biological-Effects Data

Multiple investigations have identified biomarkers of exposures to PAHs in various Puget Sound fishes. Bile and stomach content of outmigrant juvenile Chinook salmon were found to contain various PAHs (Johnson et al., 2007), demonstrating that diet pathways are important for PAH exposures. The authors suggest that the levels of exposure may result in immunosuppression and other health effects. These results expanded and confirmed previously documented PAH exposures (Stein et al., 1992; Stein et al., 1995). A dietary feeding study on juvenile Chinook documented growth and physiological responses from dietary exposures to PAHs at concentrations that were environmentally realistic in the Puget Sound (Meador et al., 2006).

Biomarkers of PAH exposures were confirmed in Puget Sound English sole, rock sole, and starry flounder collected from up to five sites in Puget Sound (Stein et al., 1992). Stein et al. (1992) found the biomarkers of exposure were related to the degree of sediment contamination. Further field study by Johnson (2000) resulted in recommended various threshold sediment concentrations of PAHs to protect English sole against liver lesions, DNA adducts in liver, and other effects. The causal relationship between elevated sediment PAH concentrations and English sole liver effects was confirmed by Myers et al. (2003). In a study of English sole from the Hylebos Waterway and Colvos Passage, Sol et al. (2008) found no correlation between PAH exposure and age and little correlation between reproductive end points and PAH exposure. However, Pacific herring embryos were found to be affected by tricyclic PAHs in weathered crude oil (Incardona et al., 2009; Carls et al., 1999).

Several laboratory studies have documented that developmental defects in fish are associated with exposures to PAHs released by weathered crude oil, notably the tricyclic-PAHs (Incardona et al., 2005; Incardona et al., 2006; Carls et al., 2008). Carls and Meador (2009) developed a description of the oil weathering, PAH toxicity, and embryo exposures to explain the observed toxicity from PAHs in weathered oil at relatively low levels. Driscoll et al. (2010) developed a framework for describing PAH exposure as a dose to fishes in order to understand the mechanisms of exposure and toxicity.

Bis(2-ethylhexyl) phthalate

Phthalates are 87% of the 10.4 billion pounds per year world market for plasticizers, with bis(2-ethylhexyl) phthalate (a.k.a. di(2-ethylhexyl) phthalate [DEHP]) as the most common plasticizer for polyvinylchloride (PVC), constituting more than 50% of the phthalates produced (SPWG, 2007; ECB, 2008). Approximately 90-95% and possibly as much as 97% of DEHP is used to plasticize PVC products, which may contain up to 60% DEHP, although 30% may be more typical (ECB, 2008). Roughly 200-300 million pounds of DEHP are produced in the U.S. each year, with unknown amounts contained in imported products. According to ATSDR (2002), relatively little is imported (4 million pounds in 1998) or exported (14-27 million pounds per year in 1994-1998).

Phthalates are not covalently bound to PVC polymer chains and migrate out over time (Rudel and Perovich, 2009). The amount of phthalates that leach out of the PVC into air is quite variable. Factors that affect the rates of volatilization include the temperature and surface area of the material; emission rates among phthalates may also be quite variable. At a certain point the PVC undergoes glassification when it becomes stiff and brittle, and very little phthalate continues to off-gas. Once emitted, phthalates tend to adhere to dust particles rather than remain in vapor phase (ATSDR, 2002; SPWG, 2007).

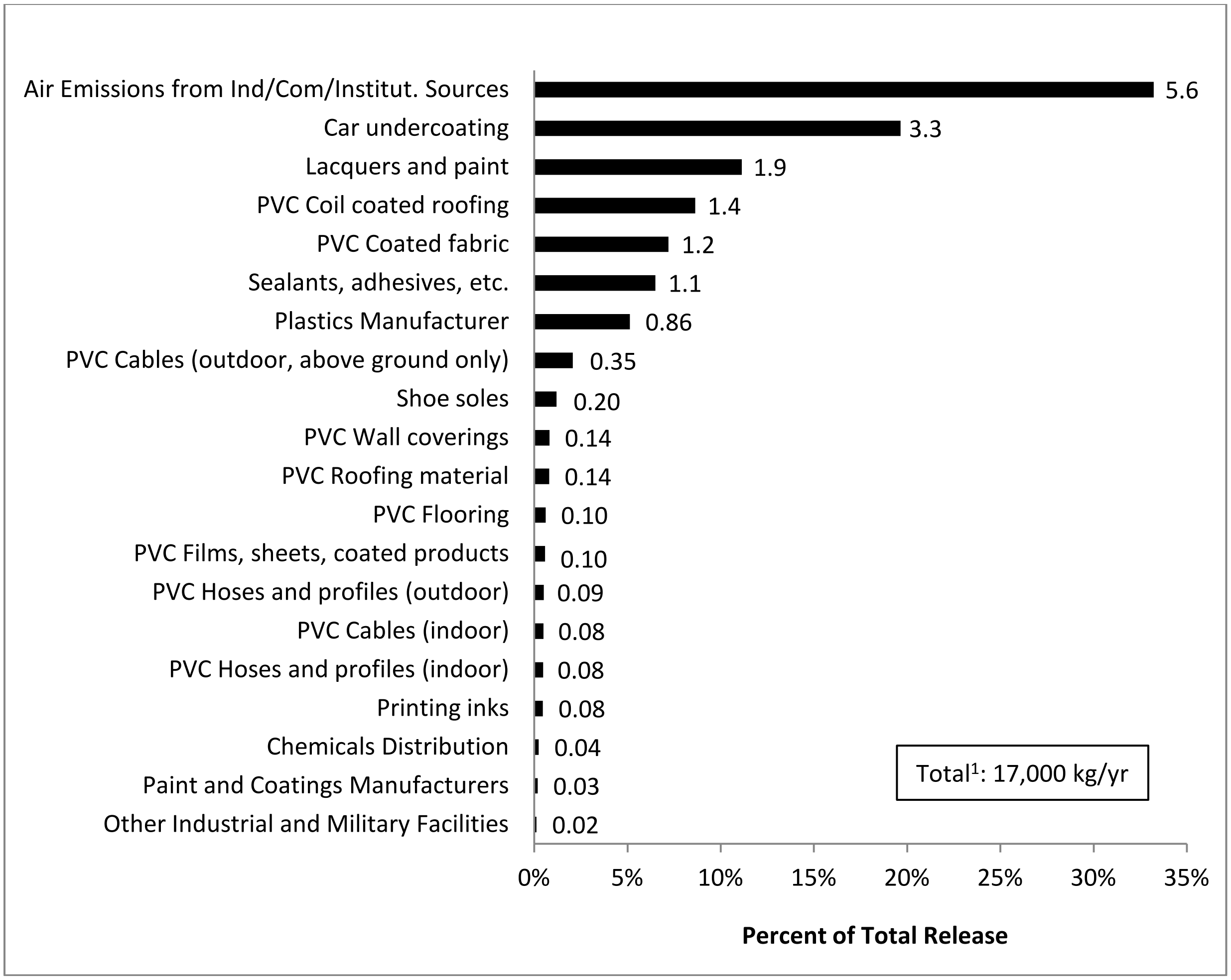
Plasticized PVC products are widely used and include exterior siding and roofing materials, automobiles, wires/cabling, advertizing banners, flooring, weather stripping, upholstery, garden hoses, swimming pool liners, footwear, clothing, food containers, tablecloths, shower curtains, rainwear, and toys. Rigid PVC products, such as pipes and windows, do not contain phthalates or other plasticizers. In Western Europe, DEHP emissions were estimated at 300 tons per year from indoor uses and 2,600 tons per year from exterior uses after measuring emissions to the air from PVC products (ATSDR, 2002). The population of Western Europe is about 400 million people, compared to about six million people in Washington State.

Non-plasticizer (non-polymer) uses of DEHP are a small percentage (<10%) of overall use. These uses include PCB replacement (dielectric fluids for electric capacitors), de-foaming agents in paper manufacturing and detergents, as well as chemical intermediates for insect repellent, cosmetics, lacquers, munitions, ceramics, printing inks, adhesives, sealants, and industrial lubricants.

Once in the environment, DEHP biodegrades in water but may accumulate in aquatic organisms to some degree (PTI, 1991). It has relatively low solubility and may resist degradation once bound to soil particles. In the aquatic environment, DEHP is nearly always detectable at concentrations >0.01 ug/l in freshwater and marine waters, and >0.01 mg/kg (dw) in sediment (PTI, 1991; Appendices D-3 and D-4).

Major Releases from Primary Sources

The inventory of phthalate sources in the Puget Sound basin suggests that total phthalate release is approximately 34,000 kg/yr for the six phthalates assessed (Ecology, 2011). Emissions from plasticized PVC products are the primary source of phthalates (SPWG, 2007), with DEHP as the dominant phthalate used as a plasticizer. In addition to releases from polymer use, DEHP releases occur through non-polymer uses, for a total annual release estimated to be 17,000 kg/yr (Figure 40).



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 40. Total DEHP Release in the Puget Sound Basin (values shown are thousands kg/yr).

DEHP release from polymer use accounts for approximately 40% of the total annual release, with large contributions from car undercoating (3,300 kg/yr), coil coated roofing (1,400 kg/yr), and coated fabric (1,200 kg/yr). Indoor polymer use accounts for approximately 5% of the loss through polymers.

Releases of DEHP from non-polymer uses include loss from lacquers, paints, sealants, adhesives, and printing inks. Combined, these account for approximately 15 - 20% of DEHP release in the Puget Sound basin. The remaining 40% (6,600 kg/yr) of DEHP releases are through industrial, commercial, and institutional point-source air emissions.

Total phthalate releases from personal care products combined are approximately 11,000 kg/yr including releases from fragrances, deodorant, nail polish, hair spray, and body lotion. According to testing data from U.S. and Swedish non-governmental organizations, diethyl phthalate (DEP) is the primary phthalate used in cosmetics and personal care products, with some products also containing di-n-butyl phthalate (DBP). However, there appears to be little if any use of DEHP in these products (DiGangi and Norin, 2002; Houlihan et al., 2002; EPA, 2009).

Major Delivery Pathways and Loading

Data on major delivery pathways for DEHP loading were obtained from PSTLA loading studies and are included in Table 22. No effort was made to assess DEHP loads from direct atmospheric deposition of DEHP to Puget Sound during Phase 3 (Brandenberger et al., 2010). As mentioned previously, loadings through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Table 22. DEHP Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater (a)	14	227	440
Air Deposition	NA	NA	NA
Surface Runoff (b)	1,746	1,777	1,863
POTWs	216	439	904
Ocean Exchange	NC	NC	NC

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in residential areas only during baseflows
NA=not analyzed
NC=not calculated due to insufficient data

The estimated range of groundwater DEHP loads is large (14 – 440 kg/yr). The range in estimates is due primarily to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude. DEHP load estimates in groundwater should be used with caution since they are based primarily on non-detect data and numerous assumptions, including unconfirmed assumptions about DEHP mobility in groundwater.

DEHP loads discharged from POTWs are approximately 220 – 900 kg/yr. Although field blank contamination and higher-than-desired laboratory quantitation limits for some samples resulted in uncertainty regarding some of the results, it appears that detection frequencies and concentrations between seasons (wet and dry) were not substantially different. However, seasonal first-flush concentrations were evident in agricultural sub-basins.

Surface water runoff loads for DEHP are estimated to be approximately 1,750 – 1,860 kg/yr for the entire Puget Sound basin based on the surface runoff study. DEHP was rarely detected during baseflows (3% frequency of detection) but was detected at a moderate frequency (30%) during storm flows, mostly in commercial/industrial areas. Although overall concentrations were slightly higher in commercial/industrial areas, loads generally mirror the amount of land cover represented by each land cover category; forested areas account for approximately 84% of the total DEHP load in surface runoff load to Puget Sound.

Loads of DEHP in marine waters were not calculated due to the low frequency of detection. DEHP was detected in only three samples – two in Hood Canal and one in the northern boundary waters – at low levels (≤0.06 ug/l).

The net sum of DEHP loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sum for the 25th, median (50th), and 75th percentile values is 2,000, 2,400, and 3,200 kg/yr, respectively. Under all of these estimates, there is a net DEHP load to Puget Sound.

DEHP loadings from pathways that may represent *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange) are shown as ranges (25th -75th percentiles) and medians for each pathway in Figures 41. Each pathway represented as a contribution to the total load is displayed in Figures 42.

Total DEHP loading from the major pathways assessed is 2,000 – 2,800 kg/yr. Surface runoff accounts for the largest pathway (58 – 88%), followed by POTWs (11 – 28%) and groundwater (1 – 14%).

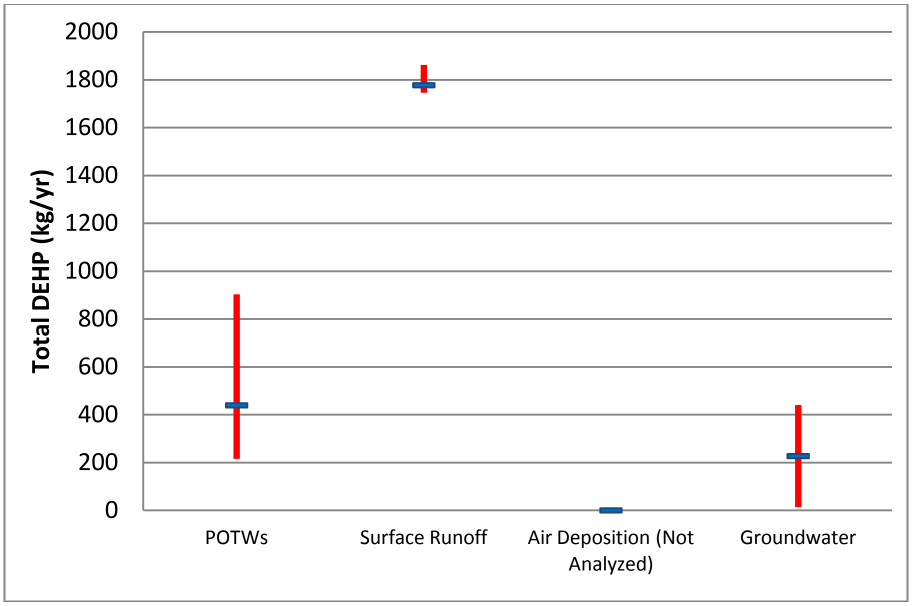


Figure 41. Range (25th - 75th percentiles) and Median DEHP Loads to Puget Sound from Each Major Delivery Pathway.

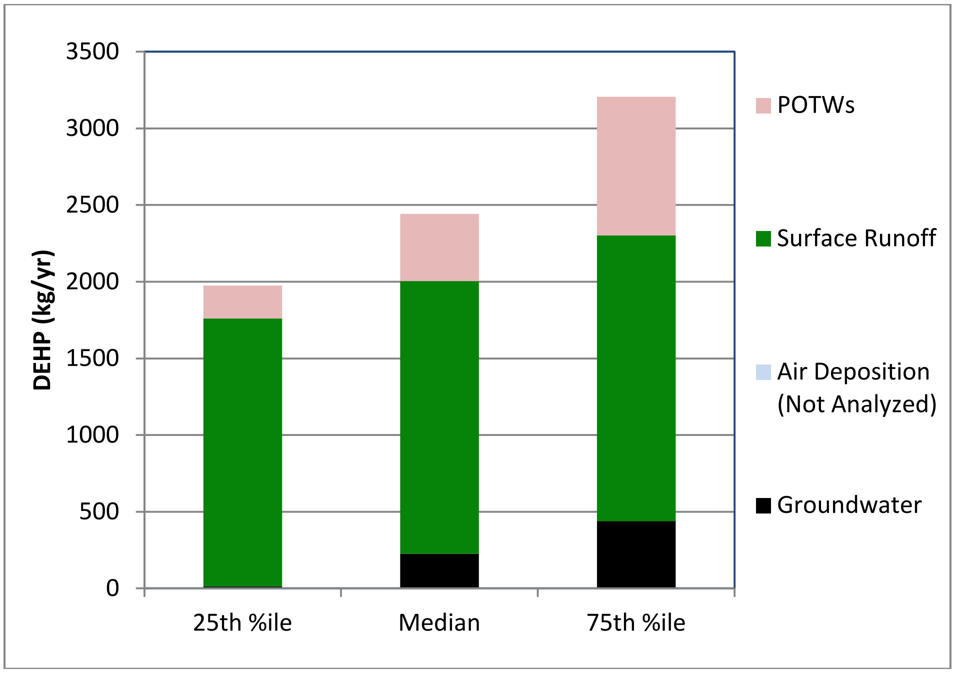


Figure 42. Total DEHP Loads to Puget Sound as the Sum of Major Delivery Pathways.

Relationship Between Sources and Pathways/Loading

The conceptual model of DEHP transport and fate following release is limited due to the low frequency of detection in surface runoff and the lack of data on DEHP in atmospheric deposition. DEHP depositions to Puget Sound of 310 to 16,000 kg/yr were estimated during the Phase 1 project, but these data were based largely on very limited sampling in an urban area (Hart Crowser et al., 2007) and may not have been representative for the Puget Sound basin. Data on atmospherically deposited DEHP loads would have proven particularly useful since the primary sources of DEHP suggest that volatilization is the principal release mechanism, followed by attachment to dust particles. In this respect, DEHP may be similar to PBDEs in their transport and fate in the environment. If DEHP follows a similar mode of fate and transport to that of PBDEs, the air deposition and POTW pathways would be expected to deliver loads of the same relative magnitude as those for surface runoff.

A portion of the DEHP emitted from both polymer and non-polymer sources is likely to occur indoors, and some fraction of these releases is presumably delivered to the sanitary sewer system. Estimates of DEHP delivered to POTWs through washing machine rinse water were conducted by the Washington Toxics Coalition and People for Puget Sound during 2009 (WTC/PPS, 2009). They estimated that roughly 960 kg/yr DEHP are delivered to Puget Sound POTWs each year due to dust-bound DEHP which becomes attached to clothing and is subsequently rinsed down the drain during the washing process.

As pointed out by the authors of the POTW loading report (Ecology and Herrera, 2010), it is difficult to draw conclusions regarding seasonal differences in COC loads from POTWs. However, it appears that POTWs discharge fairly constant DEHP loads between seasons, suggesting the presence of a steady source of DEHP. This appears to be consistent with the indoor emissions of this compound, followed by attachment to dust particles and delivery to POTWs through sanitary drains. The inventory of sources suggests that approximately 500 kg/yr DEHP is released indoors through loss from polymers, although some of the DEHP release from non-polymer use (3,000 kg/yr) may also occur indoors. While neither the WTC/PPS report nor the present loading studies provide enough information to calculate a mass-balance of DEHP delivered to, and discharged from, POTWs, the information suggests that the amount delivered to POTWs is likely on the order of thousands of kg per year.

Some of the DEHP delivered to POTWs is likely to be from stormwater, but the contribution from this pathway may be comparatively small. Surface runoff DEHP loads in areas where stormwater is potentially diverted to POTWs (i.e. commercial/industrial and residential areas) are calculated to be approximately 120 – 230 kg/yr during storm events (Herrera, 2011). Therefore, the DEHP load in stormwater diverted to POTWs is presumed to be only a fraction of that amount.

Hazard Evaluation

The hazard evaluation indicates that DEHP is a Priority 1 level of concern for sediments and for human health (Table 23). Hazard due to tissue residue effects and effects to wildlife were not evaluated.

In fresh, nearshore marine, and offshore marine surface waters, the 90th percentile of DEHP concentrations (approx. 4 ug/l, 2 ug/l, and 3 ug/l, respectively) is two orders of magnitude below the lowest 10% of effects for freshwater, and perhaps even a greater magnitude below 10% of the effects data for marine waters, although the latter is difficult to assess due to a paucity of effects data. No water quality criteria are available for comparison.

More than 25% of the DEHP concentrations in freshwater exceed the floating percentile SQS as well as the CSL, and the median concentration (approx. 120 ug/kg dw) approaches the SQS. In marine sediments, the 90th percentiles of the nearshore and marine sediments (approx. 10,000 ug/kg organic carbon and 5,000 ug/kg organic carbon, respectively) exceed the SQS, and the 90th percentile of nearshore DEHP concentrations exceed the CSL as well.

DEHP is rarely detected in freshwater or marine fish tissue, but it is detected more frequently in bivalves and (in the case of freshwater) other invertebrates as well. The 90th percentile of DEHP concentrations in freshwater bivalves (approx. 240 ug/kg) is slightly above the NTR criterion. The nearshore marine fish tissue concentration exceeds the NTR criterion as well (at the 90th percentile level), but this is based on few data (n=8 detected concentrations). The 95th percentile concentrations in other freshwater invertebrate tissues approach the NTR criterion, and the median values for freshwater bivalves, other freshwater invertebrates, and marine nearshore bivalves all exceed the criterion adjusted to the EPA-recommended subsistence rate (142.4 g/d).

Table 23. Summary of Hazard Evaluation for DEHP.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	1,484	84%	No	Priority 2
	Nearshore Marine	12	92%	INS	U
	Offshore Marine	84	54%	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	548	87%	Yes	Priority 1
	Nearshore Marine	513	74%	Yes	Priority 1
	Offshore Marine	474	67%	Yes	Priority 1
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Freshwater	a	>57%[b]	yes	Priority 1
	Nearshore Marine	c	>9%	yes	Priority 1
	Offshore Marine	d	>24%[b]	INS	U

Yellow highlight indicates Priority 1 Level of Concern
FOD=Frequency of detection
INS=Insufficient observed data available for comparison to effects data
[a] N range is 9 – 99 and varies for organism type and tissue type
[b] FOD=0% for fish tissue
[c] N range is 42 – 79 and varies for organism type and tissue type
[d] N range is 16 – 33 and varies for organism type and tissue type

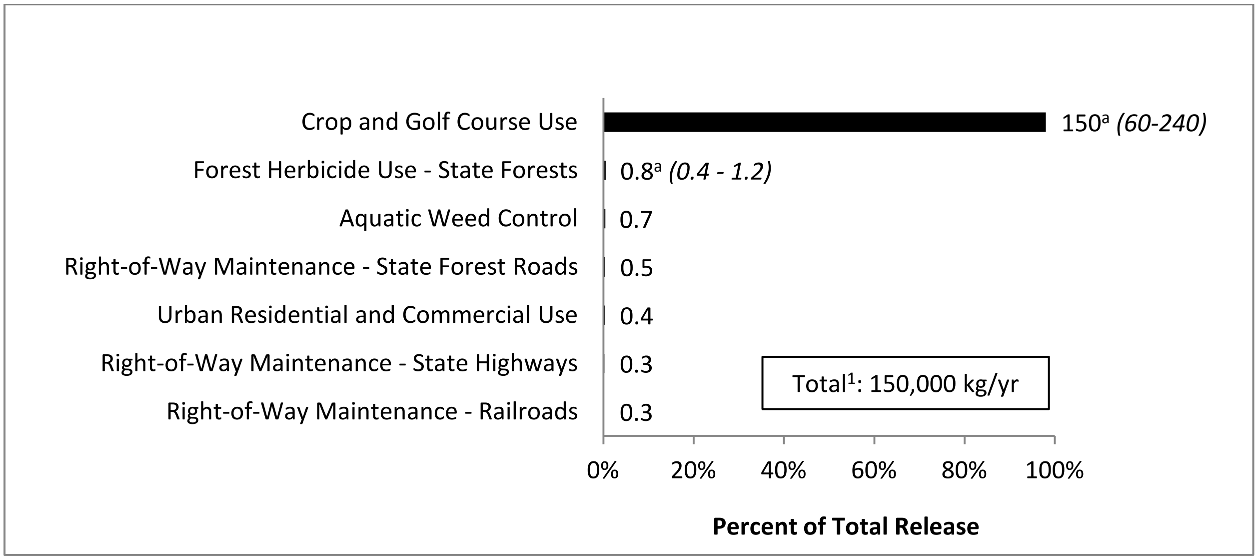
Triclopyr

Triclopyr [((3,5,6-trichloro-2-pyridinyl)oxy)acetic acid)] is a broad-leaf herbicide used primarily for rice, pasture and rangeland, rights-of-way, and turf, including home lawns and gardens (EPA, 1998). It also has limited use to control nuisance vegetation in freshwater lakes and is permitted under Ecology’s Aquatic Pesticide General Permit. Triclopyr is applied as the triethylamine (TEA) salt and butoxyethyl ester (BEE) formulations and is sold under the product names Garlon® and Crossbow® for terrestrial use and Renovate® for aquatic use.

Following release in the environment, triclopyr formulations are dissociated to the triclopyr anion which is readily soluble in water. Half-lives of triclopyr formulations following terrestrial applications are generally 10 days or less. In terrestrial soils, triclopyr undergoes microbial degradation with a half-life generally less than 20 days. Once in the aquatic environment, photolysis is the major degradation mechanism and occurs rapidly, generally with a half-life of several days or less (EPA, 1998). Although triclopyr half-lives tested under laboratory conditions suggest little persistence, environmental studies conducted following applications indicate that actual half-lives may be substantially greater, perhaps on the order of 100 days or more. Due its solubility, absorption to soil particles is not a major fate process, and triclopyr does not bioaccumulate in aquatic organisms.

Major Releases from Primary Sources

Total triclopyr release from the seven application categories assessed totaled approximately 150,000 kg/yr (Figure 43). Agricultural uses of triclopyr accounted for 76 – 94% of the total release, with golf course use accounting for 5 – 22%. The remaining triclopyr use was primarily for right-of-way maintenance (0.7%), aquatic weed control (0.5%), and domestic use (0.3%). The small amounts calculated for domestic and forestry use likely underestimate actual usage rates.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 43. Total Triclopyr Release in the Puget Sound Basin (values shown are thousands kg/yr).

Major Delivery Pathways and Loading

Data on major delivery pathways for triclopyr loading were obtained from PSTLA loading studies and are included in Table 24. No effort was made to assess triclopyr loads from direct atmospheric deposition or exchange of ocean waters. Due to the low frequency of detection in POTW effluent, loads were not calculated (Ecology and Herrera, 2010). No triclopyr data were found for groundwater, and therefore no attempt was made to calculate loading from direct groundwater discharge to Puget Sound (Pitz, 2011).

Table 24. Triclopyr Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff (a)	641	652	686
POTWs	NC	NC	NC
Ocean Exchange	NA	NA	NA

(a) Detected in commercial/industrial and agricultural areas only during baseflows
NA=not analyzed
NC=not calculated due to insufficient data

Surface runoff loads for triclopyr are estimated to be approximately 641 – 686 kg/yr for the entire Puget Sound basin based on the surface runoff study. Triclopyr was detected more frequently during storm flows than during baseflows, with the highest rate of detection during the fall first flush (63%). Overall detection rates were highest in commercial/industrial areas (47%), followed by residential areas (41%), agricultural areas (31%), and forests (16%). Detected concentrations varied little across different land covers, with typical levels in surface water of 0.03 ug/l. Although triclopyr was detected only in commercial/industrial and agricultural areas during baseflows, overall loads generally mirrored the amount of land cover represented by each land cover category. As a result, forested areas account for approximately 90% of the total surface runoff load to Puget Sound.

Hazard Evaluation

Triclopyr hazards were difficult to evaluate due to the lack of observed environmental data for all media except fresh surface waters (Table 25). Over 1,600 measurements of triclopyr are available for fresh surface waters, with a detection frequency of 33%. The 90th percentile of these detected triclopyr values is approximately 0.1 ug/l, three orders of magnitude lower than the 10th percentile of the effects data.

Table 25. Summary of Hazard Evaluation for Triclopyr.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	1,632	33%	No	Priority 2
	Nearshore Marine	0	--	INS	U
	Offshore Marine	0	--	INS	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	0	--	INS	U
	Nearshore Marine	0	--	INS	U
	Offshore Marine	0	--	INS	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection
INS=Insufficient effects or observed data available for evaluation

Nonylphenol

Nonylphenol is a commonly detected organic chemical of particular interest due to its potential to disrupt endocrine function in aquatic organisms. While there are limited uses for the direct use of nonylphenol, it has a broad range of applications as a precursor for nonionic surfactants, particularly alkylphenol ethoxylates which are used in domestic and industrial cleaning products and emulsifiers. These include uses as domestic and commercial laundry detergents, pulp and paper processing, and as deicers and in firefighting foams and gels. Annual production in the U.S. is on the order of 200 to 300 millions of pounds per year (EPA, 2005).

The breakdown of alkylphenol ethoxylates to nonylphenol occurs largely during the sewage treatment process, although the breakdown process does not require active sewage sludges for this to occur (EPA, 2005). Nonylphenol is more resistant to further degradation than its parent compounds. It has low solubility in water and partitions to sediments where its resistance to degradation is generally increased. Based on log K_{ow} values, nonylphenol's capacity for bioaccumulation is considered moderate, but it is generally found at low concentrations in fish tissue, possibly suggesting fish have some ability to metabolize nonylphenol (EPA, 2005).

Major Releases from Primary Sources

Total nonylphenol release in the Puget Sound basin totals approximately 180 kg/yr. All of the inventoried nonylphenol releases were from point-source air emissions. No effort was made to calculate nonylphenol releases from diffuse sources. Nonylphenol is not a required reporting requirement under the TRI, and therefore no industrial or institutional nonylphenol releases were catalogued (Ecology, 2011).

Major Delivery Pathways and Loading

Major delivery pathways for nonylphenol loading are included in Table 26. No effort was made to assess nonylphenol loads from direct atmospheric deposition. Due to the low frequencies of detection, nonylphenol was not detected in POTW effluent or in exchange of ocean waters (Ecology and Herrera, 2010; Gries and Osterberg, 2011). No nonylphenol data were found for groundwater, and therefore no attempt was made to calculate loading from direct groundwater discharge to Puget Sound (Pitz, 2011).

Table 26. Nonylphenol Loads (kg/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff (a)	23	23	24
POTWs	NC	NC	NC
Ocean Exchange	NC	NC	NC

(a) Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows
NA=not analyzed
NC=not calculated due to insufficient data

Surface water runoff loads for nonylphenol are estimated to be approximately 23 – 24 kg/yr for the entire Puget Sound basin based on the surface runoff study. There was a single detection of nonylphenol among the 126 samples analyzed in surface runoff, and therefore no patterns related to land cover types or hydrological conditions could be established.

Hazard Evaluation

Nonylphenol hazards were difficult to evaluate due to the lack of observed environmental data in some media and a lack of effects data for other media (Table 27). For fresh surface waters, however, there are ample observed data as well as effects data. The 90th percentile of observed concentrations of 4-nonylphenol (approx. 4 ug/l) is slightly below the chronic water quality criterion and several times lower than the 10th percentile of effects data; the 95th percentile of observed values exceeds the chronic criterion.

Table 27. Summary of Hazard Evaluation for 4-Nonylphenol.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	539	26%	No	Priority 2
	Nearshore Marine	11	27%	INS[a]	U
	Offshore Marine	84	17%	No	Priority 2
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	251	12%	INS[b]	U
	Nearshore Marine	67	15%	INS[b]	U
	Offshore Marine	91	29%	INS[b]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
				Not Analyzed	
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
				Not Analyzed	
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
				Not Analyzed	

FOD=Frequency of detection
INS[a]=Insufficient observed data available for comparison to effects data
U=Unknown level of concern due to lack of sufficient data for adequate comparison
INS[b]=Insufficient effects data available for comparison to observed data

In marine surface waters, there are few observed data of 4-nonylphenol for the nearshore (n= 11) dataset. For the offshore marine waters, there are sufficient data (n=84) for comparisons to effects and criteria. Concentrations for all marine water data are well below the water quality criteria and at least 95% of the effects data.

Of the guidelines used to assess hazards in sediments, the Canadian TEL was the only guideline with numerical values for nonylphenol; no SQS was available for freshwater or marine sediments. In freshwater sediments at least 5% of the values exceeded the TEL and over 10% of the values exceeded the TEL in marine sediments. However, the TEL was above the 95th percentile values in offshore sediments.

There are no data to assess residue effects levels in aquatic organisms, or to evaluate nonylphenol hazards to wildlife or humans. This likely reflects nonylphenol's low to moderate accumulation potential in fish and macroinvertebrates.

Oil & Grease and Petroleum Hydrocarbons

Unlike other COCs, oil & grease does not refer to one or more specific chemicals, but instead refers to a group of chemicals with loosely related chemical and physical properties. These chemicals include non-volatile hydrocarbons, vegetable oils, animal fats, waxes, soaps, greases, and related materials. The working definition of oil & grease is based on the analytical method to determine the concentration in water or soil. More specifically, oil & grease is the fraction of a sample that is extractable by n-hexane, and is sometimes referred to as hexane extractable material (HEM).

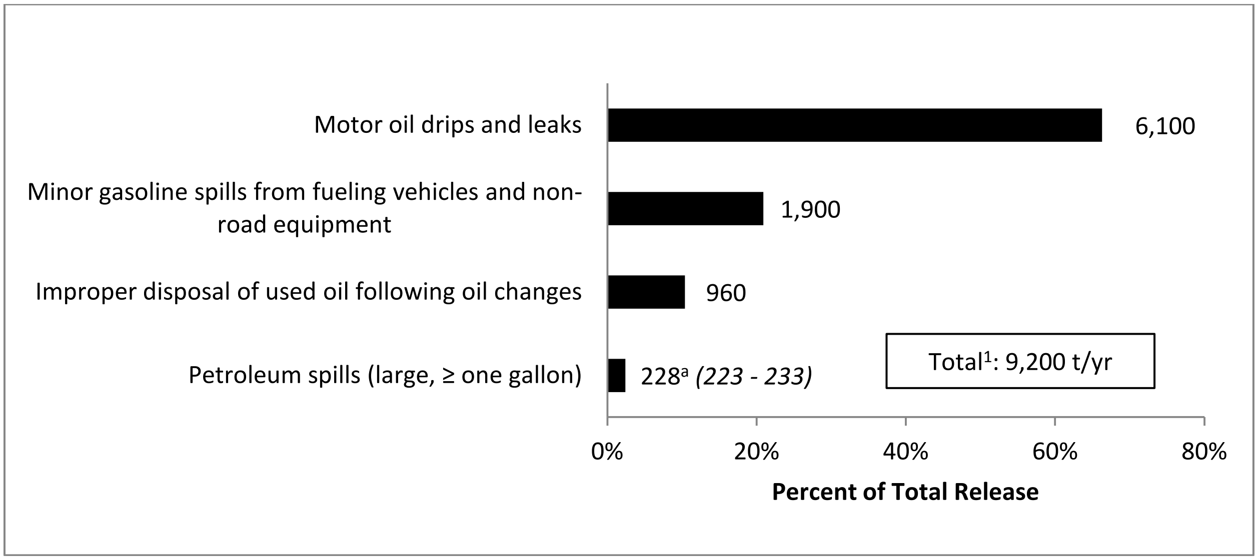
Like oil & grease, petroleum is not a specific compound, but may instead contain hundreds of chemicals, with crude oil as the ultimate source. Petroleum hydrocarbons – sometimes referred to as total petroleum hydrocarbons (TPH) – may further be divided into fractions based on the number of carbons of the major constituents. Gasoline range organics (GRO) refers to the methanol extractable fraction of organic chemicals with six to ten carbons, and generally includes a variety of gasoline alkanes, naphtha, mineral spirits, stoddard solvent, and other volatile petroleum products. Diesel range organics (DRO) are the fraction extracted with methylene chloride and have 11 – 28 carbons, and covers semi-volatile petroleum products (jet fuels through heavy fuel oils). Lube oils are typically characterized by molecules containing 29 – 35 carbons.

Major Releases from Primary Sources

The report on COC sources in the Puget Sound basin (Ecology, 2011) estimated releases of petroleum from various sources. Oil & grease releases were not estimated. Petroleum releases in the Puget Sound basin were inventoried by estimating releases from four major categories: petroleum spills ≥ one gallon, motor oil drips and leaks, minor gasoline spills, and improper disposal of used motor oil. The total quantity of petroleum released annually in the Puget Sound basin was estimated to be 9,200 metric tons (t)/yr (Figure 44). Other possible sources of petroleum including aviation fuel leaks and uncombusted oil and fuel discharged from marine engines were not estimated.

Petroleum spills ≥ one gallon were estimated to account for releases of 230 t/yr, with approximately one-half of the releases occurring in freshwaters, much of which occurs in the Lake Washington-Lake Union-Ship Canal system. Of the remaining spills, most of the volume was released to soils, followed by unknown receiving media and marine waters. Approximately one-quarter of the spill quantity was diesel fuel, followed by gasoline (approx. 10%) and jet fuel (approx. 3%), although these estimates vary depending on the source of information (see Ecology, 2011).

Although large spills may garner a host of attention, it appears that small motor oil and gasoline spills, drips, and minor leaks are responsible for approximately 87% of the total petroleum released annually in the Puget Sound basin. Improper disposal of used motor oil accounts for an additional 11% of petroleum releases inventoried.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 44. Total Petroleum Release in the Puget Sound Basin (values shown are t/yr).

Approximately 6,100 t/yr of motor oil is estimated to drip and leak from vehicle crankcases during the 58 billion kilometers travelled each year on roads in the region. Another 960 t/yr of motor oil is disposed of improperly following oil changes.

Minor gasoline spills that occur during fueling of vehicles and other motorized equipment, as well as spills that occur during the transport of portable fuel containers, lead to an estimated annual gasoline release of approximately 1,900 t, including 570 t released annually during on-road vehicle fueling at the pump. This estimate only includes liquid spillage; gasoline released through volatilization of fuel and vapor displacement during fueling was not included in this estimate.

Major Delivery Pathways and Loading

Data on major delivery pathways for oil & grease and petroleum loading were obtained from PSTLA loading studies and are included in Table 28. No effort was made to assess oil & grease or petroleum loads from direct atmospheric deposition, exchange of ocean waters, or POTWs. As mentioned previously, loading through direct groundwater discharge was estimated from literature values, whereas loadings through other pathways were estimated from field studies specifically designed to estimate loads.

Surface runoff was the only major loading pathway where oil & grease was analyzed. Loads were estimated to be approximately 8,500 – 10,600 t/yr for the entire Puget Sound basin based on the surface runoff study. Due to the low frequency of detection (<50%) in all land uses under both base and storm flow conditions, comparisons among land covers and flow conditions are not meaningful. However, seasonal first-flush elevations in oil & grease concentrations were evident for all but the forest land covers. Absolute loads calculated from surface runoff were a reflection of land cover proportions and flows, with forest lands making up the bulk of the load.

Table 28. Oil & Grease and Petroleum Loads (t/yr) to Puget Sound from Major Pathways.

	25th %ile	Median	75th %ile
		Oil & Grease	
Groundwater	NA	NA	NA
Air Deposition	NA	NA	NA
Surface Runoff	8,469	8,469	10,598
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
		Lube Oil	
Groundwater (a)	6.03	34.2	62.4
Air Deposition	NA	NA	NA
Surface Runoff (b)	320	345	360
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
		Diesel Range	
Groundwater (a)	1.84	18.0	34.2
Air Deposition	NA	NA	NA
Surface Runoff	ND	ND	ND
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA
		Gasoline Range	
Groundwater (a)	2.85	23.8	44.7
Air Deposition	NA	NA	NA
Surface Runoff	ND	ND	ND
POTWs	NA	NA	NA
Ocean Exchange	NA	NA	NA

(a) Lowest, highest, and mid-point of estimated loads
(b) Detected in agricultural areas only during baseflows
NA=not analyzed
ND=not detected

Lube oil loads were estimated for the groundwater pathway (6-62 t/yr) and for the surface runoff pathway (320 – 360 t/yr). The range of estimates for groundwater was due to differences in flows used to establish the possible range of loads; groundwater discharges used in the groundwater loading study ranged by an order of magnitude.

For surface water runoff, lube oil was detected at the highest concentrations and frequencies (75%) in commercial/industrial sub-basins during storm events. Lube oil was also detected in other land covers during storms – residential and agricultural areas displayed seasonal first-flushes – yet overall detection frequencies were low (<50%) in all but commercial/industrial land covers. Detections of lube oil were even more infrequent during baseflow sampling, where only agricultural streams had measurable lube oil.

Diesel and gasoline loads were estimated for groundwater (1.8 – 34 t/yr and 2.8 – 45 t/yr, respectively), but neither of these petroleum fractions were detected in surface water. The range of estimates for the groundwater loads was primarily due to differences in flows used to establish the possible range of loads.

The net sum of total oil & grease and petroleum loads to Puget Sound through the pathways assessed may be calculated by summing the loads for a specified percentile value for each of the pathways. The net sums for the following parameters at the respective 25th, median (50th), and 75th percentile values are: Oil and grease - 8,500, 8,500, and 10,600 t/yr; diesel – 1.8, 18, and 34 t/yr; and gasoline 2.8, 24, and 45 t/yr.

Loads for lube oil, the only form of petroleum with loading estimates available for more than one pathway, are shown in Figure 45. Total lube oil loads at the respective 25th, median (50th), and 75th percentile values are 330, 380, and 420 t/yr. Surface runoff accounts for 85 – 98% of the lube oil loads with the remaining 2 – 15% contributed by groundwater (Figure 46).

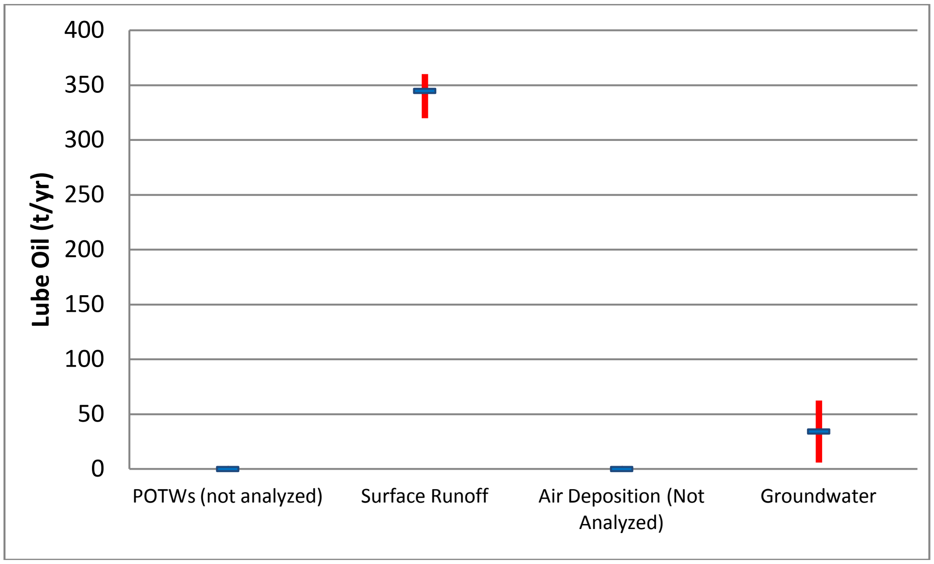


Figure 45. Range (25th - 75th percentiles) and Median Petroleum Lubricating Oil Loads to Puget Sound from Each Major Delivery Pathway.

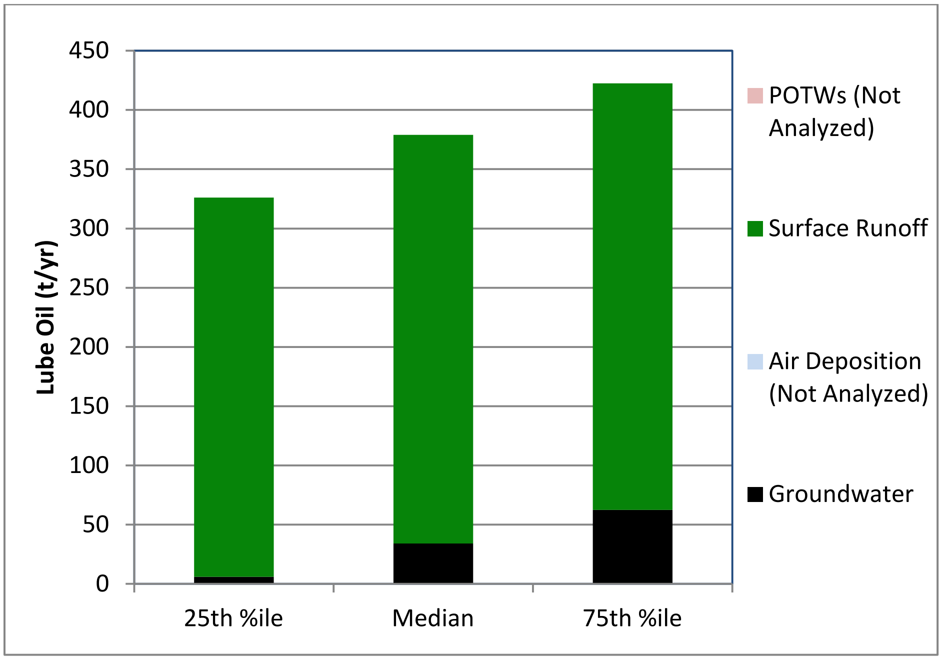


Figure 46. Petroleum Lubricating Oil Loads to Puget Sound from Each Major Delivery Pathway.

Relationship Between Sources and Pathways/Loading

The transport of oil & grease and petroleum hydrocarbons is not likely to be complex compared to many of the COCs addressed in this report. However, the lack of data in a variety of pathways, particularly the presumed major pathway (surface runoff), leaves little information on which to develop a conceptual model based on the data specific to this project.

Based on the source inventory, it appears that thousands of metric tons (t) of petroleum are released in the watershed each year. Most of the petroleum release is in the form of motor oil from engine drips and leaks (6,100 t/yr) and therefore likely to be directly to pavement. Since motor oil is only slowly degraded and is non-volatile, most would be expected to remain on the pavement until entrained by stormwater. Once entrained in stormwater, the oil may be transported directly or indirectly to surface waters, to POTWs, or become sequestered in reservoirs such as soil, detection ponds, and roadside ditches. Once bound to soil, lube oil may not be further transported unless the soil is dislodged.

Overall, it appears that approximately 6% of the motor oil released annually in the Puget Sound basin is transported to Puget Sound in surface water runoff. Diesel released on land from large spills (approximately 20 t/yr) or gasoline released during small fueling and transport spills (1,900 t/yr) was not reflected in measurable surface runoff loads. While it is possible that surface runoff estimates grossly underestimate petroleum loads due to analytical or sampling design errors, petroleum indicator PAHs (e.g. fluoranthene, naphthalene, pyrene) were also rare in surface runoff samples (Herrera, 2011), suggesting limited contribution from petroleum as a source.

Of the POTWs sources inventoried, only improper disposal of used oil (960 t/yr) is likely to be released in a constrained pathway. Petroleum was not assessed in POTWs, so there are no estimates available for this pathway. As mentioned previously in the PAH section, some of the PAHs associated with petroleum were frequently detected in POTW discharge samples, suggesting a possible petroleum source.

Hazard Evaluation

Toxicological evaluation of petroleum mixtures have historically been conducted in the aftermath of oil spills and similar events, and as a result, the majority of available petroleum toxicity data for are for crude oils. However, crude oil toxicity data are generally unsuitable for the Puget Sound basin hazard evaluation since none of the available observed environmental data are for crude oils.

A large amount of observed oil and grease data were available although these data are non-specific and do not describe a specific petroleum product; they encompass waxes, greases and other fatty acid substances from both animal, vegetable and petroleum origins. Because these environmental data are non-specific, and potentially toxic components may vary within the same concentration measured by this method; these data were considered unusable for this assessment.

Both toxicity data and environmental data were only available for four petroleum products in freshwater; heavy fuel/bunker oil, diesel fuel, gasoline, and lube oil. Toxicity data for these fours products were almost entirely based on lethal concentrations to 50% of the exposed population (LC_{50}s). Environmental data for these four products were plotted against the available toxicity data (Appendix D-3).

Only two petroleum product data results were available for marine waters and these were insufficient to estimate priority levels. No observed or effects data were available for sediment evaluation. Hazards due to tissue residue effects, effects to wildlife, and effects to human health were not evaluated.

For fresh surface waters, the median concentration for gasoline (approx. 2.5 mg/l) is above the 10th percentile of effects data, and the 75th percentile of gasoline concentrations exceeds the median effects concentration. For lube oil, the 90th percentile of concentrations (approx. 4 mg/l) exceeds at least 95% of the effects data. For heavy fuel oil and diesel, all of the observed concentrations were two- to five-fold below effects data. In all cases, however, there were not sufficient effects data for an adequate evaluation.

Only toxicity results assessing the water soluble fraction (without free product) were used to assess these data. A more complete evaluation of petroleum would require analysis of parent and alkyl PAHs as well as issues such as phototoxicity which were beyond the scope of this assessment. In addition to the specific assessment for petroleum described here, a suite of individual and high/low molecular weight PAHs were evaluated in both the water and sediment assessments previously described above.

There are no sediment guidelines to assess the level of concern posed by petroleum in sediments, and neither effects data nor observed data are available to evaluate hazards due to residues in aquatic organisms or to evaluate petroleum hazards to wildlife or humans. Due to the uncertainties discussed above and the lack of effects data, there is a high level of uncertainty for the petroleum evaluation.

Table 29. Summary of Hazard Evaluation for Petroleum.

Surface Water		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.* *or* *Acute WQC* *or* *Chronic WQC*	**Level of Concern**
	Freshwater	a	>8%	INS[b]	U
	Nearshore Marine	0	--	INS[c]	U
	Offshore Marine	0	--	INS[c]	U
Sediment		**Total N**	**FOD**	**90th%ile Observed Conc. >** *SQS*	**Level of Concern**
	Freshwater	0	--	INS[c]	U
	Nearshore Marine	0	--	INS[c]	U
	Offshore Marine	0	--	INS[c]	U
Tissue Residue Effects		**Total N**	**FOD**	**90th%ile Observed Conc. >** *10th %ile Effects Conc.*	**Level of Concern**
	Not Analyzed				
Wildlife		**Total N**	**FOD**	**Daily Dose >** *10% of Lowest Effects Dose*	**Level of Concern**
	Not Analyzed				
Human Health		**Total N**	**FOD**	**90th%ile Observed Conc. >** *NTR Criterion*	**Level of Concern**
	Not Analyzed				

FOD=Frequency of detection[a] N = 17 for heavy fuel oil, N = 295 for diesel, N = 359 for gasoline, and N = 894 for lube oil

INS[b]=Insufficient effects data available for comparison to observed data

INS[c]=Insufficient observed or effects data available for comparison

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Summary of Findings

General Considerations

Projects conducted under Phase 3 of the PSTLA which evaluated sources, loading, pathways, and relative hazards of selected COCs in the Puget Sound basin were the key sources of data used for this assessment.

The PSTLA focused on the narrow list of COCs shown below. This list was developed during Phase I of the project based on the COCs' presence in Puget Sound and their potential to cause harm, and to ensure that a broad variety of delivery pathways would be represented. There is a wide variety of chemicals in the Puget Sound basin which have the potential to cause biological and ecological harm, yet environmental data are lacking for many of them. Therefore, this assessment should be viewed as the starting point for development of a much larger toxic chemical assessment.

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs)
- Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE
- Low molecular weight polycyclic aromatic hydrocarbons (LPAHs)
- High molecular weight polycyclic aromatic hydrocarbons (HPAHs)
- Carcinogenic polycyclic aromatic hydrocarbons (cPAHs)
- Diethylhexyl phthalate (DEHP, a.k.a. bis(2-ethylhexyl) phthalate)
- Triclopyr
- Nonylphenol
- Oil and petroleum hydrocarbons

The geographical area addressed in this *Assessment Report* includes the Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits (Figure 1). This is consistent with all of the land-based PSTLA loading projects (e.g. Hart Crowser et al., 2007; Envirovision et al., 2008a), except the groundwater loading analysis which excludes loads from the western Strait of Juan de Fuca.

Summary of COC Releases

A summary of the total estimated release and largest potential source for each COC is shown in Table 30. A complete list of individual sources is shown in Appendix C.

Table 30. Summary of Estimated Anthropogenic Releases of COCs in the Puget Sound Basin.

Chemical of Concern (Release Rate)	**Major Sources**	**Modes of Release**
Arsenic (0.8 t/yr)	Industrial sources	Releases to air.
	CCA-treated wood, roofing materials	Leaching from precipitation.
Cadmium (1 t/yr)	Roofing materials	Leaching from precipitation.
Copper (180 – 250 t/yr)	Urban lawn & garden use as pesticides[a]	Direct application to soil.
	Plumbing components, roof material	Leaching from precipitation or directly to POTWs.
	Brake pads	Abrasion leading to fugitive dust emission or loss directly to roadway.
	Vessel anti-fouling paint	Leaching directly to marine water.
Lead (520 t/yr)	Ammunition and hunting shot use, loss of fishing sinkers, loss of wheel weights	Release of solid metallic lead.
	Roofing materials	Leaching from precipitation.
	Aviation fuel	Combustion emissions.
Mercury (0.5 t/yr)	Thermostats, fluorescent lamps, button cells batteries, other mercury-containing material	Volatilization and leaching following disposal.
	Crematoria, industrial plants	Combustion and other air emissions.
Zinc (1,500 t/yr)	Roofing materials	Leaching from precipitation.
	Vehicle tires	Abrasion leading to fugitive dust emission or loss directly to roadway.
Total PCBs (2 t/yr)	Electrical equipment[a]	Spills and leaks.
	Residential trash burning	Combustion emissions.
	Building sealant (caulk)	Volatilization, abrasion to dust and larger particles.
Total PBDEs (0.7 t/yr)	Furniture, computer monitors, and other components of residential and commercial indoor environments	Indoor air and dust.
PCDD/Fs (9 grams TEQ/yr)	Backyard burn barrels	Combustion emissions.
Total PAHs (310 t/yr)	Woodstoves and fireplaces, light and heavy-duty vehicles	Combustion emissions.
	Creosote-treated piling, railroad ties, and utility poles	Leaching and washout, volatilization.
DEHP (17 t/yr)	Industrial, commercial, and institutional point sources	Air emissions.
	Car undercoating, roofing	Leaching, volatilization.
Triclopyr (150 t/yr)	Crop and golf course use as herbicides	Direct application to plants or soil.
Nonylphenol (0.2 t/yr)[b]	Industrial, commercial, and institutional facilities	Air emissions.
Petroleum (9,200 t/yr)	Vehicle crankcase oil	Motor oil drips, leaks, and improper disposal of used oil.
	Vehicle and off-road equipment fueling	Gasoline (minor) spillage.
DDT	NA	NA

t=metric ton (appr. 2.2 tons)

TEQ= Toxic Equivalents

NA=Not analyzed

[a] Estimate is highly uncertain

[b] Sources were not fully assessed

Information on COC releases from primary sources in the Puget Sound basin can be summarized as follows:

- Petroleum is estimated to be released in the largest quantity, followed by zinc which is the only other COC estimated to be released at a rate greater than 1,000 metric tons (t) per year. Lead, PAHs, and copper are estimated to be released at rates greater than 200 t/yr, triclopyr is released at an estimated rate over 100 t/yr, and approximately 15 t of DEHP is released annually. PCBs, cadmium, mercury, PBDEs, arsenic, nonylphenol, and PCDD/Fs are generally released at rates near one t/yr or less.

- In general, industrial, commercial, and institutional point sources do not account for large releases of COCs. Instead, a variety of diffuse sources account for a majority of the COC releases.

- Runoff and leaching of chemicals from roofing materials is potentially an important source of metals. For two metals – cadmium and zinc – release from roofing materials were estimated to account for the majority of total release, and nearly one-third of arsenic is released through roof runoff. Zinc releases from roof runoff are particularly large (>1,000 t/yr). There were also estimated to be substantial releases of copper and lead from roof materials. Leaching of metals from rooftop runoff is likely to be largely in the form of unconstrained releases. Leaching of metals from plumbing components also accounts for substantial releases of copper and zinc, but these are likely to be constrained to sanitary sewer systems and POTWs.

- Vehicle and road-related COC releases occur primarily through wear of vehicle components, combustion of fuel, and leaks of motor oil and fuel. Abrasion of brake pads account for up to one-third of the total release of copper. The second largest source of zinc was estimated to come from tire wear. Vehicle-related fuel combustion releases large quantities of COCs, accounting for about 10% of the total PAH release due to gasoline and diesel combustion, and about 5% of the total PCDD/F release, primarily due to heavy-duty diesel vehicles.

- Petroleum represents the largest COC quantity released to roadways and other impervious surfaces. Motor oil lost via drips and leaks appears to account for over 60% of the total petroleum release and likely occurs along roadways or impervious surfaces – parking lots and driveways – connected to roadways. In addition, gasoline is released at a rate of approximately 1,900 t/yr, including 570 t/yr released during on-road vehicle fueling at the pump. PAHs contained in uncombusted petroleum are also released along with the leaked petroleum at a rate of approximately 10 t/yr.

- Emissions from backyard burn barrels account for about three-quarters of the total PCDD/Fs released, and nearly all of the PCDD/F release is from combustion sources.

- Woodstoves were estimated to be the largest source of PAHs (about one-third of the total PAH release), and aside from zinc roof runoff and petroleum leakage, represent the only COC source exceeding 100 t/yr.

- About 10% of the total PCBs released in the basin was estimated to come from residential trash burning.

- Since few of the COCs assessed are typically associated with agriculture, releases from agricultural uses are generally small. Triclopyr, the only COC used exclusively as a pesticide (herbicide), is an exception with over 100 t/yr applied to crops in the Puget Sound basin. There are also some releases of metals in fertilizers applied to agricultural crops, with zinc releases being the largest (>40 t/yr).

- Copper is used in agriculture as both a pesticide and a micronutrient. Approximately 10 t/yr of copper is used as an agricultural pesticide alone, with an additional 5 t/yr used as a micronutrient. Use of copper as an urban pesticide is potentially substantial, as much as 70 t/yr by some estimates, but other estimates put it at a much lower rate (1 t/yr). The high level of uncertainty in these estimates underscores the limited information available regarding pesticide use.

- PAH releases from creosote-treated wood (railroad ties, marine pilings, and utility poles) appear to account for over one-third of the PAHs released annually in the Puget Sound basin.

- PCB leakage from electrical equipment appears to be substantial, possibly as much as 2,000 kg/yr. However, this amount may also include indoor leakage or that which is immediately cleaned up and contained, and so may overestimate actual PCB release in the Puget Sound basin by a large degree.

- PBDE and phthalate releases are difficult to assess due to the passive nature of their emissions from the materials and products in which they are used. For PBDEs, their loss via air emissions and dust particles into air and dust from commercial offices and homes, followed by subsequent release to the outdoor environment, appears to be the major release pathway. Phthalates may be released in the same manner, although releases from domestic products – including personal care products – may be more important sources. For both of these COCs, attachment to indoor dust and subsequent release to sanitary sewers may also be an important release pathway.

COC Loading and Pathways

One component of the PSTLA was to assess chemical loading from various pathways. The chemical-specific information focused on surface water runoff, atmospheric deposition, and groundwater discharge directly to marine waters, and POTWs as the major pathways for COC delivery. The flux of COCs across the Puget Sound – ocean boundary was also assessed where data were available.

For each of the pathways mentioned above, separate projects were conducted to assess loading. Descriptions of these projects are described in the introductory sections of this report. Tables 31-33 show summaries of the COC amounts loaded from each of the pathways that have *partially controllable* sources of COCs (i.e. all of the major pathways except ocean exchange).

Summaries of the relative COC contributions delivered from each of the loading pathways are shown in Figures 47 and 48.

Caution should be used interpreting Figures 47 and 48 since load estimates are incomplete for some COCs. In particular, air deposition and groundwater loading data are not available for most of the organic COCs.

Table 31. Summary of Metals Loading to Puget Sound through Major Pathways.

Metals		**POTWs**	**Surface Runoff**	**Air Deposition**	**Ground-water***	**SUM**
Total Arsenic (t/yr)	25th %ile	NA	13.5	0.2	0.1	14
	Median	NA	16.9	0.4	0.4	18
	75th %ile	NA	23.4	0.5	0.8	25
Total Cadmium (t/yr)	25th %ile	NA	0.01 [a]	0.03	0.01	0.05
	Median	NA	0.01 [a]	0.05	0.22	0.28
	75th %ile	NA	0.02 [a]	0.07	0.43	0.53
Total Copper (t/yr)	25th %ile	2.5	28.4	1.9	0.1	33
	Median	4.3	35.7	2.7	2.2	45
	75th %ile	5.5	66.1	4.1	4.3	80
Total Lead (t/yr)	25th %ile	0.1	2.8	0.6	0.1	3.6
	Median	0.2	4.7	1.1	1.1	7.0
	75th %ile	0.3	7.6	1.5	2.1	12
Total Mercury (t/yr)	25th %ile	NR	0.09	0.01	0.005	0.11
	Median	0.002 [b]	0.14	0.02	0.05	0.21
	75th %ile	NR	0.24	0.03	0.09	0.37
Total Zinc (t/yr)	25th %ile	16	113	11	2	140
	Median	19	122	18	11	170
	75th %ile	24	134	26	20	200

* Lowest, highest, and mid-point of estimated loads
t=Metric ton (appr. 2.2 tons)
NA=Not analyzed
NR=Not reported
[a] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows
[b] Estimate from Phase 2 (Envirovision et al., 2008b)

Table 32. Summary of Organic Chemical Loading to Puget Sound through Major Pathways.

Organics		POTWs	Surface Runoff	Air Deposition	Ground-water*	SUM
Total PCBs (kg/yr)	25th %ile	0.1	2.5	0.7	NA	3.4
	Median	0.3	5.3	1.3	NA	6.9
	75th %ile	1.8	15.8	3.7	NA	21
Total PBDEs (kg/yr)	25th %ile	7.0	5.1	15.7	NA	28
	Median	10.6	5.7	20.3	NA	37
	75th %ile	20.7	10.0	23.8	NA	54
PCDD/Fs (g TEQ/yr)	25th %ile	NA	NA	NA	NA	NA
	Median	NA	NA	NA	NA	NA
	75th %ile	NA	NA	NA	NA	NA
Total DDT (kg/yr)	25th %ile	ND	2.2 [a]	NA	0.3	2.5
	Median	ND	23.9 [a]	NA	3.8	28
	75th %ile	ND	25.1 [a]	NA	7.3	32
LPAH (kg/yr)	25th %ile	3	102 [b]	NA	7	110
	Median	8	104 [b]	NA	159	270
	75th %ile	35	190 [b]	NA	311	540
HPAH (kg/yr)	25th %ile	4	25 [c]	49	6	84
	Median	5	36 [c]	96	124	260
	75th %ile	7	51 [c]	153	243	450
cPAH (kg/yr)	25th %ile	NC	18 [d]	21	5	44
	Median	NC	24 [d]	43	83	150
	75th %ile	NC	34 [d]	70	161	260
Total PAH (kg/yr)	25th %ile	8	119 [b]	49	13	190
	Median	18	224 [b]	96	284	620
	75th %ile	46	244 [b]	153	554	1,000
DEHP (kg/yr)	25th %ile	216	1,750 [e]	NA	14	2,000
	Median	439	1,780 [e]	NA	227	2,400
	75th %ile	904	1,860 [e]	NA	440	3,200
Triclopyr (kg/yr)	25th %ile	NC	641 [f]	NA	NA	640
	Median	NC	652 [f]	NA	NA	650
	75th %ile	NC	686 [f]	NA	NA	690
Nonylphenol (kg/yr)	25th %ile	NC	23 [g]	NA	NA	23
	Median	NC	23 [g]	NA	NA	23
	75th %ile	NC	24 [g]	NA	NA	24

* Lowest, highest, and mid-point of estimated loads.

NA=Not analyzed; ND=Not detected; NC=Not calculated due to insufficient data.

[a] Detected in commercial/industrial areas only during baseflows and in commercial/industrial, agricultural, and forest areas only during storm flows.

[b] Detected in commercial/industrial areas only during baseflows.

[c] Detected in commercial/industrial areas only during baseflows and in commercial, residential, and agricultural areas only during storm flows.

[d] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial, residential, and agricultural areas only during storm flows.

[e] Detected in residential areas only during baseflows.

[f] Detected in commercial/industrial and agricultural areas only during baseflows.

[g] Surface runoff loads based on storm flows only; not detected in any land covers during baseflows and in commercial/industrial areas only during storm flows.

Table 33. Summary of Petroleum and Oil & Grease Loading to Puget Sound through Major Pathways.

Petroleum and Oil & Grease		POTWs	Surface Runoff	Air Deposition	Ground-water*	SUM
Oil & Grease (t/yr)	25th %ile	NA	8,470	NA	NA	8,500
	Median	NA	8,470	NA	NA	8,500
	75th %ile	NA	10,600	NA	NA	11,000
Lube Oil (t/yr)	25th %ile	NA	320 [a]	NA	6	330
	Median	NA	345 [a]	NA	34	380
	75th %ile	NA	360 [a]	NA	62	420
Diesel (t/yr)	25th %ile	NA	ND	NA	2	2
	Median	NA	ND	NA	18	18
	75th %ile	NA	ND	NA	34	34
Gasoline (t/yr)	25th %ile	NA	ND	NA	3	3
	Median	NA	ND	NA	24	24
	75th %ile	NA	ND	NA	45	45

* Lowest, highest, and mid-point of estimated loads
t=Metric ton (appr. 2.2 tons)
NA=Not analyzed
ND=Not detected
[a] Detected in agricultural areas only during baseflows

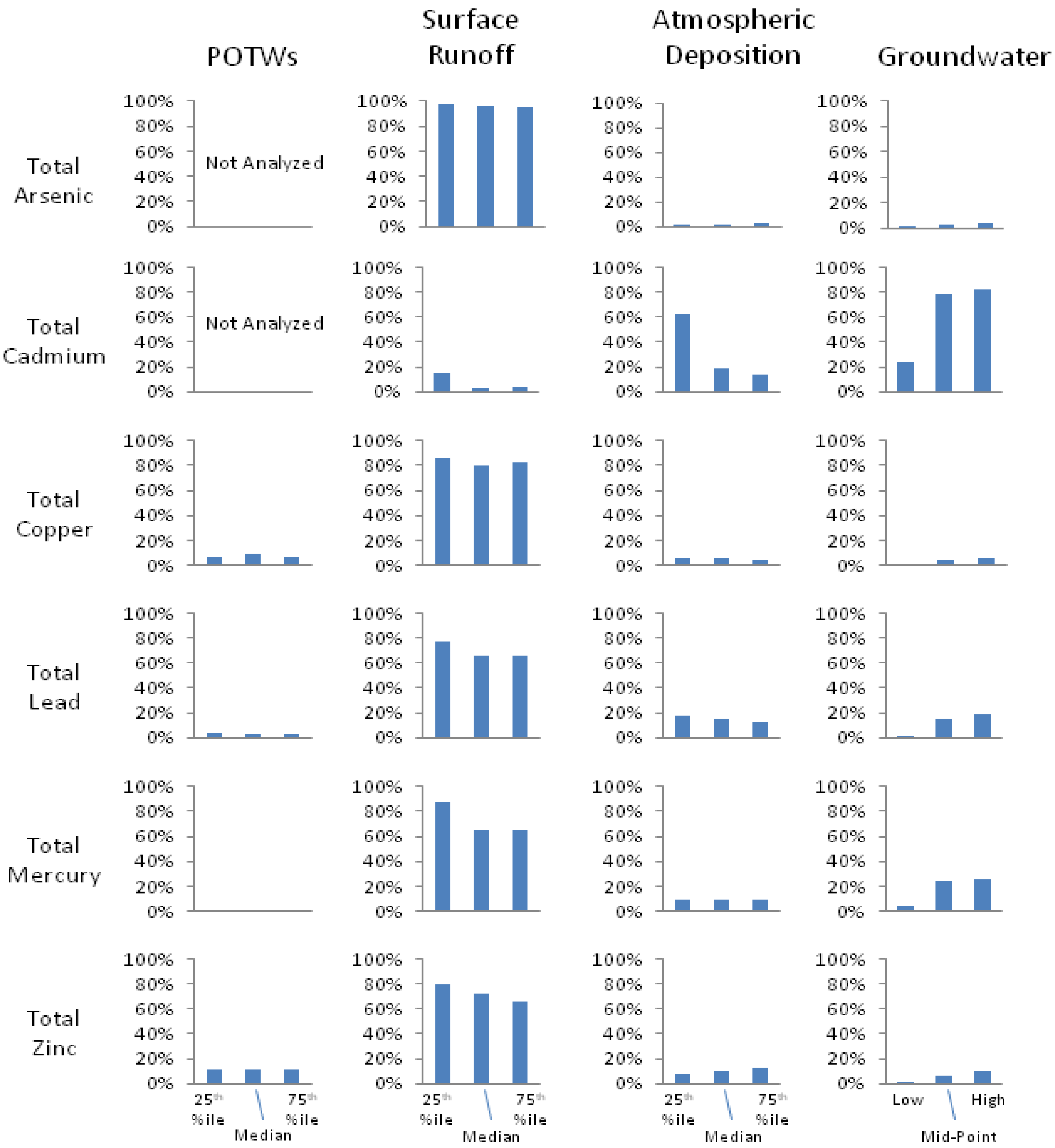

Figure 47. Percent Contribution of Major Pathways to Metals Loading in Puget Sound.

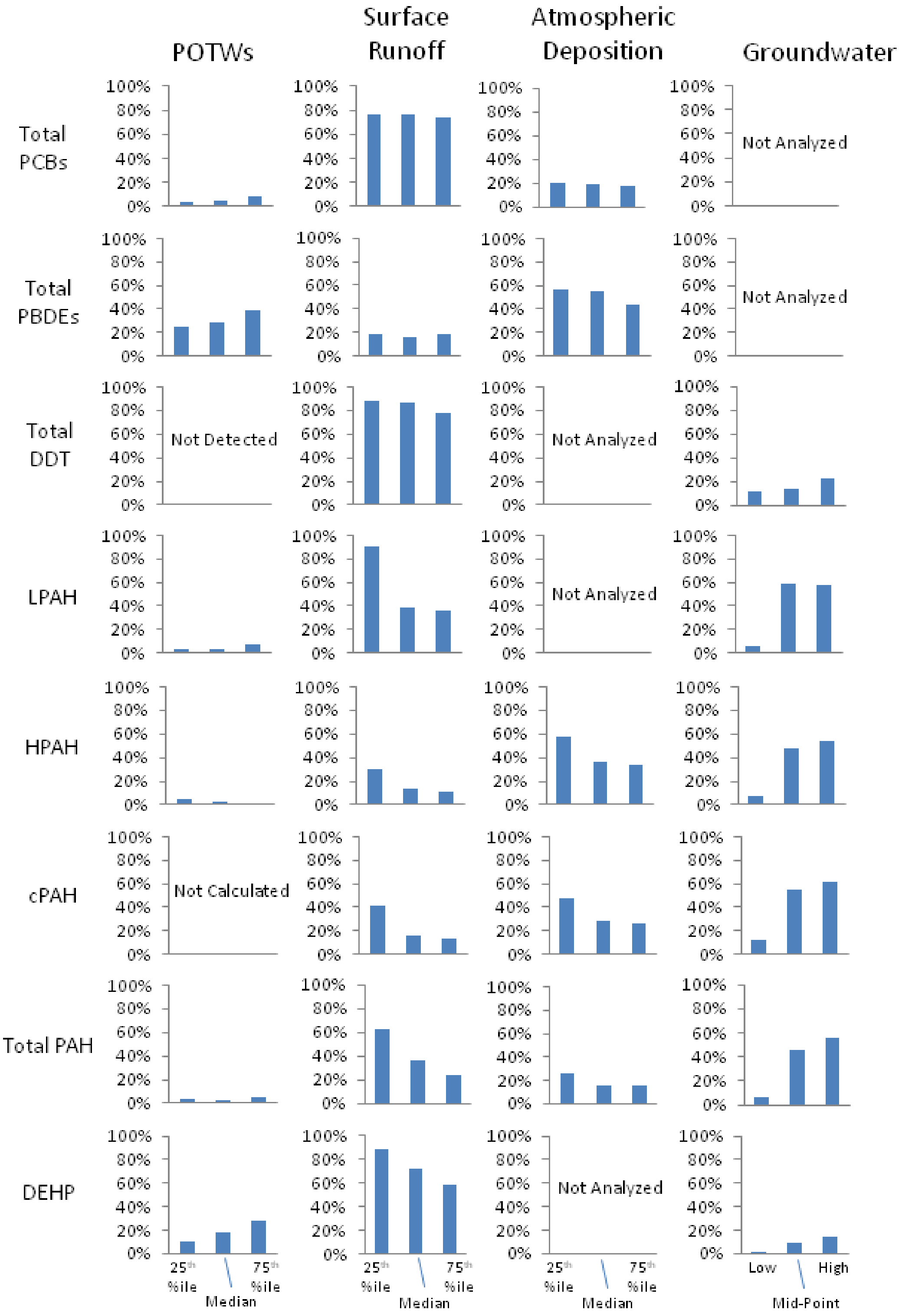

Figure 48. Percent Contribution of Major Pathways to Organic Chemical Loading in Puget Sound.

Information on the COC loading and pathways can be summarized as follows:

- Surface water runoff is the dominant pathway for all metals except cadmium. Metals are generally found at the highest concentrations in commercial/industrial and agricultural land covers, followed in decreasing order by residential and forested areas. However, forest areas account for the bulk of metals loading simply due to the comparatively high proportion of forest land cover (83% of land area) which generates the majority of the flow entering Puget Sound.

- PCBs, PBDEs, and DEHP are present in surface runoff from all land covers at very low concentrations, but tend to be highest in commercial/industrial areas, particularly during storm flows. Like most other COCs in surface runoff, the largest absolute loads are from forested areas as a result of the high proportion of forest cover and associated volume of water.

- POTW-delivered loads were comparatively small for all metals assessed. Cadmium and arsenic were not measured in POTW effluent. To some degree, loads of copper, lead, and zinc mirrored the discharge volume of POTWs relative to other annual land-based water discharge (groundwater and surface runoff) to Puget Sound (~4% of total).

- Groundwater loads directly to marine waters were estimated to constitute approximately 5-10% of the total loading for most metals. The greatest mass of metals loaded annually to Puget Sound through groundwater are from non-urban ambient areas, followed by urban ambient areas and impacted areas. The volume of groundwater discharge alone does not account for the relative contribution of groundwater loads, since groundwater accounts for only 0.2 – 2% of the total annual land-based water discharged to Puget Sound.

- Atmospheric deposition directly to the marine waters of Puget Sound is estimated to account for approximately 5 –15% of the total annual loads of copper, lead, mercury, and zinc. Lower relative loads were estimated for arsenic (≤3%), and higher loads were estimated for cadmium (14 – 62%). For all metals, the atmospheric deposition flux measured was much greater at a particular sampling location within a high-density urban area that was proximal to a nearby pulp mill, metal refiners, other industrial activities, and also close to major roadways including interstate highways. In most cases, the flux at this location was at least five-fold higher than at other locations without these urban influences.

- PBDE loads deposited directly to marine waters from the atmosphere are roughly equal to loads from surface runoff and POTWs combined. With the possible exception of HPAH, PBDEs are the only COC with the predominant load contributed by direct atmospheric deposition. In addition, PBDEs are the only COC with higher loads delivered through POTWs compared with surface runoff.

- Fluxes at the ocean boundary generally show a net export of metals out of Puget Sound. However, for cadmium and lead, there is a net import to Puget Sound from oceanic waters. Marine fluxes of cadmium and lead into Puget Sound are greater than loads from all other pathways combined.

- For most organic compounds, patterns of pathway loading are more difficult to assess than for metals due to inconsistencies in analysis among loading studies and comparatively low frequencies of detection. For instance, HPAHs were the only organic constituents analyzed in the four major pathways assessed.

- For PAHs, accurate delivery patterns and loads are difficult to assess due to infrequent detection in surface runoff. A cursory examination of loads delivered by surface runoff, atmospheric deposition, and groundwater does not reveal a clear loading pattern or principal transport pathway. However, the atmospheric deposition data appear to be the most reliable; surface runoff and groundwater loading data are derived largely from sample data with non-detect results.

- Among all of the COCs assessed, oil & grease was estimated to be delivered to Puget Sound in the largest quantity (>8,000 t/yr) with surface runoff accounting for the entire load. Although oil & grease is relatively meaningless in terms of chemical specificity – it simply refers to the fraction of a sample extractable by n-hexane – some portion of the oil & grease measured in surface runoff may be petroleum product, particularly in urban areas.

Limitations and Uncertainty of Data on COC Sources and Loads

All of the individual projects in the PSTLA contain a degree of uncertainty in the reported results, and all have limitations due to study design or due to the nature of the data collected. Limitations and uncertainty affect the usefulness of the individual projects as well as the ability to compare results among projects. Some of the most common and intractable issues are mentioned here. However, the reader is encouraged to review the individual reports in order to fully gauge uncertainty and understand how results were derived.

The projects were not designed to analyze for an identical suite of COCs. This resulted in a limited ability to fully gauge each pathway's contribution to overall loading and to compare COC quantities released among studies. For instance, air deposition and groundwater loading data are not available for most of the organic COCs, and the source inventory (Ecology, 2011) did not fully account for all major sources of zinc, nonylphenol, and petroleum releases. Appendix B shows a summary of the COCs analyzed for each project.

Much of the uncertainty surrounding the reported results for loading projects is due to sample results below reporting or detection limits (i.e. non-detects). In many cases, results were derived using datasets where more than one-half of the concentration values were reported as non-detects. There was a particularly heavy reliance on non-detects in the surface runoff and groundwater loading projects. For the assessment, this source of uncertainty is compounded by the lack of consistency in the assumptions and rules for handling data (including non-detects) among projects. It should be noted, however, that estimates of releases and loads used for this assessment are those reported in the original projects, and no attempt was made to recalculate or "normalize" results according to a common set of rules. A summary of project-by-project rules used to handle non-detects is shown in Appendix B.

It is also notable that the study conducted to assess loads from surface runoff, the major delivery pathway for most COCs, did not actually measure COC concentrations at the point of delivery to Puget Sound. The authors of the surface runoff study (Herrera, 2011) offer a detailed explanation of this and other potential sources of bias in the surface runoff project.

Relationship between Sources and Pathways/Loading

The relationships between COC sources and loading/pathways discussed in this report essentially provide a framework for the first step in developing mass-balances for COCs, from their initial release to their delivery to Puget Sound. Information provided for these conceptual models include the identification of the primary sources of COCs, mechanisms of release (e.g. combustion emission), estimates of the quantities released, COC prevalence and levels in delivery pathways, and characteristics of loading pathways (e.g. land cover) related to COC prevalence and levels.

Although the conceptual models of the relationships between sources and loading/pathways were developed on a very broad scale, some patterns appear to emerge. For instance, it appears that for most of the COCs assessed there is a one to two order of magnitude difference between the quantity released and the quantity estimated to be loaded to Puget Sound through various pathways. Notable exceptions are PCBs, triclopyr, and PAHs which have estimated releases three orders of magnitude higher than loads. However, the loading estimates for PAHs do not include the estimated release directly to marine waters from creosote-treated marine pilings.

Arsenic is the only COC with estimated loads to Puget Sound larger than releases from primary sources. This appears to be due to the natural enrichment of soils and surface runoff with arsenic, coupled with comparatively low arsenic releases from ongoing anthropogenic sources.

Estimated quantities of COCs released do not necessarily translate to equivalent loads in transport/delivery pathways due to a variety of factors affecting their behavior and fate once released in the environment. For instance, the case studies of copper mass-balance in small watersheds (Paulson et al., 2011-Draft) showed a much greater relative difference between releases and loading at the small (watershed) scale compared to the relative difference between releases and loading at the large (Puget Sound basin) scale.

In many cases, the specific COC source and the mechanism of release may have more environmental relevance then the absolute quantity released. The relationships between COC releases and their presence in specific pathways were therefore examined at finer scales than simply comparing the total annual mass released to the total annual mass loading to Puget Sound.

The following patterns reveal consistencies between releases and pathways at finer scales:

- Overall it appears that there is approximately an order of magnitude decrease between the quantity of a COC discharged to a POTW and the reported load discharged from POTWs to Puget Sound. This appears to be the case for copper, lead, and DEHP. Sources of PBDEs suggest a substantial proportion is released to POTWs as well.

- Metals (copper, lead, and zinc) and PAHs associated with vehicle sources are highly elevated in air deposition samples located near high-density urban areas and interstate highways.

- COCs which are released to air through combustion or volatilization have large load contributions from atmospheric deposition directly to marine waters compared to other pathways. PBDEs and PAHs are two primary examples. DEHP and PCDD/Fs might be expected to follow this pattern as well, but they were not measured in air deposition samples. Mercury, which is largely emitted to the air through combustion or volatilization, does not exhibit the comparatively large load from direct deposition as might be expected.

Hazard Evaluation

As noted in the introductory sections of this report, the hazards posed by different COCs are not simply associated with the quantities released to the environment or loaded to Puget Sound, but are rather more appropriately evaluated by comparing their concentrations in various environmental media to reported effects levels. To assess the relative toxic hazard posed by COCs in various media and for various receptors, the hazard evaluation assessed COCs in various media by comparing observed concentrations to data on effects or guidelines, standards, and criteria for the following categories:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water, and sediment
- Hazard to human health through fish/seafood consumption

The results of these comparisons were grouped into three broad "level of concern" categories: a Priority 1 level of concern, a Priority 2 level of concern, or unknown (U) level of concern due to lack of sufficient data for an assessment. Results were classified as a Priority 1 when high observed concentrations (e.g. 90th percentile values) exceeded low effects concentrations (e.g. 10th percentile values), selected criteria, or other threshold values. A Priority 2 level of concern was assigned in cases where high observed concentrations were below threshold values. In cases where there were not sufficient data to make a meaningful comparison, results were assigned a U. Appendix D-1 details the thresholds used for comparisons and the minimum data required for the comparisons. Table 34 provides a summary of the hazard evaluation for all of the categories and sub-categories assessed.

The hazard evaluation has several limitations that should be considered prior to acting on the results. In particular, the hazard evaluation is not a risk assessment but is instead designed to assess the *relative* level of concern of COCs *across the entire Puget Sound basin*. Although a COC may be assigned Priority 2 or U for a particular sub-category, this should not be interpreted to mean there are no hazards associated with that COC. Locally, concentration hot spots exist near major sources and may cause localized toxicity to aquatic organisms or lead to violations of standards.

In addition to the systematic methodology used to evaluate relative hazards for each COC, a review of relevant regional studies was conducted. This review was conducted to capture information on regionally important biological effects that may not have been included among the data used for the hazard evaluation. Information from these reviews was not used to assign the Priority levels summarized in Table 34, but instead was used as an additional line of evidence for assessing priorities for toxic chemical reduction and control strategies.

Table 34. Summary of the Hazard Evaluation Based on the Priority Levels of Concern for Each Sub-Category (see text for definitions of Priority levels).

COC	Surface Water			Sediment			Tissue Residue			Wildlife		Human Health			Regional Effects Data?
	Frsh.	Marine		Frsh.	Marine		Frsh.	Marine		Frsh.	Marine	Frsh.	Marine		
		nr.	off.		nr.	off.		nr.	off.				nr.	off.	
Arsenic	U	U	U	1	2	2	NA	NA	NA	NA	NA	U	U	U	No
Cadmium	2	U	U	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
Copper	1	1	2	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	Yes
Lead	2	U	2	2	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
Mercury	1	U	U	1	1	1	U	U	U	1	1	2	2	2	Yes
Zinc	2	U	1	1	2	2	NA	NA	NA	NA	NA	NA	NA	NA	No
PCBs	1	U	2	1	2	1	1/U	2/U	2/U	1	1	1	1	1	Yes
PBDEs	U	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	Yes
PCDD/Fs	U	U	U	U	U	U	U	U	U	1/U	2/U	1	1	1	Yes
DDT	1	U	U	U	U	U	2	U	U	1/2	1/2	1	1	2	Yes
PAHs	2/U	U	2/U	1/2	2	2	NA	NA	NA	NA	NA	1/2	1/2	2/U	Yes
DEHP	2	U	U	1	1	1	NA	NA	NA	NA	NA	1	1	U	No
Triclopyr	2	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No
Nonylphenol	2	U	2	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No
Petroleum	U	U	U	U	U	U	NA	NA	NA	NA	NA	NA	NA	NA	No

1=Priority 1 (highlighted in yellow)
Frsh.=freshwater
nr.=nearshore
off.=offshore
2=Priority 2
U=Unknown
NA=not analyzed

Information on the hazard evaluation and regionally important biological-effects data can be summarized as follows:

- All of the COCs except lead, PBDEs, triclopyr, nonylphenol, and petroleum were assigned a Priority 1 level of concern for at least one category. Of the COCs not assigned a Priority 1, only lead had sufficient effects data to conduct a meaningful evaluation.

- PCBs are the only COC assigned a Priority 1 in all five categories evaluated. Mercury and DDT were the only other COCs evaluated for all five categories, and they were assigned Priority 1 for four categories and three categories, respectively.

- COC concentrations in surface waters and sediments – particularly freshwater – resulted in the most COCs assigned Priority 1. Tissue residue, wildlife, and human health evaluations resulted in the fewest COCs receiving Priority 1. However, only bioaccumulative chemicals were evaluated for these latter categories, and so fewer Priority 1 assignments were expected.

- Reviews of regionally important biological-effects data showed that levels of copper, mercury, PCBs, PBDEs, PCDD/Fs, DDT, and PAHs found in the Puget Sound basin result in documented or potentially adverse effects to a variety of aquatic organisms.

Chemical-by-Chemical Summary

Arsenic

Arsenic was found to be a Priority 1 level of concern for freshwater sediments, one of the three categories for which data were sufficient for an adequate hazard evaluation. Observed data were lacking to conduct adequate evaluations for surface waters and human health.

Releases of anthropogenic arsenic are small compared to loads. The largest ongoing anthropogenic source appears to be point-source air emissions, although these are relatively small on a basin-wide scale. Loading to Puget Sound is substantial, presumably due to natural sources and possibly from historical releases, and is dominated by surface runoff. The finding that, unlike most metals, arsenic concentrations in surface runoff decrease during storm events may support the notion that a substantial portion of the arsenic in surface waters is due to natural or historic sources.

Cadmium

Cadmium was found to be a Priority 1 level of concern for freshwater sediments, one of the two categories for which data were sufficient for an adequate hazard evaluation. Observed data were lacking to conduct adequate evaluations for surface waters.

Both anthropogenic releases and loading of cadmium appear to be small. The largest ongoing source of cadmium to the environment appears to be leaching of cadmium from roofing material, constituting more than one-half of the total estimated release. Releases of cadmium from roofing material do not appear to translate to appreciable levels in surface runoff, which represents the smallest delivery pathway.

Copper

Sufficient data were available for full hazard evaluations of copper in surface waters and sediments. Copper was found to be a Priority 1 level of concern for freshwater sediments, nearshore marine sediments, and fresh surface waters. Regionally relevant biological-effects data provide additional lines of evidence to indicate a comparatively high hazard potential for copper. These regional studies have demonstrated that dissolved copper concentrations commonly found in urban and agricultural area streams reduce olfactory function in salmonids (e.g., Hecht et al., 2007).

The source inventory (Ecology, 2011) suggests that large ongoing copper releases occur from a variety of sources including abrasion of vehicle brake pads, leaching from roofing materials, leaching of vessel anti-fouling paint, pesticide applications and micronutrient use in agricultural applications, and possibly pesticide use in urban areas. These releases result in elevated levels in surface water at locations where they are most likely to occur such as agricultural, commercial/industrial, and high-density urban areas, as well as nearshore marine waters.

Lead

Sufficient data were available for full hazard evaluations of lead in surface waters and sediments except nearshore marine waters. Lead was not a Priority 1 level of concern for any of the categories assessed. There is no indication that lead concentrations in fish and shellfish from the Puget Sound basin pose a risk to human health, although lead was not evaluated in tissue due to lack of National Toxics Rule criteria for lead. Lead remains a human health hazard through exposures from material such as lead house paint (Ecology and WDOH, 2009), but no hazard emerges from exposures that include an aquatic environment pathway.

Ongoing anthropogenic releases of lead are substantial, largely due to ammunition use, fishing sinkers, and wheel weight loss. These solid metallic lead sources and mechanisms of release appear to result in limited enrichment of the aquatic environment except in some localized instances. Surface water runoff represents the largest delivery pathway, but loads are small compared to the estimated releases from ongoing anthropogenic sources.

Mercury

Mercury was found to be a Priority 1 level of concern for fresh surface waters, freshwater and marine sediments, and all freshwater and marine wildlife species evaluated. There were not sufficient observed data to conduct adequate hazard evaluations for marine sediments or effects data to evaluate tissue residue effects. Mercury concentrations in freshwater and marine seafood were generally well below the threshold used to assign a Priority 1 level of concern for the human health evaluation. However, consumption advisories for various fish species and locations in the Puget Sound basin have been established by the Washington State Department of Health due to mercury residues in tissues.

There appears to be a variety of ongoing anthropogenic mercury releases, although a number of the regional sources have been addressed in the Mercury Chemical Action Plan (Ecology and WDOH, 2003) and it is unclear if release estimates accurately portray current releases to the environment. Mercury releases due to improper disposal of materials appear to be the largest category of ongoing anthropogenic release, followed by industrial emissions. Surface runoff is the largest delivery pathway for mercury and to some degree may reflect entrainment of mercury deposited atmospherically.

Zinc

Zinc was found to be a Priority 1 level of concern for offshore marine surface waters and for freshwater sediments. Sufficient data were available for surface water and sediment, except nearshore marine surface waters.

Zinc appears to be released at high rates from a variety of roofing materials and to a lesser degree in vehicle tire wear. The study of primary sources (Ecology, 2011) indicated that zinc releases may have been underestimated because many sources (e.g. leaching from galvanized materials) were not assessed. Surface runoff was the dominant pathway for zinc loading to Puget Sound. Zinc levels in streams from commercial/industrial areas were found to be highly elevated, possibly as a reflection of galvanized material leachate and vehicle tires where these sources are likely to be most prevalent.

Polychlorinated biphenyls (PCBs)

PCBs are a Priority 1 level of concern for all five categories assessed and for nine of the 13 sub-categories where sufficient data were available. In addition, regional data show PCB levels that may result in adverse effects to locally important fish and marine mammals. Fish consumption advisories to protect human health have also been issued for both marine and freshwaters of the Puget Sound basin.

Release estimates are highly uncertain and may overestimate the degree to which PCBs continue to be released from ongoing anthropogenic sources (Ecology, 2011). Electrical equipment (capacitors and transformers) leakage is the largest PCB source category, with loss from sealants and release from residential trash burning also making up a substantial portion. However, loading is small compared with releases and may support the notion that releases were overestimated.

Due to their persistence, PCBs that were released from historical sources and continue to be released from highly contaminated areas, such as the lower Duwamish River, continue to cycle in the aquatic environment of Puget Sound. The substantial accumulation in biological tissues may be primarily a result of legacy contamination as opposed to ongoing releases.

While PCBs remain a concern, levels appear to be declining in Puget Sound harbor seals (Noel et al., 2011) and mussels (Mearns et al., 2009). While temporal PCB trends in fish do not show a clear trend (West and O'Neill, 2007), modeling results suggest that substantial declines in English sole should be expected by 2020 at current loading rates (Pelletier and Mohamedali, 2009).

Polybrominated diphenyl ethers (PBDEs)

PBDEs are an unknown level of concern due to a lack of environmental effects data, criteria, and guidelines for PBDEs. However, the lack of effects data used in the hazard evaluation methodology should not be interpreted as a low level of concern since a growing body of evidence suggests environmental concentrations may cause adverse effects to humans (Ecology and WDOH, 2006), marine animals (Ross, 2006), and birds (Fernie et al., 2009). The historical record is generally not adequate to assess PBDE trends in the Puget Sound environment since PBDEs have not typically been included in sampling investigations prior to the beginning of the 21st century.

Cessation of production through voluntary actions and bans since the mid-2000s has removed major PBDE formulations from new consumer products such as mattresses, televisions, computers, and residential upholstered furniture. However, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources.

PBDE loading patterns are different than for other COCs assessed. Direct atmospheric deposition represents the largest delivery pathway, followed by POTWs and surface runoff. The high proportion of PBDE loading through atmospheric deposition and POTWs appears to be consistent with the major sources and release mechanisms.

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs)

PCDD/Fs are a Priority 1 level of concern for a species representing a freshwater mammal (river otter) and for human health due to residue levels in freshwater and marine seafood. In addition, regional data show that Puget Sound harbor seal and southern resident killer whale prey items have higher PCDD/Fs compared to the same prey from the Strait of Georgia and the British Columbia coast. There were not sufficient data to conduct hazard evaluations for surface waters or sediments, or to evaluate tissue residue effects.

In the Puget Sound region, the major historical sources of PCDD/Fs – use of elemental chlorine in pulp bleaching, pentachlorophenol wood treatment operations, and combustion of saltwater-infused hog fuel – have been eliminated to a large extent (EPA, 1991; Yake et al., 1998; EPA, 2006), and ongoing releases are from combustion sources such as backyard burn barrels. Accumulation in biota is likely to be mainly a result of historical releases which continue to cycle in the aquatic environment, although no loading analyses were conducted to corroborate the small releases estimated for the Puget Sound basin.

Dichlorodiphenyltrichloroethane (DDT) and metabolites DDD and DDE

DDT compounds were found to be a Priority 1 level of concern for fresh surface waters, the freshwater and saltwater bird species evaluated, and human health. There were not sufficient observed or effects data to conduct hazard evaluations for marine surface waters, sediments (fresh and marine), or direct effects based on marine tissue residues. Regional data show high levels in accumulation for a variety of Puget Sound fish and marine mammals, including evidence to show apparent links between high levels in top-level, marine-mammal predators and their prey items.

Although a ban on DDT use in the United States has been in effect for decades, DDT compounds continue to exceed numerous documented effects levels due to their persistence, particularly in freshwater. DDT also persists in tissues of aquatic biota due to its highly bioaccumulative nature, but concentrations in Puget Sound basin fish are generally low, particularly when compared to watersheds with intensive agricultural use outside of the basin, such as watersheds in eastern Washington (e.g. Schneider and Coots, 2006; Johnson et al., 2010b). There are no apparent ongoing anthropogenic releases of DDT in the Puget Sound basin, and overall loading to Puget Sound appears to be low. DDT compounds will likely persist in the aquatic environment due to mobilization of DDT-bound soil particles and continued cycling in the aquatic environment due to historical releases.

Polycyclic aromatic hydrocarbons (PAHs)

PAHs are a Priority 1 level of concern for freshwater sediments and human health. There were not sufficient observed data to conduct adequate hazard evaluations for all individual PAHs in surface waters or for human health due to residues in offshore marine seafood. Localized areas with high degrees of PAH-contaminated sediments have also been a historical problem in Puget Sound, and resulting liver lesions in English sole have been a well-documented pathology associated with exposure to these sediments (Malins et al., 1987; Landahl et al., 1990; Myers et al., 1990). Recent evidence suggests that risks of liver lesions dramatically decrease when PAH-contaminated sediments are capped or removed (PSAT, 2007), and liver disease in English sole is currently being proposed as a Puget Sound-wide indicator of ecosystem health (PSP, 2011b).

There appears to be large, ongoing anthropogenic releases of PAHs in the Puget Sound basin. Generally speaking, PAH sources may be broken down into two categories: combustion emissions and releases from creosote-treated materials. PAH loads to Puget Sound are three orders of magnitude lower than estimated releases, but these loading estimates do not take PAH releases from creosote-treated pilings directly to marine waters into account. Groundwater appears to be the largest delivery pathway for PAHs, but the groundwater loading estimates are based largely on estimates derived from non-detects and should be viewed with caution. Estimated PAH loads through surface runoff are comparatively small due to the infrequency at which PAHs were detected in surface water samples.

To some degree, the lack of detectable PAHs in surface runoff may reflect the major sources and release mechanism. Few of the major sources would be expected to release PAHs directly to impervious surfaces, with the possible exception of PAH releases through motor oil loss.

Bis (2-ethylhexyl) phthalate (DEHP)

DEHP is a Priority 1 level of concern for freshwater sediments and human health. There were not sufficient observed data to conduct adequate hazard evaluations for marine surface waters or human health due to residues in offshore marine seafood.

There appears to be large ongoing anthropogenic DEHP releases in the Puget Sound basin. In addition to air emissions from point sources, DEHP is released from a variety of materials, particularly those containing PVC, although DEHP may be released from some non-polymer products as well. Surface runoff is the largest delivery pathway, although DEHP was not measured in air deposition samples. The major DEHP sources and mechanisms of release suggest that atmospheric deposition may be a major pathway. DEHP loads from POTW discharge are also relatively high and may indicate loss through indoor consumer products, and subsequent rinsing down the drain is another important delivery pathway.

Triclopyr

Triclopyr was evaluated only for hazards in fresh surface waters and was assigned a Priority 2 level of concern. There were not sufficient observed or effects data to conduct adequate hazard evaluations for marine surface waters or sediments.

Unlike other COCs evaluated, triclopyr’s only intended use is as an herbicide, and therefore its usage equates to environmental release. Major uses in the Puget Sound basin appear to be from crop and golf course use, with minor applications to road and railroad right-of-ways and for limited aquatic weed control. Surface runoff was the only delivery pathway where loads were calculated; loads were estimated to be two orders of magnitude below release estimates.

Nonylphenol

Nonylphenol was evaluated only for hazards in fresh surface waters and marine offshore waters; it was assigned a Priority 2 level of concern for both. There were not sufficient observed or effects data to conduct adequate hazard evaluations for nearshore marine surface waters or sediments. Nonylphenol is not highly toxic, but instead may exert effects at the sub-lethal level, and has documented endocrine-disrupting effects (King County, 2007). Compared with most other COCs, little is known about nonylphenol sources and effects in the Puget Sound basin.

There were limited release and loading estimates calculated for nonylphenol. Sources of nonylphenol were not fully assessed in the study of primary sources (Ecology, 2011), and loading was calculated only for surface runoff. Although a major source of nonylphenol is reported to be the breakdown of alkylphenol ethoxylates during the sewage treatment process (EPA, 2005), it was not detected frequently enough in POTW effluent to calculate loads from this pathway.

Oil and petroleum hydrocarbons

Petroleum was not fully evaluated for hazards in surface waters or sediments due to a lack of effects data (fresh surface waters) or observed data (sediments, marine surface waters). Evidence suggests that weathered crude oil has specific toxic effects to marine organisms, such as cardiac impairment and other effects in fish (Incardona et al., 2005; Incardona et al., 2006; Carls et al., 2008). However, these effects are due to specific components of weathered oil, most notably PAHs. The complexities of evaluating effects from petroleum exposure are discussed in the report section dealing specifically with petroleum.

The source inventory indicates that petroleum is released to the Puget Sound basin in much larger quantities than other COCs addressed, and the loading studies indicate it is loaded in the largest quantity. Most of the petroleum release is in the form of motor oil from engine drips and leaks and therefore likely to be directly to pavement. The large lube oil loads in surface water runoff may be a reflection of these releases. However, the large estimated releases of gasoline due to small fueling and transport spills did not translate to measurable loads in surface runoff.

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Prioritizing Chemicals of Concern and Recommendations to Fill Data Needs

Prioritizing COCs

A lines-of-evidence approach was used to move further toward a goal of deciding how best to prioritize actions and resources for controlling toxic chemicals in the Puget Sound basin. This lines-of-evidence approach considers information on the four major components of the assessment (sources, loading, pathways, and the relative hazards), but the approach mostly relies on COC sources and the relative hazards posed by COCs as determined by the hazard evaluation and review of other regional studies.

This approach adopts the rationale that chemicals with the greatest potential to elicit toxic effects at existing concentrations should be an important factor in determining the priority for source control efforts. Priority was also given to COCs associated with large opportunities for source control. This reflects the extent to which there are existing regulatory actions to control releases, such as bans, management of materials, or other permanent actions which reduce releases to the environment. Given the uncertainty associated with individual estimates of releases or loadings, this lines-of-evidence approach provides a supportable rationale for establishing relative priorities for control actions.

Opportunities for source control are considered large where the major sources of a COC have not been addressed by control actions; where some of the major sources have been addressed, opportunities may be considered medium. In cases where actions have been implemented to control and reduce all or most of the major sources and this appears to have resulted in low rates of loading to Puget Sound, the opportunities for controlling a COC are considered small. This assessment relies principally on the *Sources Report* (Ecology, 2011) with limited input by Ecology staff and management to gauge the opportunities for source control; this assessment was not intended to be a detailed review of management initiatives. Table 35 summarizes major sources for each COC and possible opportunities for reducing those sources.

Based on the lines-of-evidence approach, copper, PAHs, DEHP, and petroleum sources were rated as have the highest priority for early actions. The reasoning for this determination is as follows:

- A substantial portion of the fresh and marine water copper data observed basin-wide falls within concentrations where effects have been documented (including reduced olfactory function in salmonids). Copper is released in large quantities from a variety of sources which appear to translate to substantial loads to the Puget Sound ecosystem. The use of copper in pesticide applications and the release of copper from roofing materials are sources which warrant further investigation. In addition, the effectiveness of recent legislation to limit copper in brake pads and vessel anti-fouling paint should be evaluated.
- A number of individual PAHs surpass (do not meet) freshwater sediment guidelines and human health criteria. In addition, a variety of studies have demonstrated links between PAH

exposure and adverse effects to regionally relevant aquatic species. There appear to be numerous opportunities for control actions, primarily for combustion sources and for creosote-treated wood.

- Observed DEHP concentrations in both freshwater and marine environments exceed (do not meet) criteria for protection of benthic species and human health. Substantial amounts of DEHP are released in the Puget Sound basin, much of which occurs initially through releases to air from off-gassing of plasticized polymers and point-source air emissions. Several non-polymer uses of DEHP may also provide opportunities for source reduction.
- The relative hazard posed by petroleum in the Puget Sound basin was not able to be evaluated due primarily to the lack of biological-effects data and the absence of criteria to protect aquatic organisms, wildlife, or human health. However, some of the COCs addressed in this assessment are components of petroleum and may be released in substantial quantities along with the release of petroleum. In particular, substantial releases of PAHs are estimated to be released from petroleum. The major sources of petroleum are diffuse, such as motor oil drips and leaks and minor gasoline spillage during vehicle fueling, and therefore offer ample opportunities for reduction efforts.

Several COCs were found to be a Priority 1 level of concern based on the hazard evaluation but were not determined to be among the highest priorities for reduction actions since the major sources have been addressed through regulatory programs or other efforts. For instance, mercury poses a relatively high hazard to freshwater and marine aquatic organisms and wildlife based on doses calculated from observed data. However, many of the historical regional sources of mercury to the Puget Sound basin have been eliminated or are being addressed by the Mercury Chemical Action Plan (Ecology and WDOH, 2003). Similarly, PCBs are a Priority 1 level of concern for all hazard evaluation categories, but PCBs have been banned for decades, the major sources (use in electrical equipment) are highly regulated, and current loads to Puget Sound appear to be small.

Although the systematic prioritization approach identified four COCs for early actions, other factors should be considered to determine the need and feasibility for developing control and reduction strategies for other COCs. For instance, PBDEs are ubiquitous environmental contaminants, and although voluntary actions and bans have removed major PBDE formulations from new consumer products, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources. The hazard evaluation was not able to adequately assess the relative hazards associated with PBDEs due to a lack of environmental standards, although there is evidence in the available literature to suggest this COC may pose a hazard at observed concentrations.

Additional research is needed to assess the relative hazards posed by PBDEs and other COCs for which there are only limited environmental data. By the same token, COCs with limited source information should be further evaluated to assess additional opportunities for source control. Of the COCs addressed in this report, PBDEs and nonylphenol were the COCs that should receive top attention for further research on potential hazard as well as possible opportunities for source control.

Table 35. Summary of Possible Actions to Reduce COCs in the Environment.

COC	Opportunities for Source Control	Major Ongoing Anthropogenic Sources	Possible Actions for Reductions
Arsenic	Medium	Industrial air emissions	Maintain existing permit controls.
		CCA-treated wood leaching	Continue ban for most non-structural uses.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Cadmium	Medium	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Copper	Large	Pesticides use on urban lawns and gardens	More data needed on actual pesticide use.
		Residential plumbing component leaching	Continue to implement Lead and Copper Rule.
		Brake pad abrasion	Continue to implement legislation enacted to reduce source.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vessel anti-fouling paint leaching	Continue to implement legislation enacted to reduce source.
Lead	Small	Ammunition and hunting shot use	Implement CAP and enforce existing regulations.
		Loss of fishing sinkers and wheel weights	Implement CAP and enforce existing regulations.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Aviation fuel combustion	Implement CAP and enforce existing regulations.
Mercury	Medium	Consumer product improper disposal	Continue to implement CAP and enforce existing regulations.
		Crematoria and industrial air emissions	Continue existing permit limits.
Zinc	Large	Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
		Vehicle tire abrasion	Investigate source where it poses local concern.
Total PCBs	Small	Electrical equipment spills and leakage	Continue programs for management and disposal.
		Residential trash burning	Continue enforcing existing ban.
		Building sealant (caulk) volatilization and abrasion	Investigate source where it poses local concern.
Total PBDEs	Medium	Furniture, computer monitors, and other components of residential and commercial indoor environments	Enforce ban on new products but consider control actions to reduce the release from existing products.
PCDD/Fs	Small	Backyard burn barrels	Continue enforcing existing ban.
Total DDT	Small	None apparent	Investigate source where it poses local concern.
Total PAHs	Large	Woodstoves and fireplace combustion emissions	Continue change out programs, investigate catalysts/capture devices, promote alternatives to wood heat.
		Vehicle combustion emissions	Anti-idling programs, continue/expand engine retrofits for private section engines, enforce existing vehicle controls.
		Creosote-treated piling, railroad ties, and utility poles	Control actions needed, gather information to identify highest priority areas.
DEHP	Large	Polymer (primarily PVC) off-gassing	Gather additional information on extent of releases.
		Industrial, commercial, and institutional air emissions	Maintain existing permit controls.
		Roofing material leaching	Possible opportunity for source control, but more data needed on extent of releases.
Triclopyr	Medium	Herbicide use on crops and golf courses	More data needed on pesticide use.
Nonylphenol	Unknown	Industrial, commercial, and institutional air emissions	More information needed on emissions from these sources and unidentified releases.
Petroleum	Large	Motor oil drips and leaks Used motor oil improper disposal	Expand existing education/workshop programs.
		Gasoline spillage (minor) during fueling	Possible opportunity for source control, but more data needed on extent of releases.

Bold=Recommended as priority for near-term actions based on lines-of-evidence approach.

Strategies to control toxic chemicals in the environment may be accomplished using two general approaches: control at the source and control along the pathway. Source control strategies aimed at limiting or eliminating the initial release of chemicals may be achieved through education, chemical alternatives, release prevention technologies, or banning the use of specific chemical products. Control of a chemical once it has been released in the environment is typically more difficult and expensive, involving the use of a management actions (e.g. timing of pesticide application) or physical and technological resources (e.g. grass-lined ditches along roadways, wastewater treatment plants).

Prevention is the preferred option for controlling toxic chemicals in the environment, but source control options are not always feasible or necessary. While finding, reducing and eliminating primary releases of COCs at their source is critical to a clean and sustainable Puget Sound, so too is ensuring compliance with hazardous waste regulations, inspecting permitting facilities to ensure air and water quality, responding to spills, and cleaning up toxic messes when COCs are mismanaged.

The following recommendations provide a mixture of possible source and pathway control priorities for the target list of COCs addressed in this report.

General Recommendations

- Use results from the Puget Sound Toxics Loading Analysis (PSTLA) to help develop a long-term strategy to reduce toxic threats to Puget Sound. This strategy should include a comprehensive list of actions to prevent and manage chemical releases in conjunction with cleanup actions to reduce overall contaminant levels.

- Develop a list of specific control actions or source/pathway investigations to support control actions that may be incorporated into Ecology's long-term strategy for control of toxic chemicals under the National Estuary Program. These actions or investigations should be consistent with the findings and broader recommendations presented in this report. Examples of specific actions or investigations might be (1) adopting Low Impact Development Best Management Practices in commercial/industrial areas or (2) assessing the effectiveness of piling removal programs to reduce PAHs in the aquatic environment.

- Couple source control actions with effectiveness monitoring to assess if and how source control actions are actually reducing contaminant levels. This information is needed for an adaptive management framework to evaluate which actions should continue or be discontinued in favor of more effective actions.

Specific Recommendations

- Roofing materials appear to be an important source of metals and possibly DEHP in the Puget Sound basin. Monitoring should be conducted to further evaluate the release of these contaminants from roofing materials.

- Reduce copper loads to freshwater streams and marine waters, and assess the impacts of these efforts. The impact of recently passed legislation limiting copper and other metals in brake pads will not likely be known for at least a decade. In the meantime, the impact of copper released from pesticide/micronutrient use and vessel bottom paint should be investigated. One of the largest sources of copper is potentially from the urban use of copper in agricultural products by homeowners. Better information should be collected to evaluate the importance of these releases.

- Strategies to control the release of petroleum should be a high priority. Results of the *Sources* study (Ecology, 2011) indicate that over 9,000 metric tons of petroleum is released annually in the Puget Sound basin. Petroleum is generally released to impervious surfaces, enhancing its capacity to become mobilized in stormwater. There appears to be considerable opportunities for controlling sources of lube oil and gasoline since they are primarily released from crankcase drips and leaks and from minor spillage during fueling operations. The importance of these sources should be further evaluated.

- Strengthen existing programs to remove creosote pilings and bulkheads from the aquatic environment. Information analyzed on PAH sources and loading suggests creosote pilings may account for the largest overall PAH release to surface waters. The feasibility and need to remove creosote-treated rail ties that are over water or adjacent to sensitive aquatic areas should also be evaluated.

- Evaluate the effectiveness of actions implemented to reduce the amount of mercury released to the environment. Multiple programs and agencies have been involved with mercury disposal and recycling programs since the initiation of the Mercury Chemical Action Plan in 2003 (Ecology and WDOH, 2003). Organizing and streamlining information would make it easier to track ongoing releases to the environment and end-of-life data for mercury-containing materials.

- Evaluate the factors that appear to be effectively attenuating metals from roof and road runoff in low-density residential areas, and consider how to apply these factors in commercial/industrial areas where there appears to be little attenuation between sources and streams. If warranted, consider applying these attenuation mechanisms to high-density residential areas.

- Conduct inspections once every three years at those businesses in Washington that routinely handle large amounts of COCs. Washington is failing to find and resolve environmental threats from millions of pounds of hazardous waste in the Puget Sound basin. Hazardous wastes are toxic, flammable, or reactive, and when mismanaged, they contaminate soil, air, and water. Ten years ago, hazardous waste inspectors found serious environmental threats at 27% of businesses; the current rate is 63% (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication). Additional resources are needed to reduce the environmental threat rate to 30% by 2015, resulting in less contamination to soil, air, and water.

Data Needs

PSTLA was a four-year effort primarily focused on assessing chemical loads to Puget Sound from all of the major delivery pathways (groundwater, air deposition, surface water runoff, publicly-owned treatment works, and ocean exchange). This assessment will be useful in putting other scientific studies and action priorities into perspective, and possibly helping to shape new ones. However, there appears to be little value in pursuing further refinements to basin-wide loading assessments for the chemicals already addressed. This does not preclude the need for additional loading refinements for particular pathways or basin-wide modeling exercises, but it appears that current data needs should be conducted at a finer resolution to focus specific actions. These smaller scales may be geographical (e.g. watersheds, specific land covers, urban bays), specific pathways (e.g. stormwater), related to specific sources (e.g. the contribution of emissions sources to chemicals in stream runoff), or assessments of hazards (e.g. local hazard evaluation).

The following recommendations are provided to fill these finer-scale data needs:

- Characterize the factors that lead to high COC concentrations in streams draining commercial/industrial and agricultural areas. Assess runoff in high-density urban areas and, if warranted, assess the factors leading to high COC concentrations as well.

- Collect information on agricultural and urban usage of copper-based products in the Puget Sound basin.

- Evaluate concentrations of cadmium, copper, lead, zinc, and DEHP released from various roofing materials.

- For any work conducted to assess PAHs and petroleum hydrocarbons in surface runoff or POTWs, include sampling and analytical methods better suited to detection of these chemicals.

- Stormwater discharges directly to Puget Sound or to major rivers near their mouths should be assessed for chemicals to evaluate the importance of this loading pathway. Although the surface runoff study (Herrera, 2011) theoretically encompassed these conveyances, they were likely underestimated since high-density urban areas were under-represented in the study. Much of the information required for such an assessment may soon be available through data collection and reporting requirements of the Phase 1 municipal stormwater permit. A detailed analysis of this dataset should be conducted.

- Incorporate the data collected under PSTLA into the Puget Sound Box Model for the purpose of evaluating reductions needed to meet the Puget Sound “dashboard indicators” and other appropriate environmental targets. In addition to PCBs, selected metals, PBDEs, and PAHs appear to be good candidates for modeling since there are ample opportunities for control actions and the model may be able to predict conditions needed to meet reduction targets.

- More information is needed to help distinguish natural and legacy sources of contaminants in environmental pathways such as surface water runoff. This will help gauge the feasibility and effectiveness of actions taken to reduce releases of chemicals from contemporary anthropogenic (human-caused) releases.

- Continue to identify and assess chemicals that may be more detrimental to the Puget Sound ecosystem than the COCs addressed in PSTLA studies. Current-use pesticides, pharmaceuticals, and personal care products are examples of chemicals that are delivered to fresh and marine surface waters of the Puget Sound basin, yet their potential for effects is poorly understood (Lubliner et al., 2010).

- In general, industrial, commercial, and institutional point sources do not account for large releases of COCs. Instead, a variety of diffuse (nonpoint) sources account for a majority of the COC releases. However, it will be important to ensure that both the results of this *Assessment Report* and data on existing prevention and management controls help guide future actions and investments on Puget Sound clean-up and restoration work.

  Lack of investment in existing programs designed to safely manage COCs produced by commerce can let otherwise controlled and contained COCs "out of their bottle," where they become a threat to Puget Sound. For example, Washington ranks near the bottom of states in the U.S. for safe hazardous waste management (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication). Hazardous wastes are persistent, toxic, flammable, or reactive, and when mismanaged, they contaminate land, air, and water. Environmental threats are posed from millions of pounds of hazardous waste and hazardous products in the Puget Sound basin. Conducting fewer hazardous waste inspections results in more violations that directly contaminate land and water. So it is not surprising that as state inspection resources have diminished, the chance of finding spills of COCs and other significant environmental threats are at historic highs (Darin Rice, Ecology Hazardous Waste and Toxics Reduction Program, written communication).

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Appendices

Appendix A. Summary of the Puget Sound Toxics Loading Analysis (PSTLA) Projects

Project	Ref	Preparer	Status	Type of Study
Phase 1				
Initial Estimate of Toxic Chemical Loadings to Puget Sound	Hart Crowser et al., 2007	Hart Crowser, Ecology, EPA, Partnership	Completed - 2007	Loading estimates. Simple model using available data.
Phase 2				
Improved Estimates of Loadings from Surface Runoff and Roadways Addendum 1 (related to oil and petroleum) Addendum 2 (related to loading calculation method)	Envirovision et al., 2008a Ecology, 2009 Herrera, 2010	EnviroVision, Herrera, Ecology Ecology Herrera	Completed - 2008 Addendum 1 - 2009 Addendum 2 - 2010	Loading estimates. Simple model using available data. Clarification of oil and petroleum definitions. Revised and improved methodology for estimating runoff volumes.
Improved Estimates of Loadings from Dischargers of Municipal and Industrial Wastewater	Envirovision et al., 2008b	EnviroVision, Herrera, Ecology	Completed - 2008	Loading estimates. Simple model using available data.
Sediment Flux/Puget Sound Sediments Bioaccumulation Model – Derived Concentrations for Toxics	Ecology and Environment, 2009	Ecology and Environment	Completed - 2009	Criteria Evaluation. Bioaccumulation model using available data.
Identification and Evaluation of Water Column Data for Puget Sound and Its Ocean Boundary	Serdar, 2008	Ecology	Completed - 2008	Inventory and evaluation of existing data.
Studies to Support a Human Health Risk Assessment		EPA/Kissinger	Status unknown	Studies using available information to assess risks to human health from ingestion of toxicants in seafood.
Development of Simple Numerical Models – The Long-Term Fate and Bioaccumulation of Polychlorinated Biphenyls in Puget Sound	Pelletier, and Mohamedali, 2009	Ecology	Completed - 2009	Fate and bioaccumulation model for PCBs. Complex model using available data.
A Toxics-Focused Biological Observing System for Puget Sound	Johnson et al., 2010a	NOAA, UC Davis, WDFW	Completed - 2010	Proposal to monitor toxicants. Based on review of existing data.
Phase 3				
Characterize Toxic Chemical Loadings via Surface Runoff	Herrera, 2011	Herrera, Ecology	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Modeling Surface Runoff in Two Pilot Watersheds	Under Development	Under Development	Under development	Under Development
Study of Atmospheric Deposition of Air Toxics to the Waters of Puget Sound	Brandenberger et al., 2010	Battelle, Ecology	Completed - 2010 (PCBs complete 2011)	Sampling and loading estimates. Simple model using newly acquired field data.
Characterization of Toxic Chemicals in Marine Waters and Selected Tributaries to Puget Sound	Gries and Osterberg, 2011	Ecology	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Refine Numerical Model of Toxics in Puget Sound and Evaluate Pollution Reduction Scenarios	Under Development	Under Development	Under development	Under Development
Priority Pollutant Scans of Ten POTWs	Ecology and Herrera, 2010	Ecology, Herrera	Completed - 2011	Sampling and loading estimates. Simple model using newly acquired field data.
Primary Sources of Selected Toxic Chemicals and Quantities Released in the Puget Sound Basin	Ecology, 2011	Ecology	Completed - 2011	Inventory of chemical releases using available data.
Pharmaceuticals and Personal Care Products in Wastewater Treatment Systems	Lubliner et al., 2010	Ecology, EPA	Completed - 2010	Evaluation of POTW treatment efficacy and sampling. Evaluation based on newly acquired field data.
Persistent Organic Pollutants in Three Guilds of Pelagic Marine Species from the Puget Sound	West, et al., 2011a and b; Noel et al., 2011	WDFW	Completed - 2011	Assessment of bioaccumulative chemicals in plankton, fish, and harbor seals. Based on newly acquired field data
Toxic Chemical Loadings via Groundwater Discharge Directly to Puget Sound	Pitz, 2011	Ecology	Completed - 2011	Loading estimates. Simple model using available data.
Assessment Report	Present Report	Ecology, King County DNR	Completed - 2011	Synthesis of existing PSTLA loading and sources information, hazard evaluation

Appendix B. Chemicals Analyzed for Loading Studies and Methods Used to Handle Non-Detects

Table B-1. Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Metals						
	Aluminum	X				
	Arsenic	**X**	**X**		**X**	**X**
	Barium	X				
	Beryllium	X				
	Cadmium	**X**	**X**		**X**	**X**
	Cobalt	X				
	Copper	**X**	**X**	**X**	**X**	**X**
	Lead	**X**	**X**	**X**	**X**	**X**
	Manganese	X				
	Mercury	**X**	**X**			**X**
	Monomethyl mercury		X			
	Nickel	X				
	Selenium	X				
	Thallium	X				
	Tin	X				
	Zinc	**X**	**X**	**X**	**X**	**X**
Polychlorinated Biphenyls (PCBs)						
	21 "NOAA Status & Trends" Congeners[f]		**X**			
	209 PCB Congeners	**X**		**X**	**X**	
Polybrominated Diphenyl Ethers (PBDEs)						
	14 PBDE Congeners[g]		**X**			
	38 PBDE Congeners[h]	**X**		**X**	**X**	
Polychlorinated Dibenzodioxins and Furans (PCDD/Fs)						
	Total TCDD					**X**
	Total TCDF					**X**

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Chlorinated Pesticides						
	2,4'-DDD	**X**		**X**	**X**	**X**
	2,4'-DDE	**X**		**X**	**X**	**X**
	2,4'-DDT	**X**		**X**	**X**	**X**
	4,4'-DDD	**X**		**X**	**X**	**X**
	4,4'-DDE	**X**		**X**	**X**	**X**
	4,4'-DDT	**X**		**X**	**X**	**X**
	Aldrin	X		X	X	
	alpha-BHC	X		X	X	
	beta-BHC	X		X	X	
	delta-BHC	X		X	X	
	gamma-BHC (Lindane)	X		X	X	
	Chlorpyriphos	X		X	X	
	cis-Chlordane	X		X	X	
	trans-Chlordane	X		X	X	
	Chlordane	X		X	X	
	Dacthal (DCPA)	X		X	X	
	DDMU			X		
	Dieldrin	X		X	X	
	Endosulfan I	X		X	X	
	Endosulfan II	X		X	X	
	Endosulfan sulfate	X		X	X	
	Endrin	X		X	X	
	Endrin Aldehyde	X		X	X	
	Endrin Ketone	X		X	X	
	Heptachlor	X		X	X	
	Heptachlor epoxide	X		X	X	
	Hexachlorobenzene	X		X	X	
	Methoxychlor	X		X	X	
	Mirex	X		X	X	
	cis-Nonachlor	X		X	X	
	trans-Nonachlor	X		X	X	
	Oxychlordane	X		X	X	
	Toxaphene	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Low Molecular Weight Polycyclic Aromatic Hydrocarbons (LPAHs)						
	Acenaphthene	**X**		**X**	**X**	**X**
	Acenaphthylene	**X**		**X**	**X**	**X**
	Anthracene	**X**	**X**	**X**	**X**	**X**
	Fluorene	**X**		**X**	**X**	**X**
	Naphthalene	**X**		**X**	**X**	**X**
	Phenanthrene	**X**	**X**	**X**	**X**	**X**
High Molecular Weight Polycyclic Aromatic Hydrocarbons (HPAHs)						
	Benzo(a)anthracene*	**X**	**X**	**X**	**X**	**X**
	Benzo(a)pyrene*	**X**	**X**	**X**	**X**	**X**
	Benzo(b)fluoranthene*	**X**	**X**	**X**	**X**	**X**
	Benzo(g,h,i)perylene	**X**	**X**	**X**	**X**	**X**
	Benzo(k)fluoranthene*	**X**	**X**	**X**	**X**	**X**
	Chrysene*	**X**	**X**	**X**	**X**	**X**
	Dibenzo(a,h)anthracene*	**X**	**X**	**X**	**X**	**X**
	Fluoranthene	**X**	**X**	**X**	**X**	**X**
	Indeno(1,2,3-cd)pyrene*	**X**	**X**	**X**	**X**	**X**
	Pyrene	**X**	**X**	**X**	**X**	**X**
Phthalate Esters						
	Bis(2-ethylhexyl)phthalate	**X**		**X**	**X**	**X**
	Butylbenzylphthalate	X		X	X	
	Diethylphthalate	X		X	X	
	Dimethylphthalate	X		X	X	
	Di-N-butylphthalate	X		X	X	
	Di-N-octylphalate	X		X	X	
Herbicides						
	2,3,4,5-Tetrachlorophenol	X		X		
	2,3,4,6-Tetrachlorophenol	X		X		
	2,4,5-T	X		X		
	2,4,5-TP (Silvex)	X		X		
	2,4,5-Trichlorophenol	X		X	X	
	2,4,6-Trichlorophenol	X		X	X	
	2,4-D	X		X		
	2,4-DB	X		X		
	3,5-Dichlorobenzoic acid	X		X		
	Acifluorfen	X		X		

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies.

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Herbicides						
	Bentazon	X		X		
	Bromoxynil	X		X		
	Clopyralid	X		X		
	Dicamba I	X		X		
	Dichlorprop	X		X		
	Diclofop-methyl	X		X		
	Dinoseb	X		X		
	Ioxynil	X		X		
	MCPA	X		X		
	MCPP (Mecoprop)	X		X		
	Pentachloroanisole	X		X	X	
	Pentachlorophenol	X		X	X	
	Picloram	X		X		
	Triclopyr	**X**		**X**		
Semivolatile Organics						
	1,2,4,-Trichlorobenzene	X		X	X	
	1,2-Dichlorobenzene	X		X	X	
	1,2-Diphenylhydrazine	X			X	
	1,3-Dichlorobenzene	X		X	X	
	1,4-Dichlorobenzene	X		X	X	
	1,7-Dimethylphenanthrene		X			
	1-Methylnaphthalene	X		X	X	
	2,2'-Oxybis[1-chloropropane]				X	
	2,4-Dichlorophenol	X		X	X	
	2,4-Dimethylphenol	X		X	X	
	2,4-Dinitrophenol	X		X	X	
	2,4-Dinitrotoluene	X		X	X	
	2,6-Dimethylphenanthrene		X			
	2,6-Dinitrotoluene			X	X	
	2-Chloronaphthalene	X		X	X	
	2-Chlorophenol	X		X	X	
	2-Methylnaphthalene	X		X	X	
	2-Methylphenol			X	X	
	2-Nitroaniline	X		X	X	
	2-Nitrophenol	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Semivolatile Organics						
	3,6-Dimethylphenanthrene		X			
	3,3'-Dichlorobenzidine	X		X	X	
	3B-Coprostanol			X	X	
	3-Nitroaniline	X		X	X	
	4,6-Dinitro-2-methylphenol	X		X	X	
	4-Bromophenylphenylether	X		X	X	
	4-Chloro-3-methylphenol	X		X	X	
	4-Chloroaniline	X		X	X	
	4-Chlorophenyl-Phenylether	X		X	X	
	4-Methylphenol	X		X	X	
	4-Nitroaniline	X		X	X	
	4-Nitrophenol	X		X	X	
	4-Nonylphenol	**X**		**X**	**X**	
	Benzoic acid			X	X	
	Benzyl alcohol			X	X	
	bis(2-Chloroethoxy)methane	X		X	X	
	bis(2-Chloroethyl)ether	X		X	X	
	Bisphenol A	X		X	X	
	Caffeine	X		X	X	
	Carbazole	X		X	X	
	Cholesterol	X		X	X	
	Dibenzofuran	X		X	X	
	Ethanol, 2-chloro, phosphate (3:1)	X		X	X	
	Hexachlorobutadiene	X		X	X	
	Hexachlorocyclopentadiene	X		X	X	
	Hexachloroethane	X		X	X	
	Isophorone	X		X	X	
	Nitrobenzene	X		X	X	
	N-Nitrosodimethylamine	X		X		
	N-Nitrosodi-N-propylamine	X		X	X	
	N-Nitrosodiphenylamine	X		X	X	
	Perylene		X			
	Phenol	X		X	X	
	Retene	X	X	X	X	
	Triclosan	X		X	X	
	Triethylcitrate	X		X	X	

Table B-1 (Cont'd). Chemicals Analyzed for Loading Studies

Chemical Class		Loading Study				
	Chemical	Surface Runoff [a]	Atm. Dep.[b]	POTWs[c]	Ocean Exch.[d]	Ground-water[e]
Oil & Grease and Petroleum Hydrocarbons						
	Oil & Grease	**X**				
	TPH-Gas range	**X**				**X**
	TPH-Diesel range	**X**				**X**
	TPH-Lube oil range	**X**				**X**
Anhydrosugars						
	Galactosan		X			
	Levoglucosan		X			
	Mannosan		X			
Perfluorinated Compounds						
	Perfluorodecanoate			X		
	Perfluoroheptanoate			X		
	Perfluorohexanoate			X		
	Perfluorononanoate			X		
	Perfluorooctanoate			X		
	Perfluorooctane sulfonate			X		
	Perfluoropentanoate			X		
	Perfluorohexane sulfonate			X		
	Perfluorobutanoate			X		
	Perfluorobutane sulfonate			X		
	Perfluorooctane sulfonamide			X		
	Perfluoroundecanoate			X		
	Perfluorododecanoate			X		

Bolded and Shaded cells indicate Chemicals of Concern

[a] Herrera, 2011

[b] Brandenberger et al., 2010

[c] Ecology and Herrera, 2010

[d] Gries and Osterberg, 2011

[e] Pitz, 2011

[f] 21 "NOAA Status & Trends" Congeners = PCB-8, -18, -28, -44, -52, -66, -77, -101, -105, -118, 126, -128, -138, -153, -170, -180, -187, -195, -200, -206, and -209

[g] 14 PBDE Congeners = PBDE-17, -28, -47, -66, -71, -85, -99, -100, -138, -153, -154, -183, -190, -209

[h] 38 PBDE Congeners = PBDE-7, -10, -15, -17, -28, -30, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -139, -140, -153, -154, -156/159, -171, -180, -183, -184, -191, -196, -197/204, -201, -203, -205, -206, -207, -208, -209

[*] Carcinogenic PAHs (cPAHs)

Table B-2. Methods Used to Derive Representative Chemical of Concern (COC) Concentrations Where Sample Results Include Non-Detects.

Loading Study	**Scenario and Substitution Methods for Non-Detects (NDs)**		**Rules for Summing Constituents for Groups (e.g. PAHs, PCBs)**	**Possible Bias as a Result of Method**	**COCs Where Substitution Method Was Used**
	All of data set NDs	**Part of data set NDs**			
Surface Runoff (Herrera, 2011)	Maximum RL used and the final derived values were presented as "<" and flagged with a "U"	Where ≥ 50% of results were ND, ½ MRL assigned to NDs and final value flagged as "E" Where < 50% of results were ND, ½ MRL assigned to NDs with no flag for final value	Only detected results were summed (zero assigned to NDs) Where all results were ND, the highest MRL was used to represent the sum	Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 70% results are ND[a]. At higher rates of ND, this substitution method may yield conservative results (biased high) For summed parameters, the procedure used yields minimum or near-minimum possible values	Arsenic and copper were 0% ND (detected in 100% of samples) ½ MRL assigned to NDs for Lead, mercury, zinc, total PCBs, and total PBDEs were <50% ND (detected in ≥ 50% and < 100% of samples) ½ MRL assigned to NDs for cadmium, total PAHs, cPAH, LPAH, HPAH, DEHP, triclopyr, nonylphenol, and lube oil were ≥ 50% ND (detected in < 50% of samples)
Atmospheric Deposition (Brandenberger et al., 2010)	Not applicable	For all parameters except PCBs, MDLs assigned to NDs For PCBs, zero assigned to NDs	For all parameters except PCBs, detected results and NDs (assigned MDLs) were summed For PCBs, only detected results were summed (zero assigned to NDs)	For all parameters except PCBs, the procedure used yields maximum possible values For PCBs, the procedure used yields minimum possible values	MDLs assigned to NDs for PBDEs Zero assigned to NDs for PCBs
POTWs (Ecology and Herrera, 2010)	No attempt was made to derive representative concentration where FOD < 50%	Where n ≥ 10 and FOD ≥ 50%, ROS used to calculate representative concentration Where n < 10 and FOD ≥ 65%, ½ MRL assigned to NDs	Only detected results were summed (zero assigned to NDs) Where all results were ND, the highest MRL was used to represent the sum	Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 50% results are ND[a]. This substitution procedure was not used at higher FODs. ROS method was found to yield similar results when compared to substitution of ND with ½ MRL. For summed parameters, the procedure used yields minimum or near-minimum possible values	Copper, lead, and zinc were 0% ND (detected in 100% of samples) ROS used for some PBDE congeners, some individual PAHs, and DEHP ½ MRL assigned to NDs for some PCB congeners Representative concentrations not calculated for DDT compounds, some individual PAHs, triclopyr, nonylphenol

Table B-2 (Cont'd). Methods Used to Derive Representative Chemical of Concern (COC) Concentrations Where Sample Results Include Non-Detects.

<table>
<tr><th rowspan="2">Loading Study</th><th colspan="2">Scenario and Substitution Methods for Non-Detects (NDs)</th><th rowspan="2">Rules for Summing Constituents for Groups (e.g. PAHs, PCBs)</th><th rowspan="2">Possible Bias as a Result of Method</th><th rowspan="2">COCs Where Substitution Method Was Used</th></tr>
<tr><th>All of data set NDs</th><th>Part of data set NDs</th></tr>
<tr><td>Ocean Exchange (Gries and Osterberg, 2011)</td><td>No attempt was made to derive representative concentrations where all of data set were NDs</td><td>When most of the samples had detected results, only detected values were used to calculate representative concentrations
When few of the samples had detected results, ½ RL assigned to NDs</td><td>Only detected results were summed (zero assigned to NDs)
Where all results were ND, the highest MRL was used to represent the sum</td><td>Using only detected concentrations yields maximum possible values
Substitution of ND with ½ MRL appears to be reasonable estimate in cases where up to 70% results are ND [a]. At higher rates of ND, this substitution method may yield conservation results (biased high)
For summed parameters, the procedure used yields minimum or near-minimum possible values</td><td>Arsenic, cadmium, copper, and zinc were 0% ND (detected in 100% of samples)
Only detected values were used to calculate representative lead concentrations
½ MRL assigned to NDs for PCBs and PBDEs
Representative concentrations not calculated for DDT compounds, PAHs, DEHP, nonylphenol</td></tr>
<tr><td>Groundwater (Pitz, 2011)</td><td>Two methods were used: (1) ½ RL assigned to NDs, and (2) the minimum RL of the data set was assigned to NDs</td><td>Two methods were used: (1) ½ RL assigned to NDs, and (2) the minimum RL of the data set was assigned to NDs</td><td>All values were summed after values for individual chemicals were generated using the ND substitution procedures (½ RL or minimum RL)</td><td>Results appear to be biased low when comparing to results generated from using only detected values. Assignment of the minimum RL to NDs generates the most downward bias.</td><td>All procedures applied to all COCs</td></tr>
</table>

ND=non-detected
RL=reporting limit
MRL=maximum reporting limit
MDL=method detection limit
FOD=frequency of detection
ROS=regression on order statistics

[a] Antweiler, R.C. and H.E Taylor, 2008. Evaluation of statistical treatments of left-censored environmental data using coincident uncensored data sets: I. Summary statistics. Environmental Science and Technology 42: 3732-3728.

Appendix C. Summary of Release Estimates for All COCs (from Ecology, 2011)

Table C-1. Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Arsenic	**TOTAL**					**0.79 (0 - 1.7)**	**t/yr**	**100%**
Arsenic	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.28	t/yr	36% (0% - 100%)
Arsenic	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.27 (0.04 - 0.5)	t/yr	34% (0% - 100%)
Arsenic	Buildings and Grounds	Roofing materials - asphalt shingle	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.15 (0 - 0.84)	t/yr	19% (0% - 100%)
Arsenic	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.06	t/yr	8% (0% - 100%)
Arsenic	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Water	0.01	t/yr	2% (0% - 100%)
Arsenic	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1% (0% - 100%)
Cadmium	**TOTAL**					**0.96 (0.84 - 1.2)**	**t/yr**	**100%**
Cadmium	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.59 (0.5 - 0.7)	t/yr	61% (53% - 68%)
Cadmium	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.26	t/yr	27% (22% - 31%)
Cadmium	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release	Dust, Vapor	Air	0.06	t/yr	6% (5% - 7%)
Cadmium	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	3% (<1% - 6%)
Cadmium	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	1% (<1% - 7%)
Cadmium	Buildings and Grounds	Residential Fuel Use, except Wood	Dispersal of dust following wear	Particulate matter, Fugitive dust	air	0.01	t/yr	<1%
Cadmium	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Cadmium	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Cadmium	Industrial and Institutional Point Sources	Specialty Glass Manufacturer	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	<0.01	t/yr	<1%
Copper	**TOTAL**					**180 - 250 (120 - 390)**	**t/yr**	**100.0%**
Copper	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	1.1 - 73	t/yr	0.6% - 29% (0.3% - 38%)
Copper	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	39 (8.6 - 130)	t/yr	16% - 22% (4% - 45%)
Copper	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	37	t/yr	15% - 21% (10% - 31%)
Copper	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	27 (12 - 43)	t/yr	11% - 16% (3% - 29%)
Copper	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	25	t/yr	10% - 14% (6% - 21%)
Copper	Pesticides and Wood Preservation	Antifouling paint - total	Leaching, Ablation	Solubilized in water, Particulate matter in water	Marine surface water, Marine sediment	23 (12 - 54)	t/yr	9% - 13% (3% - 34%)
Copper	Pesticides and Wood Preservation	Agricultural use of pesticides - total	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	10	t/yr	4% - 6% (2% - 8%)
Copper	Miscellaneous Material Use	Micronutrients	Direct application to soil	Solid, Liquid	Soil	5.4	t/yr	2% - 3% (1% - 5%)
Copper	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release, Undefined release to surface water	Dust, Vapor, Undefined form released to surface water	Air, Surface water, Other	5.1	t/yr	2% - 3% (1% - 4%)
Copper	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.9 (0.02 - 5.4)	t/yr	<1% - 1% (<1% - 4%)
Copper	Pesticides and Wood Preservation	Aquatic-use algaecides in pools, fountains, spas, etc.	Direct application to water in contained pools (swimming pools, fountains, etc.)	Solid, Liquid	POTWs, Soils	1.5	t/yr	<1% - 1% (<1% - 1%)

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Copper	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.83	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.44	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Metal Foundries	Fugitive air release	Dust, Vapor	Air	0.22	t/yr	<1%
Copper	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.06 (0.04 - 0.08)	t/yr	<1%
Lead	**TOTAL**					**520 (150 - 1,000)**	**t/yr**	**100%**
Lead	Outdoor Product Use	Ammunition, Hunting shot use	Intentional loss	Soilid metal	Soil, Surface water	370 (27 - 820)	t/yr	72% (13% - 87%)
Lead	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	39	t/yr	8% (4% - 25%)
Lead	Outdoor Product Use	Fishing sinker loss	Unintentional loss	Soilid metal	Surface water, Aquatic sediment	36 (32 - 54)	t/yr	7% (3% - 31%)
Lead	Vehicles and Roads	Wheel weight loss	Unintentional loss	Solid metal	Impervious surfaces, Roadside areas	28 (20 - 29)	t/yr	5% (2% - 18%)
Lead	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	18 (15 - 20)	t/yr	3% (2% - 12%)
Lead	Non-Point Combustion Sources	Aviation fuel combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	16	t/yr	3% (2% - 10%)
Lead	Industrial and Institutional Point Sources	Various Industrial Facilities, not including pulp mills	Unknown release	Unknown form	Unknown	2.3	t/yr	<1% (<1% - 2%)
Lead	Industrial and Institutional Point Sources	Naval Shipyard	Unknown release	Unknown form	Unknown	1.8	t/yr	<1% (<1% - 1%)
Lead	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	2.6 (0.04 - 13)	t/yr	<1% (<1% - 8%)
Lead	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.2 (0.01 - 1.8)	t/yr	<1% (<1% - 1%)
Lead	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Water	0.66	t/yr	<1%
Lead	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.53	t/yr	<1%
Lead	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	0.21 (0.2 - 0.9)	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Lead	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.04	t/yr	<1%
Lead	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
Lead	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%
Mercury	**TOTAL**					**0.54 (0.47 - 0.61)**	**t/yr**	**100%**
Mercury	Miscellaneous Material Use	Thermostat Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.13 (0.11 - 0.16)	t/yr	24% (20% - 31%)
Mercury	Miscellaneous Material Use	Fluorescent Lamp Disposal	Volatilization	Vapor	Air	0.10	t/yr	18% (16% - 20%)
Mercury	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.05	t/yr	9% (8% - 11%)
Mercury	Industrial and Institutional Point Sources	Crematoria Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05 (0.02 - 0.07)	t/yr	9% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Cement Plants	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.04	t/yr	8% (7% - 9%)
Mercury	Miscellaneous Material Use	Auto Convenience Switch Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.04 (0.02 - 0.06)	t/yr	7% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Petroleum Refineries	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.03	t/yr	6% (5% - 7%)
Mercury	Personal Care Products	Dental Amalgam Excretion	Human Excretion	Excrement	POTWs, Groundwater	0.02	t/yr	4% (3% - 4%)
Mercury	Miscellaneous Material Use	Button Cell Batteries	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.02	t/yr	4% (3% - 4%)
Mercury	Industrial and Institutional Point Sources	Steel Mills	Unknown release	Unknown form	Unknown	0.02	t/yr	3%
Mercury	Industrial and Institutional Point Sources	Residual Fuel Oil Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	2%
Mercury	Personal Care Products	Dental Office Amalgam Waste	Wastewater	Liquid	POTWs, Groundwater	0.01	t/yr	2%
Mercury	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1%
Mercury	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	0.01	t/yr	1%
Mercury	Miscellaneous Material Use	Thermometers (Household)	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	<0.01	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Mercury	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	<0.01	t/yr	<1%
Mercury	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	<0.01	t/yr	<1%
Mercury	Vehicles and Roads	Gasoline and Diesel Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Zinc	**TOTAL**					**1,500 (300 - 3,200)**	**t/yr**	**100%**
Zinc	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	1,330 (210 - 2,800)	t/yr	87% (37% - 97%)
Zinc	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	82 (4.8 - 150)	t/yr	5% (<1% - 33%)
Zinc	Miscellaneous Material Use	Fertilizers and Micronutrients	Direct application to soil	Solid, Liquid	Soil	41	t/yr	3% (1% - 13%)
Zinc	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	30 (20 - 93)	t/yr	2% (<1% - 25%)
Zinc	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	12	t/yr	<1% (<1% - 4%)
Zinc	Vehicles and Roads	Motor oil leaks and improper disposal				7.9 (5.7 - 8.9)	t/yr	<1% (<1% - 3%)
Zinc	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	7.1 (0.22 - 44)	t/yr	<1% (<1% - 13%)
Zinc	Industrial and Institutional Point Sources	Steel Mills	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	4.3	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	3.7	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release	Dust, Vapor	Air	1.8	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Petroleum Refineries	Unknown release	Unknown form	Unknown	1.1	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.77	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Steel Galvanizers	Unknown release	Unknown form	Unknown	0.73	t/yr	<1%
PCBs	**TOTAL**					**2,100 (1,500 - 2,800)**	**kg/yr**	**100%**
PCBs	Miscellaneous Material Use	Large capacitors	Leakage	Liquid	Soil, Impervious surfaces	1,100	kg/yr	52% (40% - 75%)
PCBs	Miscellaneous Material Use	Small capacitors	Leakage	Liquid	Soil, Impervious surfaces	500 (1 - 1,000)	kg/yr	24% (<1% - 41%)

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCBs	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	280	kg/yr	13% (10% - 19%)
PCBs	Miscellaneous Material Use	Transformers	Leakage	Liquid	Soil, Impervious surfaces	130 (7 - 250)	kg/yr	6% (<1% - 15%)
PCBs	Buildings and Grounds	Sealants (Caulking)	Volatilization, Abrasion and fragmentation from weathering	Vapor, Sorption to dust particles	Air, Fugitive dust	110 (71 - 140)	kg/yr	5% (3% - 9%)
PBDEs	**TOTAL**					**680 (220 - 2,300)**	**kg/yr**	**100%**
PBDEs	Buildings and Grounds	Indoor office space air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	430 (120 - 750)	kg/yr	64% (7% - 88%)
PBDEs	Buildings and Grounds	Indoor residential dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	160 (100 - 320)	kg/yr	23% (5% - 72%)
PBDEs	Buildings and Grounds	Indoor office space dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	78 (<0.01 - 1,200)	kg/yr	12% (<1% - 84%)
PBDEs	Buildings and Grounds	Indoor residential air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	9.5 (0.6 - 18)	kg/yr	1% (<1% - 8%)
PCDD/Fs	**TOTAL**					**9.4**	**g TEQ/yr**	**100%**
PCDD/Fs	Buildings and Grounds	Backyard Burn Barrels	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	7.3	g TEQ/yr	77%
PCDD/Fs	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.81	g TEQ/yr	9%
PCDD/Fs	Industrial and Institutional Point Sources	Pulp and Paper Mills	Combustion, Undefined release to surface water	Aerosols, Vapor, Undefined form released to surface water	Air, Surface water	0.49	g TEQ/yr	5%
PCDD/Fs	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.38	g TEQ/yr	4%
PCDD/Fs	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.18	g TEQ/yr	2%
PCDD/Fs	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.08	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Cement Plants	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	g TEQ/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCDD/Fs	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Petroleum Refineries	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water	0.02	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Unknown	<0.01	g TEQ/yr	<1%
PAH	**TOTAL**					**310**	**t/yr**	**100%**
PAH	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	110	t/yr	34%
PAH	Pesticides and Wood Preservation	Creosote Treated Marine pilings - total	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Surface water, Air	54	t/yr	18%
PAH	Pesticides and Wood Preservation	Creosote Treated Railroad ties	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	43	t/yr	14%
PAH	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	29	t/yr	10%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Pesticides and Wood Preservation	Creosote Treated Utility poles	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	17	t/yr	6%
PAH	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	11	t/yr	3%
PAH	Vehicles and Roads	Petroleum spills, leaks, and improper motor oil disposal	Leakage, Spillage, Direct release, Improper disposal	Liquid	Impervious surfaces, Soils, Stormwater, POTWs, Landfills	11	t/yr	3%
PAH	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	6.5	t/yr	2%
PAH	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Combustion, Volatilization	Aerosols, Vapor, Particulate matter	Air	5.2	t/yr	2%
PAH	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	5.0	t/yr	2%
PAH	Industrial and Institutional Point Sources	Pulp and Paper Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	3.2	t/yr	1%
PAH	Industrial and Institutional Point Sources	Aluminum Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	2.7	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Petroleum Refineries	Fugitive air release, Volatilization, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	2.3	t/yr	<1%
PAH	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	2.0	t/yr	<1%
PAH	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.8	t/yr	<1%
PAH	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.3	t/yr	<1%
PAH	Vehicles and Roads	Gas Station Emissions	Volatilization	Vapor	Air	1.2	t/yr	<1%
PAH	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.98	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.94	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Coal tar sealants	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.92 (0.17 - 1.7)	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.86	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water, Other	0.58	t/yr	<1%
PAH	Buildings and Grounds	Roofing materials - total	Leaching	Particulate matter, Solubilized in water	Surface water, POTWs	0.57	t/yr	<1%
PAH	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.49	t/yr	<1%
PAH	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.30	t/yr	<1%
PAH	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.21	t/yr	<1%
PAH	Buildings and Grounds	Residential Yard Waste Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.15	t/yr	<1%
PAH	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	t/yr	<1%
PAH	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Non-Point Combustion Sources	Cigarette smoke	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03 (0.02 - 0.03)	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Asphalt - total	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Railroad Maintenance Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Phthalates	**TOTAL**					**34**	**t/yr**	**100%**
Phthalates	Personal Care Products	Fragrance	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	11	t/yr	32%
Phthalates	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	9.6	t/yr	28%
Phthalates	Vehicles and Roads	Car undercoating	Washout, Vapor	Liquid, Vapor	Surface water, Soil, Air	3.3	t/yr	10%
Phthalates	Miscellaneous Material Use	Lacquers and paint	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.9	t/yr	5%
Phthalates	Buildings and Grounds	PVC Coil coated roofing	Leaching, Volatilization	Liquid, Vapor	Surface water, POTWs, Air	1.5	t/yr	4%
Phthalates	Personal Care Products	Nail polish	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	1.4	t/yr	4%
Phthalates	Miscellaneous Material Use	PVC Coated fabric	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.2	t/yr	4%
Phthalates	Miscellaneous Material Use	Sealants, adhesives, etc.	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.1	t/yr	3%
Phthalates	Industrial and Institutional Point Sources	Plastics Manufacturer	Volatilization	Vapor	Air	0.86	t/yr	3%
Phthalates	Personal Care Products	Hair spray (aerosol and pump spray)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.4	t/yr	1%
Phthalates	Miscellaneous Material Use	PVC Cables (outdoor, above ground)	Volatilization	Vapor, Sorption to soil	Air	0.35	t/yr	1%
Phthalates	Personal Care Products	Deodorant (solid)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.29	t/yr	<1%
Phthalates	Miscellaneous Material Use	Shoe soles	Abrasion	Dust particles	Surface runoff, POTWs, Fugitive dust	0.2	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Wall coverings	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Roofing material	Leaching, Volatilization	Solubilized in water, Vapor	Surface water, POTWs, Air	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Flooring	Volatilization, Abrasion	Vapor, Dust particles	Air, Fugitive dust	0.1	t/yr	<1%
Phthalates	Miscellaneous Material Use	PVC Films, sheets, coated products	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.1	t/yr	<1%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Phthalates	Buildings and Grounds	PVC Hoses and profiles (outdoor)	Volatilization	Vapor	Air	0.09	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Cables (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Hoses and profiles (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Miscellaneous Material Use	Printing inks	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.08	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Boat Manufacturer	Volatilization	Vapor	Air	0.05	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Chemicals Distribution	Volatilization	Vapor	Air	0.04	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Paint and Coatings Manufacturers	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.03	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Volatilization	Vapor	Air	0.02	t/yr	<1%
Triclopyr	**TOTAL**					**150 (63 - 240)**	**t/yr**	**100%**
Triclopyr	Pesticides and Wood Preservation	Crop and Golf Course Use	Direct application to vegetation	Liquid	Vegetation and soils	150 (60 - 240)	t/yr	98% (95% - 99%)
Triclopyr	Pesticides and Wood Preservation	Forest Herbicide Use - State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.8 (0.4 - 1.2)	t/yr	<1% (<1% - 1%)
Triclopyr	Pesticides and Wood Preservation	Aquatic Weed Control	Direct application to surface water	Liquid or granular	Surface water	0.68	t/yr	<1% (<1% - 2%)
Triclopyr	Pesticides and Wood Preservation	Right-of-Way Maintenance--State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.5	t/yr	<1%
Triclopyr	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Liquid	Soil, Vegetation	0.43	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - State Highways	Direct application to vegetation	Liquid	Vegetation and soils	0.3	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - Railroads	Direct application to vegetation	Liquid	Vegetation and soils	0.1	t/yr	<1%
Nonylphenol	**TOTAL**					**0.18**	**t/yr**	**100%**
Nonylphenol	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.18	t/yr	100%

Table C-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Petroleum	**TOTAL**					**9,300**	**t/yr**	**100%**
Petroleum	Vehicles and Roads	Motor oil drips and leaks	Leakage	Liquid	Impervious surfaces	6,100	t/yr	66%
Petroleum	Vehicles and Roads	Minor gasoline spills from fueling vehicles and non-road equipment	Spillage	Liquid	Impervious surfaces, Soils	1,900	t/yr	21%
Petroleum	Vehicles and Roads	Improper disposal of used oil following oil changes	Direct release, Improper disposal	Liquid	Stormwater, Soils, POTWs, Landfills	960	t/yr	10%
Petroleum	Miscellaneous Material Use	Petroleum spills (large)	Spillage	Liquid	Surface water, Soil, Impervious surfaces	228 (223 - 233)	t/yr	3% (2% - 3%)

Appendix D. Hazard Evaluation Summary

Appendix D-1. Description of Methodology and Data Assessed (see the following pages)

The following sections of Appendix D are available only online as links to this *Assessment Report:* www.ecy.wa.gov/biblio/1103055.html

Appendix D-2. Hazard Evaluation – Results of Prioritization

Appendix D-3. Hazard Evaluation – Plots of Observed and Effects Data for Surface Waters

Appendix D-4. Hazard Evaluation – Plots of Observed Data and Threshold Values for Sediments

Appendix D-5. Hazard Evaluation – Plots of Observed and Effects Data for Tissue Residues

Appendix D-6. Hazard Evaluation – Plots of Estimated and Effects Doses for Wildlife

Appendix D-7. Hazard Evaluation – Plots of Observed Data and Criteria for Human Health

Appendix D-8. Hazard Evaluation – Summary Statistics for Environmental (Observed) Data

Appendix D-9. Hazard Evaluation – Water Effects Summary Data

Appendix D-10. Hazard Evaluation – Sediment Guidelines

Appendix D-11. Hazard Evaluation – Tissue Residue Effects Data

Appendix D-12. Hazard Evaluation – Wildlife Effects Data

Appendix D-13. Hazard Evaluation – ECOTOX QA Summary

Description of Contents for Appendix D

Appendix D-1
Description of the methodology and data assessed for the hazard evaluation. Includes a narrative summary of the results.

Appendix D-2
Tables showing results for the hazard evaluation. Each table shows a summary of whether the observed concentrations exceed threshold values, and notes on the data used for the comparisons.

Appendix D-3
Plots comparing observed environmental concentrations to effects concentrations for surface water.

Appendix D-4
Plots comparing observed environmental concentrations to guidelines for sediment.

Appendix D-5
Plots comparing observed environmental concentrations to effects concentrations for tissue residue.

Appendix D-6
Plots comparing calculated environmental doses to effects doses for wildlife.

Appendix D-7
Plots comparing observed environmental tissue concentrations to criteria for human health.

Appendix D-8
Tables showing summary statistics for the observed environmental concentrations used in the hazard evaluation.

Appendix D-9
Folder containing tables with summaries of ECOTOX data used in the hazard evaluation (the petroleum effects data are not from ECOTOX). A file containing ECOTOX codes is also included in this folder.

Appendix D-10
Table showing guidelines and other threshold values for sediment.

Appendix D-11
Tables showing summaries of the tissue residue effects for Lower Willamette River and the Lower Duwamish River Remedial Investigations

Appendix D-12
Folder containing tables with summaries of wildlife effects data. A list of references reviewed for the wildlife evaluation is also included in this folder.

Appendix D-13
Folder containing tables with summaries of the quality assurance (QA) review of the ECOTOX data and units.

Appendix D-1

Hazard Evaluation for Chemicals of Concern in the Puget Sound Basin – Description of Methodology and Data Assessed

Introduction

This chapter outlines the methods and results of the assessment conducted to estimate the relative hazard posed by exposure to the chemicals of concern (COCs) assessed in the Puget Sound Toxics Loading Analysis (PSTLA) studies. The primary purpose of this assessment was to provide a general overview of the potential for these COCs to cause adverse effects (to aquatic life and select wildlife species), and to a lesser extent, human health effects through seafood consumption within the Puget Sound basin. The assessment described here was not intended to identify specific ecological effects or quantify risk. The primary purpose of the assessment was to provide an additional weight of evidence (WOE), along with the loadings and sources information (documented in the main body of this report), to prioritize COCs for further action.

The large scale regional focus was intended to evaluate COCs at a broad level. A key goal of this effort was to provide information to help prioritize COCs based on their potential to cause adverse effects. The assessment used some of the chemical concentration data generated by the PSTLA studies discussed in this report; however, readily available environmental data for water, sediment and tissue from other sources were the primary basis of this prioritization process. To estimate the potential for effects, environmental data were compared to readily available toxicity data obtained primarily from established databases, sources and regulations. The outcome of this process was used to establish a general “priority” for management of each of the COCs.

This assessment included the following evaluations:

- Direct hazard to aquatic life through surface water exposure
- Direct hazard to benthic organisms through sediment exposure
- Direct hazard to aquatic life based on tissue residue levels
- Hazard to wildlife based on ingestion of prey, water and sediment
- Hazard to human health through fish/seafood consumption

Although some elements of the “Risk Assessment” process were applied to the hazard prioritization presented here, this effort is not intended to serves as a risk assessment. Conducting such an assessment for the Puget Sound region was beyond the scope of this effort. The remainder of this section describes the process used to acquire both the observed environmental data and toxicity data, in addition to the assumptions used to access and use this information. The section also documents the methods used to conduct the effects prioritization, including a description of the process used to determine the priority for each COC. Finally, the results of the assessment (organized by COC) and a discussion of the uncertainty and limitations associated with this process are presented.

Methods

The following sections describe the process used to identify and acquire both the observed environmental data and the toxicity data used for this assessment. Also described is the approach used to assess effects to wildlife, including selection of representative species ("receptors") and appropriate COCs, derivation of daily doses (to estimate toxicity from ingestion pathways) and the model used to estimate exposure to COCs by wildlife. Lastly, the processes used to summarize the environmental and effects data, determine the potential for effects and priority are described.

Environmental Data Collection

Environmental data, collected from a variety of sources, were used to estimate the general range of possible exposure concentrations to COCs. With the exception of a few specific sources, data collection was limited to readily available public databases and only those data collected between January 1, 2000 and July 2010 were considered "recent" and included. When available, surface water, sediment and tissue data were collected from the sources outlined in Table 1. Due to the different purposes for which some of these data were collected, not all sources included data for all matrices in both fresh water and marine environments.

Table 1. Summary of data sources and data types used in this assessment. All data were accessed from their respective sources in July 2010.

Data Source	Matrix					
	Water		Sediment		Tissue	
	Fresh	Marine	Fresh	Marine	Fresh	Marine
Ecology's EIM System [1]	X	X	X	X	X	X
King County's LIMS[2]	X	X	X	X	X	X
US Geological Survey [3]	X	N/A	See Footnote[3]	N/A	X	N/A
Puget Sound Ambient Monitoring Program	N/A	N/A	N/A	N/A	N/A	X
Regional EMAP [4]	N/A	N/A	N/A	X	N/A	X
ENNVEST Study [5]	N/A	N/A	N/A	N/A	N/A	X
Toxics Loading Studies [6]	X	X	N/A	N/A	N/A	X
WDFW[7]	N/A	N/A	N/A	N/A	N/A	X

N/A – data not available

1 - EIM – Ecology's Environmental Information Management System

2- LIMS - Laboratory Information Management System, King County data not previously submitted to EIM

3 - USGS data obtained from online database. Sediment data were obtained but not used; USGS only analyzes the <63µ sediment fraction, which is not comparable to the remainder of the data used in this assessment.

4 - Includes NOAA's mussel watch data (Valerie Partridge, Environmental Assessment Program, written communication, 2010).

5- US Department of Defense (Johnston, R.K. 2007)

6 - Includes Ecology's Ocean Exchange/River Mouth Loading study, Fish Tissue Assessment and Surface Runoff studies discussed in this document.

7 – Washington Department of Fish and Wildlife (West et al. 2011).

The datasets described in Table 1 were combined into a composite database. This process required a number of "rules" and assumptions to ensure that data were consistently formatted (similar naming conventions, units etc.) and in a chemical form appropriate for later comparison to effect concentrations. Table 2 outlines the key rules and assumptions used to combine and process the environmental data.

Table 2. Summary of rules and assumptions used to acquire and summarize observed environmental data.

All Data
• The COC list was expanded to include all forms of these chemicals.
• Data from all sources were standardized to reflect parameter and qualifier names as defined by EIM. When no EIM parameter (for certain co-eluting PBDE congeners) was available the closest match was selected.
• Only data collected between January 1, 2000 and July, 2010 were acquired.
• All data were standardized to common units.
• **Qualified** ”B” qualified data and “estimated data” were included. Data with the <u>following qualifiers were not included</u> in the assessment: ▪ “Rejected Data” ▪ The following “U” qualified non-detect data - “U”, “U?”, “UJ”, “UJG”, “UJK”,” UJL”.
• **Summing** - Polychlorinated biphenyl’s (PCBs), polycyclic aromatic hydrocarbons (PAHs) and dioxins in all matrices were summed based on SMS rules: ▪ For summed compounds, only compounds detected in a sample were summed.
• **PAHS** ▪ LPAHs include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene ▪ HPAHs include fluoranthene, pyrene, benz(a)anthracene, chrysene, total benzofluoranthenes (B, J and K), Benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene, and benzo(g,h,i)perylene. ▪ cPAHs were not summed for any media because standards and toxicity information for the sum of cPAHs whereas not available for water, sediment, tissue, or in the NTR.
• **Petroleum Compounds** – data for the following petroleum related compounds were identified and collected: TPH as heavy fuel oil, Diesel range TPH, Gasoline range TPH, Lube oil range TPH.
Sediment Data
• Organic carbon (OC) normalization was conducted for marine sediment data when the corresponding sediment quality value was OC-normalized. Otherwise, all sediment data were dry-weight normalized.
• USGS freshwater sediment data were presented as the chemical concentration in the <63 µm fraction. These sediment data were not used due to incompatibility with the majority of the available sediment data.
Tissue Data
• Tissue data were grouped into common tissue type designations for the tissue residue, wildlife and human health assessments. For example, mussel tissue data labeled as “somatic” and “visceral” were categorized as “whole body no shell”. Whole body tissue data labeled as “no-gut”, “no exoskeleton” were classified as “whole body”. Fillet data classified as “skin on”, “no skin” were combined and classified as “fillet”. Lipid-normalized tissue data were not used in this assessment due to the inconsistencies and availability of lipid data for all tissue concentrations.

The majority of data used in the assessment were obtained from EIM, followed by the King County LIMs. The remaining datasets were relatively small in comparison, but were included because the parameters and matrices measured complemented the EIM and LIMS datasets. While it is recognized that there may be other sources of environmental data (e.g., various research publications, NOAA, USFW) that could have been included in this assessment, due to the scope and timeline associated with this task, it was necessary to focus on the largest and most readily available electronic sources of primarily ambient data that did not require significant data review or re-entry. Since the intent of this effort was to better understand general regional conditions and not identify "hot spots", these data are assumed to provide reasonable estimates of exposure.

A review of the NOAA database (http://response.restoration.noaa.gov/type_subtopic_entry.php?RECORD_KEY%28entry_subtopic_type%29=entry_id,subtopic_id,type_id&entry_id(entry_subtopic_type)=751&subtopic_id(entry_subtopic_type)=5&type_id(entry_subtopic_type)=1) indicated that a significant proportion the applicable data were already included in EIM and LIMs and much of the data had been collected before 2000. A large proportion of the remaining data were primarily collected from the Duwamish River CERCLA site. It was decided that inclusion of these data in the assessment would possibly skew the results.

The primary goal of this assessment was to provide a high level summary evaluation of relative hazard; therefore, the data were differentiated into three course spatial scales; (1) freshwater, (2) marine nearshore, and (3) marine offshore. The freshwater to nearshore boundary was defined by the original data. If data were classified as "freshwater" by the original data source, they were retained as such; if classified as marine/saltwater the data were further parsed between near and offshore. Nearshore was defined as all marine areas less than 10 meters deep (based on MLLW); offshore was defined as all other marine areas.

Data records not meeting the intent of the nearshore/offshore designation were adjusted accordingly (e.g., marine locators >10m in depth in estuaries like the Duwamish were classified as nearshore). Some sample depths varied due to tidal influences. However, relatively few samples were attributed close enough to the 10m depth (e.g. 9m or 11m) to potentially fall into a different marine area based on depth of tide at the time of sampling.

Comparison of sediment data to sediment guidelines (described below) required that the marine sediment data for nonionic/non-polar organic chemicals be organic carbon (OC) -normalized. Dry-weight concentrations were used for marine sediment samples when OC was outside the range of 0.5 to 3.0%. No associated OC data were available for approximately 35% of the sediment samples. To utilize these samples, these data were OC-normalized using the mean nearshore (2.22%) or mean offshore (1.74%) OC percentages from the remaining sediment results. The process of correcting sediment data for OC resulted in some differences in the total number of measurements (N) for OC and dry weight normalized data presented in the summary tables and figures. For example, for a given COC the N for dry weight-based measurements may be 5, while the N for the OC-normalized measurements is 1.

Tissue data were grouped differently depending on which assessment was being conducted. Tissue samples were segregated into fresh, nearshore and offshore samples based on the location they were collected. Table 3 attributes tissue to freshwater or marine species for informational purposes.

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Asian clam	*Corbicula fluminea*	FW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bay ghost shrimp	*Neotrypaea californiensis*	SW	Decapod	Other invertebrate	not included
Bay mussel	*Mytilus trossulus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bent-nose macoma	*Macoma nasuta*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Black bullhead	*Ameiurus melas*	FW	Fish	Fish	Fish
Black crappie	*Pomoxis nigromaculatus*	FW	Fish	Fish	Fish
Blackmouth (Resident) Chinook salmon	*Oncorhynchus tshawytscha*	SW	Fish	Fish	Fish
Blue mussel	*Mytilus edulis*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Bluegill	*Lepomis macrochirus*	FW	Fish	Fish	Fish
Brook trout	*Salvelinus fontinalis*	FW	Fish	Fish	Fish
Brown bullhead	*Ameiurus nebulosus*	FW	Fish	Fish	Fish
Brown Rockfish	*Sebastes auriculatus*	SW	Fish	Fish	Fish
Brown trout	*Salmo trutta*	FW	Fish	Fish	Fish
Bull trout	*Salvelinus confluentus*	FW	Fish	Fish	Fish
Butter clam	*Saxidomus giganteus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
California mussel	*Mytilus californianus*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Catworm genus	*Nephtys*	SW	Bivalve + other invertebrates	Other invertebrate	not included
Channel catfish	*Ictalurus punctatus*	FW	Fish	Fish	Fish
Chinook salmon	*Oncorhynchus tshawytscha*	SW	Fish	Fish	Fish
Chum salmon	*Oncorhynchus keta*	SW	Fish	Fish	Fish
Coho salmon	*Oncorhynchus kisutch*	SW	Fish	Fish	Fish
Common carp	*Cyprinus carpio*	FW	Fish	Fish	Fish
Copper rockfish	*Sebastes caurinis*	SW	Fish	Fish	Fish
Cutthroat trout	*Oncorhynchus clarkii*	FW	Fish	Fish	Fish
Dabs	*Pleuronectidae*	SW	Fish	Fish	Fish
Dock shrimp	*Pandalus danae*	SW	Decapod	Other invertebrate	not included
Dungeness crab	*Cancer magister*	SW	Decapod	Other invertebrate	Invertebrate
English sole	*Parophrys vetulus*	SW	Fish	Fish	Fish
Fat gaper	*Tresus capax*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Flathead sole	*Hippoglossoides elassodon*	SW	Fish	Fish	Fish
Gaper clam	*Tresus sp.*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Graceful rock crab	*Cancer gracilis*	SW	Decapod	Other invertebrate	Invertebrate
Hake	*Merluccius*	SW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
	productus				
Japanese littleneck	*Tapes philippinarum*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Kokanee	*Oncorhynchus nerka*	FW	Fish	Fish	Fish
Largemouth bass	*Micropterus salmoides*	FW	Fish	Fish	Fish
Largescale sucker	*Catostomus macrocheilus*	FW	Fish	Fish	Fish
Lingcod	*Ophiodon elongatus*	SW	Fish	Fish	Fish
Longnose sucker	*Catostomus catostomus*	FW	Fish	Fish	Fish
Lumbriculus oligochaete	*Lumbriculus*	FW	Bivalve + other invertebrates	Other invertebrate	not included
Macoma clams	*Macoma sp.*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Mediterranean mussel	*Mytilus galloprovincialis*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Melita amphipods	*Melitidae*	SW	Decapod	Other invertebrate	not included
Milky venus	*Compsomyax subdiaphana*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Mountain whitefish	*Prosopium williamsoni*	FW	Fish	Fish	Fish
Northern pikeminnow	*Ptychocheilus oregonensis*	FW	Fish	Fish	Fish
Pacific Dover sole	*Microstomus*	SW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
	pacificus				
Pacific geoduck	*Panopea abrupta*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific herring	*Clupea pallasii*	SW	Fish	Fish	Fish
Pacific littleneck	*Protothaca staminea*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific oyster	*Crassostrea gigas*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pacific staghorn sculpin	*Leptocottus armatus*	SW	Fish	Fish	Fish
Pacific Tomcod	*Microgadus proximus*	SW	Fish	Fish	Fish
Peamouth	*Mylocheilus caurinus*	FW	Fish	Fish	Fish
Pile perch	*Rhacochilus vacca*	SW	Fish	Fish	Fish
Pink salmon	*Oncorhynchus gorbuscha*	SW	Fish	Fish	Fish
Pollock	*Theragra chalcogramma*	SW	Fish	Fish	Fish
Prickly sculpin	*Cottus asper*	SW	Fish	Fish	Fish
Pumpkinseed sunfish	*Lepomis gibbosus*	FW	Fish	Fish	Fish
Purple mahogany-clam	*Nuttallia obscurata*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Pygmy whitefish	*Prosopium coulterii*	FW	Fish	Fish	Fish
Quillback rockfish	*Sebastes maliger*	SW	Fish	Fish	Fish
Rainbow trout	*Oncorhynchus mykiss*	FW	Fish	Fish	Fish

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Ratfish	*Hydrolagus colliei*	FW	Fish	Fish	Fish
Red rock crab	*Cancer productus*	SW	Decapod	Other invertebrate	Invertebrate
Redside shiner	*Richardsonius balteatus*	FW	Fish	Fish	Fish
Reticulate sculpin	*Cottus perplexus*	FW	Fish	Fish	Fish
Rock bass	*Ambloplites rupestris*	FW	Fish	Fish	Fish
Rock sole	*Lepidopsetta bilineata*	SW	Fish	Fish	Fish
Sand sole	*Psettichthys melanostictus*	SW	Fish	Fish	Fish
Scorpion fishes (Order)	Scorpaeniformes	SW	Fish	Fish	not included
Sea cucumber	*Molpadia intermedia*	SW	Bivalve + other invertebrates	Other invertebrate	Invertebrate
Shiner perch	*Cymatogaster aggregata*	SW	Fish	Fish	Fish
Signal crayfish	*Pacifastacus leniusculus*	FW	Decapod	Other invertebrate	Invertebrate
Sixgill shark	*Hexanchus griseus*	SW	Fish	Fish	not included
Slender sole	*Eopsetta exilis*	SW	Fish	Fish	Fish
Smallmouth bass	*Micropterus dolomieui*	FW	Fish	Fish	Fish
Softshell clam	*Mya arenaria*	SW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Spot prawn	*Pandalus platyceros*	SW	Decapod	Other invertebrate	not included

Table 3. Fish and invertebrate tissues and their groupings for the assessments using tissue data.

Common name	Latin Name	Fresh vs Marine	Tissue Assessment	Human Health Assessment	Wildlife Assessment
Staghorn Sculpin	*Leptocottus armatus*	SW	Fish	Fish	Fish
Starry flounder	*Platichthys stellatus*	SW	Fish	Fish	Fish
Striped seaperch	*Embiotoca lateralis*	SW	Fish	Fish	Fish
Western Pearlshell	*Margaritifera falcata*	FW	Bivalve + other invertebrates	Bivalve-clam	Invertebrate
Yellow perch	*Perca flavescens*	FW	Fish	Fish	Fish

Summary Statistics

All environmental data were combined by matrix (water, sediment, tissue) and spatial location (freshwater, marine near- and offshore). Tissues were also grouped into “bivalve”, “fish” and “other invertebrate” categories for the human health assessment. Both fillet and whole-body samples were used for the human health assessment. For the tissue residue assessment, tissues were divided into “fish”; “non-decapod invertebrates”, and “decapods” (crabs and shrimps); all tissue residue comparisons were based on whole-body tissue concentrations. Summary statistics (min, max, mean, median, total number of samples, and frequency of detection) for these data were calculated using MSAccess and Total Access Statistics. Summary statistics for each matrix are presented in Appendix E.

Identification of Effects Concentrations

Unless noted otherwise, all toxicity data used in this assessment were obtained from readily available databases. A literature search was conducted to identify studies of contaminant impacts to northwest regional species which may not have been included in available toxicity databases. In many cases the regional data were not dose-response effects data and inappropriate to directly compare with observed environmental concentrations (i.e. data were lipid normalized, study included multiple chemical exposures, field based studies, etc.). These data are primarily discussed as an additional WOE when evaluating the overall hazard for each COC. The following sections describe the process used to obtain the effects data and any assumptions used in their selection.

Surface Water – Direct Effects to Aquatic Life

To determine the potential for effects to aquatic life from direct exposure to COCs in surface water, relevant effects concentrations were identified to compare with the observed environmental data. EPA’s ECOTOXicology (ECOTOX) database (http://cfpub.epa.gov/ecotox) was the primary source of surface water effects data. The “Advanced Database Query” option in ECOTOX was used to obtain the majority of the data which allows for selection of specific taxonomic, chemical, result, condition, publication, and report formats to suit the project needs.

The following rules were used to identify the appropriate toxicological effects data.

- Both aquatic plant and animal data were included in the search process (animal data were accessed in July 2010; plant data were accessed and added in June 2011).
- Effect concentrations classified as EC0, LC0, NOEC, NOEL, and NR-ZERO were not included because they were considered “no effect” results.
- Concentration units based on area (e.g., AI kg/ha, ae kg/ha), or any unit other than volume were excluded. Molar-type units (i.e., M, uM, nM) were converted to ug/L.
- The following endpoint types were included from the ECOTOX database: Lethal Concentration (LC)/Lethal Dose (LD), Effect Concentration (EC)/Effect Dose (ED), Lowest Observable Effect Concentration (LOEC), Lowest Observable Effect Level (LOEL), and NR-LETH (Near Lethal) values, and all effect measurements for both fresh and saltwater organisms. Endpoint types such as bioaccumulation factor, inhibition concentration, and time to mortality were not used. Bioaccumulation was addressed to some degree in the tissue,

wildlife and human health assessments described below. A detailed evaluation of the potential for effects associated with bioaccumulation was beyond the scope of this assessment.

- Washington State water quality criteria were included for comparison when available. Concentration types were selected to match the water quality criteria where available. Thus, for most metals, effects associated with the dissolved concentration type were used for comparison to freshwater and marine observed data. Total and dissolved mercury were used due to the different forms used by the acute and chronic WQC. Formulation type (F) was excluded for COCs except triclopyr, DDTs, and PCBs.
- If “NR” (not reported) was the result for concentration or media type (freshwater or saltwater), then that effect value was excluded.

A summary of the data derived from the ECOTOX database used in this assessment can be found in Appendix D-9.

Although efforts are made by EPA to accurately represent toxicity data in the ECOTOX database, the data are not thoroughly vetted through a detailed quality control process. It was beyond this effort’s capacity to review each of the thousands of original papers and documents from which ECOTOX was derived. However, to provide additional confidence in the quality of the ECOTOX data, 125 randomly selected documents were obtained and reviewed for accuracy and correspondence with ECOTOX. These papers represented approximately 5% of those identified by this assessment.

To evaluate the accuracy of molar unit conversions by ECOTOX, 25 papers were reviewed. Molar units were found to be converted correctly by 24 of the 25 randomly selected papers. One paper did not measure metallic zinc as reported by ECOTOX, it was instead evaluating zinc pyrithione, an organic zinc antifouling compound. If this paper is considered as reporting error the “unit error” rate is 3%.

To evaluate the ability of the ECOTOX database to accurately represent the data presented in the original source, 100 journal articles were reviewed representing 821 individual toxicity values. The review resulted in identification of 171 values that were incorrectly represented by ECOTOX (20% error rate) and would have an impact on the outcome of the assessment. A number of other errors were identified (e.g., misclassification of effects types and test species), but they did not impact the outcome of this assessment. The majority of errors were associated with use of the salt concentration of a COC to represent the effect concentration, rather than the active ingredient concentration.

Other common errors were associated with the classification of NOEC values as effect concentrations and the use of mixture concentrations to represent a single chemical exposure. Use of the salt concentration as the effect concentration rather than the active ingredient would likely underestimate the potential for effects, while use of NOEC values would likely overestimate the potential. A summary of the results of the ECOTOX QA/QC process can be found in Appendix D-13.

Sediment – Direct Effects to Benthic Organisms

To determine potential hazard to benthic organisms from direct exposure to sediment COCs, relevant sediment guidelines and thresholds were identified for comparison with the observed environmental data. The primary standards and guidelines used in this assessment were the Washington State Marine Sediment Management Standards (SMS) and the Floating Percentile (FP) based freshwater sediment guidelines developed by Ecology (Avocet Consulting 2003; Avocet Consulting and SAIC 2002; RESET 2009). A number of other sediment guidelines were also used to provide additional context to assess the sediment data. Because this effort was not exhaustive, not all available guidelines were included.

Three sets each of freshwater and marine sediment guidelines (total of 6 sets of guidelines) were selected for comparison to observed sediment concentrations. It is acknowledged that sediment pore water may be an additional important route of exposure for benthic organisms; however, readily accessible pore water toxicity data and observed pore water concentration data for the Puget Sound regional were not available. In addition, variability in the methods used to extract and analyze pore water makes comparison across studies challenging.

Marine sediment data were compared to the following guidelines/standards:

- The Washington State SMS (Chapter 173-204 WAC), which consist of two levels, a “Sediment Quality Standard” (SQS) and “Cleanup Screening Level” (CSL) and the “Apparent Effects Thresholds” (AETs) including the “Lowest AET” (LAET) and the “Second Lowest AET” (2-LAET)
- The Canadian Marine Sediment Guidelines (CCME 2001) which consist of a “threshold effect level” (TEL) and a “probable effects level” (PEL).

The SMS SQS was the primary standard used to evaluate the marine sediment data; the remainder of the guidelines presented and described here were intended to provide additional context and included as part of the WOE discussion.

The SMS SQS represents the concentration below which no adverse effects to biological resources are expected; the CSL is less stringent and corresponds to the concentration at which minor adverse effects to biological resources are expected. For comparison to the SMS, all nonionic/nonpolar organic compounds were normalized to percent total organic carbon (TOC) content. However, if TOC content was outside the range considered appropriate for normalization, (i.e., less than 0.5 or greater that 3.0 percent), these data were only compared with the Puget Sound AETs. An AET represents the chemical concentration above which adverse biological effects have been demonstrated to always occur. The LAET was used as the equivalent of the SQS, and the 2LAET was used to represent the CSL.

The Canadian Marine Sediment Guidelines consist of two thresholds. The TEL represents the concentration below which adverse biological effects are expected to rarely occur, while the PEL defines the level above which adverse effects are expected to frequently occur. The TELs and PELs represent three effect ranges:

- The minimal effect range within which adverse effects rarely occur (i.e., fewer than 25% of samples have adverse effects occur below the TEL)
- The possible effect range within which adverse effects occasionally occur (i.e., the range between the TEL and PEL)
- The probable effect range within which adverse biological effects frequently occur (i.e., more than 50% of samples have adverse effects above the PEL) (CCME 2001).

The freshwater sediment data were compared to three sets of sediment guidelines. Ecology's FP based freshwater sediment guidelines Avocet Consulting 2003; Avocet Consulting and SAIC 2002; RESET 2009, the Canadian Freshwater Sediment Guidelines (CCME 2001, Smith et al. 1996), and the Consensus-based Guidelines (MacDonald et al. 2000) were used; all guidelines consist of a set of two thresholds.

The 2003 Draft Washington FP guidelines obtained from Ecology's EIM database were used in this assessment and include the "Sediment Quality Standard" (FP-SQS) and a "Cleanup Screening Value" (FP-CSL). The FP-SQS was the primary threshold used to evaluate the freshwater sediment data; the remainder of the thresholds/guidelines described here were used to provide additional context and included as part of the WOE discussion. The FP thresholds were developed based on bioassay hit definitions from Washington's marine SQS and the CSL. The FP thresholds include a third guideline based on a statistically significant difference (STAT); the STAT guideline was not used in this prioritization process.

The FP SQS defines a biological effect when the difference between the mortality rate in the test and control is greater than 10%, when the growth test/control ratio is less than 0.8 and when the decrease in Microtox® luminescence test/control ratio is less than 0.85. The FP CSL defines a biological effect when the test results for the same bioassays are greater than 25%, less than 0.7, and less than 0.75, respectively. A more detailed description of the derivation of these thresholds can be found in Avocet Consulting (2003) and Avocet Consulting and SAIC (2002).

Similar to the marine guidelines described above, the Canadian Freshwater Sediment guidelines (CCME 2001) consist of a "Threshold Effect Level" or TEL that represents the concentration below which effects are infrequently observed and a "Probable Effects Level" or PEL, which represents the concentration above which effects are frequently observed.

The Consensus Based Freshwater Sediment Guidelines (MacDonald et al. 2000) consist of a "Threshold Effects Concentration" or TEC, the level below which effects are not expected and a "Probable Effects Concentration" or PEC, the level above which effects are expected. In this context the term "consensus" does not mean agreement among scientists on the best guideline, but rather that a variety of sediment quality guidelines from different sources were combined to generate the thresholds. A more detailed discussion of the derivation of all of these sediment thresholds (except the FP's) can be found in Wenning et al. 2005. A summary of the sediment guidelines used in this assessment are presented in Appendix D-10,

Tissue – Direct Effects to Aquatic Life

To determine the potential for effects to aquatic life via waterborne or dietary exposure to the COCs, relevant tissue residue effect concentrations were identified for comparison with the observed environmental tissue residue data described above. Initially, the Environmental Residue Effects Database (ERED) (http://el.erdc.usace.army.mil/ered/) was identified as the source of effects data for the tissue assessment.

Due to the complexities associated with tissue residue data and concern for using data appropriate for this assessment, 25% of the 100 original papers on which the ERED data were based were reviewed for accuracy. Review of the original literature resulted in an unacceptable error rate (~50%). Numerous errors were identified and included use of the dose concentration to represent the effect concentration, a value not represented by a statistically significant effect, and incorrect values (e.g., paper did not measure concentration in tissue, a lipid normalized value presented as a wet weight value). Due to the high error rate, use of the ERED database would require review of all original data sources, which was beyond the scope of this project. A summary of the results of the ERED QA/QC process can be found in Appendix D-13.

As an alternative, two regional efforts that evaluated tissue residue effects were identified. Both the Lower Duwamish Waterway Group and the Lower Willamette Group recently completed final and draft, Remedial Investigation Baseline Ecological Risk Assessments respectively, which included review and evaluation of tissue residue effects data (Windward 2010, Windward 2009). Both groups identified numerous tissue residue effects concentrations that were subsequently reviewed by toxicologists from various agencies and groups. Since these data had been previously reviewed and vetted by numerous professionals, they were identified as a readily available reliable source of tissue residue effects data for this assessment.

The tissue residue effects concentrations are intended to estimate the direct effect of a COC on an organism via waterborne or dietary exposure. Although considerable effort has been expended over the years to relate tissue metal residues to effects, with the exception of selenium, mercury and tributyl-tin, these efforts have achieved only limited success (Adams et al. 2010).

Due to the disparate physical/chemical characteristics of metals, their environmental presence in multiple forms and states, the fact that some are essential micronutrients and some are controlled by metabolic processes, metals and inorganics as a group continue to be more toxicologically challenging than organics when trying to apply the tissue residue approach. Residue approaches for metals require detailed consideration of metal specific and species specific details and determination of the toxicologically active fraction of the total body/organ tissue residue (Luoma and Rainbow 2005 in McCarty 2010).

The development of the Biotic Ligand Model (BLM) for some metals demonstrates the successful use of a tissue residue based approach for metals that can be used when the target organ and receptors have been identified and the amount of metal necessary to produce toxicity has been established. However, this is not necessarily the case for whole-body tissue residue concentrations for most metals. Aquatic organisms use a variety of storage, detoxification and excretion mechanisms to address metal exposure. As a result, measuring the total metal in an organism provides limited information regarding the biologically active metal concentrations within an organism (Adams et al. 2010; Meador et al. 2008). Due to the complexity of this type of assessment, a meaningful evaluation of metal tissue residues requires a much more detailed evaluation of the available toxicity data than could be conducted here. Therefore, tissue residue data for metals were not evaluated in this hazard prioritization process.

Although there are similar limitations in the use of tissue residue effects concentrations for organic chemicals, the relationships for some compounds (e.g., bioaccumulative and persistent) are much more established (Meador et al. 2008). Thus, the tissue assessment was restricted to bioaccumulative organic compounds that are not readily metabolized and for which data (effects and observed environmental) were available (mercury, PCBs, PBDEs, DDTs, and dioxins). When available, whole-body tissue residue effects data for fish, non-decapod invertebrates and decapods were used in the assessment. All of the available tissue residue effects data were based on wet weight concentrations. Where possible, a qualitative assessment of regionally based lipid normalized effects thresholds are discussed as part of the WOE discussion. A summary of the effects data used in this assessment can be found in Appendix D-11.

Wildlife

Four wildlife receptors were selected for evaluation by this assessment; harbor seal, osprey, river otter and great blue heron (A detailed description of species selection is described in the subsequent section on wildlife receptor and COC selection). The wildlife effects thresholds are based on the daily dose (mg chemical/kg-body weight/day) of a COC known to cause adverse effects to test species of birds or mammals. There are no state or federal standards to evaluate contaminants in wildlife; wildlife effects doses were obtained from published dose-response studies. These studies typically expose test animals to a COC through ingestion of food or water containing known contaminant concentrations and observe any effects on growth, reproduction, development or survival.

Effect doses for this assessment were obtained from multiple publications, some being compilations of effect doses from EPA efforts and included the Great Lakes Water Quality Initiative Criteria Documents (EPA 1995), the Ecological Soil Screening Levels (EPA 2007), Toxicological Benchmarks for Wildlife (Sample et al. 1996), the Draft Lower Willamette River Remedial Investigation Baseline Ecological Risk Assessment (Windward 2009) and the Lower Duwamish Waterway Remedial Investigation Baseline Ecological Risk Assessment (Windward 2010). Individual publications supplemented these sources as available. Effects doses were applied as they were presented in the source documents unless errors of interpretation were found (e.g., the dose causing an effect is incorrect, typographical errors, unit conversion errors). Safety or uncertainty factors were not used to estimate potentially hazardous levels in the assessment.

Due to data availability issues, it is common to use data from published dose-response studies conducted on test species (birds or mammals) other than those of interest in an assessment. All daily doses associated with either bird or mammal species were grouped for use with the appropriate bird or mammal receptor identified for this study. For the effects daily doses calculated directly from published data, safety factors were not applied to adjust for interspecies, lowest effect to no effect value, or any other uncertainty. This decision was made because there is no knowledge of which direction, and to what magnitude uncertainty would be biased. For example, the sensitivity of one species to chemical exposure may be greater or lesser than another. Until each species is tested, it is impossible to predict which species will be more sensitive and the degree of difference in sensitivity between species. In addition, safety factors are not standardized and therefore, when applied, are not consistent in magnitude.

A number of field studies have documented bioaccumulation of PCBs and dioxins/furans in Puget Sound harbor seals and orcas, conducted biopsies and examined immune suppression in these organisms (Ross et al. 1995, de Swart et al. 1996, Ross et al. 1996, Ross et al. 2004, Levin et al. 2005, Cullon et al. 2009). However, to date, a dose-response study has not been conducted to provide the necessary information to develop a toxicity-based daily ingestion dose for marine mammals. Because these results are not dose-response studies, they could not be used in the quantitative part of this assessment. They are instead discussed qualitatively as part of the WOE discussion. Published research on wildlife exposure and effects to COCs conducted in or near Puget Sound are summarized in the Results Section. A summary of the quantitative effects data used in the wildlife assessment can be found in Appendix D-12.

Human Health

The effect threshold used for the human health assessment was based on the National Toxics Rule (NTR) 40CFR§131.36. A number of national and regional fish consumption rates (Table 5) were calculated using the same methods to provide additional perspective and because the NTR specified rate is considered under protective for certain populations and ethnic groups such as Asians, Pacific Islanders and Native Americans. The tissue thresholds were derived by back calculating intake rates from existing water quality criteria using the applicable bioconcentration factors (BCFs) presented in the original water quality criteria development documents. COCs not part of the NTR were not assessed for human health hazards, e.g. lead, triclopyr, and the sum of cPAHs although individual PAHs were prioritized.

Table 5. List of consumption limits assessed in this document.

Guidance/Reference	Rate
NTR Standard Rate(64 FR 61184)	6.5 gm/day
EPA Recreational Rate (EPA 2000)	17.5 gm/day
EPA Subsistence Rate (EPA 2000)	142.4 gm/day
Tulalip Tribal/King County Asian Pacific Islander Rate (Toy 1996; Sechena 1999)	242.5 gm/day
Suquamish Tribal Rate (Suquamish Tribe 2000)	769 gm/day

There are no applicable human health standards for sediment. Both the Washington Sediment Management Standards and the Model Toxics Control Act (MTCA) use site specific assessments of exposure to assess human health risk; however, this process was not practical to conduct on a Puget Sound regional scale. Potential effects associated with exposure to sediment through recreational, shell fishing, or beach use would require parameterizing a human health risk assessment which was beyond the scope of this project. Similarly, water was not evaluated because recreational or consumptive water uses would require developing regional estimates of lifetime human water exposures which was also beyond the scope of this project.

Petroleum

Total petroleum hydrocarbons (TPH) is a term used to describe a large family of chemical compounds that originate from crude oils, coal tars, oil shales, and similar materials. The specific composition of petroleum products varies depending upon (1) the source of the crude oil and (2) the refining practices used to produce the product. TPH and "TPH gasoline" represent a mixture of petroleum compounds and serve as coarse estimates of the presence of the individual constituents that may cause toxicity to aquatic organisms. The ECOTOX database used for the water assessment only includes data for individual chemical components and does not include toxicity data for complex mixture compounds such as TPH. As a result, it was necessary to use a slightly different approach to assess TPH mixtures.

Toxicological evaluation of petroleum mixtures have historically been conducted in the aftermath of oil spills and similar events. Sources consulted for toxicity information on TPH and petroleum products were API, 1994; Barron, et al. 1999a; Barron, et al. 1999b; Tsvetnenko, 1998; and Woodward et al. 1983. The majority of the available toxicity data were for crude oils; however, these studies were deemed unsuitable since none of the available observed environmental data were for crude oils.

Based on availability of both observed and effects data, four classifications of petroleum products from the available environmental data were evaluated: heavy fuel/bunker oil, diesel fuel, gasoline, lube oil. Only toxicity results assessing the water soluble fraction (without free product) were used to assess these data. A more complete evaluation of petroleum would require analysis of parent and alkyl PAHs as well as issues such as phototoxicity which were beyond the scope of this assessment. In addition to the specific assessment for petroleum described here, a suite of individual and high/low molecular weight PAHs were evaluated in both the water and sediment assessments previously described above.

Selection of Wildlife Receptors and COCs

The following section describes the process used to select wildlife species and COCs evaluated in the wildlife assessment. When conducting a wildlife effects assessment, it is necessary to select a finite number of species or "receptors". In part, this is due to the impracticality of trying to assess effects to all wildlife species that reside in the Puget Sound region.

Receptors are generally assumed to be conservative representatives of other species with similar diets, trophic status and biology. A bird and a mammal species were evaluated in freshwater habitats and marine habitats. Two bird (osprey and great blue heron) and two mammal species (harbor seal and river otter) were selected based on their position as top level predators, a diet consisting primarily of fish, and their use of different feeding strategies, all of which make them at greatest risk of exposure. In addition, a substantial amount of ecological information has been published about these species allowing reasonable exposure modeling assumptions.

The four selected receptors are not intended to represent all wildlife species that may be exposed to COCs in the Puget Sound area. However, these receptors do represent other piscivorous species which tend to bioaccumulate chemicals to a greater degree than lower trophic level species.

The great blue heron was selected as the freshwater bird species because it is a common, piscivorous bird in Puget Sound freshwater and marine habitats. Some populations exclusively reside and feed in freshwater habitats as evidenced by large, established heron rookery sites on lakes and rivers. This large wading bird consumes fish of a variety of sizes and species including large predatory fish such as largemouth bass and trout. Also, some are year-round residents of this region. The river otter was selected as the freshwater mammal species because it is a common mammal in freshwater habitats and consumes primarily fish (EPA 1993). There is also evidence that river otter in Washington State accumulate mercury, PCBs, and dioxins/furans (Grove and Henny 2008). Although river otter may migrate between marine and freshwater habitats, some are exclusive to freshwater habitats. For the purposes of this assessment, it was assumed that both the great blue heron and river otter consume 100% of their prey from freshwater habitats and reside year-round in the Puget Sound region.

Osprey and harbor seal were selected as marine habitat receptors. The osprey was selected over the bald eagle, another common top predator of fish, for a number of reasons. Osprey primarily consume fish as compared to the bald eagle with consumes a large portion of birds and mammals; the osprey is much smaller than the bald eagle, making their relative ingestion rate and exposure higher. Osprey consistently hunt by diving or grasping prey from the water, whereas the bald eagle may scavenge for food, exclusively hunt salmon during salmon runs, or hunt for birds. Osprey migrate south during the winter, but breed in western Washington.

The harbor seal was selected to represent an aquatic marine mammal because it is a top predator feeding exclusively on aquatic prey, primarily fish, and has been reported to have elevated tissue concentrations of PCBs. The harbor seal was selected over the orca because the harbor seal is a smaller mammal with relatively higher ingestion rate and exposure, and because much more ecological information is available for the harbor seal. For the purposes of this assessment, it was assumed that the diet of both the osprey and harbor seal was 100% fish and that they reside only in Puget Sound.

The COCs evaluated by the wildlife assessment were limited to bioaccumulative compounds for which sufficient effects and environmental data were available and included mercury, PCBs, DDTs, and dioxins/furans. Many of the COCs are neither detected in fish (wildlife prey) nor bioaccumulative. PAHs are metabolized by fish, and therefore are not bioconcentrated (or analytically detected very frequently). Triclopyr is broken down quickly in the environment, and is not bioaccumulative. Bis(2-ethylhexyl)phthalate (DEHP) was infrequently detected (<10% FOD) in only two species of fish collected from nearshore habitats. Nonylphenol is also not considered to be bioaccumulative. Although nonylphenol is a potential endocrine disruptor, describing the potential endocrine disruption of nonylphenol to wildlife populations has never been done before.

Metals have naturally high FOD in the environment; however, the toxicity of some metals is complicated by their need as essential trace minerals. At the same time, many metals are moderately bioaccumulative and toxic to wildlife. Lead poisoning of birds exposed to lead shot and fishing weights has been a long standing concern. Although lead shot was banned for use in waterfowl hunting in 1991, it continues to be used in upland game hunting, posing risk to non-waterfowl bird species (USGS 2009). Because this assessment is focused on the aquatic environment, assessment of upland exposure of birds to lead shot was outside the scope of this assessment and not evaluated. Based on the chemical characteristics of these COCs, it was assumed that the potential for chronic wildlife exposure was very low and effort was focused on the more persistent and bioaccumulative COCs.

Exposure Model for Wildlife Receptors

Daily doses of COCs were estimated using a simple exposure model that included a body weight normalized sum of daily food intake, drinking water intake and incidental sediment ingestion during foraging and other behaviors. This is represented by the following algorithm:

$$\text{TDD} = \frac{[C_w * IR_w] + [C_s * IR_s] + [C_f * IR_f]}{BW}$$

Where:

TDD = Total daily dose (mg/kg-BW/d)

C_w = 95% UCL of mean chemical concentration in water (mg/L)

C_s = 95% UCL of mean chemical concentration in sediment (mg/kg)

C_f = 95% UCL of mean chemical concentration in food (mg/kg)

IR_w = Ingestion rate of water (L/day)

IR_s = Incidental ingestion rate of sediment (kg/day)

IR_f = Ingestion rate of food (kg/day)

BW = Body weight (kg)

The 95% upper confidence limit of the mean (95% UCL) concentration of COCs in water, sediment and food was used to estimate the amount of each COC ingested by wildlife. The datasets from which the 95% UCL statistic was calculated were the same as those used in the water, sediment, and tissue assessments with the exception of the fish tissue dataset. The 95% UCL fish tissue concentrations are summarized in Appendix D-8. The diet of each receptor was conservatively assumed to be 100% fish. Although a proportion of the receptor's actual diet may include invertebrates, the 95% UCL fish tissue concentration is higher than the invertebrate tissue concentration. Therefore, assuming a diet of 100% fish results in a conservative approach, and meets the screening goal of this wildlife assessment. Data for all fish species, with the exception of six gill sharks, were assumed to be prey. A number of sources were reviewed to characterize the harbor seal (Cullon et al. 2005, EPA 1993), osprey (EPA 1993), great blue heron (EPA 1993, Butler 1992, Alexander 1977), and river otter (EPA 1993) diets.

The wildlife receptors used in this assessment prey on a wide variety of species; based on published information on their prey consumption, none show consistent preference for some prey species over others. Because prey size data were unavailable for observed concentrations, size was not a criterion used to screen fish tissue data for inclusion in the assessment.

The daily food ingestion rates were estimated using the allometric equations of Nagy (1987) which relate food ingestion rate to body weight. These equations are:

Birds: $FI = 0.0582 * BW^{0.651}$
Mammals: $FI = 0.0687 * BW^{0.822}$
Where:
FI = food ingestion rate (kg/day dry weight)
BW = body weight (kg)

Calder and Braun (1983) also use this type of relationship to estimate water ingestion rates.

These allometric equations were used to estimate drinking water intake for receptors and are presented below.

Birds: $WI = 0.0598 * BW^{0.67}$
Mammals: $WI = 0.099 * BW^{0.90}$
Where:
WI = water intake (L)
BW = body weight (Kg)

Sediment ingestion rates have not been empirically measured in wildlife studies, but have been estimated using acid-insoluble ash measurement in scat or digestive tracts of animals. Beyer et al. (1994) used ash measurements in scat to estimate the relative proportion of inorganic solids (i.e. sediments and soils) in the diet of multiple wildlife species. None of the four wildlife receptors were subjects of the Beyer et al. study; however, these data are useful to develop rough estimates of sediment ingestion rates for species that share similar feeding strategies. Empirical measurement of sediment ingestion rate is challenging and difficult to model due to ecological variability between and within species. Thus, the sediment ingestion rates were established using best professional judgment and relied heavily on the Beyer et al. (1994) which published estimates varying from <2% to 9% of the daily food ingestion rate for mammals[1] and <2% to 30% of total food ingestion rate for birds[2].

Since total exposure is sensitive to sediment ingestion, separate daily doses for the species evaluated in this assessment were estimated assuming a low and high sediment ingestion rate based on their similarities in feeding strategy and foraging habitat to species from Beyer et al. (1994). Best professional judgment was used to select a low and a high sediment ingestion rate intended to bound the range of realistic potential sediment ingestion rates for each receptor (Table 6).

Where possible, the body weight assumed for each receptor was based on local information summarized in Table 6. The average body weight for the smaller sex, if applicable, was used in the model, because food intake for smaller-bodied animals is proportionately greater than for the larger-bodied cohorts resulting in a larger daily dose and a more conservative estimate. Body weights and ingestion rates for adult life stages were applied to represent the majority of the animal's reproductive lifetime. However, effects dose studies included dosing of immature

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[1] Only four species of mammals were included in the study and only one, the raccoon, forages in aquatic habitat.

[2] Most species of birds in the study were either shorebirds or herbivorous birds. Sediment ingestion rates for dabbling and diving ducks ranged from <2 to 3% of food ingestion.

individuals. Therefore, the effects doses reflect the most sensitive lifestage and should be protective of younger lifestages.

Table 6. Wildlife body weights and ingestion rate assumptions.

Receptor	Body weight (kg)	Source	Food IR (kg/day dw)	Sediment IR (% of Food IR)	Water IR (L/day)
Osprey	1.45	EPA 1993	0.075	1 (4)	0.078
Harbor Seal	77.0	Assuncao et al. 2007	0.985	2 (4)	1.098
Great Blue Heron	2.1	Simpson 1984 as cited in Butler 1992	0.094	2 (4)	0.098
River Otter	7.9	EPA 1993	0.376	4.5 (9.0)	0.636

IR = ingestion rate. Two different sediment ingestion rates were applied for each receptor while holding all other variables constant because this variable is the greatest source of uncertainty in the daily dose model. The higher rate is in parentheses.
Food ingestion rates are from Nagy (1987); sediment ingestion rates are from Beyer et al. (1994); water ingestion rates are from Calder and Braun (1983).

Hazard Assessment

As previously discussed, the intent of this assessment was to provide a general, high level overview of the potential for the COCs to cause deleterious effects in the matrices evaluated. To conduct the quantitative portion of the hazard evaluation, the observed environmental data were compared to the respective effects concentrations. It should be noted that these comparisons are based on single chemical exposures in a single matrix (water, sediment and tissue) and do not account for any effects associated with exposure to chemical mixtures or other physical stressors or conditions (i.e., temperature, dissolved oxygen, hardness etc.) that may be present and influence bioavailability. Sufficient data for both effects and observed environmental concentrations were not available to evaluate the priority for every COC for each type of evaluation.

Water – Direct Effects to Aquatic Life

To assess potential effects to aquatic life through direct exposure to surface waters, observed surface water concentrations for COCs were presented as box plots of percentiles (5th, 10th, 25th, 50th, 75th, 90th and 95th %iles) plotted adjacent to box plots of percentiles representing the available effects concentrations in a series of figures. If water quality criteria (WQC) were available, these values were also presented. In some cases (e.g., DDT and mercury), the WQCs are well below the available effect concentrations.

Some WQC, particularly those for bioaccumulative chemicals, are derived based on a “final residue value” and the potential to bioaccumulate which typically results in a value that is lower than effects concentrations based on direct exposure. In some cases (e.g., nonylphenol) chronic effects data are limited and the chronic WQC is derived using an acute to chronic ratio (ACR). Use of the ACR can also result in a value that may be below effects concentrations presented in ECOTOX. For a more detailed description of how WQC were derived for these COCs see http://water.epa.gov/scitech/swguidance/waterquality/standards/criteria/aqlife/index.cfm. It is also important to note that water quality criteria are not derived to protect aquatic organisms through the bioaccumulation pathway. The tissue residue assessment described below was intended to help address this pathway for a select group of COCs.

The total number of analytical measurements and number of observed detected concentrations are also presented on each figure. Most freshwater and marine (near- and offshore) data were presented on the separate figures; results for some COCs for which data were limited are combined into a single figure. If a COC was not detected (or measured), or effects data were not available, a figure was not prepared. All data were plotted on a log scale.

Sediment – Direct Effects to Benthic Organisms

To assess the potential for effects to benthic organisms, sediment COC concentrations were presented as box plots of percentiles plotted against the sediment guidelines (represented as horizontal lines on each figure). The standard or guideline use as the primarily comparison is presented as a solid red line, while the remainder of the guidelines provided for additional context are presented as dotted blue lines. When appropriate, based on the guidelines used for comparison, both dry weight and OC normalized concentrations were presented. In general, separate graphics were generated for marine and freshwater sediment data; results for some COCs for which there were limited data are combined into a single figure. If a COC was not detected (or measured), or effects data were not available, a figure was not prepared. All data were plotted on a log scale.

Tissue Residues – Direct Effects to Aquatic Life

The tissue assessment was similar to that described above for water. Observed tissue residue concentrations were presented as box plots of percentiles along with the available tissue residue effects concentrations; all data were plotted on a log scale. Graphics were generated for whole-body tissue types (fish, non-decapod invertebrates, and decapods) where both effects and observed environmental data were available. Marine and freshwater tissue data were graphed separately. If a COC was not detected, or effects data were not available, a figure was not prepared. As discussed above, the tissue residue assessment was limited to four PBTs (DDTs, dioxins, PCBs and mercury). Although PBDEs were not assessed quantitatively, the current effects literature and regional studies are reviewed in the WOE discussion.

Wildlife

The wildlife assessment compared literature-based daily effects doses for birds and mammals to the estimated daily doses of COCs for the four receptors (great blue heron, osprey, river otter and harbor seal). The literature-based daily effects doses were rank ordered and plotted against the estimated daily doses; data were plotted separately for birds and mammals. Two estimated daily doses were calculated for each receptor based on a low and a high estimate of sediment ingestion rates. Both estimates are shown on the figures.

Observed PCBs, DDTs and dioxins/furans data were assessed as sums. PCBs were assessed as Aroclor® sums for tissue and sediment and as PCB congener sums in water because too few Aroclor® detections occurred in the observed water data. DDT sums included DDT, DDE, and

DDD isomers. Dioxins and furans were converted to TEQs and summed to a total TEQ. The effects doses included individual Aroclors®, or DDTs, DDEs, and DDDs and their mixtures. Dioxin and furan effects doses were treated the same as observed data by converting to TEQs and summing to a total TEQ.

Human Health

The human health assessment utilized the National Toxics Rule (NTR) (40CFR§131.36) to establish default assumptions of body weight, toxicity and daily fish/shellfish consumption rates. The NTR uses a national average fish tissue consumption rate of 6.5 gm/day; this consumption rate was used to calculate a screening threshold and used as the primary basis of this assessment. However, this rate is suspected to be under-protective of various other users and ethnic groups (EPA 2007).

To account for different groups who may consume fish/shellfish at higher rates, five consumption rates were also used to derive alternative levels for informational purposes. Two consumption rates (242 gm/day King County API (Sechena 1999) and 243 gm/day Tulalip Tribe (Toy 1996) were almost identical so they were averaged to create one consumption scenario of 242.5 gm/day. In addition to the NTR standard rate and the average of the King County API/Tulalip rate, the EPA recommended recreational and subsistence consumption rates (EAP 2000) and the Suquamish ingestion rate (Suquamish Tribe 2000) were also used. The levels calculated based on these additional consumption rates are presented to provide additional context and are discussed as an additional WOE.

This methodology was used to evaluate bivalve, fish (whole-body and filet) and other invertebrate tissue data for human health consumption risks. The NTR is the only regulatory standard in Washington State applicable for human health risks related to consumption of surface water. However the surface water standards in the NTR are predominantly influenced by bioaccumulation by fresh water fish tissue and not the consumption of water alone. Consumption and/or dermal exposure to water alone would require development of a human health risk assessment for the entire Puget Sound region. A water risk assessment would need to include an estimate of freshwater exposure point concentrations or probabilistic estimates of exposure; such an evaluation was deemed to be beyond the scope of this assessment.

Tissue criteria were backcalculated from the NTR based water quality criteria as shown below in Table 7. The bioconcentration factors (BCFs) shown are from the original criteria documentation. These BCFs and the applicable fresh and marine water quality criteria were used along with the original 6.5 gm/day and modified consumption rates to derive tissue concentrations deemed protective at a range of tissue consumption rates as shown in Table 5.

Table 7. Summary of NTR water quality criteria and bioconcentration factors (BCF) used in the human health assessment.

COC	BCF	NTR Freshwater Human Health Criteria - Water and Organisms (µg/L)	NTR Marine Water Human Health Criteria - Organisms Only (µg/L)
Arsenic	44	0.018	0.14
Mercury (estuarine)	3765	n/a	0.15
Mercury (freshwater)	5500	0.14	n/a
Bis(2-Ethylhexyl)Phthalate	130	1.8	5.9
2,3,7,8-TCDD (Dioxin)	5000	0.000000013	1.4E-08
Total Polychlorinated Biphenyls (PCBs)	31,200	0.00017	0.00017
4,4'-DDT	53,600	0.00059	0.00059
4,4'-DDE	53,600	0.00059	0.00059
4,4'-DDD	53,600	0.00083	0.00084
Anthracene	30	9600	110,000
Benzo(a)Anthracene	30	0.0028	0.031
Benzo(a)Pyrene	30	0.0028	0.031
Benzo(b)Fluoranthene	30	0.0028	0.031
Benzo(k)Fluoranthene	30	0.0028	0.031
Dibenzo(a,h)Anthracene	30	0.0028	0.031
Fluoranthene	1150	300	370
Fluorene	30	1300	14,000
Indeno(1,2,3-cd)Pyrene	30	0.0028	0.031
Pyrene	30	960	11,000

To account for potential rounding errors and changes in estimates of cancer toxicity since the NTR was adopted; cancer slope factors or reference doses were first derived from the NTR calculations for both freshwater and marine waters. These slope factors, or dose for mercury, were then used in the following formulas to derive tissue thresholds.

Following is the calculation of the human health criterion for freshwater organisms along with 2L of drinking water per day consumption:

$$\mathrm{Fresh-HH} = \frac{\mathrm{RF\ x\ BW\ x\ }(1{,}000\,\frac{\mu\mathrm{g}}{\mathrm{mg}})}{\mathrm{q1} * \mathrm{x}\ [\mathrm{WC} + (\mathrm{FC\ x\ BCF})]}$$

Where:

Fresh-HH = Freshwater criterion in ug/L
RF = Risk Factor = 1 x 10 (-6)
BW = Body Weight = 70 kg
q1* = Cancer slope factor/toxicity (Hg only), chemical specific
WC = Water Consumption = 2 L/day
FC = Fish and Shellfish Consumption varied according to Table 5
BCF = Bioconcentration Factor

Following is the calculation of the human health criterion for marine water organism only consumption:

$$\mathrm{Marine-HH} = \frac{\mathrm{RF\ x\ BW\ x\ (1{,}000\,\frac{\mu g}{mg})}}{\mathrm{q1*\ x\ FC\ x\ BCF}}$$

Where:

Marine-HH = Marine criterion in ug/L
RF = Risk Factor = 1 x 10 (-6)
BW = Body Weight = 70 kg
q1* = Cancer slope factor/toxicity (Hg only), chemical specific
FC = Fish and Shellfish Consumption varied according to Table 5
BCF = Bioconcentration Factor

For COCs not included in the NTR, a hazard evaluation was not conducted to evaluate their hazard level for the same reasons that water and sediment hazard evaluation were not conducted.

Petroleum

As previously discussed, due to issues associated with the non-specific nature of petroleum product measurements in surface waters, this COC was evaluated using a slightly different process than the remainder of the COCs in water. Petroleum in freshwater was evaluated using data associated with the release of products to water (spills).

Most available toxicity data were for crude oils, which were not considered applicable for this assessment because no crude oil environmental data were available. Similarly, a large amount of observed oil and grease data were available from the EIM and King County LIMS databases. However, these data do not describe a specific petroleum product; they encompass waxes, greases and other fatty acid substances from both animal, vegetable and petroleum origins. Because these environmental data are non-specific, and potentially toxic components may vary within the same concentration measured by this method; these data were considered unusable for this assessment. Thus, toxicity data and environmental data were both only available for four petroleum products: heavy fuel/bunker oil, diesel fuel, gasoline, and lube oil. Toxicity data for these fours products were almost entirely based on lethal concentrations to 50% of the exposed population (LC50s).

Environmental data for these four products were plotted against the available freshwater toxicity data. Only two petroleum product data results were available for marine waters and these were insufficient to estimate hazard priority levels.

Determination of Priority

Due to the broad screening nature of this assessment, in addition to uncertainties associated with the available data (see Uncertainty Section), a conservative approach was used to determine the potential hazard posed by each COC. While an effort was made to use a consistent approach to classify the priority for each component of the assessment, due to the nature and availability of the data used and variability of methods used for each component, there are differences between some of the approaches.

The reader is urged to use caution when comparing priorities across matrices and type of assessment. The potential for ecological effects for COCs classified as Priority 1 is much higher than that for Priority 2. However, the specific type of ecological effect is not defined for Priority 1 COCs and can vary from mortality to more subtle impacts like physiological changes. The following sections describe the process by which the potential for effects was determined for each element of the assessment. Table 8 summarizes the thresholds used to define priority levels and sufficiency of data.

Water – Direct Effects to Aquatic Life

A conservative approach was used to determine the potential for effects to aquatic life from direct exposure to surface waters. If the 90th %ile of the observed environmental concentrations of a COC was above the 10th %ile concentration of the effect concentrations, the COC was classified as “Priority 1”. If the 90th %ile of the observed environmental concentrations was less than the 10th %ile of the effect concentrations, the COC was classified as “Priority 2”. If there were insufficient data (<15 effects or <50 observed environmental values) to assess a particular COC, it was classified as “Unknown” or “U”. A classification of Priority 2 or “U” is not intended to suggest that this COC is not important. It is assumed that all of these COCs are priorities at some level.

In freshwaters, hardness can have significant influence on the toxicity of metals. Due to the large volume of data used for this assessment it was not practical to evaluate the hardness concentrations associated with each observed or effect metal concentration. The reader is cautioned to take this factor into account when evaluating the findings of the freshwater metals assessment. The WQC were calculated for metals with hardness-based standards using a hardness value of 25 mg/L-CaO_3 which is approximately the average freshwater hardness in Western Washington.

Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for the direct water exposure were limited to copper and PAHs.

Sediment – Direct Effects to Benthic Organisms

To determine the potential for effects to benthic organisms from direct sediment exposure, the 90th %ile concentration for each COC was compared to the marine SQS or freshwater FP-SQS, in addition to the other sediment guidelines described above.

A process similar to that described above for water was used to assess COC priorities for sediment. If the 90th %ile of the observed sediment concentrations was above the marine SQS or the freshwater FP-SQS the COC was classified as Priority 1; if the 90th %ile concentration was less than the sediment standard/guideline is was classified as Priority 2. COCs for which there were insufficient data (effects or observed concentrations data) to assess were classified as “Unknown” or “U”. Sediment COCs were classified as “U” if a FP-SQS or SQS value was not available or if the number of observed environmental concentrations was low (<30). As previously discussed, classification of a COC as a Priority 2 or “U” does not indicate the COC is not potentially important.

It is assumed that all of these COCs are priorities at some level. Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for the direct sediment exposure were limited to PAHs and PCBs.

The level of uncertainty associated with data availability for each COC was also summarized. Data uncertainty was based on the total number of measurements and the availability of sediment thresholds for each COC. As indicated for water above, this assessment assumes that the available environmental data are representative of the overall region; however, the lower the number of measurements for each COC, the greater the uncertainty that these data are representative.

Tissue –Direct Effects to Aquatic Life

The approach used to determine priorities in the tissue assessment was the same as that described above for water. The data for this element of the assessment were very limited; when interpreting the tissue assessment results, the reader is also encouraged to take into account the amount and type of both tissue residue effects and observed data available. A COC was classified as “Priority 2” if the 90th %ile observed concentration was below the 10th %ile effects concentration. A “U” or “Unknown” priority indicates there was insufficient effects data (<5 effects or >20 observed values) to allow assessment.

Where available, data from regional studies and other readily available thresholds were used as an additional WOE to qualitatively assess each COC. In general, the available regional data for tissue residue related impacts was limited to PAHs and PCBs.

Wildlife

A COC was classified as “Priority 1” when the estimated daily dose was greater than or within 0.1 times the lowest effect dose. Because there are far fewer published effects doses for wildlife than aquatic life, there is greater uncertainty in estimating the lowest effect threshold. For this reason, a COC was classified as “Priority 2” if its estimated daily dose was less than an order of magnitude (i.e., a factor of 10) of the lowest effect dose. A “U” or “Unknown” priority indicates there was insufficient effects data (<5 effects values) to allow assessment. Only one published effects dose was available for PBDEs in birds (Fernie et al. 2011), and none for mammals. Thus, a discussion of the estimated total PBDE daily doses for the avian receptors compared to this published effects dose is included in the WOE discussion in lieu of the full quantitative evaluation.

Human Health

If the 90th %ile of the observed tissue concentrations exceeded the NTR screening criteria the COC was classified at Priority 1; if the NTR value was below the 90th %ile concentration, the COC was classified as Priority 2. Some COCs had many environmental measurements for one tissue type, but few of other tissue types. In these instances, best professional judgment was used to determine the adequacy of all the tissue data within the fresh, near, or offshore environment to develop a hazard ranking for the COC in tissue on the whole. As discussed above, classification as a Priority 2 does not indicate that a COC is not a priority. It is assumed that all COCs are priorities at some level. Results of the comparison to other consumption rate data were used as an additional WOE and discussed qualitatively below. Most COCs were found at comparable concentrations across the bivalve, fish, and other invertebrate tissue groups suggesting that prioritization rankings are relatively robust regardless of tissue type evaluated.

Table 8. Thresholds used to define priorities and sufficiency of data.

Line of Evidence	Threshold for Priority	Threshold for Sufficient Data
Surface Water	90th%ile Observed Conc. > *10th %ile Effects Conc.* *or* *Acute WQC or Chronic WQC*	n ≥ 15 for Effects Data n ≥ 50 for Observed Data
Sediment	90th%ile Observed Conc. > *SQS*	n ≥ 100 for Observed Data
Tissue Residue Effects	90th%ile Observed Conc. > *10% of 10th%ile Effects Conc.*	n ≥ 5 for Effects Data n ≥ 20 for Observed Data
Wildlife	Daily Dose >*10% of Lowest Effects Dose*	n ≥ 5 for Effects Data
Human Health	90th%ile Observed Conc. >*NTR Criterion*	Best Professional Judgment

Results and Discussion

The following sections provide an overview of the availability of both the environmental occurrence data and effects data in addition to the outcome of the individual effects assessments and a summary of the overall outcome of the assessment.

Environmental and Effects Data Availability

Water

Surface water data were used to evaluate direct effects to aquatic life, and in the wildlife assessment as appropriate to estimate exposure through water ingestion. The number of measurements for COCs in freshwater ranged from over 5000 for copper to less than 10 for dioxins. With the exception of PCB and PBDE congeners, dioxins/furans and nonylphenol, there were more than 1200 measurements for each COC in freshwater.

Relative to freshwater data, the number of measurements in marine (near- and offshore) surface waters was significantly less; nearshore data were most limited. With the exception of copper, there were less than 50 measurements (often less than 15) for each COC in nearshore waters. In general, there were less than 100 measurements for each COC in offshore waters. Marine data were not available for petroleum, triclopyr and dioxins. The water data are summarized in Appendix D-8.

The relatively low number of measurements for many of the COCs in marine waters suggests that the spatial coverage for these data is limited and these data may not be representative of the region.

Effects data were considered “sufficient” when a range of values (at least 15 effect concentrations) representing a variety of endpoint types other than mortality were available. It is generally uncommon for ambient concentrations of COCs to be present at levels that would cause mortality; organisms are typically exposed to concentrations likely to cause more subtle effects (e.g., effects to growth and reproduction). If the effects data were limited to mortality based endpoints, it is possible that priority rank may have been underestimated because the potential for effects other than mortality could not be evaluated. For example, most of the available effects data for PCBs in marine water were based on mortality; and this COC was classified as Priority 2. However, exposure to low levels of PCBs can result in food web biomagnification and cause effects to the immune systems of higher trophic level organisms. As such, it should be noted that direct water exposure is not the most sensitive approach to assess this COC.

In freshwater, there were sufficient effects data for most of the COCs with the exception of some individual PAHs, dioxins and PBDEs. Effects data for these COCs were limited, particularly for endpoints other than mortality.

Effects data for aquatic life in marine waters were sufficient for some COCs, but were limited (especially non-mortality effect data) for a number of COCs. In marine waters effects data for arsenic, cadmium, PCBs, PBDEs and a number of individual PAHs were limited.

Sediment

Sediment data were used to evaluate direct effects to benthic organisms and were also incorporated into the wildlife assessment where appropriate to estimate exposure from incidental sediment ingestion. Sediment data were available for all COCs with the exception of triclopyr and petroleum. All sediment data are summarized in Appendix D-8. With the exception of PCB and PBDE congeners, nonylphenol and a few individual PAHs, there were more than 300 measurements for each COC in both freshwater and marine (near- and offshore) sediments.
Of the 3 sets of freshwater sediment guidelines used in this assessment, threshold values were available for the majority of sediment COCs. Only one set of freshwater guidelines was available for a number of individual PAHs, LPAHs, HPAHs and dioxin/furans; two sets of guidelines were available for DDT and some individual PAHs and only a single threshold was available for nonylphenol.

Of the 3 sets of marine sediment guidelines used in this assessment, threshold values were available for the majority of sediment COCs. However, only 2 sets of guidelines were available for some individual PAHs and one set for dioxins/furans; only one sediment guideline was available for nonylphenol.

Tissue

Tissue data were used to evaluate direct effects to aquatic life (limited to PBTs) and wildlife (select organic chemicals only) and to assess human health (select organic chemicals, mercury and arsenic only). The different assessments required the various combinations of tissue types. A summary of the tissue data used for each assessment type can be found in Appendix D-8. The number of measurements for some COCs and tissue types was very low and limited the degree to which these chemicals could be assessed.

Effects data to evaluate the direct impact of tissue residues on aquatic life were very limited for most COCs, particularly for marine fish; effects data for these tissue types were not available for mercury, dioxins/furans and DDTs. The number of tissue residue effects concentrations for some tissue types and COCs was often less than 5. The lack of tissue residue effects data poses limitations on this element of the assessment. As a result, the reader is cautioned to evaluate the available data when interpreting these results.

Wildlife

Generally, the available environmental data were sufficient to meet the needs of the wildlife assessment. Far greater uncertainty exists with the effects data due to the limited number of dose-response data for wildlife species from published studies. The low number of effect dose-response data available for birds and mammals poses limitations on the assessment, particularly for dioxins/furans. The lack of dose-response data for PBDEs precludes an estimation of potential adverse effects to wildlife from exposure to this COC.

Human Health

A number of known or potentially bioaccumulative COCs were not evaluated in the human health portion of this assessment because they are not part of the human health standards in the NTR. These chemicals include:

1. Acenaphthene
2. Acenaphthylene
3. Inorganic arsenic
4. Benzo(g,h,i)perylene
5. Cadmium
6. Chrysene
7. Lead
8. Nonylphenol
9. Phenanthrene
10. Polybrominated diphenyl ethers (PBDEs)

Generally, environmental data were sufficient to meet the needs of this assessment. All chemicals without NTR criteria were classified as “unknown” due to the lack of standards against which to compare.

Effects Prioritization

The results of the quantitative assessments for water, sediment, tissue media and effects on wildlife and human health are presented in Appendix D-2. The remainder of this section summarizes the results for each COC, including any additional evidence provided by studies of regional significance.

Metals

With the exception of mercury, the assessment of metals was limited to water, sediment and human health. The availability of NTR criteria for metals limited the assessment to mercury and arsenic. Due to data availability and scope limitations, metals were not evaluated in wildlife and tissue residue.

Arsenic

Water

Freshwater

Arsenic was classified as ”U” because there were limited effects data for dissolved arsenic. The 90th %ile observed freshwater arsenic concentration is below both the acute and chronic WQC.

Marine Water

Arsenic in nearshore and offshore waters was classified as “U” because no effect data were available for arsenic in marine waters.

Sediment

Freshwater Sediment

Arsenic was classified as Priority 1 based on comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment arsenic concentrations is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Arsenic was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment copper concentrations to the SMS SQS. The 90th %ile observed arsenic concentrations in both near and offshore sediment are below additional guidelines evaluated except for the TEL.

Tissue

Arsenic was not assessed in tissues.

Wildlife

Arsenic was not assessed for wildlife.

Human Health

The NTR is based on inorganic arsenic however; most of the environmental data were for total arsenic which includes less toxic organic forms like arsenobetaine. Both inorganic and total arsenic exceeded the NTR by several orders of magnitude; however, the inorganic dataset was extremely limited with only 15 measurements in all tissue type. Thus, arsenic was classified as "U" in both fresh and marine tissue.

Cadmium

Water

Freshwater

Cadmium was classified as Priority 2 because the 90th %ile observed freshwater concentration is below the 10th %ile of the effects data. The 95%ile observed cadmium concentration was above the chronic WQC.

Marine Water

There were insufficient observed data to prioritize cadmium in nearshore and offshore marine waters and it was categorized as "U".

Sediment

Freshwater Sediment

Cadmium was classified as Priority 1 based on comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment cadmium concentration is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of concern.

Marine Sediment

Cadmium was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed concentration of cadmium in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Cadmium was not assessed in tissue.

Wildlife

Cadmium was not assessed for wildlife.

Human Health

Cadmium was not assessed for human health because it is not included in the NTR.

Copper

Water

Freshwater

Copper in freshwater was classified as Priority 1 because the 90th %ile copper concentration exceeded both the 10th %ile effects concentration and the chronic copper WQC. The 95th %ile observed copper concentration also exceeds the acute WQC.

Marine Water

Copper in offshore waters was classified as Priority 2 because the 90th %ile observed offshore concentration is below the 10th %ile concentration of the effects data. Copper in nearshore waters was classified as Priority 1 because the 90th %ile observed nearshore concentration is above the 10th %ile effect concentration. In addition the acute and chronic water quality criteria exceeded the 90th %ile observed concentration. The 95th %ile observed offshore copper concentration was above the chronic WQC.

Sediment

Freshwater Sediment

Copper was classified as Priority 1 based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment copper concentration is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Copper was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed copper concentrations in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Copper was not assessed in tissue.

Wildlife

Copper was not assessed for wildlife.

Human Health

Copper was not assessed for human health because it is not included in the NTR.

Additional Evidence from Regional Studies

Aquatic Life

Copper is one of the most far-reaching potential priority toxicants in the Puget Sound region. This is due to its ability to alter the sensory capacity and behavior of a wide variety of aquatic organisms. A number of researchers have documented effects on regional species. Tierney et al. (2010) reviewed over 150 papers and found that avoidance behaviors were common in a variety of fresh and salt water fishes at less the 1 ug/L to concentrations ranging up to 20-30 ug/L.

Tested species included coho and Chinook salmon, as well as rainbow trout and golden shiner. Hecht et al. (2007) compiled a similar body of evidence for the disruptive effects of copper on juvenile salmonids. They used US EPA methodologies to calculate benchmark concentrations predicted to represent 10% and 50% reductions in chemosensory response at 0.18 ug/L and 2.1 ug/L respectively. These concentrations bracket a variety of other regional primary literature sources which confirm that the environmentally relevant range of <1.0 ug/L to 5.0 ug/L copper adversely impacts a variety of Puget Sound basin fish, particularly salmonids. Similar neurologic impacts were found by Linbo et al. (2006) on the mechanosensory lateral line of fish.

Sandahl et al. (2004) found copper concentrations of 4.4 ug/L produced sublethal neurotoxicity in coho salmon. In this laboratory study, copper reduced the ability of coho salmon to detect the natural oderants taurcholic acid and L-serine. Further study by Sandahl et al. (2007) confirmed that concentrations as low as 2 ug/L copper are not only affecting the neurologic systems of fish but also alter their behavioral responses to alarm pheromones. Baldwin et al. (2003) also found olfactory inhibition at the comparable, environmentally relevant, concentration of 2.3 ug/L.

Hansen et al. (1999) produced a seminal work which was utilized by several of the subsequent reviews discussed above. In it they documented Chinook salmon avoidance behaviors at concentrations as low as 0.7 ug/L dissolved copper. However, Chinook also failed to avoid concentrations >44 ug/L due to the extensive neural saturation. This window of affect potentially contributes to mortality from prolonged copper exposure or impairment of olfactory dependent behaviors such as homing.

Additional local studies by McIntyre et al. (2008) found that water hardness had little impact on copper's ability to alter olfactory function in coho salmon despite water hardness being a variable influencing the Washington State water quality standards. These regional reviews and studies provide an additional line of evidence suggesting that copper is a very important toxicant at concentrations well within the range found it the Puget Sound regional environment.

Lead

Water

Freshwater

Lead was classified as Priority 2 because the 90^{th} %ile observed freshwater concentration is below the 10^{th} %ile of the effects data.

Marine Water

Lead in offshore marine waters was classified as Priority 2 because the 90^{th} %ile observed concentrations are below the 10^{th} %ile of the effects data. In nearshore marine waters, lead was classified as "U" due to the insufficient amount of observed data.

Sediment

Freshwater Sediment

Lead was classified as Priority 2 because the 90^{th} %ile observed freshwater sediment concentration is below the FP-SQS.

Marine Sediment

Lead was classified as Priority 2 based on the comparison of the 90^{th} %ile observed near and offshore sediment concentrations to the SMS SQS. The 90^{th} %ile observed lead concentrations in both near and offshore sediment are below all of the additional guidelines evaluated.

Tissue

Lead was not assessed in tissue.

Wildlife

Lead was not assessed for wildlife.

Human Health

Lead was not assessed for human health because it is not included in the NTR.

Mercury

Water

Freshwater

The 90th %ile mercury concentration was below the 10th %ile of the effects concentration, and also exceeded the chronic WQC. Mercury in freshwater was classified as Priority 2.

Marine Water

Mercury in nearshore and offshore waters was classified as “U” because there were a limited number of environmental measurements (n = 13 and 7, respectively).

Sediment

Freshwater Sediment

Mercury was classified as a Priority 1 COC based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment lead concentrations is below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Mercury was classified as Priority 1 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed mercury concentrations in both near and offshore sediments are below all of the additional guidelines evaluated.

Tissue

Freshwater

Mercury is classified as “U” for freshwater non-decapod invertebrates and fish because insufficient observed and effects data are available for the assessment.

Marine Water

Mercury is classified as “U” for all three tissue types in near and offshore marine waters mainly due to a lack of effects concentrations. No marine effects concentrations were available for non-decapod invertebrates and fish and only one marine decapod effect concentration was available. There are observed concentrations for all tissue types although only five for offshore decapods.

Wildlife

Mercury was classified as Priority 1 for all four wildlife receptors because the estimated daily doses are at or above the lowest effect doses. Estimated daily doses of mercury to great blue heron and river otter are above 2 or more effects doses, whereas those of osprey and harbor seal are at or just above the lowest effect dose. Generally, mercury is estimated to bioaccumulate more in receptors living in freshwater habitats than marine habitats around Puget Sound.

Human Health

Freshwater

Mercury was classified as Priority 2 because the 90th percentile tissue concentration for bivalves, fish and other invertebrates did not exceed the NTR.

Marine Water

Mercury was classified as Priority 2 because the 90th percentile tissue concentration for bivalves, fish and other invertebrates did not exceed the NTR.

Additional Evidence from Regional Studies

Wildlife

Regional evaluations of mercury residues in Puget Sound wildlife in the Puget Sound are limited. Johnson et al. (2009) measured mercury concentrations in osprey eggs from the Lower Duwamish River in 2003 and again in 2006/2007. These data demonstrated that mercury concentrations in osprey eggs decreased between these sample periods.

Grove and Henny (2008) measured contaminants in the livers of river otter carcasses collected by trappers in western Oregon and western Washington, including Puget Sound. Mercury liver concentrations were higher in adult river otters from Puget Sound (mean of 7.89 mg/kg dry) than those from the northwest Washington area (mean of 5.85 mg/kg dry weight), located just east of Puget Sound and including the greater Seattle urban area. Mercury levels in Willamette River otter livers and coastal Oregon were slightly higher (mean of 9.2-9.3 mg/kg dw), but similar to Puget Sound levels. This study demonstrates that river otters living in the Puget Sound area are exposed to and bioaccumulate mercury.

Zinc

Water

Freshwater

Zinc was classified as Priority 2 because the 90th %ile observed freshwater concentration is below the 10th %ile of the effects data.

Marine Water

Zinc in offshore waters was classified as Priority 1 because the 90th %ile observed nearshore zinc concentrations is above the 10th %ile concentration of the effects data. Insufficient observed data for zinc in nearshore waters was available; zinc in nearshore waters was classified as “U”.

Sediment

Freshwater Sediment

Zinc was classified as Priority 1 based on the comparison of the 90th %ile observed sediment concentration to the FP-SQS. The 50th %ile observed sediment zinc concentration was below all additional guidelines used in this assessment, suggesting that only the highest concentrations are of greatest concern.

Marine Sediment

Zinc was classified as Priority 2 based on comparison of the 90th %ile observed near and offshore sediment concentrations to the SMS SQS. The 90th %ile observed sediment zinc concentration is above the TEL; the 90th %ile observed nearshore concentration was below the TEL.

Tissue

Zinc was not assessed in tissue.

Wildlife

Zinc was not assessed for wildlife.

Human Health

Zinc was not assessed for human health because it is not included the NTR.

PCBs

The potential for effects associated with PCB exposure was assessed for all media and pathways (water, sediment, tissue, human health and wildlife). Observed environmental PCB concentrations were assessed as the sum of Aroclors® and the sum of congeners. In general, the majority of the available toxicity data were based on individual Aroclor® exposures. It was not practical to compare observed individual Aroclor® and congener data to available effects data for individual compounds. Caution is advised in the use of Aroclor® data; these data may not be optimal due to shifts in the congener composition associated with weathering. It is also important to note that the PCB WQC is not protective of aquatic life through the bioaccumulation pathway.

Water

Freshwater

The 90th %ile observed concentrations of both PCB congeners and Aroclors® were below the 10th %ile concentration of the available effects data. However, the 90th %ile PCB Aroclor® concentration exceeded the chronic WQC. PCB Aroclors® were classified as Priority 1; PCB congeners were classified as Priority 2.

Marine Water

Extremely limited PCB concentration data are available for marine nearshore waters, with only 11 PCB Aroclor® concentrations and no PCB Congener data available; PCBs in nearshore waters were classified as “U”. Similarly, PCB Aroclor® data in offshore marine waters were not available. However, sufficient PCB Congener data were available in marine offshore waters, where the 90th %ile concentration of total PCB congeners was below the 10th %ile of the available effects concentrations; PCB congeners in offshore marine waters were classified as Priority 2.

Sediment

Freshwater Sediment

PCB Aroclors® in freshwater sediments were classified as Priority 1. The 90th %ile observed concentrations of both PCB congeners and Aroclors® were above the FP-SQS, in addition to 3 of the 5 additional guidelines evaluated. Only the PEC fell above the 90th %ile concentration of both PCB congeners and Aroclors®. With the exception of the TEL, the 50th %ile PCB Aroclor® concentration was below all remaining guidelines evaluated. In general, this suggests that areas with the highest concentrations are of concern. Sediment congener data were limited (n=26); as such they were classified as “U”.

Marine Sediment

PCB Aroclors® in marine offshore sediment were classified as Priority 1 because the 90th %ile OC normalized PCB Aroclor® concentrations in marine offshore sediments exceeded the marine SQS. The 90th %ile concentrations of OC normalized PCB Congeners in offshore sediments and PCB Aroclors® in nearshore sediments did not exceed the SQS, resulting in a Priority 2 classification. Insufficient data were available to evaluate PCB congeners in nearshore sediments.

Tissue

Freshwater

PCB Aroclors® and PCB congeners in freshwater non-decapod invertebrates are classified as Priority 1 because the 90th %ile observed concentrations are higher than the 10th %ile of the effects data. All other freshwater tissues are classified as “U” for both PCB Aroclors® and congeners due to the limited number of effects values and/or environmental concentrations. There were no observed data available for PCB congeners in decapods.

Marine

PCB Aroclors® and congeners in nearshore decapods are classified as Priority 2 because the 90th %ile observed concentration is below the 10th %ile effects concentration. PCB Aroclors® and congeners in nearshore non-decapod invertebrates are also classified as Priority 2. PCBs in fish are classified as "U" due to the limited amount of available effects data.

PCB Aroclors® and congeners in offshore decapods and fish were classified as "U" due to a lack of effects values or insufficient quantity of observed concentrations. PCB Aroclors® in offshore non-decapod invertebrates were classified as Priority 2 but PCB congeners are classified as "U" because of insufficient numbers of observed and effect concentrations in non-decapod invertebrates (< 20 and < 5 respectively).

Wildlife

PCBs were classified as Priority 1 for all four wildlife receptors because the estimated daily doses are more than 10 times lower than the lowest effect doses. Generally, PCBs are estimated to bioaccumulate to a greater degree in receptors living in marine habitats than freshwater habitats around Puget Sound. Estimated daily PCB doses to osprey, river otter and harbor seal are above several effects doses. Those of the great blue heron hover near the three lowest effect doses.

Human Health

PCBs were classified as a Priority 1 human health concern; multiple freshwater and near- and offshore tissues types exceeded the NTR PCB concentration standard. The range of observed PCB concentrations analyzed by Aroclor® and congener methods vary from one another. This variability is likely the result of multiple projects using different analytical methods for different suspected levels of contamination.

Additional Evidence from Regional Studies

Aquatic Life

PCBs have been detected in outmigrant juvenile salmon (Johnson et al., 2007) from multiple northwest estuaries and hatcheries, including three in the Puget Sound. Whole-body juvenile Chinook salmon from the Duwamish River contained the highest PCB concentration (103 ng/g wet weight or 3100 ng/g lipid) of any of the locations tested. Johnson et al. (2007) note that this concentration is higher than NOAA's estimated threshold for adverse health effects of 2400 ng/g lipid. Separately, juvenile salmonid PCB exposures were documented as occurring via food source by an analysis of stomach content of outmigrants at three locations in Puget Sound (Stein et al. 1995). Meador et al. (2010) found that PCB tissue concentrations in outmigrant juvenile Chinook from the Duwamish estuary varied by time and location within the estuary, suggesting that localized heterogeneity of sediment concentrations may substantially impact accumulation in fishes.

PCB concentrations in adult Puget Sound Chinook salmon tissues were found to be 3 – 5 times higher than those measured in six other populations of Chinook salmon on the West Coast of North America (O'Neill and West 2009). O'Neill and West note that these elevated tissue concentrations have resulted in consumption advisories, and have implications for the viability of these fish and southern resident killer whales. Cullon et al. (2009) found PCBs in adult Chinook returning to the Duwamish River, as well as in Puget Sound Chinook smolts.

PCBs concentrations in Puget Sound herring and Puget Sound flatfish have also been evaluated. Puget Sound herring were found to contain 3 to 9 times higher concentrations of PCBs than herring from the Strait of Georgia, with Puget Sound whole-body concentrations ranging from about 120 to 160 ng/g wet weight (West et al. 2008). Analyses of various biomarkers of pollution exposures in benthic flatfish were shown to successfully differentiate between sites with differing degrees of sediment contamination (Stein et al. 1992). Cullon et al. (2005) also found about seven times higher levels of PCBs in a mixture of fishes designed to represent the diet of Puget Sound harbor seals than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia. Sol et al. (2008) found a statistically significant correlation between PCB concentrations in English sole livers and two biological effects parameters.

Wildlife

A number of studies have been conducted in the Puget Sound region investigating exposure and/or effects of PCBs and other persistent and bioaccumulative contaminants on wildlife, particularly marine mammals. The salient information on PCBs in wildlife is summarized here.

Johnson et al. (2009) measured PCB concentrations in osprey eggs from the Lower Duwamish River and compared them to those sampled from the upper Willamette River. Total PCB residues were significantly higher in Lower Duwamish River osprey eggs (geometric mean = 897 ug/kg wet weight) compared to those from the Willamette River (geometric mean = 182 ug/kg ww). These results demonstrate that adult osprey bioaccumulation and maternal transfer of PCBs is occurring in osprey nesting in PCB contaminated areas of Puget Sound. This study also compared egg residues over time and determined that PCB concentrations in osprey eggs from the Lower Duwamish River had decreased 53% between 2003 and 2007.

Grove and Henny (2008) also demonstrated the bioaccumulation of PCBs in river otter livers from Puget Sound. The Puget Sound area river otters accumulated more PCBs (as total PCBs) than otters from other areas in western Washington.

PCBs and other organochlorines have been shown to cause immunosuppression, thyroid disruption and possibly cancer in harbor seals (Tabuchi et al. 2006, Ylitalo et al. 2005; Simms et al. 2000; Ross et al. 1996; Ross et al. 1995; de Swart et al. 1996; de Swart et al. 1995; Van Loveren et al. 1994). Vitamin A disruption has also been observed in harbor seal pups found on the Washington State coast whose mothers contained high PCB residues in their blubber (Simms et al. 2000). This effect on seal pups is suspected to result from exposure to contaminated milk.

There is substantial evidence that Puget Sound harbor seals and killer whales are bioaccumulating PCBs at very high concentrations in their blubber. The prey items of Puget Sound harbor seals were measured to have 7 times higher concentrations of PCBs than prey from Strait of Georgia on a lipid basis (Cullon et al. 2005), which corresponds to PCB concentrations measured in harbor seal blubber.

PCB tissue concentrations are often reported as dioxin toxicity equivalents (i.e. TEQs) which represent the toxicity of dioxin-like PCBs, dioxins and furans relative to the most toxic dioxin - 2,3,7,8-TCDD. Puget Sound harbor seals have significantly higher TEQs in blubber (158 ng/kg lipid weight) compared to seals from the Strait of Georgia (33 ng/kg lipid weight) (Ross et al. 2004); the TEQ contribution was greater from PCBs than dioxins and furans. Levin et al. (2005) also found that the majority of TEQs in harbor seal pups (from southern B.C.) were from PCBs, not dioxins and furans.

Ross et al. (2000) reported measured mean total PCB concentrations in transient and Southern resident male killer whales of 251 and 146 mg/kg -lipid, respectively. The authors concluded these marine mammals are among most contaminated in the world. Further research on the northern, southern and transient killer whale communities have discovered that males bioaccumulate more PCBs than females; female offload a portion of their tissue burden to their young through maternal transfer (Krahn et al. 2007; Krahn et al. 2009). PCB concentrations in the blubber of mothers decrease temporarily during nursing and can reach concentrations below those in their calves. Mothers initiate bioaccumulation again after calves are weaned. Total PCB concentrations in southern resident killer whales range from about 5,000 to 180,000 µg /kg lipid. For all but three recent mothers, the measured concentrations exceed a marine mammal threshold for blubber concentrations (17,000 µg /kg lipid) (Krahn et al. 2007).

Although environmental concentrations of PCBs are gradually declining, one modeled estimate of southern resident killer whale recovery projects that blubber concentrations will not decrease to the marine mammal threshold until 2063 (Hickie et al. 2007). The reviewed studies suggest that marine mammals in Puget Sound are accumulating PCBs in their blubber to very high concentrations. The results of the quantitative assessment are congruent and classify PCBs as Priority 1.

PBDEs

The PBDE assessment was limited due to the lack of effects data, guidelines or criteria. While PBDEs have been measured in a variety of media, appropriate effects data were insufficient to fully asses this COC.

Water

Surface water data for PBDEs in freshwater (n=255) and marine offshore waters (n=126) were available; however, appropriate effects data were not available in the ECOTOX database, nor is there a WQC for PBDEs. Due to the lack of effects data, PBDEs in both fresh and marine waters were classified as “U”.

Sediment

PBDEs in freshwater and marine sediments were classified as "U". A limited number of observed concentration data for PBDEs in marine (n=46) and freshwater (n=77) sediments were available. However sediment guidelines are not available for PBDEs in either fresh or marine sediments.

Tissue

PBDEs were not assessed in tissue due to a lack of effects thresholds.

Wildlife

PBDEs were not assessed quantitatively in wildlife due to a lack of effects doses. See the Additional Evidence from Regional Studies below for a discussion of available effects information.

Human Health

PBDEs are prioritized as unknown, "U" because NTR criteria are not available.

Additional Evidence from Regional Studies

Aquatic Life

Sloan et al. (2010) detected PBDEs in outmigrant Chinook salmon tissue and stomach content from four sites in Puget Sound. Concentrations in wild outmigrant juveniles were higher than in hatchery fish. PBDE concentrations in Puget Sound juvenile fishes ranged from 67 to 13,000 µg /kg lipid, which was generally comparable to those measured in the Lower Columbia River and Estuary. Sloan et al. (2010) conclude that PBDEs may be contributing to reduced health and fitness in outmigrant juvenile Chinook salmon.

PBDEs have also been detected in adult Chinook salmon returning to the Duwamish River; however, they were not detected in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or the Deschutes River (Cullon et al. 2009). Lema et al. (2008) demonstrated that dietary exposures of certain PBDEs by adult fathead minnows alter thyroid status and thyroid hormone-regulated gene transcription. Arkoosh et al. (2010) found that juvenile Chinook salmon exposed to moderate doses of PBDEs through their diet may be at increased risk of disease relative to those exposed to higher or lower doses of PBDEs in their diet potentially indicative of a complex U-shaped dose response curve for PBDEs in Chinook salmon. PBDE levels in a mixture of fishes designed to represent the diet of Puget Sound harbor seals were found to be about four to five times higher than in a similar mixture of fish designed to represent the diet of harbor seals from the Strait of Georgia (Cullon et al. 2005).

Wildlife

Because little information on PBDE toxicity to wildlife is available and a quantitative assessment could not be conducted, the publications available are reviewed here regardless of their geographic location. Few studies have been conducted examining effects of PBDEs on birds.

The studies reviewed indicate that PBDEs impact the reproduction and endocrine system similarly to PCBs. Fernie et al. 2005 showed that American kestrel egg injection and oral gavage exposure to PBDE congeners caused hepatic oxidative stress and altered thyroid hormone and vitamin A concentrations and glutathione metabolism. Exposure to PBDE congener 71 for 75 days adversely impacted courtship and mating behavior of American kestrels (Fernie et al. 2008). These birds also displayed significant delays in clutch initiation and produced smaller eggs (Fernie et al. 2009). Eggshell thinning and reduced hatching success also resulted.

A study of species sensitivity to PBDEs (PBDE-71) observed that pentabrominated diphenyl ether (Penta BDE) exposure to eggs at 0.01 to 20 mg/kg caused decreased pipping and hatching success in American kestrels but not chickens or Mallard ducks (McKernan et al. 2009). Species sensitivity was concluded to be Mallard ducks <chickens <American kestrels.

Total PBDE concentrations in osprey eggs and nestling plasma were significantly lower in birds from the Lower Duwamish River (eggs: 321 ug/kg ww; plasma: 6 ppb ww) compared to (Johnson et al. 2009) those from the upper Willamette River (eggs: 897 ug/kg ww; plasma: 22 ug/kg ww). The total PBDE concentrations in the osprey eggs did not change significantly between 2003 and 2007. Reproductive failure was observed in four of nine nests in the Lower Duwamish area. A small dataset from this study suggests that some nestlings may have experienced immunosuppression. However, the results were inconclusive due to the small sample size.

One study (Fernie et al. 2011) was acquired for which a dietary effect dose could be determined. Fernie et al. (2011) exposed American kestrels to PBDEs (Hexa-BDE) via dietary exposure and measured reproductive parameters. Adult kestrels exposed to 0.51 mg/kg/d PBDE, an environmentally relevant dose, through their diet displayed less courtship behaviors, earlier egg laying, a greater clutch size and smaller eggs; however, there were no significant differences in the fledging or hatching rates compared to control.

To compare exposures of birds feeding in Puget Sound, a PBDE daily dose was estimated using the same methods as the other COCs for wildlife. At the higher sediment ingestion rate (4%), the estimated daily dose of total PBDEs for the osprey is 0.006 mg/kg/d. The estimated daily dose for the great blue heron is also 0.006 mg/kg/d. If the threshold from Fernie et al. 2011 is considered an effect dose, piscivorous birds in the Puget Sound watershed are estimated to experience lower exposure by approximately a factor of 100.

Compared to birds, a larger but still limited number of publications exist on the effects of PBDEs in mammals. Rodent exposure studies have demonstrated thyroid hormone disruption (Hallgren et al. 2001, Zhou et al. 2002), developmental neurotoxic and behavioral effects (Ericksson et al. 2001, Viberg et al. 2003a, Viberg et al. 2003b). A study of grey seal pups and juveniles observed a relationship between circulating thyroid hormones, transport proteins and PBDE uptake (Hall et al. 2003).

Similar to PCBs, there is evidence of PBDE bioaccumulation in the blubber of marine mammals at high concentrations. However, absolute total PBDEs concentrations appear to be lower than total PCBs. Cullon et al. (2005) measured PBDE concentrations 5 times higher in harbor seal prey from Puget Sound than the Strait of Georgia; however, the mean PBDE concentration was 5 times lower than that measured for PCBs. Krahn et al. (2009) and Rayne et al. (2004) found the same pattern of killer whale blubber concentrations in males, mothers and calves as they found for PCBs with males having the highest concentrations and females experiencing fluctuations due to maternal transfer. Krahn et al. (2005) measured total PBDE concentrations in killer whale blubber ranging from 680 to 15,000 ug/kg lipid. Mean PBDE concentrations in northern male killer whale blubber have been found to be significantly lower (203 ug/kg lw) than those of southern resident (942 ug/kg lw) and transient males (1015 ug/kg lw).

Although a full quantitative effects assessment was not conducted for PBDE exposure to wildlife, published research demonstrates that PBDEs are bioaccumulating to high concentrations in Puget Sound marine mammals. This coupled with the growing evidence that PBDE exposure can cause thyroid and developmental effects in mammals strongly suggest that PBDEs should be classified Priority 1.

Dioxins and Furans

Both observed environmental concentrations and effects data for dioxins and furans (PCDD/Fs) were limited. As a result the assessment was limited to evaluation of potential effects to wildlife and human health.

Water

Observed surface water data for PCDD/Fs were limited to 7 measurements in freshwater for 3 compounds (1,2,3,4,7,8 HxCDD, 1,2,3,7,8 PeCDD and 2,3,7,8 TCDD). No observed data were available for marine waters; effects data for these compounds were very limited. Due to the lack of data to assess these compounds, dioxins and furans in surface waters were classified as “U”.

Sediment

PCDD/Fs in freshwater and marine sediments were classified as “U”. A moderate number (n >700) of observed concentration data for PCDD/Fs in marine and freshwater sediments were available. However, FP SQS and SQS values are not available for PCDD/Fs. The 90th %ile observed freshwater sediment concentration exceeded both the PEL and TEL; while the 50th %ile concentration was below both the PEL and TEL. The 90th %ile observed nearshore marine sediment concentration was just above the PEL; while the 50th %ile concentration was above the TEL. The 90th %ile observed offshore was below the PEL, but above the TEL.

Tissue

PCDD/Fs were not evaluated in freshwater or marine tissues due to the lack of effects data or the lack of observed concentrations. No observed PCDD/Fs concentrations are available in freshwater or marine tissues.

Wildlife

PCDD/Fs were classified as Priority 1 for the great blue heron and river otter because the estimated daily dose was greater than 0.1 times the lowest effects dose. For harbor seal, PCDD/Fs were classified as Priority 2 because adequate effects data were available and the estimated daily doses are more than 10 times lower than the lowest effects dose. The osprey daily doses are estimated to be almost 1,000 times lower than the lowest effects dose; however, substantial uncertainty exists around the effects of PCDD/Fs on birds so the resulting classification was "U". The estimated daily doses for great blue heron are close to one of the two existing effects doses for birds, so the assumption was made that reproductive effects were likely occurring.

Human Health

2,3,7,8-Tetrachlorodibenzo-p-dioxin (2,3,7,8 TCDD) was classified as Priority 1; NTR criteria were not available for other PCDD/Fs. The 90th %ile of the observed 2,3,7,8 TCDD concentrations exceeded the NTR criteria for all three tissue groups (bivalves, fish and other invertebrates). Tissue from both near and offshore areas exceeded the NTR criteria for one or more tissue types, although sample sizes were generally smaller than for freshwater tissues.

Additional Evidence from Regional Studies

Aquatic Life

Generally similar concentrations of PCDD/Fs were found in adult Chinook salmon returning to the Duwamish River, Johnstone Strait, Lower Fraser River, and the Deschutes River (Cullon et al. 2009).

Wildlife

PCDD/Fs TEQ residues in river otter livers from Puget Sound contributed one third to the total TEQs (Grove and Henny, 2008) indicating that PCBs accumulate in river otter livers to a concentration that is twice as toxic as dioxins and furans.

Studies in Puget Sound of harbor seal and southern resident killer whale prey items have shown that prey of these marine mammals are higher in dioxins and furans compared to the same prey from the Strait of Georgia and British Columbia coast (Cullon et al. 2005, Cullon et al. 2009). PCBs in Harbor seal prey were 3-4 times higher on a lipid basis than prey from the Strait of Georgia (Cullon et al. 2005). However, Ross et al. (2000) found that dioxin and furan concentrations in killer whale blubber were much lower than PCBs and there were no differences between whales from the northern and southern resident and transient communities. This was suspected to be due to metabolic removal of dioxins and furans.

DDTs and metabolites

Observed environmental data for DDTs were available for water, sediment and tissue; data in marine waters were very limited. Effects data were available for water, marine sediment, tissue residue, wildlife and human health. Freshwater sediment guidelines were not available.

Water

Freshwater

The 90th %ile observed concentration of DDTs and metabolites was below the 10th %ile of the available effects data; however, the 90th %ile concentration was above both the acute and chronic DDT WQC. DDTs in freshwater were classified as Priority 1.

Marine

DDTs were measured, but not detected, in a limited number of samples (n=11) in marine nearshore waters; there were no DDT measurements in offshore waters. DDTs in marine waters were classified as “U”.

Sediment

Freshwater Sediment

A FP-SQS is not available for DDT in freshwater sediment; as such this COC was classified as “U”. The 90th %ile observed DDT freshwater sediment concentration was well below both the PEL and PEC, while the 50th %ile concentration was above both the TEC and TEL.

Marine Sediment

A SQS is not available for DDT; as such, this COC was classified as “U”. The 90th %ile observed concentration in nearshore marine sediment was above the PEL, 2LAET, LAET and TEL; the 50th %ile concentration was below all four of these guidelines. In offshore sediments, both the 90th %ile and 50th %ile were below the PEL, 2LAET and LAET; and above the TEL.

Tissue

Freshwater

Total DDTs are classified as Priority 2 in freshwater non-decapod and fish tissue because the 90th %ile concentration is below the 10th %ile effects data. DDTs could not be assessed in freshwater decapods due to a lack of observed concentrations.

Marine

Total DDTs are classified as "U" in all marine nearshore and offshore tissues due to insufficient (< 5 values) effects data.

Wildlife

DDT and metabolites were assessed as the sum of DDT, DDE, and DDD isomers and were classified as Priority 1 for great blue heron and osprey because the estimated daily doses are greater than 0.1 times the lowest effect dose. The mammals, river otter and harbor seal, were both classified as Priority 2 because the estimated daily doses are more than 10 times lower than the lowest effect dose.

Human Health

4,4' DDT was classified as Priority 2 in all freshwater and offshore tissues types. In the nearshore area, the 90th %ile concentration of 4,4' DDT for both fish and other invertebrates exceeded the NTR threshold and were classified as Priority 1. For the DDT metabolite 4,4' DDE, only the 90th %ile of other invertebrate tissues in freshwater exceeded the NTR threshold and was classified as Priority 1. All tissues in near and offshore areas were classified as Priority 2. For the DDT metabolite 4,4' DDD, other invertebrates tissues in freshwater were classified as Priority 1, while all other areas and tissues were classified as Priority 2.

Additional Evidence from Regional Studies

Aquatic Life

Total DDT (sum of DDT, DDE, and DDD) concentrations in stomach contents of outmigrant juvenile Chinook salmon from the Duwamish Estuary and Commencement Bay were found to be elevated relative to the stomach content concentrations of fish from the Nisqually Estuary (Stein et al. 1995). Whole-body total DDT (sum of DDT, DDE, and DDD) concentrations in juvenile Chinook salmon were found to be relatively high (over 1000 ng/g lipid or 25 ng/g wet weight) in fish from the Nisqually, Duwamish and Columbia River Estuaries (Johnson et al. 2007).

Johnson et al. (2007) also found detectable levels of DDTs in stomach contents, with stomach content concentrations substantially higher in Columbia River and Grays Harbor juvenile Chinook than in Duwamish and Nisqually Estuary juvenile Chinook. Johnson et al. (2007) suggest that at the observed levels, DDTs are unlikely to cause adverse effect by themselves; however, they may contribute via additive or synergistic effects with other contaminants. Substantially higher levels of DDTs were found in adult Chinook salmon returning to the Duwamish River than in adult Chinook returning to the Johnstone Strait, Lower Fraser River, or Deschutes River (Cullon et al. 2009).

Analysis of DDT concentrations in Pacific herring indicated that concentrations from Puget Sound herring were 1.5 to 2.5 times higher than those from Strait of Georgia (West et al. 2008), with Puget Sound concentrations ranging from 19 to 27 ng/g wet weight (240 to 330 ng/g lipid).

Cullon et al. (2005) found similar levels of DDTs in a mixture of fishes designed to represent the diets of Puget Sound and Strait of Georgia harbor seals.

Wildlife

Grove and Henny (2008) evaluated DDTs in river otters. They did not detect DDT and detected only low concentrations of DDE (mean of 0.004-0.28 mg/kg ww) in river otter livers from Puget Sound which were much lower than those found in animals residing near the Columbia River (mean of 0.12-1.65 mg/kg ww).

Lipid based concentrations of total DDT concentrations in Harbor seal prey in Puget Sound are 1.6 times higher than those from the Strait of Georgia (Cullon et al. 2005). Puget Sound Chinook, the primary prey of southern resident killer whales, have higher body residues of DDTs and lower lipids compared to Chinook from British Columbia coast (Cullon et al. 2009). Krahn et al. (2009) found the same pattern of killer whale blubber concentrations as found for PCBs in males, mothers and calves. That is, males have the highest DDT concentrations in their blubber and female blubber concentrations vary with their maternity status due to maternal transfer. Total DDT concentrations in killer whales ranged from 1,000 to 160,000 ug/kg lipid.

PAHs

PAHs were evaluated in water, sediment and for human health.

Water

The majority of the toxicity data available for PAHs in surface waters is based on individual PAHs. As such, Total PAHs, HPAH and LPAH were not directly evaluated here.

Freshwater

While there were sufficient observed concentration data (N>1500 measurements) for the individual PAHs evaluated here, there were limited effects data for a number of PAH compounds. The 90th %ile observed concentrations of acenaphthene, anthracene, benzo(a)pyrene, fluoranthene, fluorene, naphthalene, phenanthrene, and pyrene were below the 10th %ile of the available effects concentrations; these COC s were classified as Priority 2. Effects data were insufficient to evaluate benzo(a)anthracene, benzo(k)fluoranthene, benzo(g,h,i)perylene, chrysene, and dibenzo(a,h)anthracene; these COCs were classified as “U”.

Marine Water

Observed concentrations of individual PAHs in nearshore waters were very limited (N=12). Due to the lack of sufficient measured concentrations, in addition to the limited availability of effects data, individual PAHs in nearshore waters were classified as “U”.

While there were sufficient observed nearshore marine concentration data for some individual PAHs, marine effects data were limited for a number of COCs. The 90th %ile observed concentrations of acenaphthene, fluoranthene, naphthalene and phenanthrene in nearshore waters were below the 10th %ile of the available effects data; these COCs were classified as "U". The remainder of the individual PAHs in nearshore waters (anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(k)fluoranthene, chrysene, dibenzo(a,h,)anthracene, fluorene and pyrene) were classified as "U" due to insufficient data to fully assess these COCs. There were insufficient data to evaluate individual PAHS in offshore waters; these COCs were classified as "U".

Sediment

Freshwater Sediment

Both LPAH and HPAHs in freshwater sediments were classified as Priority 1. The 90th %ile observed LPAH concentration was also above the FP-CSL; however, the 75th %ile concentration was below both the FP SQS and FP CSL. The 90th %ile HPAH concentration was below the FP-CSL and the 75th %ile concentration was below .both the FP-CSL and FP-SQS. These data suggest that only some of the highest detected concentrations are likely to be of concern. With the exception of benzo(a)anthracene which was classified as Priority 2, all of the individual PAHs evaluate were classified as Priority 1.

Marine Sediment

Both LPAH and HPAH in near and offshore sediments were classified as Priority 2; the 90th %ile observed concentrations were below the SQS. All of the individual PAHS in near and offshore sediments were also classified as Priority 2.

Tissue

Tissue residue concentrations of PAHs were not evaluated because these chemicals typically to not accumulate in the tissue of vertebrates and are rapidly metabolized in fish. It was beyond the scope of this effort to evaluate PAHs in those invertebrates that are not capable of metabolizing PAHs.

Wildlife

PAHs were not evaluated because these chemicals typically do not bioaccumulate in wildlife.

Human Health

Five if the nine individual PAHS (benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene) evaluated in freshwater tissue exceeded the NTR criteria and were classified as Priority 1. Concentrations of the remaining (anthracene, fluorene, fluoranthene, and pyrene) PAHs evaluated were below the NTR criteria and classified as Priority 2.

The same pattern of Priority classification was observed in nearshore tissues as was found for the freshwater tissues. Tissue data from nearshore waters was limited; as a result, five of the individual PAHS were classified as "U" (benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-cd)pyrene). The remainder of offshore marine tissue was classified as Priority 2 for human consumption.

Additional Evidence from Regional Studies

Aquatic Life

Multiple investigations have identified biomarkers of PAH exposure in various Puget Sound fishes.

Bile and stomach content of outmigrant juvenile Chinook salmon were found to contain various PAHs (Johnson et al. 2007), demonstrating that diet pathways are important PAH exposure pathways. The authors suggest that exposure may result in immunosuppression and other health effects. These results expanded and confirmed previously documented PAH exposures (Stein et al. 1992; Stein et al. 1995). A dietary feeding study on juvenile Chinook documented growth and physiological responses from dietary exposures to PAHs at concentrations that were environmentally realistic in the Puget Sound (Meador et al. 2006).

Biomarkers of PAH exposure were confirmed in Puget Sound English sole, rock sole, and starry flounder collected from up to five sites in Puget Sound (Stein et al. 1992). Stein et al. found that biomarkers of exposure were related to the degree of sediment contamination. Further field study (Johnson 2000) resulted in recommended a sediment threshold of 1000 ppb total PAHs to protect English sole against liver lesions, DNA adducts in liver, and other effects. The causal relationship between elevated sediment PAH concentrations and English sole liver effects was confirmed by Meyers et al. (2003). In a study of English sole from the Hylebos Waterway and Colvos Passage, Sol et al. (2008) found no correlation between PAH exposure and age and little correlation between reproductive end points and PAH exposure. Pacific herring embryos were found to be affected by tricyclic PAHs in weathered crude oil (Incardona et al. 2009; Carls et al. 1999).

Several laboratory studies have documented that developmental defects in fish are associated with exposure to PAHs released by the weathered crude oil, notably the tricyclic-PAHs (Incardona et al. 2005; Incardona et al. 2006; Carls et al. 2008). Carls and Meador (2009) developed a description of the oil weathering, PAH toxicity, and embryo exposures to explain the observed toxicity of PAHs in weathered oil at relatively low levels. Driscoll et al. (2010) developed a framework for describing PAH exposure as a dose to fishes in order to understand the mechanisms of exposure and toxicity.

Bis(2-Ethylhexyl) Phthalate

The assessment of bis(2-ethylhexyl) phthalate was limited to water and sediment.

Water

The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in freshwater was below the 10th %ile of the available effects data; this COC was classified as Priority 2. Insufficient observed and effects data for bis(2-ethylhexyl) phthalate in marine nearshore waters were available; as such it was classified as “U”. The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in offshore marine water was below the 10th %ile of the available effects data; as such it was classified as Priority 2.

Sediment

The 90th %ile observed concentration of bis(2-ethylhexyl) phthalate in freshwater sediments was above the FP-SQS and the FP-CSL; this COC was classified as Priority 1. The OC-normalized 90th %ile concentrations in both marine nearshore and offshore sediments exceeded the marine SQS, resulting in Priority 1 classification.

Tissue

Bis(2-ethylhexyl)phthalate tissue concentrations were not evaluated because phthalates typically do not accumulate to a significant degree in tissues.

Wildlife

Bis(2-ethylhexyl) phthalate was not evaluated.

Human Health

The 90th %ile of the observed freshwater bivalve tissue concentration of bis(2-ethylhexyl) phthalate exceeded the NTR criteria. While there were a number of measurements (>100) for the other tissue types in marine and freshwaters, there were too few detections to calculate a 90th percentile. Thus for freshwater, bis(2-ethylhexyl)phthalate was classified as Priority 1, while for marine tissues bis(2-ethylhexyl)phthalate was classified as priority “U”.

Triclopyr

The assessment of triclopyr was limited to water.

Water

The 90th %ile observed triclopyr concentration in freshwater was below the 10th %ile concentration of the available effects concentrations and was classified as Priority 2. No observed triclopyr data were available in marine nearshore or offshore waters, resulting in a classification of “U”.

Sediment

No observed concentrations of triclopyr are available in freshwater or marine sediments. This COC was classified as “U” in sediments.

Tissue

Tissue concentrations were not evaluated because these types of pesticides typically do not accumulate in tissues.

Wildlife

Triclopyr was not evaluated because it is not bioaccumulative in wildlife.

Human Health

Triclopyr is not listed in the NTR and was not evaluated.

Nonylphenol

The assessment of nonylphenol was limited to water and sediment. Effects data are limited for this COC.

Water

Nonylphenol was classified as Priority 2 in freshwater because the 90th %ile concentration is below the acute and chronic WQC and below the 10th %ile of the available effects data. However, the 95th %ile observed concentration was above the chronic WQC for nonylphenol. Nonylphenol was classified as "U" in marine nearshore water because insufficient effects and observed data were available. Nonylphenol was classified as Priority 2 in offshore marine water because the 90th %ile concentration is below the acute and chronic WQC and below the 10th %ile of the available effects data.

Sediment

Nonylphenol was classified as "U" in sediments because no FP-SQS is available for freshwater sediments and no SQS or CSL are available for marine sediments.

Tissue

Nonylphenol tissue concentrations were not evaluated because this COC does not typically bioaccumulate to a significant degree in tissues.

Wildlife

Nonylphenol was not evaluated because it is not bioaccumulative in wildlife.

Human Health

Nonylphenol is not listed on the NTR and was not evaluated.

Petroleum

Water

All four of the individual petroleum products were classified as "U" because the number of effect concentrations for each product was insufficient (< 15) and the number of observed concentrations for heavy fuel/bunker oil was insufficient (< 50).

Sediment

Petroleum was not assessed in sediment because there are no sediment guidelines nor observed sediment data.

Tissue

Petroleum was not assessed for tissue residues because there are no effects nor observed tissue data.

Wildlife

Petroleum could not be evaluated for wildlife because there are no effects data.

Human Health

Petroleum was not evaluated because it is not listed on the NTR.

Combined Prioritization for all Elements of the Assessment

The specific COCs evaluated in the quantitative assessments varied; a summary of the media and pathways evaluated for each COC is presented in Table 9. The chemicals assessed in this report were placed into three groups based on the likelihood that they may currently be causing widespread environmental effects (Table 10). This grouping was based on a review of the individual priority classification for each line of evidence evaluated.

COCs with multiple Priority 1 and different lines of evidence

Those chemicals with two or more Priority 1 classifications for the different lines of evidence were categorized as "Multiple Priority 1". These chemicals represent the COCs with the most compelling evidence that they may be causing widespread environmental effects in the Puget Sound region. Chemicals in "Multiple Priority 1" are likely to warrant action to reduce the potential for widespread environmental affects.

COCs with a single Priority 1 or line of evidence

Those chemicals with one priority 1 classification were placed in "Single Priority 1". These COCs represent those with strong evidence that they may be causing widespread environmental effects in the Puget Sound region, but where the evidence is limited to one line evidence from the many evaluated. Chemicals in “Single Priority 1” are likely to warrant action to reduce the potential for widespread environmental affects.

COCs with no Priority 1 and no additional lines of evidence

COCs that were not classified in any media for pathway were categorized as “No Priority 1”. These COCs represent those with ongoing concern about their effects, but for which limited evidence is available to indicate they may be causing widespread environmental impacts in the Puget Sound region. Some “No Priority 1” COCs were not evaluated for some lines of evidence due to limited availability of observed data and/or the lack of effects data. Chemicals in “No Priority 1” may warrant action based on existing concerns that the current assessment was unable to capture and the extent and nature of the potential effects from these chemicals.

Table 9. Summary of COCs Assessed.

COC	Water	Sediment	Tissue	Wildlife	Human Health
Arsenic	X	X	NA	NA	X[1]
Cadmium	X	X	NA	NA	NA
Copper	X	X	NA	NA	NA
Lead	X	X	NA	NA	NA
Mercury	X	X	X	X	X
Zinc	X	X	NA	NA	NA
PCBs[2]	X	X	X	X	X
PBDEs	NA	NA	NA	X[3]	NA
PCDD/Fs[4]	X	NA	NA	X	X
DDT and Metabolites[5]	X	X	X	X	X
LPAHs[6]	NA	X	NA	NA	NA
HPAH[6]	NA	X	NA	NA	NA
Acenaphthene	X	X	NA	NA	NA
Anthracene	X	X	NA	NA	X
Benzo(a) anthracene	X	X	NA	NA	X
Benzo(a)pyrene	X	X	NA	NA	X
Benzo(b) fluoranthene[7]	NA	X	NA	NA	X
Benzo(k) fluoranthene[7]	X	X	NA	NA	X
Chrysene	X	X	NA	NA	NA
Dibenzo(a,h) anthracene	X	X	NA	NA	X
Fluoranthene	X	X	NA	NA	X
Fluorene	X	X	NA	NA	X
Indeno(1,2,3)pyrene	X	X	NA	NA	X
Naphthalene	X	X	NA	NA	NA
Phenanthrene	X	X	NA	NA	NA
Pyrene	X	X	NA	NA	X
Bis(2-Ethylhexyl) Phthalate	X	X	NA	NA	X
Triclopyr	X	NA	NA	NA	NA
Nonylphenol	X	NA	NA	NA	NA
Petroleum - Heavy Fuel Oil	X	NA	NA	NA	NA
Petroleum - Diesel Fuel Oil	X	NA	NA	NA	NA
Petroleum - Gasoline	X	NA	NA	NA	NA
Petroleum - Lube Oil	X	NA	NA	NA	NA

NA - not evaluated in the hazard assessment for one or more reasons.

[1] Specifically the inorganic arsenic form was assessed for human health.

[2] Assessed as individual Aroclors and/or congeners or total PCBs.

[3] In the WOE discussion for PBDEs, limited effects data are discussed in the context of estimated daily doses for wildlife.

[4] Assessed as individual congeners, 2,3,7,8-TCDD or TEQs.

[5] Assessed as DDT, DDE, and DDD or as a sum.

[6] LPAHs and HPAHs are assessed as individual PAHs in all assessments except sediment.

[7] Assessed as part of total benzofluoranthenes in sediment assessment.

Table 10. Overall chemical groupings based on evaluation of all lines of evidence, including regional studies.

Multiple Priority 1 Classifications	Single Priority 1 Classifications		No Priority 1 Classifications
Copper	Arsenic*	Chrysene*	Lead*
Mercury*	Cadmium*	Fluoranthene*	Benzo(a)anthracene*
Zinc*	PBDEs*	Fluorene*	Triclopyr* ’
PCBs*	LPAHs	Naphthalene*	Nonylphenol*
Dioxins/Furans*	HPAHs	Phenanthrene*	Petroleum – Diesel*
DDT/DDE/DDD*	Anthracene*	Pyrene*	Petroleum – Heavy Fuel Oil*
Bis(2-ethylhexyl)phthalate*	Benzo(ghi)perylene*		Petroleum – Gasoline*
Benzo(a)pyrene*	Acenaphthene*		Petroleum – Lube Oil*
Dibenzo(ah)anthracene*	Acenaphthylene		
Indeno(123-cd)pyrene*			
Benzo(b)fluoranthene*			
Benzo(k)fluoranthene*			

Notes: COCs with an “*” were not prioritized in at least one media or pathway due to insufficient data (observed or effect data). It is important to note that not all COCs were evaluated in all media or pathways; the reader is encouraged to review Table 9 which summarizes the assessments that were conducted on each COC.

Uncertainty

While this hazard evaluation provides a broad general overview of the potential for the COCs evaluated to cause adverse ecological and human health effects, a number of uncertainties associated with the assessment process should be taken into consideration when interpreting the results. The following sections highlight the key uncertainties associated with the various elements of the assessment.

Environmental Data

This assessment included collection of a large number of environmental measurements for COCs in sediment, water and tissue from throughout the region. The EIM database likely includes the majority of observed ambient environmental data available for the Puget Sound region. Combined with the additional data included in this assessment, the dataset likely represents a reasonable representation of conditions in the Puget Sound region for many of the COCs. However, as previously indicated, there are likely data associated with special studies and research that have not been incorporated into readily accessible databases that were not included here.

Due to the broad nature of this assessment, the appropriateness of the analytical detection limits for the available environmental data were not evaluated. It is not anticipated that detection limits are a significant source of uncertainty for most COCs. However, for some COCs, such as PCB Aroclors® in surface waters, insufficient detection limits are a likely significant source of uncertainty. While there were a relatively large number of measurements for PCB Aroclors® (N>1200 in freshwater) the FOD was very low (3.5%). Based on a comparison to the much higher FOD for PCB congeners in water (58%), it is likely that Aroclor® measurements in water may represent an underestimate of the observed water concentration of this COC and PCBs as congeners better represents PCB concentrations and thus priority.

The assessment methodology is focused on COCs, which due to high concentrations (90th %ile) in some areas may pose a threat to Puget Sound. Including estimates of non-detected concentrations (e.g. detection limits, ½ detection limits, or zero) would result in lowering the 90th %ile for infrequently detected compounds, although it would not likely influence 90th %iles for frequently detected chemicals. Thus, commonly measured but rarely detected COCs such as DDT/DDD/DDE in water would be less likely to rank as Priority 1, while the rank of commonly detected COCs such as zinc in water would be unchanged. By considering only detected concentrations, some COCs with low FODs may be conservatively included in Priority 1.

For the Human Health priority classifications, the greatest source of uncertainty is associated with COCs that were not evaluated because there are no NTR criteria for these chemicals; the priority for these COCS is unknown. An additional uncertainty is associated with the exposure pathways that were not assessed here (e.g., dermal water exposures as well as air, inhalation, and dust exposures). These various other exposure pathways result in an additive exposure to COCs.

Ecological Data

The wildlife assessment required a number of ecological parameters to estimate daily doses of COC s for wildlife receptors. The most important of these parameters are the food and sediment ingestion rates because bioaccumulation is driven mainly by prey and sediment ingestion. Although a model was applied to estimate prey ingestion rates, the uncertainty associated with this parameter is small relative to the sediment ingestion rates. This is due to the significant relationship between body size and food ingestion rate. However, sediment ingestion is less predictable from body size and is more dependent on feeding strategies and foraging habitat. The total daily dose estimated using the upper and lower sediment ingestion rates for each receptor in the wildlife assessment demonstrated that there is little relative sensitivity to this parameter. Thus, the high uncertainty associated with sediment ingestion rates appears to have minimal impact on the results of this assessment.

Effects Data

Due to the variety of effects data used for this assessment there are a number of uncertainties that should be considered when interpreting the results. The water assessment relied primarily on the effect data obtained from the ECOTOX database. While an effort was made to screen out effects data that were inappropriate for use, the sheer volume of effect concentrations precluded a detailed review of these data. The QA/QC process used to evaluate the accuracy of the ECOTOX data base suggests that the use of these data is likely a source of uncertainty. It is unclear if the errors associated with the ECOTOX database would result in an over- or underestimate of the potential for effects to occur.

Some of the greatest uncertainty in the surface water assessment was associated with lack of effects data for some COCs, particularly for marine organisms and some dissolved metals. Uncertainties associated with the effects data used for the surface water assessment include, but are not limited to differences in the following variables: species sensitivity, exposure conditions (water quality - hardness and pH, light regime, temperature, feeding regime if any, chemical form of the COC and whether the test was static or flow through), test duration, appropriateness of endpoints evaluated and type of endpoint. These differences make comparability of effects data challenging at best.

An additional source of uncertainty was the lack of non-mortality based effects data for some COCs; this was especially an issue for some of the marine COCs. Only evaluating effects associated with COC exposure at concentrations that cause mortality may underestimate the potential for these COCs to cause more subtle impacts (e.g., growth, reproduction etc.) to some aquatic organisms. Uncertainty is also associated with the comparison of surface water concentrations of bioaccumulative COCs (PCBs, mercury, DDTs) to effects data based on direct water exposure only. This comparison does not account for indirect impacts associated with bioaccumulation; the potential for adverse effects is likely underestimated for these COCs. The reader is encouraged to evaluate the types of effects data available for each COC when drawing conclusions about these results.

Hardness can have a significant influence on the toxicity of metals in freshwater; however, due to the large volume of data evaluated here it was not practical to account for study specific hardness. Due to the large number of effects data that were available for most metals, it is not expected that this would have a significant impact on the outcome of the assessment. In general, hardness in the Puget Sound region tends to be low; therefore, it is unlikely that the effects data would have significantly underestimated metal toxicity.

The sediment assessment relied on a variety of established sediment guidelines thresholds. While these guidelines have been reviewed by others and some have undergone regulatory scrutiny, there are still some uncertainties associated with their use. For some COCs the concentration of OC and sulfides in the sediment can have a significant influence on bioavailability and toxicity. While some of the thresholds used here incorporated organic carbon, most did not account for site specific conditions that could influence bioavailability and toxicity. This assumption could have resulted in both an over- or under estimate of the potential for effects. The sediment guidelines were developed based on impacts to benthic organisms. Therefore, they do not provide a direct assessment of how sediment associated COCs can indirectly impact other aquatic organisms through bioaccumulation.

There are significant uncertainties associated with the effects data used for the tissue assessment. While use of tissue residues to assess toxicity can be a useful tool in some cases, care must be taken when using these data to estimate the potential for effects, particularly when used in a screening approach. In many cases, the available tissue residue data are not based on experiments designed to directly relate tissue residue to an effect and as a result they lack dose response data. An additional limitation results from the way much of the residue effects data are reported; unlike water based toxicity data which is reported in the context of a dose response, relatively little of the tissue residue data is reported as such (Meador et al. 2008).

For some organisms and COCs, lipid content can be an important factor in interpreting the toxic response. Due to the nature of this assessment and the limited availability of data, lipid content was not incorporated into the data interpretation.

In addition, there is also significant uncertainty associated with the number and type of tissue residue effect concentrations available for some COCs and tissue types; in some cases there were only one or two tissue residue effects levels available for a COC and tissue type. Some of the tissue residue effects data were limited to data only for mortality endpoints; which likely underestimated the prioritization for these COCs and tissue types. Typically, elevated tissue levels of bioaccumulative compounds will result in more subtle effects to growth or reproduction before they cause mortality. These factors contribute to the high level of uncertainty associated with the tissue residue assessment.

The petroleum effects concentrations were limited to fuel and lubricating oils. It is suspected that actual toxicity can vary substantially based on product additives, weathering, and phototoxicity. Additionally, the studies that served as a source of effects data utilized various water-product mixtures. For the purposes of this assessment only dissolved fractions were utilized. However, there is anecdotal evidence that product spills are common and sheens of product can often be found in the environment in selected areas. Waters with free product are potentially more toxic and/or present physical hazards to organisms. These issues create a high level of uncertainty for the petroleum assessment.

As previously discussed, the limited number of published effect doses for the wildlife receptors assessed here poses the greatest level of uncertainty in assessing potential effects in wildlife. In comparison to the volume of effects data available for aquatic life, the data available for wildlife may seem very weak; however, it should be noted that the dose-response studies for wildlife are frequently very comprehensive in examining effects. In comparison to a 30-day aquatic study that measures lethal doses, wildlife dose-response studies often include exposure over a reproductive cycle and examine everything from changes in adult body weight to egg size, time to hatch, female menses, and fertility of offspring. All these observations get lumped into broad categories. Considering the relative number of effects doses available for wildlife, the greatest uncertainty is associated with the dioxin/furan effects doses, particularly for birds.

Only about one-half of the COCs in this assessment were also listed in the NTR. The remaining chemicals were not evaluated, and thus pose a significant uncertainty regarding the potential for these COCs to cause human health effects. As previously discussed, none of the COCs were evaluated to estimate potential risk from water ingestion or dermal exposure. The same issue applies to sediment exposures, as there are no adopted and published standards for human exposure to sediments. Systemic human toxicity was also not evaluated for any of the COCs.

Data Gaps

It was assumed that the data identified for this assessment would be relatively representative of the region. However, due to the broad nature of this effort, the spatial distribution of data for a given COC within an assessment area (freshwater, near and offshore areas) is not well understood. This is a key gap in the current analytical approach when making broad generalizations regarding the priority on a sound wide basis.

For some matrices and spatial areas (e.g., freshwater metals where N was >3000) the assumption that the data are representative is likely true; however, data were limited for some COCs in some matrices and spatial areas (e.g., offshore COCs where many Ns were <20). This is not to suggest that thousands of measurements are necessary to provide a reasonable estimate of priority; in some cases other factors can be taken into account to increase the confidence of this assumption. For example, since the source of many COCs to offshore waters is via upland or nearshore inputs it can be reasonably assumed that if a COC is not Priority 1 in the nearshore it is not likely to be of Priority 1 in the offshore marine waters. There may be some exceptions to this logic; for example, bioaccumulative and hydrophobic chemicals tend to be present in freshwater streams at relatively low levels, but are subsequently biomagnified to high levels in upper trophic level organisms.

Potential gaps in data availability and spatial distribution should be taken into account when interpreting these data and using them to prioritize future efforts. A spatial analysis of existing data to identify regions or areas with high and low data densities would be a first step in understanding the magnitude and distribution of environmental data gaps.

As presented in the uncertainty discussion above, there were limited effects data for a number of COCs and matrices. This data gap is particularly significant for a number COCs in marine matrices. In water, there were limited effects data for many of the COCs evaluated; in some cases the available data were primarily based on mortality effects. This data gap limited the ability to fully assess the potential for these COCs to cause effects in marine waters and was compounded by the limited number of marine surface water measurements, particularly nearshore waters, for many COCs where the number of measurements was typically less than 15. Very few surface water PCB measurements in both marine and freshwaters were available; this limited the ability to fully assess this COC in this matrix.

In comparison to surface water, the sediment dataset (both effects and observed) was more robust. The most significant sediment data gap was the lack of multiple sediment guidelines for some COCs and a lack of nonylphenol environmental data. Only one guideline was available to evaluate nonylphenol in both marine and freshwater sediments. Only one set of guidelines were available for HPAH and LPAH and a number of individual PAHs in freshwater sediments and for dioxins/furans in both freshwater and marine sediments. Sediment guidelines were also lacking for triclopyr and PBDEs.

There were significant data gaps associated with the tissue assessment. Tissue residue effects data were limited for a number of the COCs and tissue types evaluated. In some cases there were less than five tissue residue effect values (e.g., mercury in marine tissues; PCBs in freshwater decapods and marine fish; dioxins/furans in all tissue types except freshwater fish; DDTs in marine tissues). There were very few measurements of dioxins/furans in tissue; no data were available for freshwater tissue types. There were limited PBDE tissue data and no readily available effects data for this COC. These data gaps limited the ability to fully assess the potential effects of these COCs.

As previously indicated, the lack of NTR criteria limited the number of COCs that could be evaluated for human health effects. Although there were invertebrate inorganic arsenic data, there were no inorganic arsenic data for freshwater or marine fish, which limited the ability to fully assess the human health priority for this COC. Additionally triclopyr, PBDEs and the sum of cPAHs are not part of the NTR and could not be prioritized.

The limited amount of effects data for PBDEs limited the ability to assess them in tissue and wildlife.

Recommendations for Further Assessment

- The spatial distribution of data used in this assessment was not assessed beyond the general habitat types (freshwater, marine near and offshore). The data for some COCS in all matrices are not likely evenly distributed throughout the region. As a result it is unknown if the data used in this assessment are truly representative of the region or only limited areas. This is of greatest concern for those COCs for which data were limited (see data gaps listed above). To fully understand if the priority identified for a COC applies on a regional basis, or is limited to a discrete area, additional data analysis that examines the spatial distribution of the data could be done. This analysis would provide additional information to focus future hazard reduction efforts to areas of greatest concern.

- Marine surface water data were limited for a number of COCs. Expansion of the number and type of parameters measured by ambient surface water monitoring programs would provide a means to fill this environmental data gap. The parameter list should focus on COCs that were Priority 1 in freshwaters; in addition to those likely to be bioaccumulated in the marine food web (e.g., nonylphenol, mercury, PCBs, HPAHs, PBDEs, and DDTs).

- Petroleum product data was not available for marine waters and the degree to which individual PAHs represent spilled and degraded petroleum products is unknown. Future investigations targeting a more complete suite of alkylated PAHS, other PAH and petroleum products, and their degradates could more fully characterize petroleum hazards.

- Sediment data for nonylphenol, particularly in marine sediment was limited. Sediment data for PCB congeners and PBDEs was also limited, particularly in marine nearshore areas. PCB Aroclor data is likely sufficient to identify priority; however additional PBDE and nonylphenol data would improve the ability to determine the priority for these COCs

- Tissue data for inorganic arsenic were very limited. Collection of inorganic arsenic data in edible seafood tissue would provide more certainty in determining human health priority for this COC.

- Effects data to assess surface waters were limited for a number of COCs, particularly for effects other than mortality. It is possible that additional data may be identified through a literature search and review for those COCs that are of greatest concern (this was beyond the scope of the current assessment). Additional effects data for some COCs would provide better certainty in determining the priority.

- Sediment guidelines were not available or limited for a number of COCs (PBDEs, dioxins, and nonylphenol). It is possible that additional effects data may be identified through a literature search. Additional effects data for these COCs would provide better certainty in determining priority.

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Appendix E. Glossary, Acronyms, and Abbreviations

Glossary

Ambient area: Un-impacted area; away from point sources of contamination.

Anthropogenic: Human-caused.

Aquatic life: Aquatic life refers to any organism which spends all of, the majority of, or significant portions of its life stage in water.

Attenuation: The reduction in the concentration, mass, toxicity, mobility, or volume of a chemical due to chemical, biological, or physical processes.

Baseflow: Groundwater discharge to a surface stream or river. The component of total streamflow that originates from direct groundwater discharges to a stream.

Basin: A drainage area or watershed in which all land and water areas drain or flow toward a central collector such as a stream, river, or lake at a lower elevation.

Benthic: Bottom-dwelling.

Bioaccumulative chemicals: Chemicals that build up and become concentrated in organisms.

Bivalves: Mollusks having a shell consisting of two hinged valves. Examples are clams, mussels, and oysters.

Built-up roof: A general term that can be applied to many flat, membrane, or torch-down roof types.

Catch basin: Large underground container for the collection of sediment and other debris from stormwater run-off. Designed to catch or collect the dirt and debris, and prevents it from entering surface water.

Congener: In chemistry, congeners are related chemicals. For example, polychlorinated biphenyls (PCBs) are a group of 209 related chemicals that are called congeners.

Constrained systems: Physical systems that generally do not allow chemicals to become mobilized in other environmental compartments or environmental pathways (e.g., sanitary sewer systems).

Dry season: In this study, May through September.

Effects data: In this study and specifically for the hazard evaluation, data from toxicity testing or derived otherwise to indicate chemical concentrations where effects are elicited.

Effluent: An outflowing of water from a natural body of water or from a man-made structure. For example, the treated outflow from a wastewater treatment plant.

Enrichment: Natural or human-caused enhancement of chemical concentrations in environmental media such as water or soils.

Exceeds (or is above) guidelines or standards: Does not meet guidelines or standards.

First flush: A rain event following an extended dry period. Runoff from a first flush can contain elevated concentrations of contaminants.

Flux: Amount that flows through a unit area in a unit of time.

Guilds: In this study, groups distinguished by their trophic level.

Hazard evaluation: A screening-level assessment of relative toxic hazards posed by chemicals at observed concentrations.

Land cover types: In this study, commercial/industrial, residential, agricultural, and forests.

Load pathways: In this study, groundwater, air deposition, surface water runoff, publicly-owned treatment works, and ocean exchange.

Loading: The input of pollutants into a waterbody.

Marine water: Saltwater

Micronutrient: Nutrients required by organisms at very low concentrations.

Nutrient: Substance such as carbon, nitrogen, and phosphorus used by organisms to live and grow. Too many nutrients in the water can promote algal blooms and rob the water of oxygen vital to aquatic organisms.

Observed data: In this study and specifically for the hazard evaluation, data on chemical concentrations in various media sampled since January 1, 2000.

Ocean boundary: Approximate area or space – theoretically defined by a vertical plane, a series of planes, or polygons – where Pacific Ocean waters are exchanged with the waters of Puget Sound.

Ocean exchange: The flux of Pacific Ocean waters and the waters of Puget Sound across the ocean boundary.

Organics: Shorthand for organic chemicals (those which contain carbon).

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

Pelagic: Open-water.

Personal care products (or toiletries): Products used for personal hygiene or beautification. Personal care includes products as diverse as chapstick, colognes, cotton swabs, deodorant, eye liner, facial tissue, hair clippers, lipstick, lotion, makeup, mouthwash, nail files, pomade, perfumes, personal lubricant, razors, shampoo, shaving cream, skin cream, toilet paper, cleansing pads and wipes, lip gloss, toothbrushes, and toothpaste, to give a few examples.

Piscivorous: Fish-eating.

Point source: Sources of pollution that discharge at a specific location from stacks, pipes, outfalls, or conveyance channels to a surface water.

Salmonid: Any fish that belong to the family *Salmonidae*. Basically, any species of salmon, trout, or char.

Sediment: Solid fragmented material (soil and organic matter) that is transported and deposited by water and covered with water (example, river or lake bottom).

Sill: A relatively shallow area of the seabed.

Source: For the purpose of the present project, the term *source* is strictly defined as: the object or activity from which a COC is initially released to environmental media (air, water, or soil) or released in a form which can be mobilized and transported in an environmental pathway.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snow melt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Study Area: The geographical study area for this *Assessments* study is Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits.

Surface runoff: In this study, surface runoff is broadly defined to include stormwater, nonpoint source overland flow, and groundwater discharge to surface waters that flow into marine waters.

Toxicant: Toxic contaminant.

Toxics: Shorthand for toxic chemical.

Water column: In a waterbody, a conceptual cylinder of water extending from the top of the sediment layer to the surface of the water.

Watershed: Basin. A drainage area in which all land and water areas drain or flow toward a central collector such as a stream, river, or lake at a lower elevation.

Wet season: In this study, October through April.

Acronyms and Abbreviations

a.k.a.	also known as
ABS	acrylonitrile butadiene styrene
AOP	Air Operating Permit
API	American Petroleum Institute or [King County] Asian and Pacific Islander
ATSDR	Agency for Toxic Substances and Disease Registry
BEE	butoxyethyl ester
CAP	Chemical Action Plan
CCA	chromated copper arsenate
COC	chemical of concern
cPAH	carcinogenic polycyclic aromatic hydrocarbon
CSL	cleanup screening level
DBP	di-n-butyl phthalate
DDT	dichlorodiphenyltrichloroethane
DEHP	di(2-ethylhexyl) phthalate [a.k.a. bis(2-ethylhexyl) phthalate]
DEP	diethyl phthalate
ECB	European Chemicals Bureau
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
Hg	mercury
HPAH	high-molecular weight polycyclic aromatic hydrocarbon
LAET	lowest apparent effects threshold
LCR	Lead and Copper Rule

LPAH	low-molecular weight polycyclic aromatic hydrocarbon
N	number
NOAA	National Oceanic Atmospheric Administration
NTR	National Toxics Rule
OC	organic carbon
PAH	polycyclic aromatic hydrocarbon
PBDE	polybrominated diphenyl ether
PBT	persistent, bioaccumulative, and toxic substance
PCB	polychlorinated biphenyl
PCDD/F	polychlorinated dibenzo-*p*-dioxin and dibenzofuran
PEC	probable effects concentration
PEL	probable effects level
POP	persistent organic pollutant
POTW	Publicly-owned treatment works
PSP	Puget Sound Partnership
PSTLA	Puget Sound Toxics Loading Analysis
PVC	polyvinyl chloride
SQS	Sediment Quality Standards
TEA	triethylamine
TEC	threshold effects concentration
TEL	thresholds effects level
TEQ	toxic equivalent
TRI	Toxics Release Inventory
UC	University of California
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WDFW	Washington Department of Fish and Wildlife
WDOH	Washington State Department of Health
WQC	water quality criteria

Units of Measurement

°C	degrees Celsius
d	day
dw	dry weight
g	gram, a unit of mass
kg	kilograms, a unit of mass equal to 1,000 grams.
kg/d	kilograms per day
km	kilometer, a unit of length equal to 1,000 meters.
m	meter
mg	milligrams
mg/kg	milligrams per kilogram (parts per million)
mg/l	milligrams per liter (parts per million)
ng/g	nanograms per gram (parts per billion)
ng/kg	nanograms per kilogram (parts per trillion)
ng/l	nanograms per liter (parts per trillion)
pg/l	picograms per liter (parts per quadrillion)

t	tonne (metric ton, equal to 1,000 kg); appr. 2.2 tons
t/yr	tonnes (metric tons) per year
ug/kg	micrograms per kilogram (parts per billion)
ug/l	micrograms per liter (parts per billion)
ww	wet weight

Treatment Technology Review and Assessment

Association of Washington Business
Association of Washington Cities
Washington State Association of Counties

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Acronyms

Acronym	Definition
AACE	Association for the Advancement of Cost Engineering
AOP	advanced oxidation processes
AWB	Association of Washington Businesses
BAC	biological activated carbon
BAP	benzo(a)pyrene
BOD	biochemical oxygen demand
BTU	British thermal unit
CEPT	Chemically-enhanced primary treatment
cf	cubic feet
CIP	clean in place
CRITFC	Columbia River Inter-Tribal Fish Commission
Ecology	Washington Department of Ecology
EPA	U.S. Environmental Protection Agency
FCR	fish consumption rate
g/day	grams per day
GAC	granular activated carbon
gal	gallon
gfd	gallons per square foot per day
GHG	greenhouse gas
gpd	gallons per day
gpm	gallons per minute
GWh	giga watt hours
HDR	HDR Engineering, Inc.
HHWQC	human health water quality criteria
HRT	hydraulic residence time
IPCC	Intergovernmental Panel on Climate Change
kg	kilogram
KWh/MG	kilowatt-hours per million gallons
lb	pound
MBR	membrane bioreactor
MCL	maximum contaminant level
MF	microfiltration
mgd	million gallons per day
mg/L	milligrams per liter
MMBTU	million British thermal units
MWh/d	megawatt-hours per day
NF	nanofiltration
ng/L	nanograms per liter
NPDES	National Pollutant Discharge Elimination System
NPV	net present value
O&M	operations and maintenance
ODEQ	Oregon Department of Environmental Quality
PAC	powdered activated carbon
PAH	polycyclic aromatic hydrocarbons
PCB	polychlorinated biphenyls
PE	population equivalents
PIX	potable ion exchange

Acronym	Definition
ppm	parts per million
RO	reverse osmosis
SDWA	Safe Drinking Water Act
sf	square feet
SGSP	salinity gradient solar pond
SRT	solids retention time
Study Partners	Association of Washington Businesses/Association of Washington Cities and Washington State Association of Counties consortium
TDS	total dissolved solids
TMDL	total maximum daily load
TSS	total suspended solids
UF	ultrafiltration
µg/L	micrograms per liter
USDA	U.S. Department of Agriculture
UV	ultraviolet
WAC	Washington Administrative Code
WAS	waste activated sludge
WLA	waste load allocation
WWTP	wastewater treatment plant
ZLD	zero liquid discharge

Executive Summary

This study evaluated treatment technologies potentially capable of meeting the State of Washington Department of Ecology's (Ecology) revised effluent discharge limits associated with revised human health water quality criteria (HHWQC). HDR Engineering, Inc. (HDR) completed a literature review of potential technologies and an engineering review of their capabilities to evaluate and screen treatment methods for meeting revised effluent limits for four constituents of concern: arsenic, benzo(a)pyrene (BAP), mercury, and polychlorinated biphenyls (PCBs). HDR selected two alternatives to compare against an assumed existing baseline secondary treatment system utilized by dischargers. These two alternatives included enhanced secondary treatment with membrane filtration/reverse osmosis (MF/RO) and enhanced secondary treatment with membrane filtration/granulated activated carbon (MF/GAC). HDR developed capital costs, operating costs, and a net present value (NPV) for each alternative, including the incremental cost to implement improvements for an existing secondary treatment facility.

Currently, there are no known facilities that treat to the HHWQC and anticipated effluent limits that are under consideration. Based on the literary review, research, and bench studies, the following conclusions can be made from this study:

- Revised HHWQC based on state of Oregon HHWQC (2001) and U.S. Environmental Protection Agency (EPA) "National Recommended Water Quality Criteria" will result in very low water quality criteria for toxic constituents.
- There are limited "proven" technologies available for dischargers to meet required effluent quality limits that would be derived from revised HHWQC.
    - Current secondary wastewater treatment facilities provide high degrees of removal for toxic constituents; however, they are not capable of compliance with water quality-based National Pollutant Discharge Elimination System (NPDES) permit effluent limits derived from the revised HHWQC.
    - Advanced treatment technologies have been investigated and candidate process trains have been conceptualized for toxics removal.
        - Advanced wastewater treatment technologies may enhance toxics removal rates; however, they will not be capable of compliance with HHWQC-based effluent limits for PCBs. The lowest levels achieved based on the literature review were between <0.00001 and 0.00004 micrograms per liter (µg/L), as compared to a HHWQC of 0.0000064 µg/L.
        - Based on very limited performance data for arsenic and mercury from advanced treatment information available in the technical literature, compliance with revised criteria may or may not be possible, depending upon site specific circumstances.
            - Compliance with a HHWQC for arsenic of 0.018 µg/L appears unlikely. Most treatment technology performance information available in the literature is based on drinking water treatment applications targeting a much higher Safe Drinking Water Act (SDWA) maximum contaminant level (MCL) of 10 µg/L.
            - Compliance with a HHWQC for mercury of 0.005 µg/L appears to be potentially attainable on an average basis, but perhaps not if effluent limits are structured on a maximum monthly, maximum weekly or maximum daily basis. Some secondary treatment facilities attain average effluent mercury levels of 0.009 to 0.066 µg/L. Some treatment facilities with effluent filters attain average effluent mercury levels of 0.002 to 0.010 µg/L. Additional

        advanced treatment processes are expected to enhance these removal rates, but little mercury performance data is available for a definitive assessment.

    - Little information is available to assess the potential for advanced technologies to comply with revised BAP criteria. A municipal wastewater treatment plant study reported both influent and effluent BAP concentrations less than the HHWQC of 0.0013 ug/L (Ecology, 2010).
  - Some technologies may be effective at treating identified constituents of concern to meet revised limits while others may not. It is therefore even more challenging to identify a technology that can meet all constituent limits simultaneously.
  - A HHWQC that is one order-of-magnitude less stringent could likely be met for mercury and BAP; however, it appears PCB and arsenic limits would not be met.
- Advanced treatment processes incur significant capital and operating costs.
  - Advanced treatment process to remove additional arsenic, BAP, mercury, and PCBs would combine enhancements to secondary treatment with microfiltration membranes and reverse osmosis or granular activated carbon and increase the estimated capital cost of treatment from $17 to $29 in dollars per gallon per day of capacity (based on a 5.0-million-gallon-per-day (mgd) facility).
  - The annual operation and maintenance costs for the advanced treatment process train will be substantially higher (approximately $5 million - $15 million increase for a 5.0 mgd capacity facility) than the current secondary treatment level.
- Implementation of additional treatment will result in additional collateral impacts.
  - High energy consumption.
  - Increased greenhouse gas emissions.
  - Increase in solids production from chemical addition to the primaries. Additionally, the membrane and GAC facilities will capture more solids that require handling.
  - Increased physical space requirements at treatment plant sites for advanced treatment facilities and residuals management including reverse osmosis reject brine processing.
- It appears advanced treatment technology alone cannot meet all revised water quality limits and implementation tools are necessary for discharger compliance.
  - Implementation flexibility will be necessary to reconcile the difference between the capabilities of treatment processes and the potential for HHWQC driven water quality based effluent limits to be lower than attainable with technology

Table ES-1 indicates that the unit NPV cost for baseline conventional secondary treatment ranges from $13 to $28 per gallon per day of treatment capacity. The unit cost for the advanced treatment alternatives increases the range from the low $20s to upper $70s on a per gallon per-day of treatment capacity. The resulting unit cost for improving from secondary treatment to advanced treatment ranges between $15 and $50 per gallon per day of treatment capacity. Unit costs were also evaluated for both a 0.5 and 25 mgd facility. The range of unit costs for improving a 0.5 mgd from secondary to advanced treatment is $60 to $162 per gallon per day of treatment capacity. The range of unit costs for improving a 25 mgd from secondary to advanced treatment is $10 to $35 per gallon per day of treatment capacity.

Table ES-1. Treatment Technology Costs in 2013 Dollars for a 5-mgd Facility

Alternative	Total Construction Cost, 2013 dollars ($ Million)	O&M Net Present Value, 2013 dollars ($ Million)***	Total Net Present Value, 2013 dollars ($ Million)	NPV Unit Cost, 2013 dollars ($/gpd)
Baseline (Conventional Secondary Treatment)*	59 - 127	5 - 11	65 - 138	13 - 28
Incremental Increase to Advanced Treatment - MF/RO	48 - 104	26 - 56	75 - 160	15 - 32
Advanced Treatment - MF/RO**	108 - 231	31 - 67	139 - 298	28 - 60
Incremental Increase to Advanced Treatment - MF/GAC	71 - 153	45 - 97	117 - 250	23 - 50
Advanced Treatment - MF/GAC	131 - 280	50 - 108	181 - 388	36 - 78

* Assumed existing treatment for dischargers. The additional cost to increase the SRT to upwards of 30-days is about $12 - 20 million additional dollars in total project cost for a 5 mgd design flow.

** Assumes zero liquid discharge for RO brine management, followed by evaporation ponds. Other options are available as listed in Section 4.4.2.

*** Does not include the cost for labor.

mgd=million gallons per day
MG=million gallons
MF/RO=membrane filtration/reverse osmosis
MF/GAC=membrane filtration/granulated activated carbon
O&M=operations and maintenance
Net Present Value = total financed cost assuming a 5% nominal discount rate over an assumed 25 year equipment life.

Costs presented above are based on a treatment capacity of 5.0 mgd, however, existing treatment facilities range dramatically across Washington in size and flow treated. The key differences in cost between the baseline and the advanced treatment MF/RO are as follows:

- Larger aeration basins than the baseline to account for the longer SRT (>8 days versus <8 days).
- Additional pumping stations to pass water through the membrane facilities and granulated activated carbon facilities. These are based on peak flows.
- Membrane facilities (equipment, tanks chemical feed facilities, pumping, etc.) and replacement membrane equipment.
- Granulated activated carbon facilities (equipment, contact tanks, pumping, granulated activated carbon media, etc.)
- Additional energy and chemical demand to operate the membrane and granulated activated carbon facilities
- Additional energy to feed and backwash the granulated activated carbon facilities.
- Zero liquid discharge facilities to further concentrate the brine reject.
    - o Zero liquid discharge facilities are energy/chemically intensive and they require membrane replacement every few years due to the brine reject water quality.
- Membrane and granulated activated carbon media replacement represent a significant maintenance cost.

- Additional hauling and fees to regenerate granulated activated carbon off-site.

The mass of pollutant removal by implementing advanced treatment was calculated based on reducing current secondary effluent discharges to revised effluent limits for the four pollutants of concern. These results are provided in Table ES-2 as well as a median estimated unit cost basis for the mass of pollutants removed.

Table ES-2. Unit Cost by Contaminant for a 5-mgd Facility Implementing Advanced Treatment using Membrane Filtration/Reverse Osmosis

Component	PCBs	Mercury	Arsenic	BAPs
Required HHWQC based Effluent Quality (µg/L)	0.0000064	0.005	0.018	0.0013
Current Secondary Effluent Concentration (µg/L)	0.002	0.025	7.5	0.006
Total Mass Removed (lbs) over 25 year Period	0.76	7.6	2,800	1.8
Median Estimated Unit Cost (NPV per total mass removed in pounds over 25 years)	$290,000,000	$29,000,000	$77,000	$120,000,000

µg/L=micrograms per liter

lbs=pounds

NPV=net present value

Collateral adverse environmental impacts associated with implementing advanced treatment were evaluated. The key impacts from this evaluation include increased energy use, greenhouse gas production, land requirements and treatment residuals disposal. Operation of advanced treatment technologies could increase electrical energy by a factor of 2.3 to 4.1 over the baseline secondary treatment system. Direct and indirect greenhouse gas emission increases are related to the operation of advanced treatment technologies and electrical power sourcing, with increases of at least 50 to 100 percent above the baseline technology. The energy and air emission implications of advanced treatment employing granulated activated carbon construction of advanced treatment facilities will require additional land area. The availability and cost of land adjacent to existing treatment facilities has not been included in cost estimates, but could be very substantial. It is worthwhile noting residual materials from treatment may potentially be hazardous and their disposal may be challenging to permit. Costs assume zero liquid discharge from the facilities.

1.0 Introduction

Washington's Department of Ecology (Ecology) has an obligation to periodically review waterbody "designated uses" and to modify, as appropriate, water quality standards to ensure those uses are protected. Ecology initiated this regulatory process in 2009 for the human health-based water quality criteria (HHWQC) in Washington's *Surface Water Quality Standards* (Washington Administrative Code [WAC] 173-201A). HHWQC are also commonly referred to as "toxic pollutant water quality standards." Numerous factors will influence Ecology's development of HHWQC. The expectation is that the adopted HHWQC will be more stringent than current adopted criteria. National Pollutant Discharge Elimination System (NPDES) effluent limits for permitted dischargers to surface waters are based on U.S. Environmental Protection Agency (EPA) and state guidance. Effluent limits are determined primarily from reasonable potential analyses and waste load allocations (WLAs) from total maximum daily loads (TMDLs), although the permit writer may use other water quality data. Water quality-based effluent limits are set to be protective of factors, including human health, aquatic uses, and recreational uses. Therefore, HHWQC can serve as a basis for effluent limits. The presumption is that more stringent HHWQC will, in time, drive lower effluent limits. The lower effluent limits will require advanced treatment technologies and will have a consequent financial impact on NPDES permittees. Ecology anticipates that a proposed revision to the water quality standards regulation will be issued in first quarter 2014, with adoption in late 2014.

The Association of Washington Businesses (AWB) is recognized as the state's chamber of commerce, manufacturing and technology association. AWB members, along with the Association of Washington Cities and Washington State Association of Counties (collectively referred to as Study Partners), hold NPDES permits authorizing wastewater discharges. The prospect of more stringent HHWQC, and the resulting needs for advanced treatment technologies to achieve lower effluent discharge limits, has led this consortium to sponsor a study to assess technology availability and capability, capital and operations and maintenance (O&M) costs, pollutant removal effectiveness, and collateral environmental impacts of candidate technologies.

The "base case" for the study began with the identification of four nearly ubiquitous toxic pollutants present in many industrial and municipal wastewater discharges, and the specification of pollutant concentrations in well-treated secondary effluent. The pollutants are arsenic, benzo(a)pyrene (BAP), mercury and polychlorinated biphenyls (PCBs), which were selected for review based on available monitoring data and abundant presence in the environment. The purpose of this study is to review the potential water quality standards and associated treatment technologies able to meet those standards for four pollutants.

A general wastewater treatment process and wastewater characteristics were used as the common baseline for comparison with all of the potential future treatment technologies considered. An existing secondary treatment process with disinfection at a flow of 5 million gallons per day (mgd) was used to represent existing conditions. Typical effluent biochemical oxygen demand (BOD) and total suspended solids (TSS) were assumed between 10 and 30 milligrams per liter (mg/L) for such a facility and no designed nutrient or toxics removal was assumed for the baseline existing treatment process.

Following a literature review of technologies, two advanced treatment process options for toxics removal were selected for further evaluation based on the characterization of removal effectiveness from the technical literature review and Study Partners' preferences. The two tertiary treatment options are microfiltration membrane filtration (MF) followed by either reverse osmosis (RO) or granular activated carbon (GAC) as an addition to an existing secondary treatment facility.

The advanced treatment technologies are evaluated for their efficacy and cost to achieve the effluent limitations implied by the more stringent HHWQC. Various sensitivities are examined, including for less stringent adopted HHWQC, and for a size range of treatment systems. Collateral environmental impacts associated with the operation of advanced technologies are also qualitatively described.

2.0 Derivation of the Baseline Study Conditions and Rationale for Selection of Effluent Limitations

2.1 Summary of Water Quality Criteria

Surface water quality standards for toxics in the State of Washington are being updated based on revised human fish consumption rates (FCRs). The revised water quality standards could drive very low effluent limitations for industrial and municipal wastewater dischargers. Four pollutants were selected for study based on available monitoring data and abundant presence in the environment. The four toxic constituents are arsenic, BAP, mercury, and PCBs.

2.2 Background

Ecology is in the process of updating the HHWQC in the state water quality standards regulation. Toxics include metals, pesticides, and organic compounds. The human health criteria for toxics are intended to protect people who consume water, fish, and shellfish. FCRs are an important factor in the derivation of water quality criteria for toxics.

The AWB/City/County consortium (hereafter “Study Partners”) has selected four pollutants for which more stringent HHWQC are expected to be promulgated. The Study Partners recognize that Ecology probably will not adopt more stringent arsenic HHWQC so the evaluation here is based on the current arsenic HHWQC imposed by the National Toxics Rule. Available monitoring information indicates these pollutants are ubiquitous in the environment and are expected to be present in many NPDES discharges. The four pollutants include the following:

- Arsenic
    - Elemental metalloid that occurs naturally and enters the environment through erosion processes. Also widely used in batteries, pesticides, wood preservatives, and semiconductors. Other current uses and legacy sources in fungicides/herbicides, copper smelting, paints/dyes, and personal care products.
- Benzo(a)pyrene (BAP)
    - Benzo(a)pyrene is a polycyclic aromatic hydrocarbon formed by a benzene ring fused to pyrene as the result of incomplete combustion. Its metabolites are highly carcinogenic. Sources include wood burning, coal tar, automobile exhaust, cigarette smoke, and char-broiled food.
- Mercury
    - Naturally occurring element with wide legacy uses in thermometers, electrical switches, fluorescent lamps, and dental amalgam. Also enters the environment through erosion processes, combustion (especially coal), and legacy industrial/commercial uses. Methylmercury is an organometallic that is a bioaccumulative toxic. In aquatic systems, an anaerobic methylation process converts inorganic mercury to methylmercury.
- Polychlorinated Biphenyls (PCBs)
    - Persistent organic compounds historically used as a dielectric and coolant in electrical equipment and banned from production in the U.S. in 1979. Available information indicates continued pollutant loadings to the environment as a byproduct from the use of some pigments, paints, caulking, motor oil, and coal combustion.

2.3 Assumptions Supporting Selected Ambient Water Quality Criteria and Effluent Limitations

Clean Water Act regulations require NPDES permittees to demonstrate their discharge will “not cause or contribute to a violation of water quality criteria.” If a “reasonable potential analysis” reveals the possibility of a standards violation, the permitting authority is obliged to develop “water quality-based effluent limits” to ensure standards achievement. In addition, if ambient water quality monitoring or fish tissue assessments reveal toxic pollutant concentrations above HHWQC levels, Ecology is required to identify that impairment (“303(d) listing”) and develop corrective action plans to force reduction in the toxic pollutant discharge or loading of the pollutant into the impaired water body segment. These plans, referred to as total maximum daily loads (TMDLs) or water cleanup plans, establish discharge allocations and are implemented for point discharge sources through NPDES permit effluent limits and other conditions.

The effect of more stringent HHWQC will intuitively result in more NPDES permittees “causing or contributing” to a water quality standards exceedance, and/or more waterbodies being determined to be impaired, thus requiring 303(d) listing, the development of TMDL/water cleanup plans, and more stringent effluent limitations to NPDES permittees whose treated wastewater contains the listed toxic pollutant.

The study design necessarily required certain assumptions to create a “baseline effluent scenario” against which the evaluation of advanced treatment technologies could occur. The Study Partners and HDR Engineering, Inc (HDR) developed the scenario. Details of the baseline effluent scenario are presented in Table 1. The essential assumptions and rationale for selection are presented below:

- Ecology has indicated proposed HHWQC revisions will be provided in first quarter 2014. A Study Partners objective was to gain an early view on the treatment technology and cost implications. Ecology typically allows 30 or 45 days for the submission of public comments on proposed regulations. To wait for the proposed HHWQC revisions would not allow sufficient time to complete a timely technology/cost evaluation and then to share the study results in the timeframe allowed for public involvement/public comments.
- Coincident with the issuance of the proposed regulation, Ecology has a statutory obligation to provide a Significant Legislative Rule evaluation, one element of which is a “determination whether the probable benefits of the rule are greater than its probable costs, taking into account both the qualitative and quantitative benefits and costs and the specific directives of the statute being implemented” (RCW 34.05.328(1)(d)). A statutory requirement also exists to assess the impact of the proposed regulation to small businesses. The implication is that Ecology will be conducting these economic evaluations in fourth quarter 2013 and early 2014. The Study Partners wanted to have a completed technology/cost study available to share with Ecology for their significant legislative rule/small business evaluations.
- The EPA, Indian tribes located in Washington, and various special interest groups have promoted the recently promulgated state of Oregon HHWQC (2011) as the “model” for Washington’s revisions of HHWQC. The Oregon HHWQC are generally based on a increased FCR of 175 grams per day (g/day) and an excess cancer risk of 10^{-6}. While the Study Partners do not concede the wisdom or appropriateness of the Oregon criteria, or the selection of scientific/technical elements used to derive those criteria, the Study Partners nevertheless have selected the Oregon HHWQC as a viable “starting point” upon which this study could be based.

- The scenario assumes generally that Oregon's HHWQC for ambient waters will, for some parameters in fact, become effluent limitations for Washington NPDES permittees. The reasoning for this important assumption includes:
    - The state of Washington's NPDES permitting program is bound by the *Friends of Pinto Creek vs. EPA* decision in the United States Court of Appeals for the Ninth Circuit (October 4, 2007). This decision held that no NPDES permits authorizing new or expanded discharges of a pollutant into a waterbody identified as impaired; i.e., listed on CWA section 303(d), for that pollutant, may be issued until such time as "existing dischargers" into the waterbody are "subject to compliance schedules designed to bring the (waterbody) into compliance with applicable water quality standards." In essence, any new/expanded discharge of a pollutant causing impairment must achieve the HHWQC at the point of discharge into the waterbody.
    - If a waterbody segment is identified as "impaired" (i.e., not achieving a HHWQC), then Ecology will eventually need to produce a TMDL or water cleanup plan. For an existing NPDES permittee with a discharge of the pollutant for which the receiving water is impaired, the logical assumption is that any waste load allocation granted to the discharger will be at or lower than the numeric HHWQC (to facilitate recovery of the waterbody to HHWQC attainment). As a practical matter, this equates to an effluent limit established at the HHWQC.
    - Acceptance of Oregon HHWQC as the baseline for technology/cost review also means acceptance of practical implementation tools used by Oregon. The HHWQC for mercury is presented as a fish tissue methyl mercury concentration. For the purposes of NPDES permitting, however, Oregon has developed an implementation management directive which states that any confirmed detection of mercury is considered to represent a "reasonable potential" to cause or contribute to a water quality standards violation of the methyl mercury criteria. The minimum quantification level for total mercury is presented as 0.005 micrograms per liter (µg/L) (5.0 nanograms per liter (ng/L)).
    - The assumed effluent limit for arsenic is taken from EPA's *National Recommended Water Quality Criteria* (2012) (inorganic, water and organisms, 10^{-6} excess cancer risk). Oregon's 2011 criterion is actually based on a less protective excess cancer risk (10^{-4}). This, however, is the result of a state-specific risk management choice and it is unclear if Washington's Department of Ecology would mimic the Oregon approach.
    - The assumption is that no mixing zone is granted such that HHWQC will effectively serve as NPDES permit effluent limits. Prior discussion on the impact of the Pinto Creek decision, 303(d) impairment and TMDL Waste Load Allocations processes, all lend support to this "no mixing zone" condition for the parameters evaluated in this study.
- Consistent with Ecology practice in the evaluation of proposed regulations, the HHWQC are assumed to be in effect for a 20-year period. It is assumed that analytical measurement technology and capability will continue to improve over this time frame and this will result in the detection and lower quantification of additional HHWQC in ambient water and NPDES dischargers. This knowledge will trigger the Pinto Creek/303(d)/TMDL issues identified above and tend to pressure NPDES permittees to evaluate and install advanced treatment technologies. The costs and efficacy of treatment for these additional HHWQC is unknown at this time.

Other elements of the Study Partners work scope, as presented to HDR, must be noted:

- The selection of four toxic pollutants and development of a baseline effluent scenario is not meant to imply that each NPDES permittee wastewater discharge will include those pollutants at the assumed concentrations. Rather, the scenario was intended to represent a composite of many NPDES permittees and to facilitate evaluation of advanced treatment technologies relying on mechanical, biological, physical, chemical processes.
- The scalability of advanced treatment technologies to wastewater treatment systems with different flow capacities, and the resulting unit costs for capital and O&M, is evaluated.
- Similarly, a sensitivity analysis on the unit costs for capital and O&M was evaluated on the assumption the adopted HHWQC (and effectively, NPDES effluent limits) are one order-of-magnitude less stringent than the Table 1 values.

Table 1: Summary of Effluent Discharge Toxics Limits

Constituent	Human Health Criteria based Limits to be met with no Mixing Zone (µg/L)	Basis for Criteria	Typical Concentration in Municipal Secondary Effluent (µg/L)	Typical Concentration in Industrial Secondary Effluent (µg/L)	Existing Washington HHC (water + org.), NTR (µg/L)
PCBs	0.0000064	Oregon Table 40 Criterion (water + organisms) at FCR of 175 grams/day	0.0005 to 0.0025[b,c,d,e,f]	0.002 to 0.005[i]	0.0017
Mercury	0.005	DEQ IMD[a]	0.003 to 0.050[h]	0.010 to 0.050[h]	0.140
Arsenic	0.018	EPA National Toxics Rule (water + organisms)[k]	0.500 to 5.0[j]	10 to 40[j]	0.018
Benzo(a)Pyrene	0.0013	Oregon Table 40 Criterion (water + organisms) at FCR of 175 grams/day	0.00028 to 0.006[b,g]	0.006 to1.9	0.0028

[a] Oregon Department of Environmental Quality (ODEQ). Internal Management Directive: Implementation of Methylmercury Criterion in NPDES Permits. January 8, 2013.

[b] Control of Toxic Chemicals in Puget Sound, Summary Technical Report for Phase 3: Loadings from POTW Discharge of Treated Wastewater, Washington Department of Ecology, Publication Number 10-10-057, December 2010.

[c] Spokane River PCB Source Assessment 2003-2007, Washington Department of Ecology, Publication No. 11-03-013, April 2011.

[d] Lower Okanogan River Basin DDT and PCBs Total Maximum Daily Load, Submittal Report, Washington Department of Ecology, Publication Number 04-10-043, October 2004.

[e] Palouse River Watershed PCB and Dieldrin Monitoring, 2007-2008, Wastewater Treatment Plants and Abandoned Landfills, Washington Department of Ecology, Publication No. 09-03-004, January 2009

[f] A Total Maximum Daily Load Evaluation for Chlorinated Pesticides and PCBs in the Walla Walla River, Washington Department of Ecology, Publication No. 04-03-032, October 2004.

[g] Removal of Polycyclic Aromatic Hydrocarbons and Heterocyclic Nitrogenous Compounds by A POTW Receiving Industrial Discharges, Melcer, H., Steel, P. and Bedford, W.K., Water Environment Federation, 66th Annual Conference and Exposition, October 1993.

[h] Data provided by Lincoln Loehr's summary of WDOE Puget Sound Loading data in emails from July 19, 2013.

[i] NCASI memo from Larry Lefleur, NCASI, to Llewellyn Matthews, NWPPA, revised June 17, 2011, summarizing available PCB monitoring data results from various sources.

[j] Professional judgment, discussed in August 6, 2013 team call.

[k] The applicable Washington Human Health Criteria cross-reference the EPA National Toxics Rule, 40 CFR 131.36. The EPA arsenic HHC is 0.018 ug/L for water and organisms.

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3.0 Wastewater Characterization Description

This section describes the wastewater treatment discharge considered in this technology evaluation. Treated wastewater characteristics are described, including average and peak flow, effluent concentrations, and toxic compounds of concern.

3.1 Summary of Wastewater Characterization

A general wastewater treatment process and wastewater characteristics were developed as the common baseline to represent the existing conditions as a starting point for comparison with potential future advanced treatment technologies and improvements. A secondary treatment process with disinfection at a flow of 5 mgd as the current, baseline treatment system for existing dischargers was also developed. Typical effluent biochemical oxygen demand (BOD) and total suspended solids (TSS) were assumed between 10 to 30 mg/L from such a facility and no nutrient or toxics removal was assumed to be accomplished in the existing baseline treatment process.

3.2 Existing Wastewater Treatment Facility

The first step in the process is to characterize the existing wastewater treatment plant to be evaluated in this study. The goal is to identify the necessary technology that would need to be added to an existing treatment facility to comply with revised toxic pollutant effluent limits. Rather than evaluating the technologies and costs to upgrade multiple actual operating facilities, the Study Partners specified that a generalized municipal/industrial wastewater treatment facility would be characterized and used as the basis for developing toxic removal approaches. General characteristics of the facility's discharge are described in Table 2.

Table 2. General Wastewater Treatment Facility Characteristics

Average Annual Wastewater Flow, mgd	Maximum Month Wastewater Flow, mgd	Peak Hourly Wastewater Flow, mgd	Effluent BOD, mg/L	Effluent TSS, mg/L
5.0	6.25	15.0	10 to 30	10 to 30

mgd=million gallons per day
mg/L=milligrams per liter
BOD=biochemical oxygen demand
TSS=total suspended solids

In the development of the advanced treatment technologies presented below, the capacity of major treatment elements are generally sized to accommodate the maximum month average wastewater flow. Hydraulic elements, such as pumps and pipelines, were selected to accommodate the peak hourly wastewater flow.

The general treatment facility incorporates a baseline treatment processes including influent screening, grit removal, primary sedimentation, suspended growth biological treatment (activated sludge), secondary clarification, and disinfection using chlorine. Solids removed during primary treatment and secondary clarification are assumed to be thickened, stabilized, dewatered, and land applied to agricultural land. The biological treatment process is assumed to be activated sludge with a relatively short (less than 10-day) solids retention time. The baseline secondary treatment facility is assumed not to have processes dedicated to removing nutrients or toxics. However, some coincident removal of toxics will occur during conventional treatment.

3.3 Toxic Constituents

As described in Section 2.3, the expectation of more stringent HHWQC will eventually trigger regulatory demands for NPDES permittees to install advanced treatment technologies. The Study Group and HDR selected four specific toxic pollutants reflecting a range of toxic constituents as the basis for this study to limit the constituents and technologies to be evaluated to a manageable level.

The four toxic pollutants selected were PCBs, mercury, arsenic, and BAP, a polycyclic aromatic hydrocarbon (PAH). Mercury and arsenic are metals, and PCBs and PAHs are organic compounds. Technologies for removing metals and organic compounds are in some cases different. Key information on each of the compounds, including a description of the constituent, the significance of each constituent, proposed HHWQC, basis for the proposed criteria, typical concentration in both municipal and industrial secondary effluent, and current Washington state water quality criteria, are shown in Table 1. It is assumed that compliance with the proposed criteria in the table would need to be achieved at the “end of pipe” and Ecology would not permit a mixing zone for toxic constituents. This represents a “worst–case,” but a plausible assumption about discharge conditions.

4.0 Treatment Approaches and Costs

4.1 Summary of Treatment Approach and Costs

Two advanced treatment process options for toxics removal for further evaluation based on the characterization of removal effectiveness from the technical literature review and Study Group preferences. The two tertiary treatment options are microfiltration MF followed by either RO or GAC as an addition to an existing secondary treatment facility. Based on the literature review, it is not anticipated that any of the treatment options will be effective in reducing all of the selected pollutants to below the anticipated water quality criteria. A summary of the capital and operations and maintenance costs for tertiary treatment is provided, as well as a comparison of the adverse environmental impacts for each alternative.

4.2 Constituent Removal – Literature Review

The evaluation of treatment technologies relevant to the constituents of concern was initiated with a literature review. The literature review included a desktop search using typical web-based search engines, and search engines dedicated to technical and research journal databases. At the same time, HDR's experience with the performance of existing treatment technologies specifically related to the four constituents of concern, was used in evaluating candidate technologies. A summary of the constituents of concern and relevant treatment technologies is provided in the following literature review section.

4.2.1 Polychlorinated Biphenyls

PCBs are persistent organic pollutants that can be difficult to remove in treatment. PCB treatment in wastewater can be achieved using oxidation with peroxide, filtration, biological treatment or a combination of these technologies. There is limited information available about achieving ultra-low effluent PCB concentrations near the 0.0000064 µg/L range under consideration in the proposed rulemaking process. This review provides a summary of treatment technology options and anticipated effluent PCB concentrations.

Research on the effectiveness of ultraviolet (UV) light and peroxide on removing PCBs was tested in bench scale batch reactions (Yu, Macawile, Abella, & Gallardo 2011). The combination of UV and peroxide treatment achieved PCB removal greater than 89 percent, and in several cases exceeding 98 percent removal. The influent PCB concentration for the batch tests ranged from 50 to 100 micrograms per liter (µg/L). The final PCB concentration (for the one congener tested) was <10 µg/L (10,000 ng/L) for all tests and <5 µg/L (5,000 ng/L) for some tests. The lowest PCB concentrations in the effluent occurred at higher UV and peroxide doses.

Pilot testing was performed to determine the effectiveness of conventional activated sludge and a membrane bioreactor to remove PCBs (Bolzonella, Fatone, Pavan, & Cecchi 2010). EPA Method 1668 was used for the PCB analysis (detection limit of 0.01 ng/L per congener). Influent to the pilot system was a combination of municipal and industrial effluent. The detailed analysis was for several individual congeners. Limited testing using the Aroclor method (total PCBs) was used to compare the individual congeners and the total concentration of PCBs. Both conventional activated sludge and membrane bioreactor (MBR) systems removed PCBs. The effluent MBR concentrations ranged from <0.01 ng/L to 0.04 ng/L compared to <0.01 ng/L to 0.88 ng/L for conventional activated sludge. The pilot testing showed that increased solids retention time (SRT) and higher mixed liquor suspended solids concentrations in the MBR system led to increased removal in the liquid stream.

Bench scale studies were completed to test the effectiveness of GAC and biological activated carbon (BAC) for removing PCBs (Ghosh, Weber, Jensen, & Smith 1999). The effluent from the

GAC system was 800 ng/L. The biological film in the BAC system was presumed to support higher PCB removal with effluent concentrations of 200 ng/L. High suspended sediment in the GAC influent can affect performance. It is recommended that filtration be installed upstream of a GAC system to reduce solids and improve effectiveness.

Based on limited available data, it appears that existing municipal secondary treatment facilities in Washington state are able to reduce effluent PCBs to the range approximately 0.10 to 1.5 ng/L. It appears that the best performing existing municipal treatment facility in Washington state with a microfiltration membrane is able to reduce effluent PCBs to the range approximately 0.00019 to 0.00063 µg/L. This is based on a very limited data set and laboratory blanks covered a range that overlapped with the effluent results (blanks 0.000058 to 0.00061 µg/L).

Addition of advanced treatment processes would be expected to enhance PCB removal rates, but the technical literature does not appear to provide definitive information for guidance. A range of expected enhanced removal rates might be assumed to vary widely from level of the reference microfiltration facility of 0.19 to 0.63 ng/L.

Summary of PCB Technologies

The literature review revealed there are viable technologies available to reduce PCBs **but no research was identified with treatment technologies capable of meeting the anticipated human health criteria based limits for PCB removal**. Based on this review, a tertiary process was selected to biologically reduce PCBs and separate the solids using tertiary filtration. Alternately, GAC was investigated as an option to reduce PCBs, although it is not proven that it will meet revised effluent limits.

4.2.2 Mercury

Mercury removal from wastewater can be achieved using precipitation, adsorption, filtration, or a combination of these technologies. There is limited information available about achieving ultra-low effluent mercury concentrations near the 5 ng/L range under consideration in the proposed rulemaking process. This review provides a summary of treatment technology options and anticipated effluent mercury concentrations.

Precipitation (and co-precipitation) involves chemical addition to form a particulate and solids separation, using sedimentation or filtration. Precipitation includes the addition of a chemical precipitant and pH adjustment to optimize the precipitation reaction. Chemicals can include metal salts (ferric chloride, ferric sulfate, ferric hydroxide, or alum), pH adjustment, lime softening, or sulfide. A common precipitant for mercury removal is sulfide, with an optimal pH between 7 and 9. The dissolved mercury is precipitated with the sulfide to form an insoluble mercury sulfide that can be removed through clarification or filtration. One disadvantage of precipitation is the generation of a mercury-laden sludge that will require dewatering and disposal. The mercury sludge may be considered a hazardous waste and require additional treatment and disposal at a hazardous waste site. The presence of other compounds, such as other metals, may reduce the effectiveness of mercury precipitation/co-precipitation. For low-level mercury treatment requirements, several treatment steps will likely be required in pursuit of very low effluent targets.

EPA compiled a summary of facilities that are using precipitation/co-precipitation for mercury treatment (EPA 2007). Three of the full-scale facilities were pumping and treating groundwater and the remaining eight facilities were full-scale wastewater treatment plants. One of the pump and treat systems used precipitation, carbon adsorption, and pH adjustment to treat groundwater to effluent concentrations of 300 ng/L.

Adsorption treatment can be used to remove inorganic mercury from water. While adsorption can be used as a primary treatment step, it is frequently used for polishing after a preliminary treatment step (EPA 2007). One disadvantage of adsorption treatment is that when the adsorbent is saturated, it either needs to be regenerated or disposed of and replaced with new adsorbent. A common adsorbent is GAC. There are several patented and proprietary adsorbents on the market for mercury removal. Adsorption effectiveness can be affected by water quality characteristics, including high solids and bacterial growth, which can cause media blinding. A constant and low flow rate to the adsorption beds increases effectiveness (EPA 2007). The optimal pH for mercury adsorption on GAC is pH 4 to 5; therefore, pH adjustment may be required.

EPA compiled a summary of facilities that are using adsorption for mercury treatment (EPA 2007). Some of the facilities use precipitation and adsorption as described above. The six summarized facilities included two groundwater treatment and four wastewater treatment facilities. The reported effluent mercury concentrations were all less than 2,000 ng/L (EPA 2007).

Membrane filtration can be used in combination with a preceding treatment step. The upstream treatment is required to precipitate soluble mercury to a particulate form that can be removed through filtration. According to the EPA summary report, ultrafiltration is used to remove high-molecular weigh contaminants and solids (EPA 2007). The treatment effectiveness can depend on the source water quality since many constituents can cause membrane fouling, decreasing the effectiveness of the filters. One case study summarized in the EPA report showed that treatment of waste from a hazardous waste combustor treated with precipitation, sedimentation, and filtration achieved effluent mercury concentrations less than the detection limit of 200 ng/L.

Bench-scale research performed at the Oak Ridge Y-12 Plant in Tennessee evaluated the effectiveness of various adsorbents for removing mercury to below the NPDES limit of 12 ng/L and the potential revised limit of 51 ng/L (Hollerman et al. 1999). Several proprietary adsorbents were tested, including carbon, polyacrylate, polystyrene, and polymer adsorption materials. The adsorbents with thiol-based active sites were the most effective. Some of the adsorbents were able to achieve effluent concentrations less than 51 ng/L but none of the adsorbents achieved effluent concentrations less than 12 ng/L.

Bench-scale and pilot-scale testing performed on refinery wastewater was completed to determine treatment technology effectiveness for meeting very low mercury levels (Urgun-Demirtas, Benda, Gillenwater, Negri, Xiong & Snyder 2012) (Urgun-Demirtas, Negri, Gillenwater, Agwu Nnanna & Yu 2013). The Great Lakes Initiative water quality criterion for mercury is less than 1.3 ng/L for municipal and industrial wastewater plants in the Great Lakes region. This research included an initial bench scale test including membrane filtration, ultrafiltration, nanofiltration, and reverse osmosis to meet the mercury water quality criterion. The nanofiltration and reverse osmosis required increased pressures for filtration and resulted in increased mercury concentrations in the permeate. Based on this information and the cost difference between the filtration technologies, a pilot-scale test was performed. The 0.04 um PVDF GE ZeeWeed 500 series membranes were tested. The 1.3 ng/L water quality criterion was met under all pilot study operating conditions. The mercury in the refinery effluent was predominantly in particulate form which was well-suited for removal using membrane filtration.

Based on available data, it appears that existing municipal treatment facilities are capable of reducing effluent mercury to near the range of the proposed HHWQC on an average basis. Average effluent mercury in the range of 1.2 to 6.6 ng/L for existing facilities with secondary treatment and enhanced treatment with cloth filters and membranes. The Spokane County plant data range is an average of 1.2 ng/L to a maximum day of 3 ng/L. Addition of advanced treatment processes such as GAC or RO would be expected to enhance removal rates. Data from the West Basin treatment facility in California suggests that at a detection limit of 7.99 ng/L mercury is not detected in the effluent from this advanced process train. A range of expected enhanced removal rates from the advanced treatment process trains might be expected to ranged from meeting the proposed standard at 5 ng/L to lower concentrations represented by the Spokane County performance level (membrane filtration) in the range of 1 to 3 ng/L, to perhaps even lower levels with additional treatment. For municipal plants in Washington, this would suggest that effluent mercury values from the two advanced treatment process alternatives might range from 1 to 5 ng/L (0.001 to 0.005 µg/L) and perhaps substantially better, depending upon RO and GAC removals. It is important to note that industrial plants may have higher existing mercury levels and thus the effluent quality that is achievable at an industrial facility would be of lower quality.

Summary of Mercury Technologies

The literature search revealed limited research on mercury removal technologies at the revised effluent limit of 0.005 µg/L. Tertiary filtration with membrane filters or reverse osmosis showed the best ability to achieve effluent criteria less than 0.005 µg/L.

4.2.3 Arsenic

A variety of treatment technologies can be applied to capture arsenic (Table 3). Most of the information in the technical literature and from the treatment technology vendors is focused on potable water treatment for compliance with a Safe Drinking Water Act (SDWA) maximum contaminant level (MCL) of 10 µg/L. The most commonly used arsenic removal method for a wastewater application (tertiary treatment) is coagulation/ flocculation plus filtration. This method by itself could remove more than 90 to 95 percent of arsenic. Additional post-treatment through adsorption, ion exchange, or reverse osmosis is required for ultra-low arsenic limits in the 0.018 µg/L range under consideration in the proposed rulemaking process. In each case it is recommended to perform pilot-testing of each selected technology.

Table 3: Summary of Arsenic Removal Technologies[1]

Technology	Advantages	Disadvantages
Coagulation/filtration	• Simple, proven technology • Widely accepted • Moderate operator training	• pH sensitive • Potential disposal issues of backwash waste • As^{+3} and As^{+5} must be fully oxidized
Lime softening	• High level arsenic treatment • Simple operation change for existing lime softening facilities	• pH sensitive (requires post treatment adjustment) • Requires filtration • Significant sludge operation
Adsorptive media	• High As^{+5} selectivity • Effectively treats water with high total dissolved solids (TDS)	• Highly pH sensitive • Hazardous chemical use in media regeneration • High concentration SeO_4^{-2}, F^-, Cl^-, and SO_4^{-2} may limit arsenic removal

Table 3: Summary of Arsenic Removal Technologies[1]

Technology	Advantages	Disadvantages
Ion exchange	• Low contact times • Removal of multiple anions, including arsenic, chromium, and uranium	• Requires removal of iron, manganese, sulfides, etc. to prevent fouling • Brine waste disposal
Membrane filtration	• High arsenic removal efficiency • Removal of multiple contaminants	• Reject water disposal • Poor production efficiency • Requires pretreatment

[1] Adapted from WesTech

The removal of arsenic in activated sludge is minimal (less than 20 percent) (Andrianisa et al. 2006), but biological treatment can control arsenic speciation. During aerobic biological process As (III) is oxidized to As (V). Coagulation/flocculation/filtration removal, as well as adsorption removal methods, are more effective in removal of As(V) vs. As (III). A combination of activated sludge and post-activated sludge precipitation with ferric chloride (addition to MLSS and effluent) results in a removal efficiency of greater than 95 percent. This combination could decrease As levels from 200 µg/L to less than 5 µg/L (5,000 ng/L) (Andrianisa et al. 2008) compared to the 0.018 µg/L range under consideration in the proposed rulemaking process.

Data from the West Basin facility (using MF/RO/AOP) suggests effluent performance in the range of 0.1 to 0.2 µg/L, but it could also be lower since a detection limit used there of 0.15 µg/l is an order of magnitude higher than the proposed HHWQC. A range of expected enhanced removal rates might be assumed to equivalent to that achieved at West Basin in 0.1 to 0.2 µg/L range.

Review of Specific Technologies for Arsenic Removal

Coagulation plus Settling or Filtration

Coagulation may remove more than 95 percent of arsenic through the creation of particulate metal hydroxides. Ferric sulfite is typically more efficient and applicable to most wastewater sources compared to alum. The applicability and extent of removal should be pilot-tested, since removal efficiency is highly dependent on the water constituents and water characteristics (i.e., pH, temperature, solids).

Filtration can be added after or instead of settling to increase arsenic removal. Example treatment trains with filtration are shown in Figures 1 and 2, respectively.

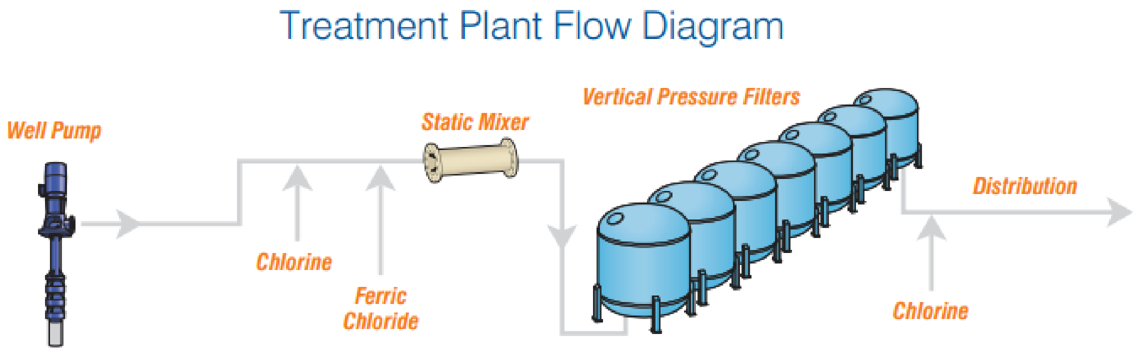

Figure 1. Water Treatment Configuration for Arsenic Removal (WesTech)

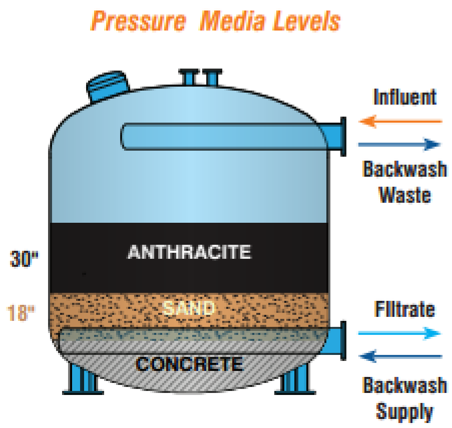

Figure 2. WesTech Pressure Filters for Arsenic Removal

One system for treatment of potable water with high levels of arsenic in Colorado (110 parts per million [ppm]) consists of enhanced coagulation followed by granular media pressure filters that include anthracite/silica sand/garnet media (WesTech). The arsenic levels were reduced to less than the drinking water MCL, which is 10 µg/L (10,000 ng/L). The plant achieves treatment by reducing the pH of the raw water to 6.8 using sulfuric acid, and then adding approximately 12 to 14 mg/L ferric sulfate. The water is filtered through 16 deep bed vertical pressure filters, the pH is elevated with hydrated lime and is subsequently chlorinated and fed into the distribution system. (http://www.westechinc.com/public/uploads/global/2011/3/Fallon%20NV%20Installation%20ReportPressureFilter.pdf).

Softening (with lime)

Removes up to 90 percent arsenic through co-precipitation, but requires pH to be higher than 10.2.

Adsorption processes

Activated alumina is considered an adsorptive media, although the chemical reaction is an exchange of arsenic ions with the surface hydroxides on the alumina. When all the surface hydroxides on the alumina have been exchanged, the media must be regenerated. Regeneration consists of backwashing, followed by sodium hydroxide, flushing with water and neutralization with a strong acid. Effective arsenic removal requires sufficient empty bed contact time. Removal efficiency can also be impacted by the water pH, with neutral or slightly acidic conditions being considered optimum. If As (III) is present, it is generally advisable to increase empty bed contact time, as As (III) is adsorbed more slowly than As (V). Alumina dissolves slowly over time due to contact with the chemicals used for regeneration. As a result, the media bed is likely to become compacted if it is not backwashed periodically.

Granular ferric hydroxide works by adsorption, but when the media is spent it cannot be regenerated and must be replaced. The life of the media depends upon pH of the raw water, the concentrations of arsenic and heavy metals, and the volume of water treated daily. Periodic backwashing is required to prevent the media bed from becoming compacted and pH may need to be adjusted if it is high, in order to extend media life. For maximum arsenic removal, filters operate in series. For less stringent removal, filters can operate in parallel.

One type of adsorption media has been developed for application to non-drinking water processes for arsenic, phosphate and for heavy metals removal by sorption (Severent Trent Bayoxide® E IN-20). This granular ferric oxide media has been used for arsenic removal from mining and industrial wastewaters, selenium removal from refinery wastes and for phosphate polishing of municipal wastewaters. Valley Vista drinking water treatment with Bayoxide® E IN-20 media achieves removal from 31-39 µg/L (31,000-39,000 ng/L) to below 10 µg/L MCL (http://www.severntrentservices.com/News/Successful_Drinking_Water_Treatment_in_an_Arsenic__Hot_Spot__nwMFT_452.aspx).

Another adsorptive filter media is greensand. Greensand is available in two forms: as glauconite with manganese dioxide bound ionically to the granules and as silica sand with manganese dioxide fused to the granules. Both forms operate in pressure filters and both are effective. Greensand with the silica sand core operates at higher water temperatures and higher differential pressures than does greensand with the glauconite core. Arsenic removal requires a minimum concentration of iron. If a sufficient concentration of iron is not present in the raw water, ferric chloride is added.

WesTech filters with greensand and permanganate addition for drinking water systems can reduce As from 15-25 µg/L to non-detect. Sodium hypochlorite and/or potassium permanganate are added to the raw water prior to the filters. Chemical addition may be done continuously or intermittently, depending on raw water characteristics. These chemicals oxidize the iron in the raw water and also maintain the active properties of the greensand itself. Arsenic removal is via co-precipitation with the iron.

Ion Exchange

Siemens offers a potable ion exchange (PIX) arsenic water filtration system. PIX uses ion exchange resin canisters for the removal of organic and inorganic contaminants, in surface and groundwater sources to meet drinking water standards.

Filtronics also uses ion exchange to treat arsenic. The technology allows removal for below the SWDA MCL for potable water of 10 µg/L (10,000 ng/L).

Reverse osmosis

Arsenic is effectively removed by RO when it is in oxidative state As(V) to approximately 1,000 ng/L or less (Ning 2002).

Summary of Arsenic Technologies

The current state of the technology for arsenic removal is at the point where all the processes target the SWDA MCL for arsenic in potable water. Current EPA maximum concentration level for drinking water is 10 ug/l; much higher than 0.0018 µg/L target for arsenic in this study. The majority of the methods discussed above are able to remove arsenic to either EPA maximum contaminant level or to the level of detection. The lowest detection limit of one of the EPA approved methods of arsenic measurements is 20 ng/l (0.020 µg/l) (Grosser, 2010), which is comparable to the 0.018 µg/L limit targeted in this study.

4.2.1 Polycyclic Aromatic Hydrocarbons

BAP During Biological Treatment

During wastewater treatment process, BAP tends to partition into sludge organic matter (Melcer et al. 1993). Primary and secondary processing could remove up to 60 percent of incoming PAHs and BAP in particular, mostly due to adsorption to sludge (Kindaichi et al., NA, Wayne et al. 2009). Biodegradation of BAP is expected to be very low since there are more than five benzene rings which are resistant to biological degradation. Biosurfactant addition to biological process could partially improve biodegradation, but only up to removal rates of 50 percent (Sponza et al. 2010). Existing data from municipal treatment facilities in Washington state have influent and effluent concentrations of BAP of approximately 0.30 ng/L indicating that current secondary treatment has limited effectiveness at BAP removal.

Methods to Enhance Biological Treatment of BAP

Ozonation prior to biological treatment could potentially improve biodegradability of BAP (Zeng et al. 2000). In the case of soil remediation, ozonation before biotreatment improved biodegradation by 70 percent (Russo et al. 2012). The overall removal of BAP increased from 23 to 91 percent after exposure of water to 0.5 mg/L ozone for 30 minutes during the simultaneous treatment process and further to 100 percent following exposure to 2.5 mg/L ozone for 60 minutes during the sequential treatment mode (Yerushalmi et al. 2006). In general, to improve biodegradability of BAP, long exposure to ozone might be required (Haapea et al. 2006).

Sonication pre-treatment or electronic beam irradiation before biological treatment might also make PAHs more bioavailable for biological degradation..

Recent studies reported that a MBR is capable of removing PAHs from wastewater (Rodrigue and Reilly 2009; Gonzaleza et al. 2012). None of the studies listed the specific PAHs constituents removed.

Removal of BAP from Drinking Water

Activated Carbon

Since BAP has an affinity to particulate matter, it is removed from the drinking water sources by means of adsorption, such as granular activated carbon (EPA). Similarly, Oleszczuk et al. (2012) showed that addition of 5 percent activated carbon could remove 90 percent of PAHs from the wastewater.

Reverse Osmosis

Light (1981) (referenced by Williams, 2003) studied dilute solutions of PAHs, aromatic amines, and nitrosamines and found rejections of these compounds in reverse osmosis to be over 99 percent for polyamide membranes. Bhattacharyya et al. (1987) (referenced by Williams, 2003) investigated rejection and flux characteristics of FT30 membranes for separating various pollutants (PAHs, chlorophenols, nitrophenols) and found membrane rejections were high (>98 percent) for the organics under ionized conditions.

Summary of BAP Technologies

Current technologies show that BAP removal may be 90 percent or greater. The lowest detection limit for BAP measurements is 0.006 µg/L, which is also the assumed secondary effluent BAP concentration assumed for this study. If this assumption is accurate, it appears technologies may exist to remove BAP to a level below the proposed criteria applied as an effluent limit of 0.0013 µg/L; however, detection limits exceed this value and it is impossible to know this for certain. A municipal wastewater treatment plant study reported both influent and effluent BAP concentrations less than the HHWQC of 0.0013 ug/L (Ecology, 2010).

4.3 Unit Processes Evaluated

Based on the results of the literature review, a wide range of technologies were evaluated for toxic constituent removal. A listing of the technologies is as follows:

- Chemically enhanced primary treatment (CEPT): this physical and chemical technology is based on the addition of a metal salt to precipitate particles prior to primary treatment, followed by sedimentation of particles in the primary clarifiers. This technology has been shown to effectively remove arsenic but there is little data supporting the claims. As a result, the chemical facilities are listed as optional.

- Activated sludge treatment (with a short SRT of approximately 8 days or less): this biological technology is commonly referred to as secondary treatment. It relies on converting dissolved organics into solids using biomass. Having a short SRT is effective at removing degradable organics referred to as BOD compounds for meeting existing discharge limits. Dissolved constituents with a high affinity to adsorb to biomass (e.g., metals, high molecular weight organics, and others) will be better removed compared to smaller molecular weight organics and recalcitrant compounds which will have minimal removal at a short SRT.
- Enhanced activated sludge treatment (with a long SRT of approximately 8 days or more): this technology builds on secondary treatment by providing a longer SRT, which enhances sorption and biodegradation. The improved performance is based on having more biomass coupled with a more diverse biomass community, especially nitrifiers, which have been shown to assist in removal of some of the more recalcitrant constituents not removed with a shorter SRT (e.g., lower molecular weight PAHs). There is little or no data available on the effectiveness of this treatment for removing BAP.

  Additional benefits associated with having a longer SRT are as follows:

  - Lower BOD/TSS discharge load to receiving water
  - Improved water quality and benefit to downstream users
  - Lower effluent nutrient concentrations which reduce algal growth potential in receiving waters
  - Reduced receiving water dissolved oxygen demand due to ammonia removal
  - Reduced ammonia discharge, which is toxic to aquatic species
  - Improved water quality for habitat, especially as it relates to biodiversity and eutrophication
  - Secondary clarifier effluent more conditioned for filtration and disinfection
  - Greater process stability from the anaerobic/anoxic zones serving as biological selectors
- Coagulation/Flocculation and Filtration: this two-stage chemical and physical process relies on the addition of a metal salt to precipitate particles in the first stage, followed by the physical removal of particles in filtration. This technology lends itself to constituents prone to precipitation (e.g., arsenic).
- Lime Softening: this chemical process relies on increasing the pH as a means to either volatilize dissolved constituents or inactivate pathogens. Given that none of the constituents being studied are expected to volatilize, this technology was not carried forward.
- Adsorptive Media: this physical and chemical process adsorbs constituents to a combination of media and/or biomass/chemicals on the media. There are several types of media, with the most proven and common being GAC. GAC can also serve as a coarse roughing filter.
- Ion Exchange: this chemical technology exchanges targeted constituents with a resin. This technology is common with water softeners where the hard divalent cations are

exchanged for monovalent cations to soften the water. Recently, resins that target arsenic and mercury removal include activated alumina and granular ferric hydroxides have been developed. The resin needs to be cleaned and regenerated, which produces a waste slurry that requires subsequent treatment and disposal. As a result, ion exchange was not considered for further.

- Membrane Filtration: This physical treatment relies on the removal of particles larger than the membranes pore size. There are several different membrane pore sizes as categorized below.
    - Microfiltration (MF): nominal pore size range of typically between 0.1 to 1 micron. This pore size targets particles, both inert and biological, and bacteria. If placed in series with coagulation/flocculation upstream, dissolved constituents precipitated out of solution and bacteria can be removed by the MF membrane.
    - Ultrafiltration (UF): nominal pore size range of typically between 0.01 to 0.1 micron. This pore size targets those solids removed with MF (particles and bacteria) plus viruses and some colloidal material. If placed in series with coagulation/flocculation upstream, dissolved constituents precipitated out of solution can be removed by the UF membrane.
    - Nanofiltration (NF): nominal pore size range of typically between 0.001 to 0.010 micron. This pore size targets those removed with UF (particles, bacteria, viruses) plus colloidal material. If placed in series with coagulation/flocculation upstream, dissolved constituents precipitated out of solution can be removed by the NF membrane.
- MBR (with a long SRT): this technology builds on secondary treatment whereby the membrane (microfiltration) replaces the secondary clarifier for solids separation. As a result, the footprint is smaller, the mixed liquor suspended solids concentration can be increased to about 5,000 – 10,000 mg/L, and the physical space required for the facility reduced when compared to conventional activated sludge. As with the activated sludge option operated at a longer SRT, the sorption and biodegradation of organic compounds are enhanced in the MBR process. The improved performance is based on having more biomass coupled with a more diverse biomass community, especially nitrifiers which have been shown to assist in removal of persistent dissolved compounds (e.g., some PAHs). There is little or no data available on effectiveness at removing BAP. Although a proven technology, MBRs were not carried further in this technology review since they are less likely to be selected as a retrofit for an existing activated sludge (with a short SRT) secondary treatment facility. The MBR was considered to represent a treatment process approach more likely to be selected for a new, greenfield treatment facility. Retrofits to existing secondary treatment facilities can accomplish similar process enhancement by extending the SRT in the activated sludge process followed by the addition of tertiary membrane filtration units.
- RO: This physical treatment method relies on the use of sufficient pressure to osmotically displace water across the membrane surface while simultaneously rejecting most salts. RO is very effective at removing material smaller than the size ranges for the membrane filtration list above, as well as salts and other organic compounds. As a result, it is expected to be more effective than filtration and MBR methods described above at removing dissolved constituents. Although effective, RO produces a brine reject water that must be managed and disposed.

- Advanced Oxidation Processes (AOPs): this broad term considers all chemical and physical technologies that create strong hydroxyl-radicals. Examples of AOPs include Fenton's oxidation, ozonation, ultraviolet/hydrogen peroxide (UV-H2O2), and others. The radicals produced are rapid and highly reactive at breaking down recalcitrant compounds. Although effective at removing many complex compounds such as those evaluated in this study, AOPs does not typically have as many installations as membranes and activated carbon technologies. As a result, AOPs were not carried forward.

Based on the technical literature review discussed above, a summary of estimated contaminant removal rated by unit treatment process is presented in Table 4.

Table 4. Contaminants Removal Breakdown by Unit Process

Unit Process	Arsenic	BAP	Mercury	Polychlorinated Biphenyls
Activated Sludge Short SRT	No removal	Partial Removal by partitioning		80% removal; effluent <0.88 ng/L
Activated Sludge Long SRT	No removal	Partial removal by partitioning and/or partially biodegradation; MBR could potentially remove most of BAP		>90% removal with a membrane bioreactor, <0.04 ng/L (includes membrane filtration)
Membrane Filtration (MF)	More than 90 % removal (rejection of bound arsenic)	No removal	<1.3 ng/L	>90% removal with a membrane bioreactor, <0.04 ng/L (includes membrane filtration)
Reverse Osmosis (RO)	More than 90% removal (rejection of bound arsenic and removal of soluble arsenic)	More than 98% removal		
Granular Activated Carbon (GAC)	No removal, removal only when carbon is impregnated with iron	90 % removal	<300 ng/L (precipitation and carbon adsorption) <51 ng/L (GAC)	<800 ng/L Likely requires upstream filtration
Disinfection	--	--	--	--

4.4 Unit Processes Selected

The key conclusion from the literature review was that there is limited, to no evidence, that existing treatment technologies are capable of simultaneously meeting all four of the revised discharge limits for the toxics under consideration. Advanced treatment using RO or GAC is expected to provide the best overall removal of the constituents of concern. It is unclear whether these advanced technologies are able to meet revised effluent limits, however these processes may achieve the best effluent quality of the technologies reviewed. This limitation in the findings is based on a lack of an extensive dataset on treatment removal effectiveness in the technical literature for the constituents of interest at the low levels relevant to the proposed criteria, which approach the limits of reliable removal performance for the technologies. As Table 4 highlights, certain unit processes are capable of removing a portion, or all, of the removal requirements for each technology. The removal performance for each constituent will vary from facility to facility and require a site-specific, detailed evaluation because the proposed criteria are such low concentrations. In some cases, a facility may only have elevated concentrations of a single constituent of concern identified in this study. In other cases, a discharger may have elevated concentrations of the four constituents identified in this study, as well as others not identified in this study but subject to revised water quality criteria. This effort is intended to describe a planning level concept of what treatment processes are required to comply with discharge limits for all four constituents. Based on the literature review of unit processes above, two different treatment trains were developed for the analysis that are compared against a baseline of secondary treatment as follows:

- Baseline: represents conventional secondary treatment that is most commonly employed nationwide at wastewater treatment plants. A distinguishing feature for this treatment is the short solids residence time (SRT) (<8 days) is intended for removal of BOD with minimal removal for the toxic constituents of concern.
- Advanced Treatment – MF/RO: builds on baseline with the implementation of a longer SRT (>8 days) and the addition of MF and RO. The longer SRT not only removes BOD, but it also has the capacity to remove nutrients and a portion of the constituents of concern. This alternative requires a RO brine management strategy which will be discussed in sub-sections below.
- Advanced Treatment – MF/GAC: this alternative provides a different approach to advanced treatment with MF/RO by using GAC and avoiding the RO reject brine water management concern. Similar to the MF/RO process, this alternative has the longer SRT (>8 days) with the capacity to remove BOD, nutrients, and a portion of the toxic constituents of concern. As a result, the decision was made to develop costs for both advanced treatment options.

A description of each alternative is provided in Table 5. The process flowsheets for each alternative are presented in Figure 3 to Figure 5.

4.4.1 Baseline Treatment Process

A flowsheet of the baseline treatment process is provided in Figure 3. The baseline treatment process assumes the current method of treatment commonly employed by dischargers. For this process, water enters the headworks and undergoes primary treatment, followed by conventional activated sludge (short SRT) and disinfection. The solids wasted in the activated sludge process are thickened, followed by mixing with primary solids prior to entering the anaerobic digestion process for solids stabilization. The digested biosolids are dewatered to produce a cake and hauled off-site. Since the exact process for each interested facility in Washington is unique, this baseline treatment process was used to establish the baseline capital and O&M costs. The baseline costs will be compared against the advanced treatment alternatives to illustrate the magnitude of the increased costs and environmental impacts.

Table 5. Unit Processes Description for Each Alternative

Unit Process	Baseline	Advanced Treatment – MF/RO	Advanced Treatment - GAC
Influent Flow	5 mgd	5 mgd	5 mgd
Chemically Enhanced Primary Treatment (CEPT); Optional	--	• Metal salt addition (alum) upstream of primaries	• Metal salt addition (alum) upstream of primaries
Activated Sludge	• Hydraulic Residence Time (HRT): 6 hrs • Short Solids Residence Time (SRT): <8 days	• Hydraulic Residence Time (HRT): 12 hrs (Requires more tankage than the Baseline) • Long Solids Residence Time (SRT): >8 days (Requires more tankage than the Baseline)	• Hydraulic Residence Time (HRT): 12 hrs (Requires more tankage than the Baseline) • Long Solids Residence Time (SRT): >8 days (Requires more tankage than the Baseline)
Secondary Clarifiers	Hydraulically Limited	Solids Loading Limited (Larger clarifiers than Baseline)	Solids Loading Limited (Larger clarifiers than Baseline)
Microfiltration (MF)	--	Membrane Filtration to Remove Particles and Bacteria	Membrane Filtration to Remove Particles and Bacteria
Reverse Osmosis (RO)	--	Treat 50% of the Flow by RO to Remove Metals and Dissolved Constituents. Sending a portion of flow through the RO and blending it with the balance of plant flows ensures a stable non-corrosive, non-toxic discharge.	--
Reverse Osmosis Brine Reject Mgmt	--	Several Options (All Energy or Land Intensive)	--
Granular Activated Carbon (GAC)	--	--	Removes Dissolved Constituents
Disinfection	Not shown to remove any of the constituents	Not shown to remove any of the constituents	Not shown to remove any of the constituents

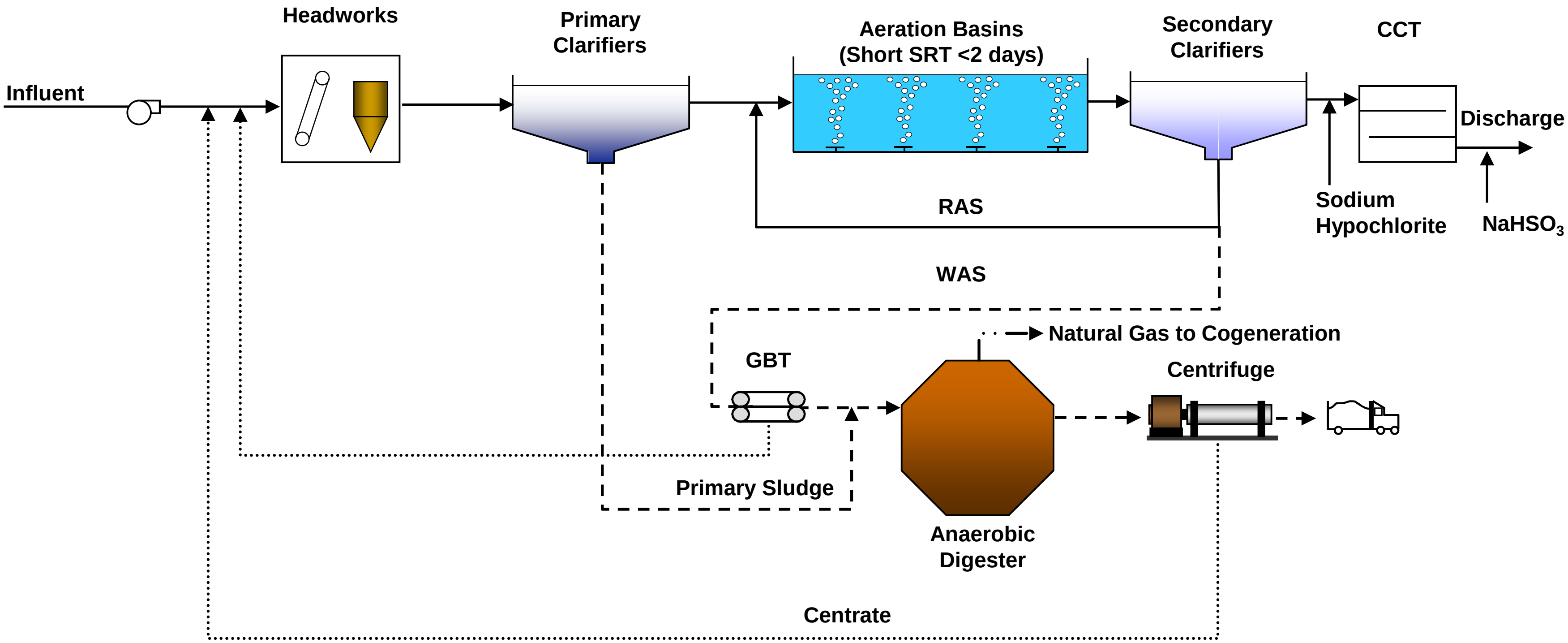

Figure 3. Baseline Flowsheet – Conventional Secondary Treatment

4.4.2 Advanced Treatment – MF/RO Alternative

A flowsheet of the advanced treatment – MF/RO alternative is provided in Figure 4. This alternative builds on the baseline secondary treatment facility, whereby the SRT is increased in the activated sludge process, and MF and RO are added prior to disinfection. The solids treatment train does not change with respect to the baseline. Additionally, a brine management strategy must be considered.

The RO process concentrates contaminants into a smaller volume reject stream. Disposing of the RO reject stream can be a problem because of the potentially large volume of water involved and the concentration of contaminants contained in the brine. For reference, a 5 mgd process wastewater flow might result in 1 mgd of brine reject requiring further management. The primary treatment/handling options for RO reject are as follows:

- Zero liquid discharge
- Surface water discharge
- Ocean discharge
- Haul and discharge to coastal location for ocean discharge
- Sewer discharge
- Deep well injection
- Evaporate in a pond
- Solar pond concentrator

Many of the RO brine reject management options above result in returning the dissolved solids to a “water of the state” such as surface water, groundwater, or marine waters. Past rulings in Washington State have indicated that once pollutants are removed from during treatment they are not to be re-introduced to a water of the state. As a result, technologies with this means for disposal were not considered viable options for management of RO reject water in Washington.

Zero Liquid Discharge

Zero liquid discharge (ZLD) is a treatment process that produces a little or no liquid brine discharge but rather a dried residual salt material. This process improves the water recovery of the RO system by reducing the volume of brine that must be treated and disposed of in some manner. ZLD options include intermediate treatment, thermal-based technologies, pressure driven membrane technologies, electric potential driven membrane technologies, and other alternative technologies.

Summary

There are many techniques which can be used to manage reject brine water associated with RO treatment. The appropriate alternative is primarily governed by geographic and local constraints. A comparison of the various brine management methods and potential costs are provided in Table 6.

Of the listed options, ZLD was considered for this analysis as the most viable approach to RO reject water management. An evaporation pond was used following ZLD. The strength in this combination is ZLD reduces the brine reject volume to treat, which in turn reduces the required evaporation pond footprint. The disadvantage is that evaporation ponds require a substantial amount of physical space which may not be available at existing treatment plant sites. It is also important to recognize that the greenhouse gas (GHG) emissions vary widely for the eight brine management options listed above based on energy and chemical intensity.

Table 6. Brine Disposal Method Relative Cost Comparison

Disposal Method	Description	Relative Capital Cost	Relative O&M Cost	Comments
Zero Liquid Discharge (ZLD)	Further concentrates brine reject for further downstream processing	High	High	This option is preferred as an intermediate step. This rationale is based on the reduction in volume to handle following ZLD. For example, RO reject stream volume is reduced on the order of 50-90%.
Surface Water Discharge	Brine discharge directly to surface water. Requires an NPDES permit.	Lowest	Lowest	Both capital and O&M costs heavily dependent on the distance from brine generation point to discharge. Not an option for nutrient removal.
Ocean Discharge	Discharge through a deep ocean outfall.	Medium	Low	Capital cost depends on location and availability of existing deep water outfall.
Sewer Discharge	Discharge to an existing sewer pipeline for treatment at a wastewater treatment plant.	Low	Low	Both capital and O&M costs heavily dependent on the brine generation point to discharge distance. Higher cost than surface water discharge due to ongoing sewer connection charge. Not an option for wastewater treatment.
Deep Well Injection	Brine is pumped underground to an area that is isolated from drinking water aquifers.	Medium	Medium	Technically sophisticated discharge and monitoring wells required. O&M cost highly variable based on injection pumping energy.
Evaporation Ponds	Large, lined ponds are filled with brine. The water evaporates and a concentrated salt remains.	Low – High	Low	Capital cost highly dependent on the amount and cost of land.
Salinity Gradient Solar Ponds (SGSP)	SGSPs harness solar power from pond to power an evaporative unit.	Low – High	Lowest	Same as evaporation ponds plus added cost of heat exchanger and pumps. Lower O&M cost due to electricity production.
Advanced Thermal Evaporation	Requires a two-step process consisting of a brine concentrator followed by crystallizer	High	Highest	Extremely small footprint, but the energy from H_2O removal is by far the most energy intensive unless waste heat is used.

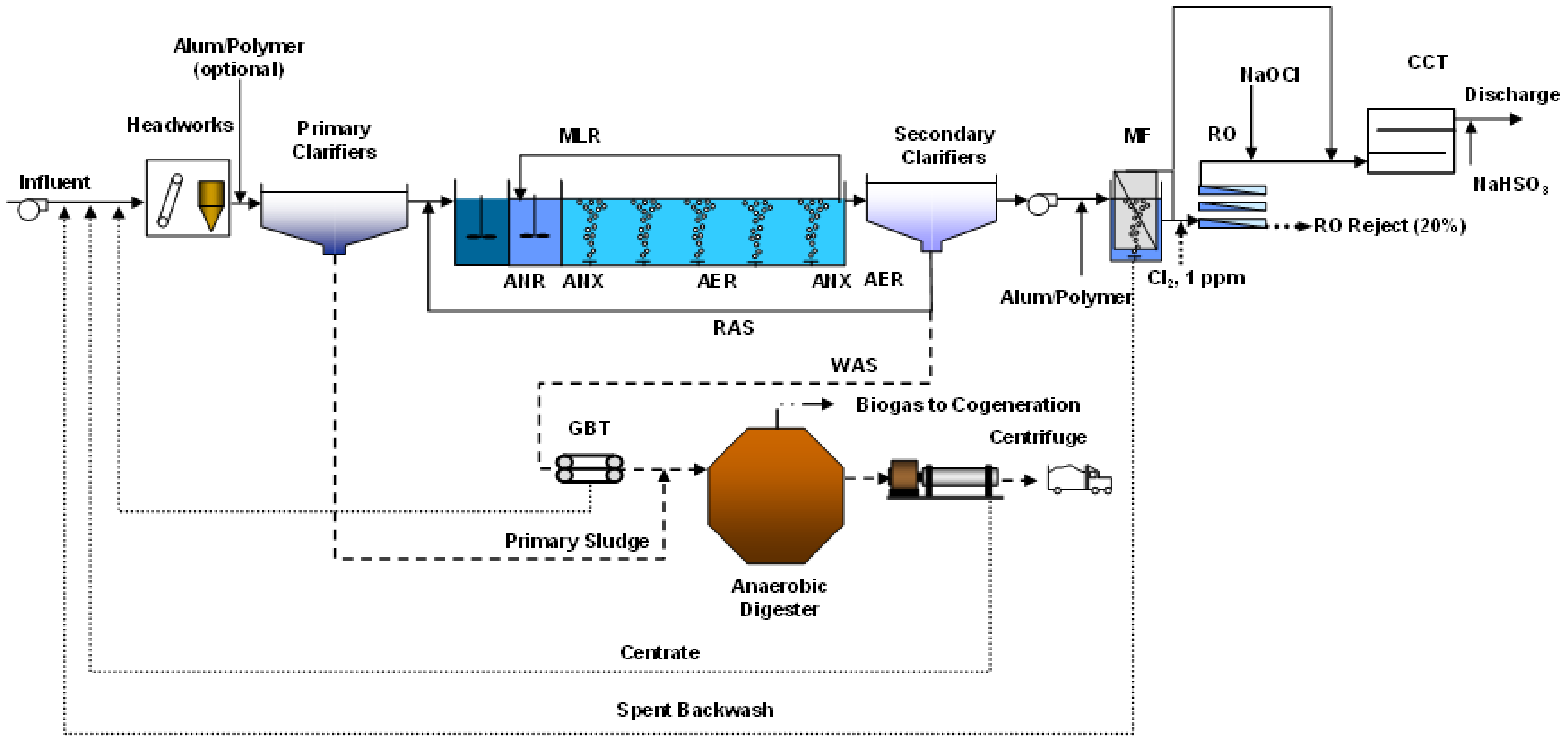

Figure 4. Advanced Treatment Flowsheet – Tertiary Microfiltration and Reverse Osmosis

4.4.3 Advanced Treatment – MF/GAC Alternative

A flowsheet of the advanced treatment – MF/GAC alternative is provided in Figure 5. Following the MF technology, a GAC contactor and media are required.

This alternative was developed as an option that does not require a brine management technology (e.g., ZLD) for comparison to the MF/RO advanced treatment alternative. However, this treatment alternative does require that the GAC be regenerated. A baseline secondary treatment facility can be retrofitted for MF/GAC. If an existing treatment facility has an extended aeration lagoon, the secondary effluent can be fed to the MF/GAC. The longer SRT in the extended aeration lagoon provides all the benefits associated with the long SRT in an activated sludge plant as previously stated:

- Lower BOD/TSS discharge load
- Higher removal of recalcitrant constituents and heavy metals
- Improved water quality and benefit to downstream users
- Less downstream algal growth
- Reduced receiving water dissolved oxygen demand due to ammonia removal
- Reduced ammonia discharge loads, which is toxic to several aquatic species
- Improved water quality for habitat, especially as it relates to biodiversity and eutrophication
- Secondary clarifier effluent more conditioned for filtration and disinfection
- Greater process stability from the anaerobic/anoxic zones serving as a selector

If an existing treatment facility employs a high rate activated sludge process (short SRT) similar to the baseline, it is recommended that the activated sludge process SRT be increased prior to the MF/GAC unit processes. The longer SRT upstream of the MF is preferred to enhance the membrane flux rate, reduce membrane biofouling, increase membrane life, and reduce the chemicals needed for membrane cleaning.

The key technical and operational challenges associated with the tertiary add-on membrane filtration units are as follows:

- The membrane filtration technology is a proven and reliable technology. With over 30 years of experience, it has made the transition in recent years from an emerging technology to a proven and reliable technology.
- Membrane durability dependent on feed water quality. The water quality is individual facility specific.
- Membranes are sensitive to particles, so upstream screening is critical. The newer generations of membranes have technical specifications that require a particular screen size.
- Membrane area requirements based on peak flows as water must pass through the membrane pores. Additionally, membranes struggle with variable hydraulic loading. Flow equalization upstream can greatly reduce the required membrane surface area and provide uniform membrane loading.

- Membrane tanks can exacerbate any foam related issues from the upstream biological process. Foam entrapment in the membrane tank from the upstream process can reduce membrane filtration capacity and in turn result in a plant-wide foam problem.
- Reliable access to the membrane modules is key to operation and maintenance. Once PLC is functionary properly, overall maintenance requirements for sustained operation of the system are relatively modest.
- The membranes go through frequent membrane relaxing or back pulse and a periodic deep chemical clean in place (CIP) process.
- Sizing of membrane filtration facilities governed by hydraulic flux. Municipal wastewaters have flux values that range from about 20 to 40 gallons per square foot per day (gfd) under average annual conditions. The flux associated with industrial applications is wastewater specific.

Following the MF is the activated carbon facilities. There are two kinds of activated carbon used in treating water: powdered activated carbon (PAC) and GAC. PAC is finely-ground, loose carbon that is added to water, mixed for a short period of time, and removed. GAC is larger than PAC, is generally used in beds or tanks that permit higher adsorption and easier process control than PAC allows, and is replaced periodically. PAC is not selective, and therefore, will adsorb all active organic substances making it an impractical solution for a wastewater treatment plant. As a result, GAC was considered for this analysis. The type of GAC (e.g., bituminous and subbituminous coal, wood, walnut shells, lignite or peat), gradation, and adsorption capacity are determined by the size of the largest molecule/ contaminant that is being filtered (AWWA, 1990).

As water flows through the carbon bed, contaminants are captured by the surfaces of the pores until the carbon is no longer able to adsorb new molecules. The concentration of the contaminant in the treated effluent starts to increase. Once the contaminant concentration in the treated water reaches an unacceptable level (called the breakthrough concentration), the carbon is considered "spent" and must be replaced by virgin or reactivated GAC.

The capacity of spent GAC can be restored by thermal reactivation. Some systems have the ability to regenerate GAC on-site, but in general, small systems haul away the spent GAC for off-site regeneration (EPA 1993). For this study, off-site regeneration was assumed.

The basic facilities and their potential unit processes included in this chapter are as follows:

- GAC supply and delivery
- Influent pumping
    - Low head feed pumping
    - High head feed pumping (assumed for this study as we have low limits so require high beds)
- Contactors and backwash facilities
    - Custom gravity GAC contactor
    - Pre-engineered pressure GAC contactor (Used for this study)
    - Backwash pumping
- GAC transport facilities
    - Slurry pumps
    - Eductors (Used for this study)

- Storage facilities
  - Steel tanks
  - Concrete tanks (Used for this study; larger plants would typically select concrete tanks)
- Spent carbon regeneration
  - On-site GAC regeneration
  - Off-Site GAC regeneration

Following the MF is the GAC facility. The GAC contactor provides about a 12-min hydraulic residence time for average annual conditions. The GAC media must be regenerated about twice per year in a furnace. The constituents sorbed to the GAC media are removed during the regeneration process. A typical design has full redundancy and additional storage tankage for spent and virgin GAC. Facilities that use GAC need to decide whether they will regenerate GAC on-site or off-site. Due to challenges associated with receiving air emission permits for new furnaces, it was assumed that off-site regeneration would be evaluated.

The key technical and operational challenges associated with the tertiary add-on GAC units are as follows:

- Nearest vendor to acquire virgin GAC – How frequently can they deliver virgin GAC and what are the hauling costs?
- Contactor selection is typically based on unit cost and flow variation. The concrete contactor is typically more cost effective at higher flows so it was used for this evaluation. The pre-engineered pressure contactor can handle a wider range of flows than a concrete contactor. Additionally, a pressure system requires little maintenance as they are essentially automated
- Periodical contactor backwashing is critical for maintaining the desired hydraulics and control biological growth
- Eductors are preferred over slurry pumps because they have fewer mechanical components. Additionally, the pump with eductors is not in contact with the carbon, which reduces wear.
- Off-site GAC regeneration seems more likely due to the challenges with obtaining an air emissions permit.

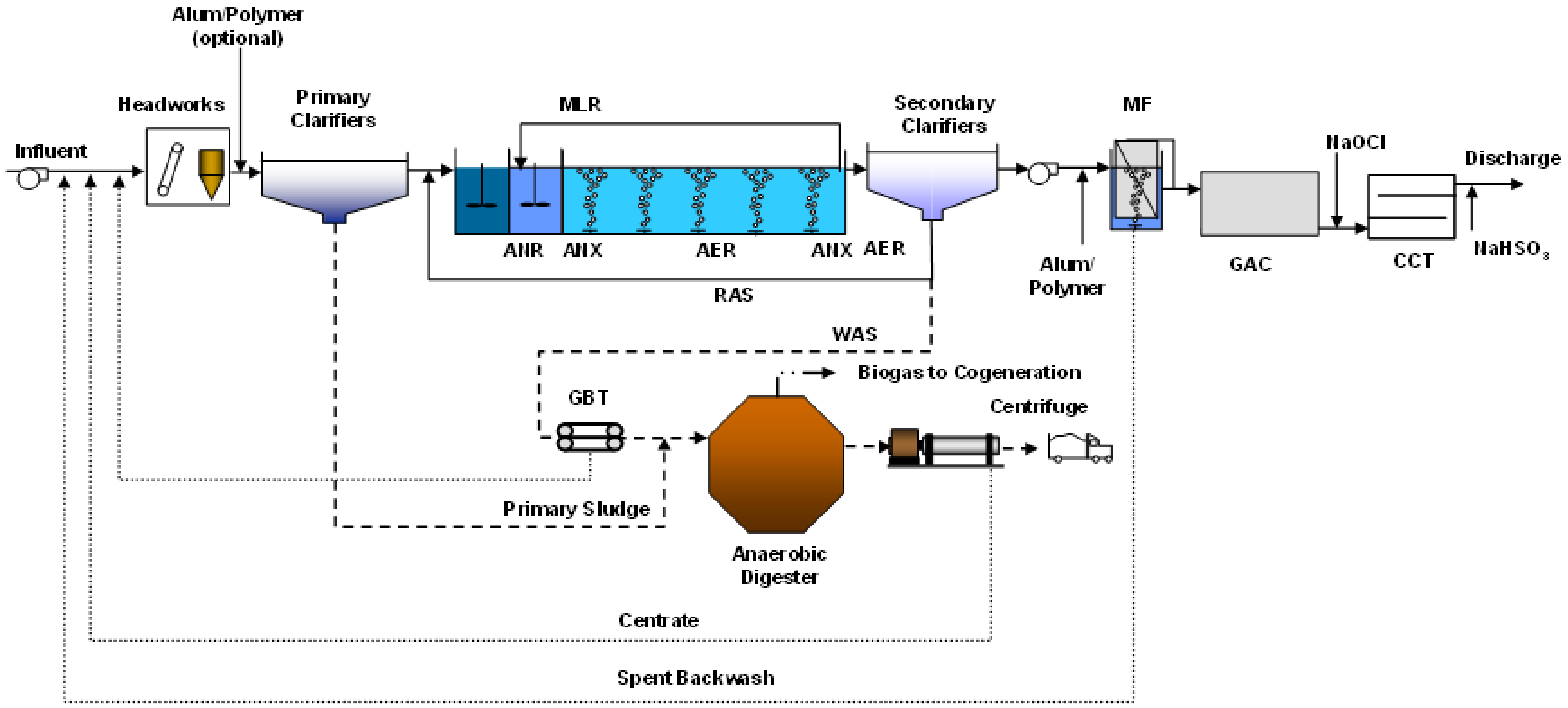

Figure 5. Advanced Treatment Flowsheet – Tertiary Microfiltration and Granular Activated Carbon

4.5 Steady-State Mass Balance

HDR used its steady-state mass balance program to calculate the flows and loads within the candidate advanced treatment processes as a means to size facilities. The design of wastewater treatment facilities are generally governed by steady-state mass balances. For a steady-state mass balance, the conservation of mass is calculated throughout the entire wastewater treatment facility for defined inputs. Dynamic mass balance programs exist for designing wastewater facilities, but for a planning level study such as this, a steady state mass balance program is adequate. A dynamic program is generally used for detailed design and is site-specific with associated requirements for more detailed wastewater characterization.

The set of model equations used to perform a steady-state mass balance are referred to as the model. The model equations provide a mathematical description of various wastewater treatment processes, such as an activated sludge process, that can be used to predict unit performance. The program relies on equations for each unit process to determine the flow, load, and concentration entering and leaving each unit process.

An example of how the model calculates the flow, load, and concentration for primary clarifiers is provided below. The steady-state mass balance equation for primary clarifiers has a single input and two outputs as shown in the simplified Figure 6. The primary clarifier feed can exit the primary clarifiers as either effluent or sludge. Solids not removed across the primaries leave as primary effluent, whereas solids captured leave as primary sludge. Scum is not accounted for.

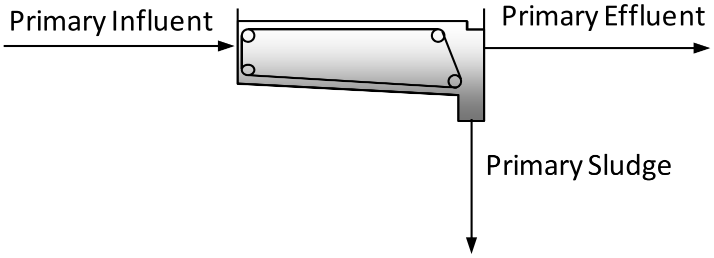

Figure 6. Primary Clarifier Inputs/Outputs

The mass balance calculation requires the following input:

- Solids removal percentage across the primaries (based on average industry accepted performance)
- Primary solids thickness (i.e., percent solids) (based on average industry accepted performance)

The steady-state mass balance program provides a reasonable first estimate for the process performance, and an accurate measure of the flows and mass balances at various points throughout the plant. The mass balance results were used for sizing the facility needs for each alternative. A listing of the unit process sizing criterion for each unit process is provided in Appendix A. By listing the unit process sizing criteria, a third-party user could redo the analysis and end up with comparable results. The key sizing criteria that differ between the baseline and treatment alternatives are as follows:

- Aeration basin mixed liquor is greater for the advanced treatment alternatives which in turn requires a larger volume
- The secondary clarifiers are sized based on hydraulic loading for the baseline versus solids loading for the advanced treatment alternatives

- The MF/GAC and MF/RO sizing is only required for the respective advanced treatment alternatives.

4.6 Adverse Environmental Impacts Associated with Advanced Treatment Technologies

The transition from the baseline (conventional secondary treatment) to either advanced treatment alternatives has some environmental impacts that merit consideration, including the following:

- Land area for additional system components (which for constrained facility sites, may necessitate land acquisition and encroachment into neighboring properties with associated issues and challenges, etc.).
- Increased energy use and atmospheric emissions of greenhouse gases and criteria air contaminants associated with power generation to meet new pumping requirements across the membrane filter systems (MF and RO) and GAC.
- Increased chemical demand associated with membrane filters (MF and RO).
- Energy and atmospheric emissions associated with granulated charcoal regeneration.
- RO brine reject disposal. The zero liquid discharge systems are energy intensive energy and increase atmospheric emissions as a consequence of the electrical power generation required for removing water content from brine reject.
- Increase in sludge generation while transitioning from the baseline to the advanced treatment alternatives. There will be additional sludge captured with the chemical addition to the primaries and membrane filters (MF and RO). Additionally, the GAC units will capture more solids.
- Benefits to receiving water quality by transitioning from a short SRT (<2 days) in the baseline to a long SRT (>8 days) for the advanced treatment alternatives (as previously stated):
  - Lower BOD/TSS discharge load
  - Higher removal of recalcitrant constituents and heavy metals
  - Improved water quality and benefit to downstream users
  - Reduced nutrient loadings to receiving waters and lower algal growth potential
  - Reduced receiving water dissolved oxygen demand due to ammonia removal
  - Reduced ammonia discharge loads, which is toxic to aquatic species
  - Improved water quality for habitat, especially as it relates to biodiversity and eutrophication
  - Secondary clarifier effluent better conditioned for subsequent filtration and disinfection
  - Greater process stability from the anaerobic/anoxic zones serving as a biological selectors

HDR calculated GHG emissions for the baseline and advanced treatment alternatives. The use of GHG emissions is a tool to normalize the role of energy, chemicals, biosolids hauling, and fugitive emissions (e.g., methane) in a single unit. The mass balance results were used to quantify energy demand and the corresponding GHG emissions for each alterative. Energy demand was estimated from preliminary process calculations. A listing of the energy demand for each process stream, the daily energy demand, and the unit energy demand is provided in Table 7. The advanced treatment options range from 2.3 to 4.1 times greater than the baseline. This large increase in energy demand is attributed to the energy required to pass water through the membrane barriers and/or the granular activated carbon. Additionally, there is energy required to handle the constituents removed as either regenerating the GAC or handling the RO brine reject water. This additional energy required to treat the removed constituents is presented in Table 7.

Table 7. Energy Breakdown for Each Alternative (5 mgd design flow)

Parameter	Units	Baseline	Advanced Treatment – MF/GAC	Advanced Treatment – MF/RO
Daily Liquid Stream Energy Demand	MWh/d	11.6	23.8	40.8
Daily Solids Stream Energy Demand	MWh/d	-1.6	-1.1	-1.1
Daily Energy Demand	MWh/d	10.0	22.7	39.7
Unit Energy Demand	kWh/MG Treated	2,000	4,500	7,900

MWh/d = megawatt hours per day
kWh/MG = kilowatt hours per million gallons

Details on the assumptions used to convert between energy demand, chemical demand and production, as well as biologically-mediated gases (i.e., CH4 and N2O) and GHG emissions are provided in Appendix B.

A plot of the GHG emissions for each alternative is shown in Figure 7. The GHG emissions increase from the baseline to the two advanced treatment alternatives. The GHG emissions increase about 50 percent with respect to baseline when MF/GAC is used and the GHG emissions increase over 100 percent with respect to baseline with the MF/RO advanced treatment alternative.

The MF/GAC energy demand would be larger if GAC regeneration was performed on-site. The GHG emissions do not include the energy or air emissions that result from off-site GAC regeneration. Only the hauling associated with moving spent GAC is included. The energy associated with operating the furnace would exceed the GHG emissions from hauling spent GAC.

The zero liquid discharge in the MF/RO alternative alone is comparable to the Baseline. This contribution to increased GHG emissions by zero liquid discharge brine system highlights the importance of the challenges associated with managing brine reject.

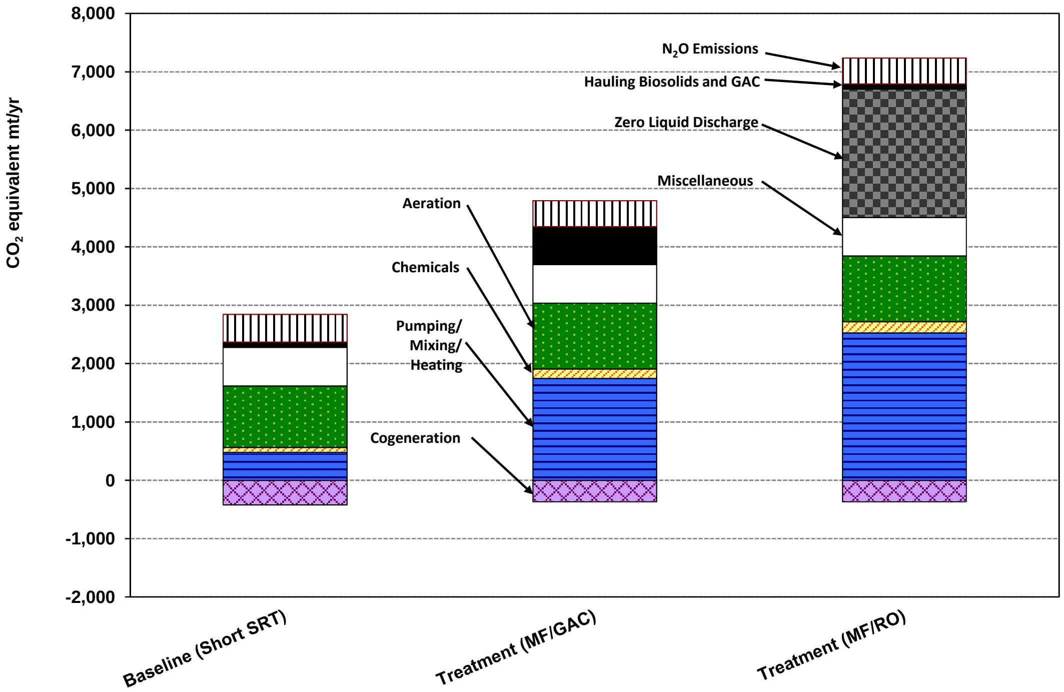

Figure 7. Greenhouse Gas Emissions for Each Alternative

The use of GHG emissions as a measure of sustainability does not constitute a complete comparison between the baseline and advanced treatment alternatives. Rather, it is one metric that captures the impacts of energy, chemical demand and production, as well as biologically-mediated gases (i.e., CH_4 and N_2O). The other environmental impacts of advanced treatment summarized in the list above should also be considered in decision making beyond cost analysis.

4.7 Costs

Total project costs along with the operations and maintenance costs were developed for each advanced treatment alternative for a comparison with baseline secondary treatment.

4.7.1 Approach

The cost estimates presented in this report are planning level opinions of probable construction costs for a nominal 5 mgd treatment plant design flow representing a typical facility without site specific details about local wastewater characteristics, physical site constraints, existing infrastructure, etc. The cost estimates are based on wastewater industry cost references, technical studies, actual project cost histories, and professional experience. The costs presented in this report are considered planning level estimates. A more detailed development of the advanced treatment process alternatives and site specific information would be required to further refine the cost estimates. Commonly this is accomplished in the preliminary design phase of project development for specific facilities following planning.

The cost opinion includes a range of costs associated with the level of detail used in this analysis. Cost opinions based on preliminary engineering can be expected to follow the Association for the Advancement of Cost Engineering (AACE International) Recommended Practice No. 17R-97 Cost Estimate Classification System estimate Class 4. A Class 4 estimate is based upon a 5 to 10 percent project definition and has an expected accuracy range of -30 to +50 percent and typical end usage of budget authorization and cost control. It is considered an "order-of-magnitude estimate." The life-cycle costs were prepared using the net present value (NPV) method.

The cost associated for each new unit process is based on a unit variable, such as required footprint, volume, demand (e.g., lb O_2/hr), and others. This approach is consistent with the approach developed for the EPA document titled "Estimating Water Treatment Costs: Volume 2- Cost Curves Applicable to 1 to 200 mgd Treatment Plants" dated August 1979. The approach has been updated since 1979 to account for inflation and competition, but the philosophy for estimating costs for unit processes has not changed. For example, the aeration system sizing/cost is governed by the maximum month airflow demand. Additionally, the cost associated constructing an aeration basin is based on the volume. The cost considers economies of scale.

The O&M cost estimates were calculated from preliminary process calculations. The operations cost includes energy and chemical demand. For example, a chemical dose was assumed based on industry accepted dosing rates and the corresponding annual chemical cost for that particular chemical was accounted for. The maintenance values only considered replacement equipment, specifically membrane replacement for the Advanced Treatment Alternatives.

4.7.2 Unit Cost Values

The life-cycle cost evaluation was based on using the economic assumptions shown in Table 8. The chemical costs were based on actual values from other projects. To perform detailed cost evaluations per industry, each selected technology would need to be laid out on their respective site plan based on the location of the existing piping, channels, and other necessary facilities.

Table 8. Economic Evaluation Variables

Item	Value
Nominal Discount Rate	5%
Inflation Rate:	
General	3.5%
Labor	3.5%
Energy	3.5%
Chemical	3.5%
Base Year	2013
Project Life	25 years
Energy	$0.06/kWh
Natural Gas	$0.60/therm
Chemicals:	
Alum	$1.1/gal
Polymer	$1.5/gal
Hypochlorite	$1.5/gal
Salt	$0.125/lb
Antiscalant	$12.5/lb
Acid	$0.35/lb
Deionized Water	$3.75/1,000 gal
Hauling:	

Table 8. Economic Evaluation Variables

Item	Value
Biosolids Hauling Distance	100 miles (one way)
Biosolids Truck Volume	6,000 gal/truck
Biosolids Truck Hauling	$250/truck trip
GAC Regeneration Hauling Distance	250 miles (round trip)
GAC Regeneration Truck Volume	$20,000 lb GAC/truck
GAC Regeneration Truck Hauling	Included in cost of Virgin GAC

kWh= kilowatt hours; lbs=pounds; GAC=granulated activated carbon; gal=gallon

4.7.3 Net Present Value of Total Project Costs and Operations and Maintenance Cost in 2013 Dollars

An estimate of the net present value for the baseline treatment process and the incremental cost to implement the advanced treatment alternatives is shown in Table 9. The cost for the existing baseline treatment process was estimated based on new construction for the entire conventional secondary treatment process (Figure 3). The incremental cost to expand from existing baseline secondary treatment to advanced treatment was calculated by taking the difference between the baseline and the advanced treatment alternatives. These values serve as a benchmark for understanding the prospective cost for constructing advanced treatment at the planning level of process development.

Table 9. Treatment Technology Total Project Costs in 2013 Dollars for a 5 mgd Facility

Alternative	Total Construction Cost, 2013 dollars ($ Million)	O&M Net Present Value, 2013 dollars ($ Million)*	Total Net Present Value, 2013 dollars ($ Million)	NPV Unit Cost, 2013 dollars ($/gpd)
Baseline (Conventional Secondary Treatment)*	59 - 127	5 - 11	65 – 138	13 - 28
Advanced Treatment – MF/RO**	108 - 231	31 - 67	139 - 298	28 - 60
Advanced Treatment – MF/GAC	131 - 280	50 - 108	181 - 388	36 - 78
Incremental Increase to Advanced Treatment MF/RO	48 - 104	26 - 56	75 - 160	15 - 32
Incremental Increase to Advanced Treatment MF/GAC	71 - 153	45 - 97	117 - 250	23 - 50

* The additional cost to increase the SRT to upwards of 30-days is about $12 - 20 million additional dollars in total project cost for a 5 mgd design flow

** Assumes zero liquid discharge for RO brine management, followed by evaporation ponds. Other options are available as listed in Section 4.4.2.

O&M=operations and maintenance; MF/RO=membrane filtration/reverse osmosis; MF/GAC=membrane filtration/granulated activated carbon; gpd=gallons per day

4.7.4 Unit Cost Assessment

Costs presented above are based on a treatment capacity of 5.0 mgd, however, existing treatment facilities range dramatically across Washington in size and flow treated. Table 9 indicates that the unit capital cost for baseline conventional secondary treatment for 5.0 mgd ranges between $13 to 28 per gallon per day of treatment capacity. The unit cost for the advanced treatment alternatives increases the range from the low $20s to upper $70s on a per-gallon per-day of capacity. The increase in cost for the advanced treatment alternatives is discussed in the sub-sections below.

Advanced Treatment MF/RO

The advanced treatment MF/RO alternative has a total present worth unit cost range of $28 to $60 million in per gallon per day of capacity. This translates to an incremental cost increase with respect to the baseline of $15 to $32 million dollars in per gallon per day treatment capacity. The key differences in cost between the baseline and the advanced treatment MF/RO are as follows:

- Larger aeration basins than the baseline to account for the longer SRT (<8 days versus >8 days).
- Additional pumping stations to pass water through the membrane facilities (MF and RO). These are based on peak flows.
- Membrane facilities (MF and RO; equipment, tanks chemical feed facilities, pumping, etc.) and replacement membrane equipment.
- Additional energy and chemical demand to operate the membrane facilities (MF and RO) and GAC.
- Zero liquid discharge facilities to further concentrate the brine reject.
- Zero liquid discharge facilities are energy/chemically intensive and they require membrane replacement every few years due to the brine reject water quality.
- An evaporation pond to handle the brine reject that has undergone further concentration by zero liquid discharge.

The advanced treatment MF/RO assumes that 100 percent of the flow is treated by MF, followed by 50 percent of the flow treated with RO. Sending a portion of flow through the RO and blending it with the balance of plant flows ensures a stable water to discharge. The RO brine reject (about 1.0 mgd) undergoes ZLD pre-treatment that further concentrates the brine reject to about 0.1-0.5 mgd. The recovery for both RO and ZLD processes is highly dependent on water quality (e.g., silicate levels).

ZLD technologies are effective at concentrating brine reject, but it comes at a substantial cost ($17.5 per gallon per day of ZLD treatment capacity of brine reject). The zero liquid discharge estimate was similar in approach to the demonstration study by Burbano and Brandhuber (2012) for La Junta, Colorado. The ability to further concentrate brine reject was critical from a management standpoint. Although 8 different options were presented for managing brine reject in Section 4.4.2, none of them is an attractive approach for handling brine reject. ZLD provides a viable pre-treatment step that requires subsequent downstream treatment. Evaporation ponds following ZLD were used for this study. Without ZLD, the footprint would be 3-5 times greater.

Roughly 30 acres of evaporation ponds, or more, may be required to handle the ZLD concentrate, depending upon concentrator effectiveness, local climate conditions, residuals accumulation, residual removal, etc. Precipitation throughout Washington is highly variable which can greatly influence evaporation pond footprint. The approach for costing the evaporation pond was in accordance with Mickley et al. (2006) and the cost was about $2.6 million.

Recent discussions with an industry installing evaporation ponds revealed that they will use mechanical evaporators to enhance evaporation rates. The use of mechanical evaporators was not included in this study, but merits consideration if a facility is performing a preliminary design that involves evaporation ponds. The mechanical evaporators have both a capital costs and annual energy costs.

Advanced Treatment MF/GAC

The advanced treatment MF/GAC alternative has a total present worth unit cost range of $36 to $78 million in per gallon per day capacity. This translates to an incremental cost increase with respect to the baseline of $23 to $50 million dollars on a per gallon per day of treatment capacity basis. The key differences in cost between the baseline and the advanced treatment MF/GAC are as follows:

- Larger aeration basins than the baseline to account for the longer SRT (<8 days versus >8 days).
- Additional pumping stations to pass water through the MF membrane and GAC facilities. These are based on peak flows.
- GAC facilities (equipment, contact tanks, pumping, GAC media, etc.)
- Additional energy to feed and backwash the GAC facilities.
- GAC media replacement was the largest contributor of any of the costs.
- Additional hauling and fees to regenerate GAC off-site.

The advanced treatment MF/GAC assumes that 100 percent of the flow is treated by MF, followed by 100 percent of the flow treated with GAC. The GAC technology is an established technology. The costing approach was in accordance with EPA guidelines developed in 1998.

The critical issue while costing the GAC technology is whether a GAC vendor/regeneration facility is located within the region. On-site regeneration is an established technology with a furnace.

However, there are several concerns as listed in Section 4.4.3:

- Ability to obtain an air emissions permit
- Additional equipment to operate and maintain
- Energy and air emissions to operate a furnace on-site
- Operational planning to ensure that furnace is operating 90-95 percent of the time. Otherwise, operations is constantly starting/stopping the furnace which is energy intensive and deleterious to equipment
- If not operated properly, the facility has the potential to create hazardous/toxic waste to be disposed

If located within a couple hundred miles, off-site regeneration is preferred. For this study, off-site regeneration was assumed with a 250-mile (one-way) distance to the nearest vendor that can provide virgin GAC and a regeneration facility.

Incremental Treatment Cost

The difference in costs between the baseline and the advanced treatment alternatives is listed in Table 10. The incremental cost to retrofit the baseline facility to the advanced treatment was calculated by taking the difference between the two alternatives. These values should serve as a planning level benchmark for understanding the potential cost for retrofitting a particular facility. The incremental cost is unique to a particular facility. Several reasons for the wide range in cost in retrofitting a baseline facility to advanced treatment are summarized as follows:

- Physical plant site constraints. A particular treatment technology may or may not fit within the constrained particular plant site. A more expensive technology solution that is more compact may be required. Alternately, land acquisition may be necessary to enlarge a plant site to allow the addition of advanced treatment facilities. An example of the former is stacking treatment processes vertically to account for footprint constraints. This is an additional financial burden that would not be captured in the incremental costs presented in Table 10.
- Yard piping. Site specific conditions may prevent the most efficient layout and piping arrangement for an individual facility. This could lead to additional piping and pumping to convey the wastewater through the plant. This is an additional financial burden that would not be captured in the incremental costs presented in Table 10.
- Pumping stations. Each facility has unique hydraulic challenges that might require additional pumping stations not captured in this planning level analysis. This is an additional financial burden that would not be captured in the incremental costs presented in Table 10.

A cursory unit cost assessment was completed to evaluate how costs would compare for facilities with lower (0.5 mgd) and higher capacity (25 mgd), as presented in Table 10. Capital costs were also evaluated for a 0.5 mgd and 25 mgd facility using non-linear scaling equations with scaling exponents. The unit capital cost for baseline conventional secondary treatment for 0.5 mgd and 25 mgd is approximately $44 and $10 per gallon per day of treatment capacity, respectively. The incremental unit costs to implement an advanced treatment retrofit for 0.5 mgd would range between $30 to $96 per gallon per day of treatment capacity and would be site and discharger specific. The incremental unit costs to implement an advanced treatment retrofit for 25 mgd would range between $10 to 35 per gallon per day of treatment capacity and would be site and discharger specific. The larger flow, 25 mgd, is not as expensive on a per gallon per day of treatment capacity. This discrepancy for the 0.5 and 25 mgd cost per gallon per day of treatment capacity is attributed to economies of scale. Cost curve comparisons (potential total construction cost and total net present value) for the baseline and the two tertiary treatment options (MF/RO and MF/GAC) are shown in Figure 8 and Figure 9 between the flows of 0.5 and 25 mgd. It is important to note that while the economies of scale suggest lower incremental costs for the larger size facilities, some aspects of the advanced treatment processes may become infeasible at larger capacities due to factors such as physical space limitations and the large size requirements for components such as RO reject brine management.

Table 10. Treatment Technology Total Project Costs in 2013 Dollars for a 0.5 mgd Facility and a 25 mgd Facility

Alternative	Total Construction Cost, 2013 dollars ($ Million)	O&M Net Present Value, 2013 dollars ($ Million)*	Total Net Present Value, 2013 dollars ($ Million)	NPV Unit Cost, 2013 dollars ($/gpd)
0.5 mgd:				
Baseline (Conventional Secondary Treatment)	15 - 32	0.5 - 1.1	15 - 33	31 - 66
Advanced Treatment – MF/RO**	27 - 58	3.2 - 6.8	30 - 65	60 - 130
Advanced Treatment – MF/GAC	33 - 70	5 - 10.8	38 - 81	76 - 162
Incremental Increase to Advanced Treatment MF/RO	12 - 26	2.7 - 5.7	15 - 32	30 - 64
Incremental Increase to Advanced Treatment MF/GAC	18 - 38	4.6 - 9.8	22 - 48	45 - 96
25 mgd:				
Baseline (Conventional Secondary Treatment)	156 - 335	25 - 54	182 - 389	7 - 16
Advanced Treatment – MF/RO**	283 - 606	157 - 336	440 - 942	18 - 38
Advanced Treatment – MF/GAC	343 - 735	252 - 541	595 - 1276	24 - 51
Incremental Increase to Advanced Treatment MF/RO	127 - 272	131 - 281	258 - 553	10 - 22
Incremental Increase to Advanced Treatment MF/GAC	187 - 401	226.9 - 486	414 - 887	17 - 35

* Does not include the cost for labor.

** Assumes zero liquid discharge for RO brine management, followed by evaporation ponds. Other options are available as listed in Section 4.4.2.

MF/RO=membrane filtration/reverse osmosis
MF/GAC=membrane filtration/granulated activated carbon
O&M=operations and maintenance
gpd=gallons per day

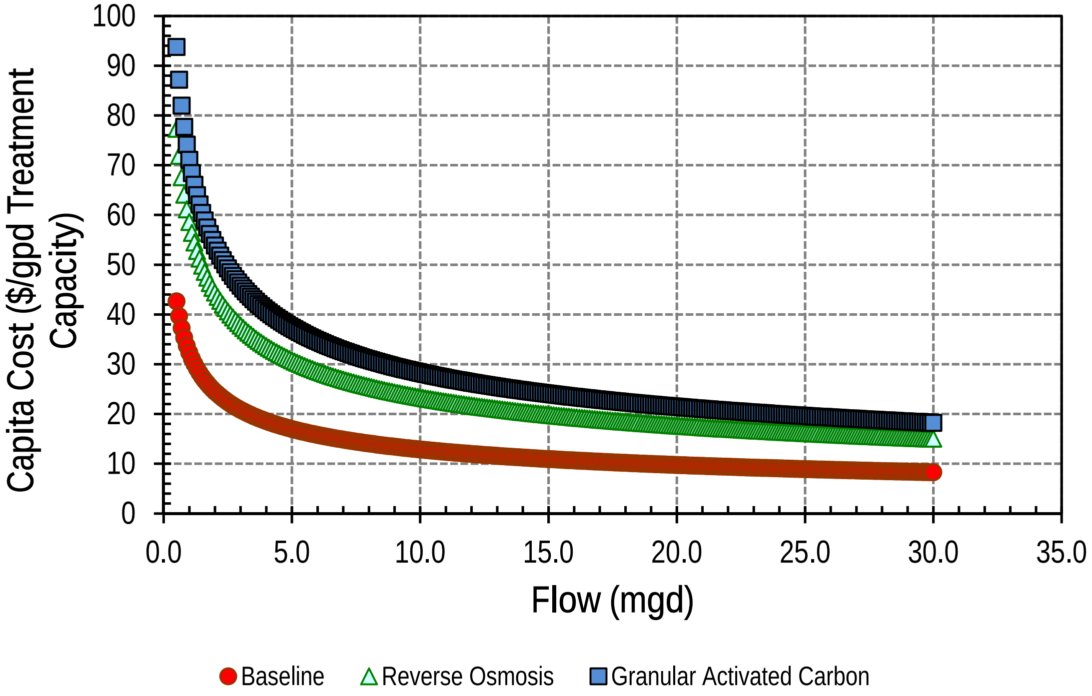

Figure 8: Capital Cost Curve Comparison for Baseline Treatment, MF/RO, and MF/GAC

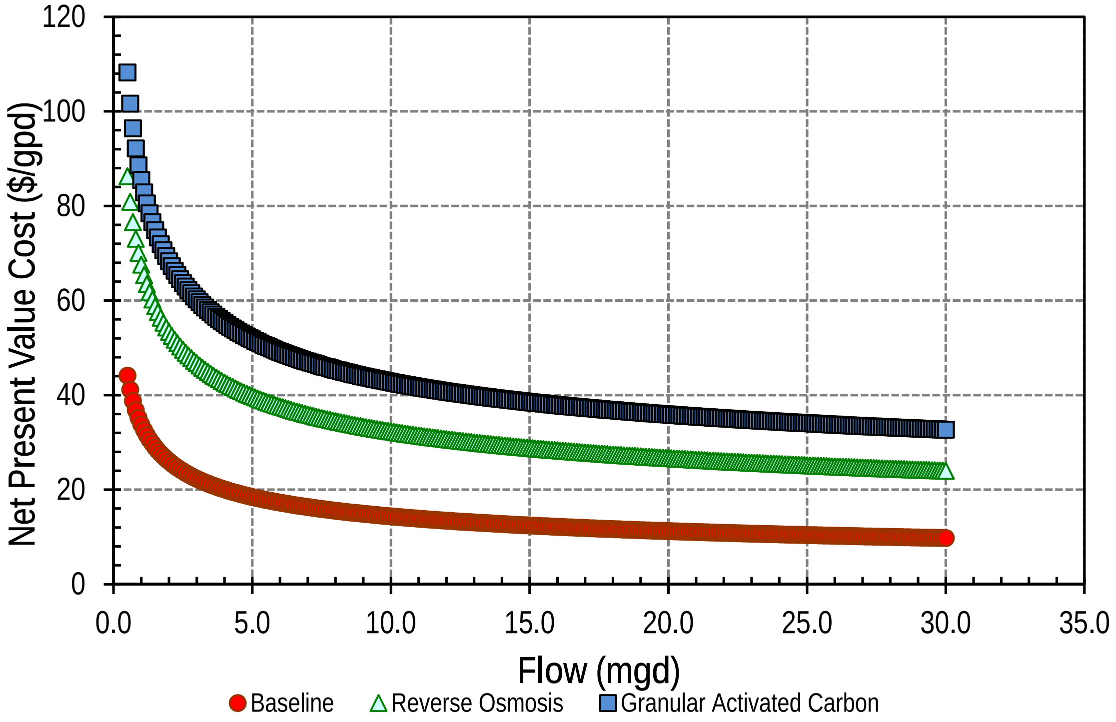

Figure 9: NPV Cost Curve Comparison for Baseline Treatment, MF/RO, and MF/GAC

4.8 Pollutant Mass Removal

An estimate of the projected load removal for the four constituents of concern was developed and is presented in Table 11. The current secondary effluent and advanced treatment effluent data is based on the only available data to HDR and is from municipal treatment plant facilities. Data is not available for advanced treatment facilities such as MF/RO or MF/GAC. Due to this lack of data, advanced treatment using MF/RO or MF/GAC was assumed to remove an additional zero to 90 percent of the constituents presented resulting in the range presented in Table 11. It is critical to note these estimates are based on limited data and are presented here simply for calculating mass removals. Current secondary effluent for industrial facilities would likely be greater than the data presented here and as a result, the projected effluent quality for industrial facilities would likely be higher as well. Based on the limited actual data from municipal treatment facilities, Table 11 indicates that mercury and BAP effluent limits may potentially be met using advanced treatment at facilities with similar existing secondary effluent quality.

Table 11. Pollutant Mass Removal by Contaminant for a 5 mgd Facility

Component	PCBs	Mercury	Arsenic	BAP
Required HHWQC based Effluent Quality (µg/L)	0.0000064	0.005	0.018	0.0013
Current Secondary Effluent Concentration (µg/L)*	0.0015	0.025	7.5	0.00031
Projected Effluent Quality (µg/L) from Advanced Treatment (MF/RO or MF/GAC)*	0.000041 – 0.00041	0.00012 – 0.0012	0.38 – 3.8	0.000029 - 0.00029
Mass Removed (mg/d)**	21 - 28	451 - 471	71,000 – 135,000	0.4 – 5.0
Mass Removed (lb/d)**	0.000045 – 0.000061	0.00099 – 0.0010	0.16 – 0.30	0.0000010 – 0.0000012

* Based on or estimated for actual treatment plant data from municipal facilities. Data sets are limited and current secondary effluent for industrial facilities would likely be greater than the data presented here.
** 1 lb = 454,000 mg
HHWQC=human health-based water quality criteria
MF/RO=membrane filtration/reverse osmosis
MF/GAC=membrane filtration/granulated activated carbon
µg/L=micrograms per liter
mg/d=milligrams per day
lb/d=pounds per day

Unit costs were developed based on required mass removal from a 5 mgd facility for each of the four constituents of concern to reduce discharges from current secondary effluent quality to the assumed required effluent quality (HHWQC). It important to note that this study concludes it is unclear if existing technology can meet the required effluent quality, however, the information presented in Table 12 assumes HHWQC would be met for developing unit costs. The unit costs are expressed as dollars in NPV (over a 25 year period) per pound of constituent removed over the same 25 year period using advanced treatment with MF/RO. The current secondary effluent quality data presented are based on typical secondary effluent quality expected for a municipal/industrial discharger. Table 12 suggests unit costs are most significant in meeting the PCB, mercury, and PAH required effluent quality.

Table 12. Unit Cost by Contaminant for a 5 mgd Facility Implementing Advanced Treatment using MF/RO

Component	PCBs	Mercury	Arsenic	PAHs
Required HHWQC based Effluent Quality (µg/L)	0.0000064	0.005	0.018	0.0013
Current Secondary Effluent Concentration (µg/L)*	0.002	0.025	7.5	0.006
Total Mass Removed (lbs) over 25-year Period	0.76	7.6	2,800	1.8
Unit Cost (NPV per total mass removed in pounds over 25 years)	$290,000,000	$29,000,000	$77,000	$120,000,000

*Derived from data presented in Table 3.
**Based on assumed 25-year NPV of $219,000,000 (average of the range presented in Table 10) and advanced treatment using MF/RO.
NPV=net present value
HHWQC=human health-based water quality criteria
µg/l=micrograms per liter

4.9 Sensitivity Analysis

The ability of dischargers to meet a HHWQC one order of magnitude less stringent (than HHWQC presented in Table 3 and used in this report) was considered. The same advanced treatment technologies using MF/RO or MF/GAC would still be applied to meet revised effluent quality one order-of-magnitude less stringent despite still not being able to meet less stringent effluent limits. As a result, this less stringent effluent quality would not impact costs. Based on available data, it appears the mercury and BAP limits would be met at a less stringent HHWQC. PCB effluent quality could potentially be met if advanced treatment with RO or GAC performed at the upper range of their projected treatment efficiency. It does not appear the less stringent arsenic HHWQC would be met with advanced treatment. It is important to note that a discharger's ability to meet these less stringent limits depends on existing secondary effluent characteristics and is facility specific. Facilities with higher secondary effluent constituent concentrations will have greater difficulty meeting HHWQC.

5.0 Summary and Conclusions

This study evaluated treatment technologies potentially capable of meeting revised effluent discharge limits associated with revised HHWQC. HDR completed a literature review of potential technologies and engineering review of their capabilities to evaluate and screen treatment methods for meeting revised effluent limits for four constituents of concern: arsenic, BAP, mercury, and PCBs. HDR selected two alternatives to compare against a baseline, including enhanced secondary treatment, enhanced secondary treatment with MF/RO, and enhanced secondary treatment with MF/GAC. HDR developed capital costs, operating costs, and a NPV for each alternative, including the incremental cost to implement from an existing secondary treatment facility.

The following conclusions can be made from this study.

- Revised HHWQC based on state of Oregon HHWQC (2001) and EPA "National Recommended Water Quality Criteria" will result in very low water quality criteria for toxic constituents.
- There are limited "proven" technologies available for dischargers to meet required effluent quality limits that would be derived from revised HHWQC.
    - Current secondary wastewater treatment facilities provide high degrees of removal for toxic constituents; however, they will not be capable of compliance with water quality-based NPDES permit effluent limits derived from revised HHWQC.
    - Advanced treatment technologies have been investigated and candidate process trains have been conceptualized for toxics removal.
        - Advanced wastewater treatment technologies may enhance toxics removal rates, however they will not be capable of compliance with HHWQC based effluent limits for PCBs. The lowest levels achieved based on the literature review were between <0.00001 and 0.00004 µg/L, as compared to a HHWQC of 0.0000064 µg/L.
        - Based on very limited performance data for arsenic and mercury from advanced treatment information available in the technical literature, compliance with revised criteria may or may not be possible, depending upon site specific circumstances.
            - Compliance with a HHWQC for arsenic of 0.018 µg/L appears unlikely. Most treatment technology performance information available in the literature is based on drinking water treatment applications targeting a much higher SDWA MCL of 10 µg/L.
            - Compliance with a HHWQC for mercury of 0.005 µg/L appears to be potentially attainable on an average basis but perhaps not if effluent limits are structured on a maximum monthly, weekly or daily basis. Some secondary treatment facilities attain average effluent mercury levels of 0.009 to 0.066 µg/L. Some treatment facilities with effluent filters attain average effluent mercury levels of 0.002 to 0.010 µg/L. Additional advanced treatment processes are expected to enhance these removal rates, but little mercury performance data is available for a definitive assessment.
        - Little information is available to assess the potential for advanced technologies to comply with revised benzo(a)pyrene criteria. A municipal wastewater treatment plant study reported both influent and effluent BAP concentrations less than the HHWQC of 0.0013 ug/L (Ecology, 2010).

  - Some technologies may be effective at treating identified constituents of concern to meet revised limits while others may not. It is therefore even more challenging to identify a technology that can meet all constituent limits simultaneously.
  - A HHWQC that is one order-of-magnitude less stringent could likely be met for mercury and PAHs however it appears PCB and arsenic limits would not be met.
- Advanced treatment processes incur significant capital and operating costs.
  - Advanced treatment process to remove additional arsenic, benzo(a)pyrene, mercury, and PCBs would combine enhancements to secondary treatment with microfiltration membranes, reverse osmosis, and granular activated carbon and increase the estimated capital cost of treatment from $17 to $29 in dollars per gallon per day of capacity (based on a 5.0 mgd facility).
  - The annual operation and maintenance costs for the advanced treatment process train will be substantially higher (approximately $5 million - $15 million increase for a 5.0 mgd capacity facility) than the current secondary treatment level.
- Implementation of additional treatment will result in additional collateral impacts.
  - High energy consumption.
  - Increased greenhouse gas emissions.
  - Increase in solids production from chemical addition to the primaries. Additionally, the membrane and GAC facilities will capture more solids that require handling.
  - Increased physical space requirements at treatment plant sites for advanced treatment facilities and residuals management including reverse osmosis reject brine processing.
- It appears advanced treatment technology alone cannot meet all revised water quality limits and implementation tools are necessary for discharger compliance.
  - Implementation flexibility will be necessary to reconcile the difference between the capabilities of treatment processes and the potential for HHWQC driven water quality based effluent limits to be lower than attainable with technology

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7.0 Appendices

- Appendix A - Unit Process Sizing Criteria
- Appendix B - Greenhouse Gas Emissions Calculation Assumptions

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APPENDIX A - UNIT PROCESS SIZING CRITERIA

Table A-1. Unit Processes Sizing Criteria for Each Alternative

Unit Process	Units	Baseline Treatment	Advanced Treatment	Comment
Influent Pumping Station	unitless	3 Times Ave Flow	3 Times Ave Flow	This is peaking factor used to size the pumps (peak flow:average flow)
Alum Dose for CEPT (optional)	mg/L	20	20	This is the metal salt upstream of the primaries
Primary Clarifiers	gpd/sf	1000	1000	This is for average annual flows
Primary Solids Pumping Station	unitless	1.25 Times Ave Flow	1.25 Times Ave Flow	This is peaking factor used to size the pumps (maximum month flow:average flow)
Aeration System Oxygen Uptake Rate (OUR)	mg/L/hr	25	25	Average annual OUR is used in tandem with mixed liquor to determine the required aeration basin volume (the limiting parameter governs the activated sludge basin volume)
Aeration Basin Mixed Liquor	mg/L	1250	2500	Average annual mixed liquor is used in tandem with OUR (see next row) to determine the required aeration basin volume (the limiting parameter governs the activated sludge basin volume)
Secondary Clarifiers Hydraulic Loading	gpd/sf	650	--	Only use for Baseline as clarifiers governed hydraulically with short SRT (<2 days)
Secondary Clarifiers Solids Loading	lb/d/sf	--	24	Only use for Advanced Treatment as clarifiers governed by solids with long SRT (>8 days)
Return Activated Sludge (RAS) Pumping Station	unitless	1.25 Times Ave Flow	1.25 Times Ave Flow	RAS must have capacity to meet 100% influent max month Flow. The influent flow is multiplied by this peaking factor to determine RAS pumping station capacity.
Waste Activated Sludge (WAS) Pumping Station	gpm	1.25 Times Ave Flow	1.25 Times Ave Flow	WAS must have capacity to meet max month WAS flows. The average annual WAS flow is multiplied by this peaking factor to determine WAS pumping station capacity.
Microfiltration (MF) Flux	gfd	--	25	Based on average annual pilot experience in Coeur D'Alene, ID
MF Backwash Storage Tank	unitless	--	1.25	Storage tanks must have capacity to meet maximum month MF backwash flows. The average annual MF backwash volume is multiplied by this peaking factor to determine required volume.

Table A-1. Unit Processes Sizing Criteria for Each Alternative

Unit Process	Units	Baseline Treatment	Advanced Treatment	Comment
MF Backwash Pumps	unitless	--	1.25	Backwash pumps must have capacity to meet maximum month MF backwash flows. The average annual MF backwash flow is multiplied by this peaking factor to determine required flows.
Reverse Osmosis (RO)	gallon per square foot per day (gfd)	--	10	
RO Reject	%	--	20	This represents the percentage of feed flow that is rejected as brine
Chlorination Dose	mg/L	15	15	
Chlorination Storage Capacity	days	14	14	
Chlorine Contact Tank	min	30	30	This is for average annual conditions.
Dechlorination Dose	mg/L	15	15	
Dechlorination Storage Capacity	days	14	14	
Gravity Belt Thickener	gpm/m	200	200	This is for maximum month conditions using the 1.25 peaking factor from average annual to maximum month
Anaerobic Digestion	Hydraulic residence time (HRT)	18	18	This is for average annual conditions
Dewatering Centrifuge	gpm	120	120	This is for maximum month conditions using the 1.25 peaking factor from average annual to maximum month

gpd=gallons per day; sf=square feet; gpm=gallons per minute

Appendix B – Greenhouse Gas Emissions Calculation Assumptions

The steady state mass balance results were used to calculate GHG emissions. The assumptions used to convert between energy demand, chemical demand and production, as well as biologically-mediated gases (i.e., CH4 and N2O) and GHG emissions are provided in Table B-1. The assumptions are based on EPA (2007) values for energy production, an adaptation of the database provided in Ahn et al. (2010) for N2O emissions contribution, Intergovernmental Panel on Climate Change (IPCC) (2006) for fugitive CH4 emissions, and various resources for chemical production and hauling from production to the wastewater treatment plant (WWTP). Additionally, the biogas produced during anaerobic digestion that is used as a fuel source is converted to energy with MOP8 (2009) recommended waste-to-energy values.

Table B-1. Greenhouse Gas Emissions Assumptions

Parameters	Units	Value	Source
N_2O to CO_2 Conversion	lb CO_2/lb N_2O	296	IPCC, 2006
CH_4 to CO_2 Conversion	lb CO_2/lb CH_4	23	IPCC, 2006
Energy Production			
CO_2	lb CO_2/MWh	1,329	USEPA (2007)
N_2O	lb N_2O/GWh	20.6	USEPA (2007)
CH_4	lb CO_2/GWh	27.3	USEPA (2007)
Sum Energy Production	lb CO_2/MWh	1336	USEPA (2007)
GHGs per BTU Natural Gas			
CO_2	lb CO_2/MMBTU Natural Gas	52.9	CA Climate Action Registry Reporting Tool
N_2O	lb N_2O/MMBTU Natural Gas	0.0001	CA Climate Action Registry Reporting Tool
CH_4	lb CO_2/MMBTU Natural Gas	0.0059	CA Climate Action Registry Reporting Tool
Sum Natural Gas		53.1	CA Climate Action Registry Reporting Tool
Non-BNR N_2O Emissions	g N_2O/PE/yr	32	Ahn et al. (2010)
BNR N_2O Emissions	g N_2O/PE/yr	30	Ahn et al. (2010)
Biogas Purity	% Methane	65	WEF, 2009
Biogas to Energy	BTU/cf CH4	550	WEF, 2009
Digester Gas to Electrical Energy Transfer Efficiency	%	32	HDR Data

Table B-1. Greenhouse Gas Emissions Assumptions

Parameters	Units	Value	Source
Chemical Production			
Alum	lb CO_2/lb Alum	0.28	SimaPro 6.0 - BUWAL250, Eco-indicator 95
Polymer	lb CO_2/lb Polymer	1.18	Owen (1982)
Sodium Hypochlorite	lb CO_2/lb Sodium Hypochlorite	1.07	Owen (1982)
Building Energy Efficiency	kBTU/sf/yr	60	Calif. Commercial End-Use Survey (2006)
Hauling Distance		-	
Local	miles	100	-
Hauling Emissions			
Fuel Efficiency	miles per gallon	8	
CO_2	kg CO_2/gal diesel	10.2	CA Climate Action Registry Reporting Tool
N_2O	kg N_2O/gal diesel	0.0001	CA Climate Action Registry Reporting Tool
CH_4	kg CH_4/gal diesel	0.003	CA Climate Action Registry Reporting Tool
Sum Hauling Fuel	kg CO_2/gal diesel	10.2	CA Climate Action Registry Reporting Tool

GWh = Giga Watt Hours
MWh = Mega Watt Hours
MMBTU = Million British Thermal Units
BTU = British Thermal Unit
PE = Population Equivalents
kBTU/sf/yr = 1,000 British Thermal Units per Square Foot per Year
cf = cubic feet
lb = pound
kg = kilogram
gal = gallon

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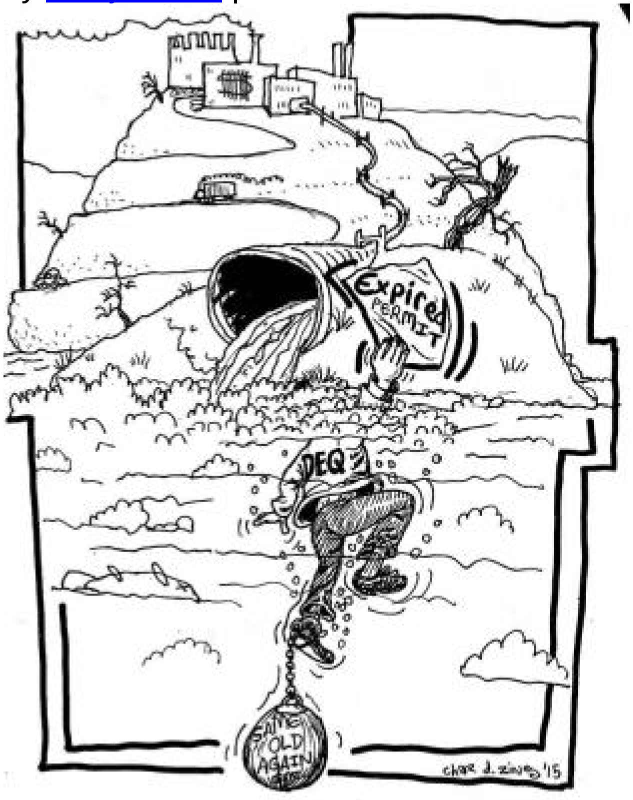
http://news.streetroots.org/2015/02/24/underwater-oregons-agency-responsible-monitoring-waterway-polluters-most-backlogged

Underwater: Oregon's agency responsible for monitoring waterway polluters is the most backlogged in the country

by Emily Green | 24 Feb 2015



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Oregon has the stiffest water pollution regulations of any state in the U.S.

Sounds impressive, right?

Think again.

Oregon is dead last nationwide when it comes to keeping wastewater permits current with state regulations. According to the EPA's most recent data on the status of each state's stormwater and wastewater permits, Oregon has the highest percentage of expired state-issued permits nationwide.

Most of the state-issued permits polluters operate under have long-passed their original expiration dates — in some cases by 20 years or more.

This means these permit holders have to be in compliance only with laws that existed when their permits were written, not with current laws. In the past two decades, numerous changes to state regulations have reduced permissible levels of toxics and temperature in wastewater.

Under the Clean Water Act of 1972, facilities and municipalities releasing wastewater directly into U.S. waterways must adhere to limits outlined in EPA or state-issued wastewater and stormwater permits. The Oregon Department of Environmental Quality issues these permits in Oregon, under the oversight of the EPA. Under the Clean Water Act, DEQ is supposed to revise water-quality standards every three years and renew each permit every five years.

But according to DEQ's current backlog count, 63 percent of existing state-issued permits for releasing stormwater and treated wastewater directly into streams and rivers are expired. An alarming 76 percent of major wastewater permits – issued to facilities with more than 1 million gallons of wastewater flowing through their treatment plants every day – also are operating on administrative extensions. Because permits are written five years before they expire, there are many municipal sewage facilities and several large industrial facilities operating under permits written in the late 1980s and early 1990s, when regulations were much less stringent than they are today.

For example, TDY Industries in Albany – a metals manufacturer that is one of the state's top polluters according to the EPA – is operating under the parameters of a wastewater permit that was written in 1988. Most of the state's municipal sewage treatment plants are operating under expired permits as well. Klamath Falls' is the oldest, written in 1990.

The amount of pollution a permit allows is based on many factors, including the number and size of other facilities contributing to the pollution of a waterway, the temperature of the water and the condition of wildlife living in the area.

"It's very circumstantial, and those circumstances can change, and they do change, as well as the scientific knowledge," says Nina Bell, an attorney who specializes in the implementation of Clean Water Act programs as the executive director of Northwest Environmental Advocates. She says when the Clean Water Act was written, Congress understood that science was going to be changing rapidly and that permits needed to keep up with it – that's why permits are supposed to be rewritten every five years.

"We have a much better idea of the science behind what pollution does to people, fish and wildlife. And that change in our scientific understanding is supposed to be reflected in our water quality standards," says Bell. "In that course of time you could also have species that have come to the brink of extinction and are listed as threatened or endangered and need the protection a new permit might afford. There could be studies that show there's contamination in fish or water that we didn't know about the last time the permit was issued. The amount of water in the stream that dilutes the pollution could change over that period of time as well – whether because there are more withdrawals or from the effects of climate change," she says.

In 2012, Oregon's waterways were ranked the 33rd most polluted in the nation, according to the Environmental Protection Agency's Toxics Release Inventory. So why is DEQ allowing polluters to operate under such outdated parameters? It's not because polluters are failing to reapply for new permits. They are applying on time, which removes their liability. According to DEQ, it just doesn't have the resources it needs to do its job.

Greg Aldrich has worked for DEQ for 15 years and is the former head of its water quality program. He's seen backlogs like the one DEQ is experiencing before. He says the program was the most backlogged in the nation in the early 2000s as well, but then DEQ ramped the water program's funding, creating more positions, and it started to catch up.

Then the recession hit.

"We took huge cuts – 19 percent cuts," says Aldrich. "It is frustrating. We get an agreement, we start ramping up, we add a stormwater program that people support, and then you start seeing cuts. Some of it is a loss of general funds, some of it is a loss of federal funds. And then the stormwater (program) – because a lot of it is paid for by industry – as they shrank, you just see the program shrink," he says.

DEQ's $52 million Water Quality Program suffered a $5.7 million reduction in the 2011-13 budget, which represented a loss of about 37 full-time positions. These cuts included seven employees among wastewater permitting and stormwater programs.

But budget isn't the only factor contributing to DEQ's backlog. Aldrich says tighter regulations make the permit writing process increasingly complex, which requires a lot more water testing and analysis. DEQ is also experiencing high turnover rates as its experts, many of whom joined the program in the early 1970s when it was created, are retiring.

Litigation in 2012 surrounding DEQ's temperature requirements added to the backlog because DEQ stopped issuing permits involving temperature regulation during that time.

Now Aldrich is DEQ's policy and analysis manager. He hopes the Legislature will help figure out a way to increase funding to the water quality program in the next biennium, because the budget package DEQ is seeking will merely sustain the program, not help it catch up, he says.

As things are, DEQ prioritizes which permits get renewed and which ones continue to be extended based on risk factors and economic needs. Aldrich says the agency always prioritizes permits for new facilities and expansions.

The last time DEQ was this far behind on water permits, it stopped inspecting and monitoring permit holders entirely and focused only on writing permits. Aldrich says this time DEQ is approaching things differently.

“We’ve been putting more effort into the compliance part,” he says. But with the workload, his environmental engineers are not always able to review all the water testing data that permit holders self-report to DEQ.

“I think what’s been challenging for us is we sometimes have this data presented to us, and we’ve not spent time looking at it to say, ‘Oh, well you even reported a problem and we didn’t follow up on it.’ That’s where it looks incredibly bad for us,” he says.

Because DEQ hasn’t been able to do its job fully, river-watching nonprofits and their attorneys have stepped in to fill the void.

Doug Quirke, founder of Oregon Clean Water Project, has been bringing litigation against facilities violating the terms of their DEQ water quality permits since 1999. He often works with nonprofits such as the Willamette Riverkeepers.

“We track permit compliance, and we bring citizen enforcement action when we find noncompliance,” says Quirke. But he can enforce only the terms of the permit any particular polluter is operating under, not current regulations.

“In a general sense, the regulations under the Clean Water Act tend to get more stringent over time, so any of these facilities with expired permits would have more stringent permits if they were issued today.” He says older permits issued in the 1990s are of particular concern. “You don’t really need to do an analysis to know that a permit that old would definitely be more stringent if it were issued today,” he says.

DEQ’s current Water Quality Program Manager Jennifer Wigal says in the last 15 years, DEQ has seen tighter regulations in several areas.

“Temperature standards have been done and redone several times over the years, which is a concern for a lot of dischargers,” she says.

Most notably, in 2011 toxic pollutants standards changed significantly, specifically regarding reductions on pollutants that affect fish and people drinking the water, she says.

The changes in 2011 came about when Oregon found that its fish consumption rate was much higher than previously thought, says Teresa Huntsinger, water program director at Oregon Environmental Council. She sits on a committee that was formed to help find solutions to DEQ’s tremendous water permit backlog in 2001. “For many years they were making progress and catching up with that backlog, but in the last few years things have been getting worse again,” she says.

Huntsinger says part of DEQ’s resource issues have to do with water permit fees.

“The wastewater plants pay an annual fee, and the increase in that fee hasn’t ever been commensurate with the staffing cost,” she says. “DEQ has a proposal this year to increase those fees, and they’re increasing them more than they usually do to catch up. The permitees have been really balking at that, which you can understand. Partly because, from their perspective, they’re saying, ‘We’re getting poor service, it’s taking you a long time to deal with our issues, and what are we going to get in response for paying increased fees? We’re still going to be getting poor service.’ So I can see that point of view, but also at the same time they’re not paying the cost of writing their permits, so their fees need to be increased.”

DEQ’s air and land programs are not backlogged in the way its water quality program is, and this is in part because funding for those programs is more reliant on fees, says Aldrich.

While most of Oregon’s water quality permits are expired, Indiana leads the nation with nearly 100 percent of its permits up to date. But it too has experienced significant backlogs. Back in 2005, there were 263 administratively extended water quality permits across the state. According to Bruno Pigott, assistant commissioner to Indiana’s Office of Water Quality, it took a statewide commitment to fixing the problem to get his department to where it is today.

When Mitch Daniels became Indiana’s governor in 2005, he made the water quality permit backlog a top priority, meeting with Indiana’s water program his second day in office. “He was very concerned about our agency’s backlog,” says Pigott. He appointed a new commissioner and required the agency report its progress to his office. “Our individual performances as well as our agency performances were judged on our ability to reduce and eliminate that backlog,” says Pigott. “We made a concerted effort – it came from the very top and came down to an organizational commitment to getting it done,” he says. His organization also ceased extended negotiation with permitees over the parameters of permits. “If they said they were going to appeal a permit, we said OK, go ahead,’” he says.

But Oregon has not given DEQ the funding that would be necessary to see the kind of progress that states like Indiana have made in reducing backlogs. And that means stringent water quality requirements are an unreachable ideal, not a reality.

Last year Siltronic Corp., a semiconductor manufacturer in Portland, made headlines when the EPA listed it as the top waterway polluter in the state for 2012. Its wastewater permit expired in June, and the DEQ regulator responsible for its permit, Mer Wiren, says she has no idea when it will be renewed.

Wiren, is one of only two employees charged with monitoring, inspecting and writing permits for all 55 major industrial wastewater permit holders in DEQ’s Northwest Region of Oregon. She says Siltronic applied for the renewal six months ahead of time like it was supposed to, and she issued an “indefinite administrative renewal” that will keep it legal until she rewrites the permit. Additionally, as a major facility, Siltronic is supposed to receive an on-sight inspection every two years, but received no inspection between 2010 and 2014.

But the pollutant that earned Siltronic its place at the top of Oregon’s polluters, nitrate compounds, isn’t even limited under the terms of its current permit.

When told about the EPA's ranking of Siltronic due to nitrates, Wiren says, "That's a new piece of information to me, but if it's a concern, we can look at that."

Wiren says in the five years she's worked as an environmental engineer at DEQ, she's never seen a water quality permit renewed on time.

Siltronic spokesman Christof Bachmair says Siltrnoic has been working at reducing its pollution. He said the company has effectively reduced nitrate compound emissions by 90 percent since 2000.

Bachmair also pointed out that many facilities with wastewater permits discharge to wastewater treatment plants, not directly into the river.

"Some of these companies are discharging more nitrates than Siltronic," he says. "In addition, the water treatment plants receive residential waste, which also contain nitrates. Again, these nitrates are typically not treated, nor are they reported to EPA as a part of the Toxic Release Reporting program. So in our view, the true picture of discharge rankings is not clearly portrayed by the EPA reports," he says.

Portland's Environmental Services spokesman Linc Mann confirmed that there are no specific requirements at the Columbia Boulevard and Tryon Creek wastewater treatment plants to remove nitrates and other nitrogen compounds. According to the EPA, nitrates can cause people to become seriously ill if high levels are present in drinking water. Nitrate compounds can also be harmful to the health of aquatic organisms and fish.

In DEQ's Northwest Region, covering Multnomah, Clackamas, Washington, Columbia, Tillamook and Clatsop counties, the most outdated permits belong to the Port of St. Helens, written in 2003, and StarLink Logistics Inc., a Portland agricultural chemicals manufacturer that's permit was renewed in 2003.

The EPA ultimately is responsible for Oregon's permit backlog. It oversees DEQ, and gave it the authority to issue and monitor water quality permits. Quirke says any lawsuit resulting from someone getting sick from pollution emitted under the parameters of an old permit ultimately would be directed at the EPA.

"The main plan of attack for this sort of thing that I've seen is to try to get EPA to pull the approval of the state program, and if it actually did happen, that means that the administration of the program would get taken away and kicked back to EPA," says Quirke.

The EPA has never fully revoked a state's authority to issue permits, and to do so in this case wouldn't be in Oregon's best interest, says Christine Psyk, associate director of the EPA's regional wastewater permitting unit. "We've had conversations with Oregon about their backlog," she says, "but we don't have a standing workforce to come in and take over permitting." She says the EPA is working with Oregon to find ways to get caught up.

As the permitting process becomes more and more complex, with some wastewater permits filling an entire six-inch binder, looking at ways to streamline the process and providing roadmaps to permit writers are areas of focus, she says.

While Oregon may be the farthest behind on issuing permits, it’s backlog problem is not unique. “Everyone has somewhat of a backlog,” says Psyk. “The EPA has a backlog, too.”

STATE OF WASHINGTON
DEPARTMENT OF ECOLOGY
15 W Yakima Ave, Ste 200 • Yakima, WA 98902-3452 • (509) 575-2490

January 7, 2010

Al Jensen
Facility Manager
U.S. Fish and Wildlife Service
12790 Fish Hatchery Road
Leavenworth, WA 98826

**RE: Final 401 Certification for the Leavenworth National Fish Hatchery
Order No. 7192**

Dear Mr. Jensen:

Enclosed is Order number 7192, in the matter of granting a Water Quality Certification to U.S. Fish and Wildlife Service. The certification includes limits and treatment requirements for wastewater discharges, and monitoring and reporting requirements pertaining to water temperature and fish habitat in Icicle Creek affected by hatchery operations.

All correspondence relating to this document should be directed to Pat Irle, Department of Ecology, 15 W. Yakima Avenue, Suite 200, Yakima, WA 98902. If you have any questions concerning the content of the document, please call Pat, at 509/454-7864.

Sincerely,

Charles McKinney

Charles McKinney, Section Manager
Water Quality Program
Central Regional Office

Enclosure: Order No. 7192
1-30-09 Flow Management Plan
Schedule of required studies and reports (Table 1)

By Registered Mail

cc: Steve Croci, Leavenworth National Fish Hatchery
Michael Lidgard, Environmental Protection Agency, Region 10
Tom Tebb, Ecology-Central

IN THE MATTER OF GRANTING A
WATER QUALITY
CERTIFICATION TO
U.S. Fish and Wildlife Service
1790 Fish Hatchery Road
Leavenworth, WA 98826
in accordance with 33 U.S.C. 1341
(FWPCA § 401), RCW 90.48.120, RCW
90.48.260 and Chapter 173-201A WAC

ORDER No. 7192
Certification of the Leavenworth National Fish
Hatchery (NPDES Permit No. WA-000-190-2)
on Icicle Creek, Chelan County, Washington.

TO: **Al Jensen, Facility Manager**
U.S. Fish and Wildlife Service
12790 Fish Hatchery Road
Leavenworth, WA 98826

The Leavenworth National Fish Hatchery (Leavenworth NFH) is required to have a Clean Water Act (CWA) National Pollutant Discharge Elimination System (NPDES) Permit issued by the U.S. Environmental Protection Agency (EPA) authorizing the discharge of wastewater. In 2005, the U.S. Fish and Wildlife Service (USFWS), which manages and operates the Leavenworth NFH, applied to EPA to renew its NPDES Permit. On June 26, 2006, EPA issued a draft NPDES Permit and associated fact sheet for the Leavenworth NFH. On January 15, 2008, Washington State Department of Ecology (Ecology) received an application from the USFWS requesting a CWA Section 401 water quality certification (401 Certification), 33 USC §1341, for the draft NPDES permit. Pursuant to Ecology's request, the USFWS prepared a Tier II Water Quality Analysis for the Leavenworth NFH and submitted it to Ecology on April 30, 2008. On January 9, 2009, Ecology received a request from the USFWS to withdraw and re-apply for a 401 Certification. Ecology has determined that the project has and is likely to continue to violate water quality standards. This Order is intended to ensure that its continued operation does not create future violations. If operated consistent with the terms of this Order, Ecology has reasonable assurance that the project will meet water quality standards. This document represents Ecology's Section 401 water quality certification and ch. 90.48 RCW order (Order) for the Leavenworth NFH. For purposes of this Order, USFWS and Leavenworth NFH will be referred to as Leavenworth NFH.

PROJECT DESCRIPTION

The Leavenworth NFH is located on Icicle Creek, a tributary to the Wenatchee River, at river mile (RM) 3.0 near Leavenworth, Washington. The Leavenworth NFH was authorized as mitigation for the construction of Grand Coulee Dam and is used to capture, spawn, and rear approximately 1.2 million spring Chinook salmon at 70,000 pounds and acclimate coho salmon for a total weight gain of less than 10,000 pounds annually. According to the draft NPDES permit, the Project has had a total daily average discharge of 26 million gallons per day and the main pollutants of concern are nitrogen, phosphorus, settleable solids (SS), total suspended solids (TSS), dissolved oxygen (DO), pH, temperature, and total residual chlorine (EPA, draft NPDES permit). Water is discharged from the hatchery operations at two locations: (1) from the rearing ponds and raceways via combined Outfalls 001 and 004; and (2) from the Pollution Abatement Pond, Outfall 002.

AUTHORITIES

In exercising authority under 33 U.S.C. § 1341, RCW 90.48.120, and RCW 90.48.260, Ecology has reviewed this application pursuant to the following:

1. Conformance with applicable water quality-based, technology-based, and toxic or pretreatment effluent limitations as provided under 33 U.S.C. §§1311, 1312, 1313, 1316, and 1317 (FWPCA §§ 301, 303, 306 and 307);

2. Conformance with the state water quality standards contained in Chapter 173-201A WAC and authorized by 33 U.S.C. §1313 and by Chapter 90.48 RCW, and with other applicable state laws; and

3. Conformance with the provision of using all known, available and reasonable methods to prevent and control pollution of state waters as required by RCW 90.48.010.

4. Conformance with RCW 90.54.020(3)(a) and (b).

WATER QUALITY CERTIFICATION CONDITIONS

Through issuance of this Order, Ecology certifies that it has reasonable assurance that the activity as proposed and conditioned will be conducted in a manner that will comply with applicable water quality standards and other appropriate requirements of state law. In view of the foregoing and in accordance with 33 U.S.C. §1341, RCW 90.48.120, RCW 90.48.260, Chapter 173-200 WAC and Chapter 173-201A WAC, water quality certification is granted to the Leavenworth NFH subject to the conditions within this Order.

Certification of this project does not authorize the Leavenworth NFH to exceed applicable state water quality standards (Chapter 173-201A WAC), ground water quality standards (Chapter 173-200 WAC) or sediment quality standards (Chapter 173-204 WAC). Furthermore, nothing in this certification shall absolve Leavenworth NFH from liability for contamination and any subsequent cleanup of surface waters, ground waters or sediments occurring as a result of project operations.

A. General Conditions

1. For purposes of this Order, the term "Applicant" shall mean the Leavenworth NFH and its agents, assignees and contractors.

2. For purposes of this Order, all submittals required as conditions shall be sent to the Section Manager, Water Quality Program, Washington State Department of Ecology, Central Regional Office, 15 West Yakima Ave., Suite 200, Yakima, WA 98902 or via e-mail (preferred), if possible, to the Section Manager, Water Quality Program for Ecology's Central Regional Office. Notifications shall be made via phone or e-mail (preferred). All submittals and notifications shall be identified with Order No. 7192 and include the Applicant's name, project name, project location, the project contact and the contact's phone number.

3. Conditions B.6, B.7, B.8, and B.9 shall be deemed denied if the EPA does not issue a NPDES Permit for the Leavenworth NFH within two years from the date of the Order.

4. Copies of this Order shall be kept on the site and readily available for reference by staff of the Leavenworth NFH, its contractors and consultants, Ecology personnel, and state and local government inspectors.

5. The Leavenworth NFH shall ensure that all project staff and other workers at the project site with authority to direct work have read and understand relevant conditions of this Order and all permits, approvals, and documents referenced in this Order.

6. The Leavenworth NFH shall provide access to the project site and all monitoring sites upon request by Ecology personnel for site inspections, monitoring, necessary data collection, and/or to ensure that conditions of this Order are being met.

7. Nothing in this Order waives Ecology's authority to issue additional orders if Ecology determines that further actions are necessary to implement the water quality laws of the state. Further, Ecology retains continuing jurisdiction to make modifications hereto through supplemental order, if additional impacts due to project operation are identified (*e.g.*, violations of water quality standards, downstream erosion, etc.), or if additional conditions are necessary to further protect water quality.

8. All applications, reports, or information submitted to Ecology shall be signed and certified.

    a. All permit applications shall be signed by the project staff or manager with authority to act for USFWS.

    b. All reports required by this permit and other information requested by Ecology shall be signed by a person described above or by a duly authorized representative of that person. A person is a duly authorized representative only if:

        i. The authorization is made in writing by a person described above and submitted to Ecology.

        ii. The authorization specifies either an individual or a position having responsibility for the overall operation of the regulated facility, such as the position of plant manager, superintendent, position of equivalent responsibility, or an individual or position having overall responsibility for environmental matters. (A duly authorized representative may thus be either a named individual or any individual occupying a named position.)

    c. Changes to authorization. If an authorization under paragraph A.8.b.ii. is no longer accurate because a different individual or position has responsibility for the overall operation of the facility, a new authorization satisfying the requirements of paragraph A.8.b.ii. shall be submitted to Ecology prior to or together with any reports, information, or applications to be signed by an authorized representative.

    d. Certification. Any person signing a document under this section shall make the following certification:

> I certify under penalty of law, that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified personnel properly gathered and evaluated the information submitted. Based on my inquiry of the person or persons who manage the system, or those persons directly responsible for gathering information, the information submitted is, to the best of my knowledge and belief, true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment for knowing violations.

9. This Order does not authorize direct, indirect, permanent, or temporary impacts to waters of the state or related aquatic resources, except as specifically provided for in conditions of this Order.

10. Failure of any person or entity to comply with the Order may result in the issuance of civil penalties or other actions, whether administrative or judicial, to enforce the terms of this Order.

B. Specific Conditions

1. <u>Icicle Creek Stream Flow</u>

    a. <u>Flow Management</u>. The Leavenworth NFH shall comply with all of the provisions of the Leavenworth National Fish Hatchery Proposed Flow Management Operations 2009-2014 (Proposed Flow Management Plan), dated January 30, 2009, until replaced with a final plan approved by Ecology in writing. A copy of the Flow Management Plan is attached as Appendix A. Compliance shall include completion of all studies to address the information needs identified in the Proposed Flow Management Plan. All studies required by the Proposed Flow Management Plan shall be submitted to Ecology for its review.

    b. <u>Ramping Rates</u>. When adjusting Structure 2, flow ramping rates shall not exceed one inch per hour. If the condition or capability of Structure 2 does not reliably allow stage adjustments as fine as one inch per hour, adjustments should be made in the finest increment possible and field observation of the historical channel or the hatchery canal, as appropriate, should be performed to verify that fish stranding is minimized.

    c. <u>Flow Monitoring</u>. The Leavenworth NFH shall prepare the following flow monitoring plans, in accordance with the approved Quality Assurance Project Plan (QAPP), described in paragraph D, and implement them upon Ecology's approval.

        i. <u>Historic channel and hatchery canal</u>. Within four months of issuance of this Order, the Leavenworth NFH shall prepare a plan describing how stream flow will be monitored in the hatchery canal and the historic Icicle Creek channel. The plan shall include a description of the monitoring equipment to be used, the monitoring frequency, and the procedures for such monitoring. The plan shall be submitted to Ecology for its review and written approval. The plan shall be implemented within 90 days of its approval by Ecology.

ii. Snow/Nada Lake. Within four months of issuance of this Order, the Leavenworth NFH shall prepare a plan for monitoring releases from Snow and Nada Lakes to supplement Icicle Creek flow below USGS Gage 12458000. The plan shall be submitted to Ecology for its review and written approval. The plan shall be implemented within 90 days of its approval by Ecology.

d. Annual Flow Monitoring Report. By December 31 of each year following approval of the flow monitoring plans, the Leavenworth NFH shall submit an annual report to Ecology compiling the flow monitoring data for the prior water year (October 1 – September 30).

e. Implementation. Within four years of the issuance of this Order, the Leavenworth NFH shall submit a Final Flow Management Plan. Once approved by Ecology, Leavenworth NFH shall implement the Final Flow Management Plan on the schedule set forth in the approved plan.

2. IFIM Study.

a. Within 18 months of issuance of this Order, the Leavenworth NFH shall prepare an Instream Flow Incremental Methodology (IFIM) study plan (IFIM Study Plan) for the historic channel consistent with the requirements of "Instream Flow Guidelines: Technical and Habitat Suitability Issues," Publication No. 04-11-007 (WDFW and Ecology, updated 2/12/2008), or an equivalent method or methodology approved by Ecology. The IFIM Study Plan shall include an implementation schedule. Upon completion, the IFIM Study Plan shall be submitted to Ecology for its review and written approval.

b. Within three months of approval by Ecology, the Leavenworth NFH shall implement the IFIM Study Plan.

c. Within three years of the issuance of this Order, the Leavenworth NFH shall submit a report of the IFIM study results to Ecology. The study results shall include flow recommendations. Once approved by Ecology, USFWS shall incorporate the flow recommendations into the proposed Final Flow Management Plan.

3. Fish Passage.

a. Within one year of issuance of this Order, the Leavenworth NFH shall submit a plan to investigate stream flow management and structural options for improving fish passage at (a) the intake structure, (b) Structure 2, and (c) Structure 5.

The investigation shall address the following:

- Determine flows needed for upstream and downstream passage of all swimming stages of native species;
- Investigate a long-term solution for year-round passage that may include modifications to the structure; and

- Analyze the potential for fish stranding due to ramping rates employed for operating Structure 2.

b. Within three years of issuance of this Order, the Leavenworth NFH shall submit a report summarizing the results of the study and recommending flow management and structural options for improving fish passage.

c. Implementation. Once approved by Ecology, USFWS shall incorporate the flow recommendations from the plan into the Final Flow Management Plan and implement the fish passage plan in accordance with the schedule set forth in the approved plan.

4. Hatchery Canal Hydrologic Functions.

a. Aquifer Recharge. The Leavenworth NFH shall prepare a plan to quantify the timing, rate, and volume of water needed in the hatchery canal to provide sufficient recharge to maintain water levels in the shallow aquifer within the range of variation exhibited during the 1945-1980 period. The Aquifer Recharge Plan shall be submitted to Ecology for its review and written approval.

b. Flood Control. The aquifer recharge plan shall also include a description of the operation of Structure 2 for the purpose of flood control.

c. Within three years of the issuance of this Order, Leavenworth NFH shall submit the Aquifer Recharge Plan and the Flood Control operating rules to Ecology for its review and approval. Once approved by Ecology, USFWS shall incorporate the flow recommendations from these two plans into the Final Flow Management Plan.

5. Navigation and Boating.

Within 24 months of issuance of this order, the Leavenworth NFH shall submit a report evaluating approaches or methods to provide navigation and boating through the natural channel reach from structure 2 through structure 5.

6. Temperature and Flow Monitoring.

a. Water used in hatchery operations. In accordance with the Quality Assurance Project Plan prepared pursuant to the requirements of paragraph D, the Leavenworth NFH shall monitor the temperature and flow of (a) each groundwater well; (b) surface water entering the hatchery; and (c) hatchery discharges to Icicle Creek. To the extent that flow monitoring required by this section coincides with or duplicates flow monitoring required in section B.1, the monitoring frequencies and locations should be reconciled with the flow monitoring plans required in section B.1.c.

i. Monitoring frequency. Temperature shall be monitored hourly from October 1st through September 30th.

ii. Monitoring report. By December 31st of each year, the Leavenworth NFH shall prepare and submit to Ecology an annual report summarizing its monitoring results. In addition to the temperature and flow data collected, the report shall include an analysis comparing the measured temperature of discharged water to the temperature calculated by flow-averaging the well water and surface water used by the hatchery.

b. Instream Temperature. Within two years of the issuance of this Order, the Leavenworth NFH shall submit a Temperature Study Plan to evaluate measures to reduce temperatures in Icicle Creek. The Temperature Study Plan shall include a QAPP consistent with the requirements of paragraph D for monitoring water temperatures at appropriate locations and frequencies and shall be submitted to Ecology for its review and written approval.

i. Plan Contents. The Temperature Study Plan shall include an evaluation of measures to:

- Lower temperatures in Icicle Creek to temperatures that would occur under natural conditions, focusing on the critical period between June and October.
- Meet the site-potential shade throughout the length of the historic river channel and hatchery canal.

ii. Plan Review and Approval. Within four years of the issuance of this Order, the Leavenworth NFH shall submit a report describing the results of the above study, including the environmental impacts, feasibility, costs, and potential schedules for implementation of each feasible alternative. Once approved by Ecology, the Leavenworth NFH shall within 180 days prepare and submit an Implementation Plan for review and written approval by Ecology.

iii. Implementation. Upon Ecology's approval of the Implementation Plan, Leavenworth NFH shall implement it in accordance with the schedule set forth in the approved Implementation plan.

7. Dissolved Oxygen and pH. Portions of the Wenatchee River, lower Icicle Creek, and other tributaries do not meet water quality standards for Dissolved Oxygen and pH during the critical period identified in the Wenatchee River pH and Dissolved Oxygen TMDL, Ecology Publication No. 08-10-062, August 2009. Phosphorus levels in the Wenatchee River watershed must be reduced to improve DO and pH levels to protect aquatic resources and designated uses. In the TMDL, the Leavenworth NFH was allocated a waste load of 0.52 kg/day total phosphorous.

a. Standards. The Leavenworth NFH shall meet a discharge limit of 0.52 kg/day total phosphorous within five years of permit issuance.

b. Monitoring. The Leavenworth NFH shall prepare and implement a QAPP consistent with the requirements of paragraph D to monitor phosphorus levels in its effluent and Icicle Creek flows upstream of the hatchery. Icicle Creek flow monitoring for calculating total phosphorous concentrations and mass should be reflected in the development of the flow monitoring required in paragraph B.5. Results shall be provided in both concentrations and total mass.

8. Toxics. The Leavenworth NFH shall prepare and implement a QAPP consistent with the requirements of paragraph D to monitor sediment deposited through hatchery operations for the presence of toxic pollutants. The QAPP shall include the following elements:

   a. Every 12 months, sample and analyze sediment in the pollution abatement pond for total PCBs, hexacholorcyclohexanes and endrin.

   b. Once during the five years subsequent to issuance of this order, sample and analyze the sediment in the pollution abatement pond, above the hatchery intake, and below the hatchery outfall for total PCB, endrin, endosulfan II and DDT.

   c. Provisions for the removal and proper disposal of all sediment in the pollution abatement pond in the event that any sample exceeds its Apparent Effects Threshold (Ecology 2003) for the identified parameter.

9. Turbidity. The Leavenworth NFH shall conduct turbidity monitoring when cleaning sediments from (a) the sand settling basin, (b) conveyance channel, (c) behind the fish screens, and (d) the pollution abatement pond.

   a. Sampling analysis and method. Turbidity analysis shall be performed with a calibrated turbidity meter (turbidimeter), either on-site or at an accredited lab. The results shall be recorded in a site log book in Nephelometric Turbidity Units (NTU).

   b. Sampling locations. Sampling is required at all discharge points where water used to clean sediments is being discharged back into surface waters. Background samples shall also be gathered to ensure that the discharge meets the requirements of WAC 173-201A-200 (discharge not to exceed five NTU above background if turbidity is 50 NTU or less, or 10 percent above background if background turbidity is greater than 50 NTU).

   c. Reporting. A copy of sampling results shall be submitted to Ecology within one month of each cleaning event.

C. Aquatic Workgroup.

1. Aquatic Workgroup Participation. Ecology will periodically convene a workgroup of federal, state, and tribal fisheries co-managers for consultation prior to its approval of the several flow- and aquatic resource-related studies, plans, or reports required by this Order. Workgroup members are not obligated to participate. A proposed workgroup member may request Ecology to consult with them separately.

2. Workgroup Composition. Members of the workgroup are USFWS, Ecology, the Yakama Nation, the Colville Confederated Tribes, National Marine Fisheries Service, and Washington Department of Fish and Wildlife.

3. Facilitation. Ecology, or another member of the workgroup, may serve as facilitator for the workgroup.

4. Study Procedures. A summary of the study plans, study reports and implementation plans required under this subsection is provided in Table 1. These documents shall be prepared according to the schedule provided in Table 1 or as modified by Ecology in writing. Where possible, the studies may be done in conjunction with other EPA-required plans and reports associated with the NPDES permit.

5. Plan Preparation. The Leavenworth NFH shall be responsible for preparing proposed study plans and study reports, with any proposed implementation measures, required by this Order. Ecology reserves the right to make the final determination as to the adequacy of the final products required by this 401 Certification.

D. Quality Assurance Project Plan

1. Plan Preparation. The Leavenworth NFH shall prepare a Quality Assurance Project Plan (QAPP) for monitoring of each parameter required to be monitored under this Order, including flow, temperature, phosphorus, turbidity and toxics.

2. QAPP Contents. The QAPP shall be prepared in accordance with the Guidelines for Preparing Quality Assurance Project Plans for Environmental Studies (Ecology Publication Number 04-03-030, July 2004) or its successor. The QAPP shall contain, at a minimum, the list of parameters to be monitored, a map of sampling locations, and descriptions of the purpose of the monitoring, sampling frequency, sampling procedures and equipment, analytical methods, quality control procedures, data handling and data assessment procedures, and reporting protocols.

3. Use of Best Available Science. In preparing the study plans and study reports, and in developing implementation measures to address Project impacts, the Leavenworth NFH shall use the most current and best available scientific information and analysis. When all implementation options are approximately equal in their biologic and hydrologic effectiveness, the Leavenworth NFH may also consider, in evaluating the options, (a) time required to achieve that success, and (b) cost effectiveness of solution.

4. Submittal to Ecology. The draft QAPPs shall be submitted to Ecology for its review and written approval. Once approved, Leavenworth NFH shall follow the approved QAPP for the collection and management of data and analyses for the reports and studies required in section B of this Order.

E. Appeal Process

You have a right to appeal this Order. To appeal this you must:

- File your appeal with the Pollution Control Hearings Board within 30 days of the "date of receipt" of this document. Filing means actual receipt by the Board during regular office hours.
- Serve your appeal on the Department of Ecology within 30 days of the "date of receipt" of this document. Service may be accomplished by any of the procedures identified in WAC 371-08-305(10). "Date of receipt" is defined at RCW 43.21B.001(2).

Be sure to do the following:

- Include a copy of this document that you are appealing with your Notice of Appeal.

- Serve and file your appeal in paper form; electronic copies are not accepted.

1. To file your appeal with the Pollution Control Hearings Board

Mail appeal to:

The Pollution Control Hearings Board
PO Box 40903
Olympia, WA 98504-0903

OR

Deliver your appeal in person to:

The Pollution Control Hearings Board
4224 – 6th Ave SE Rowe Six, Bldg 2
Lacey, WA 98503

2. To serve your appeal on the Department of Ecology

Mail appeal to:

The Department of Ecology
Appeals & Application for Relief Coordinator
PO Box 47608
Olympia, WA 98504-7608

OR

Deliver your appeal in person to:

The Department of Ecology
Appeals & Application for Relief Coordinator
300 Desmond Dr SE
Lacey, WA 98503

3. And send a copy of your appeal to:

Section Manager
Department of Ecology
Central Regional Office
15 W. Yakima Avenue, Suite 200
Yakima, WA 98902

For additional information visit the Environmental Hearings Office Website: http://www.eho.wa.gov
To find laws and agency rules visit the Washington State Legislature Website: http://www.leg.wa.gov/CodeReviser

Your appeal alone will not stay the effectiveness of this Order. Stay requests must be submitted in accordance with RCW 43.21B.320. These procedures are consistent with Ch. 43.21B RCW.

DATED this 7th day of January, 2010 at Yakima, WA 98902.

Charles McKinney

Charles McKinney, Section Manager
Water Quality Program
Central Regional Office

WASHINGTON STATE
DEPARTMENT OF
ECOLOGY

Persistent Organic Pollutants in Feed and Rainbow Trout from Selected Trout Hatcheries

April 2006

Publication No. 06-03-017

04681

This report is available on the Department of Ecology home page on the World Wide Web at www.ecy.wa.gov/biblio/0603017.html

Data for this project are available at Ecology's Environmental Information Management (EIM) website at www.ecy.wa.gov/eim/index.htm. Search User Study ID, DSER0015.

Ecology's Project Code for this study is 05-056.

For a printed copy of this report, contact:

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Persistent Organic Pollutants in Feed and Rainbow Trout from Selected Trout Hatcheries

by
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Environmental Assessment Program
Olympia, Washington 98504-7710

in cooperation with

Washington Department of Fish and Wildlife
Fish Program, Hatcheries Division
Olympia, Washington 98501-1091

April 2006

Waterbody numbers: Statewide

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Abstract

During 2005, persistent organic pollutants (POPs) were analyzed in fish feed and catchable rainbow trout from ten Washington Department of Fish and Wildlife (WDFW) hatcheries and the fish purchased by WDFW from one private hatchery operator. Fish originating from the same hatchery populations were also sampled approximately 2½ months following planting into unpolluted lakes in order to assess contaminant depuration or uptake. All feed and tissue (fillet) samples were analyzed for a variety of chlorinated pesticides, polychlorinated biphenyls (PCBs), a select group of polybrominated diphenyl ethers (PBDEs), and lipid content. A subset of feed and tissue samples was also analyzed for polychlorinated dioxins and furans (PCDD/Fs).

Feed samples had the following mean wet weight concentrations: ΣPCBs[1] – 13.8 ng/g, ΣDDT – 8.2 ng/g, ΣPBDEs – <0.25 ng/g, PCDD/F toxic equivalent – 0.75 pg/g. Fish tissue samples had the following respective mean wet weight concentrations in hatchery and planted rainbow trout: ΣPCBs – 13.0 and 3.1 ng/g, ΣDDT – 3.9 and 8.8 ng/g, ΣPBDEs – 0.66 ng/g for both. PCDD/F toxic equivalent averaged 0.032 pg/g in hatchery fish but was not analyzed in trout collected from lakes.

Other pesticides found in feed (f), hatchery fish (h), and planted fish (p) were: DDMU (f,h,p), dieldrin (f,h,p), hexachlorobenzene (f,h,p), pentachloroanisole (f,h,p), trans-nonachlor (f,h), cis-chlordane (f,h), trans-chlordane (f), methoxychlor (f), and toxaphene (f).

Results suggest that some portion of POP concentrations in trout from unpolluted waters may originate from hatcheries. In addition, some catchable trout contain POP concentrations above regulatory criteria when they are planted in lakes.

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[1] Σ = Total

Acknowledgements

The authors would like to thank the following individuals for their contributions:

Staff with the Washington Department of Fish and Wildlife:

- John Kerwin, Hatchery Division Manager, helped conceive and develop this project
- Managers and staff of the trout hatcheries provided samples and answered our inquiries
- District biologists provided valuable suggestions and information on lakes and their trout populations
- Jeff Korth and Patrick Verhey helped collect fish in Warden Lake
- John Sneva aged the specimens
- John Kerwin and Sandie O'Neill reviewed the report

Staff with the Washington State Department of Ecology:

- Karin Feddersen contracted and reviewed the dioxin/furan analysis
- Dean Momohara conducted the lipid analysis
- Will White and Pam Covey handled and tracked samples
- Paul Anderson, Randy Coots, Casey Deligeannis, Brandee Era-Miller, Brandi Lubliner, Brenda Nipp, Brian Reese, Kirk Sinclair, and Trevor Swanson assisted with the collection and processing of fish samples
- Dale Norton and Art Johnson reviewed the report
- Carol Norsen and Joan LeTourneau edited and formatted the sampling plan and final report, respectively

Other contributors:

- Corey Bray assisted with fish collection
- Dave McBride, Washington State Department of Health, provided useful comments

Introduction

Recent reports have indicated that commercially farmed salmon, hatchery-raised trout, and the feed used to grow them may contain polychlorinated biphenyls (PCBs) and other persistent organic pollutants (POPs). For instance, Hites et al. (2004) showed that salmon raised in net-pens had substantially higher PCBs than those caught wild, presumably due to PCB-contaminated feed. Carline et al. (2004) found that concentrations of PCBs in hatchery rainbow trout (*Oncorhynchus mykiss*) fillets were correlated to concentrations in feed, and nearly all the body burden was due to PCBs in the diet. Other investigations have revealed detectable concentrations of dioxins, dieldrin, and endrin as well as PCBs in hatchery broodstock salmon and trout (Millard et al., 2004). In Pennsylvania, PCB contamination of edible tissues accumulated through dietary uptake in hatcheries exceeded thresholds for issuance of consumption advisories (Carline et al., 2004).

Currently there is no statewide program in Washington to evaluate toxic chemicals in hatchery feed or hatchery fish. At the same time, low levels of POPs in fish from lakes and streams across the state are being detected at an increasing rate (e.g., Seiders 2003; Seiders and Kinney, 2004) due to increased sampling coverage and better analytical detection limits. These waterbodies are often added to the list of impaired waters as required by the federal Clean Water Act section 303(d), and subsequently require a plan to control or clean up the contaminants. Many of the POPs found in fish tissue (e.g., PCBs, dioxins) are ubiquitous environmental contaminants and may be found globally through atmospheric deposition, historical releases, or food-web cycling. Fish may accumulate low concentrations of these chemicals through one or more of these pathways, although it is nearly impossible to distinguish and quantify these diffuse sources, and control and clean-up is often unrealistic. Due to recent data, however, contamination stemming from hatcheries is now considered a possible source of POPs in fish.

Study Description

Catchable rainbow trout – fish approximately six inches or more released into lakes and streams just prior to the opening of fishing season – were sampled from ten Washington Department of Fish and Wildlife (WDFW) hatcheries. Approximately 2½ months following planting, samples from un-mixed hatchery populations were sampled from stocked lakes. All feed and tissue (fillet) samples were analyzed for a variety of chlorinated pesticides, PCB aroclors, a select group of polybrominated diphenyl ethers (PBDEs), and lipid content. A subset of feed and tissue samples was also analyzed for polychlorinated dioxins and furans (PCDD/Fs). Specific project objectives were to:

- Measure concentrations of POPs in catchable rainbow trout released to lakes by WDFW.
- Measure concentrations of POPs in feed used to raise catchable rainbow trout in WDFW hatcheries to assess the correlation between diet and contaminant burdens in fish tissue.
- Estimate the degree of contaminant depuration or uptake in catchable rainbow trout following their release into lakes.

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Methods

Study Design

Fish feed and catchable rainbow trout were sampled from ten WDFW hatcheries during March 29 - April 5, 2005. Hatchery selection was made based on consultation with John Kerwin, Hatchery Division Manager with the WDFW Fish Program. Figure 1 shows locations of hatcheries. All ten hatcheries use well or spring water for hatching and rearing, although Tucannon River water is used in the final six-month rearing phase at the Tucannon Hatchery. Vancouver and Puyallup Hatchery personnel have also observed surface runoff entering hatcheries during rainy periods, but the extent of the exposure to fish is minor. POPs are much less likely to be present in groundwater and surface water due to their low solubility and immobility in soils.

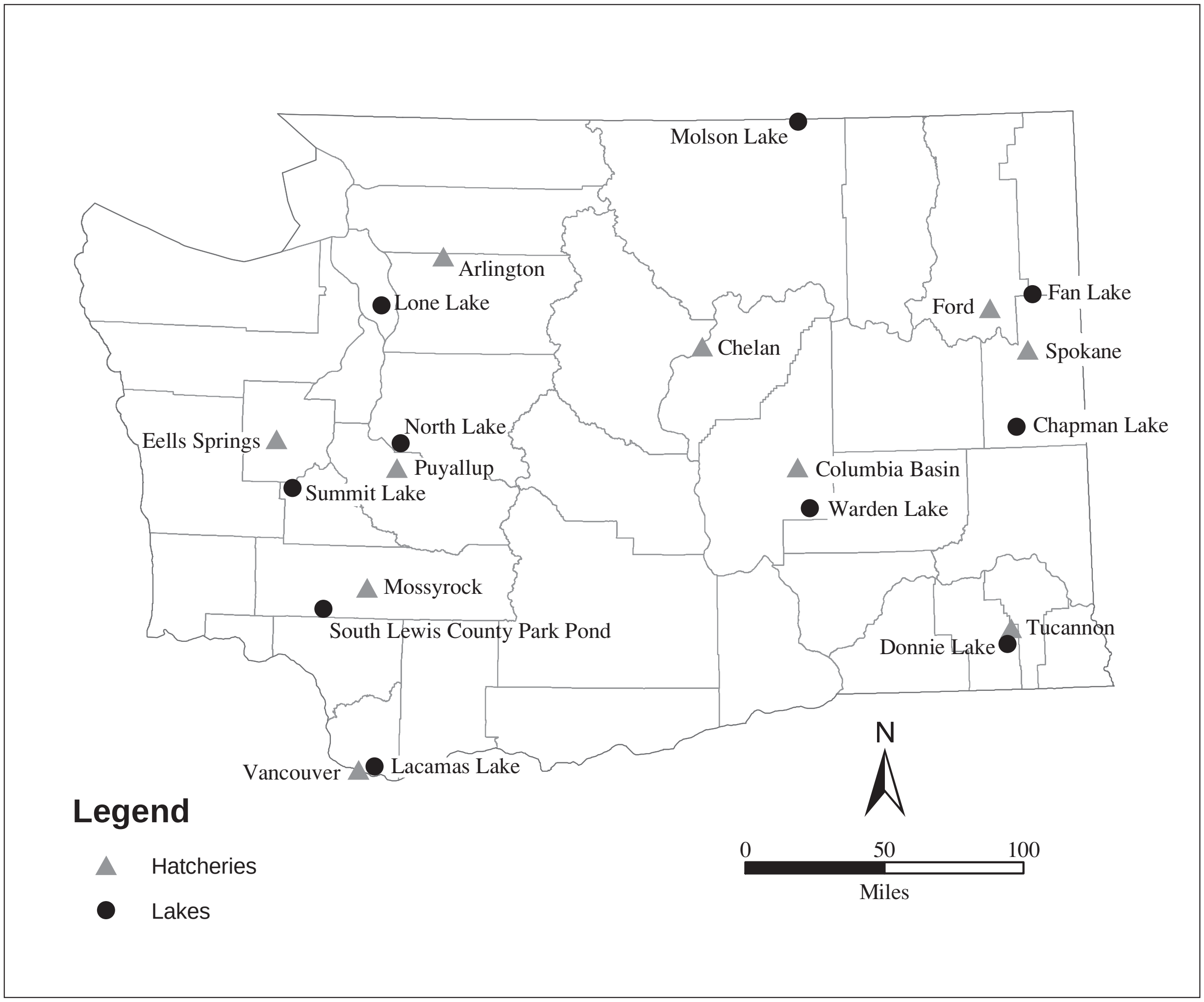

Figure 1. Locations of Hatcheries and Lakes Sampled for the 2005 Study of Persistent Organic Pollutants in Hatchery Feed and Hatchery Fish.

Hatchery personnel were interviewed about the feed used, schedule for changes in feed size and type, weight growth obtained using the sampled feed, hatchery water source, planting schedules, and other pertinent information related to the project. Feed samples consisted of material being fed to the trout at the time of sampling. In most cases, fish had been on the feed sampled for at least four months during which they had gained 50% - 80% of their mass (Table 1).

Table 1. Feed Analyzed from WDFW Hatcheries.

Hatchery	Feed Sampled	Pellet Size (mm)	Manufacturer	Period of use	Weight Gain
Arlington	Rangen	3.2	Rangen, Inc. Buhl, ID	6 mo.	76%
Chelan	Silver Cup Trout	3.0	Nelson & Sons, Inc., Murray, UT	6 mo.	76%
Columbia Basin	Silver Cup Fish Feed	3.2	Nelson & Sons, Inc., Murray, UT	6 mo.	80%
Eells Springs	Rangen	4.0	Rangen, Inc. Buhl, ID	4 mo.(a)	56%
Ford	Orient	4.0	Skretting, Vancouver, B.C.	na	50%
Mossyrock	Silver Cup Salmon	3.0	Nelson & Sons, Inc., Murray, UT	10 mo.	na
Puyallup	EWOS Vita	3.0	EWOS, Surrey, B.C.	na	na
Spokane	Silver Cup Fish Feed	3.2	Nelson & Sons, Inc., Murray, UT	na	78%
Tucannon	EWOS Pacific	3.0	EWOS, Surrey, B.C.	1 mo.(b)	70% (c)
Vancouver	Rangen	4.0	Rangen, Inc. Buhl, ID	6 mo.	82%

(a) fish fed Rangen since fry stage
(b) fish fed EWOS 2.0 for preceding 4 months, and EWOS 1.2 for 2 months prior to that
(c) weight gain during diet of EWOS 1.2, EWOS 2.0, and EWOS 3.0
na - not available

Ten rainbow trout specimens from each hatchery were randomly selected for sampling. They were from the general catchable populations which were in the process of being planted or were planned to be stocked within the subsequent weeks.

Ten triploid rainbow trout from Troutlodge, a private facility that supplies trout to WDFW, were provided by WDFW staff. Triploid trout are fish with three sets of chromosomes produced by pressure-treating the newly dividing fertilized eggs. Since they are sterile, more energy is used for somatic growth than gamete production, and the resulting triploid trout is larger than diploid fish of the same age. No feed samples or post-plant fish associated with Troutlodge were sampled for this study.

Lakes selected for sampling were based on the following criteria:

1. No known contaminant sources and low potential for appreciable contamination
2. Little or no natural rainbow trout production
3. Rainbow trout originating from a single hatchery planted between late-March and mid-April, 2005
4. Geographically dispersed to reflect a variety of ecosystem types, water chemistry, aquatic environments, and regions of the state containing differing preponderance of land use types.

Few of the approximately 380 lakes stocked annually with catchable trout have any contaminant data. Therefore, criterion 1 was assumed to be met unless a potential contaminant source was obvious. District WDFW biologists were interviewed to satisfy criterion 2. Criterion 3 was the most difficult to meet due to the common practice of multiple plantings of fish from different hatcheries. Lakes are also often planted at various intervals throughout the spring, which would yield uncertainties in fish residence periods; these lakes were avoided. Planting reports provided weekly by WDFW were reviewed in order to find lakes with single hatchery plants and fish residence times of approximately 2½ months. Criterion 4 was easily satisfied due to the geographic separation of the hatcheries and their associated lakes. Table 2 lists lakes where rainbow trout were sampled and their hatcheries of origin.

Table 2. Lakes Sampled and 2005 Rainbow Trout Plants.

Lake	County	Area (hect.)	Mean Depth (meters)	2005 Stock Date	Number	Mean Weight (grams)	Hatchery
Lone	Island	41	2.7	30-Mar	4,060	114	Arlington
				18-Apr	593*	649	Troutlodge
Molson	Okanogan	9.3	1.8	12-Apr	4,160	142	Chelan
Warden	Grant	81	8.2	15-Mar	12,723	116	Columbia Basin
				18-Mar	11,596	108	
				5-Apr	685	123	
Summit	Thurston	214	16	28-Mar	400	1,746	Eells Springs
				29-Mar	402	1,681	
				14-Apr	10,048	142	
				15-Apr	646	825	
				22-Apr	15,097	138	
				25-Apr	9,810	153	
				26-Apr	5,056	142	
Fan	Pend Oreille	32	7.6	22-Mar	3,021	86	Ford
S. Lewis Co. Park Pond	Lewis	4.5	2.7	14-Apr	3,043	134	Mossyrock
North	King	23	4.3	20-Apr	8,500	114	Puyallup
Chapman	Spokane	61	20	15-Mar	5,925	91	Spokane
Donnie	Columbia	0.4	0.9	14-Apr	420	108	Tucannon
Lacamas	Clark	129	7.3	7-Mar	2,000	227	Vancouver
				8-Apr	3,000	267	
				26-Apr	4,000	197	

*Triploid fish

Sampling Procedures

Hatchery feed and pre-plant catchable rainbow trout samples were collected with assistance of hatchery staff. Fish averaged 235 mm total length and 152 gm in weight (excluding Troutlodge samples). Feed samples were placed directly in 1-liter organics-free glass jars with Teflon lid liners and certificates of analysis. Fish from hatcheries were killed with a blow to the skull, double-wrapped in aluminum foil, sealed in zip-lock polyethylene bags, and transported on ice to Ecology headquarters where they were weighed and measured prior to being stored frozen at –20° C.

Rainbow trout from lakes were collected by hook-and-line or electrofishing. Following capture, fish were observed for signs confirming previous hatchery residence. Specimens were then killed with a blow to the skull, weighed to the nearest gram and measured to the nearest millimeter, assigned a sample number, double-wrapped in aluminum foil, placed in zip-lock polyethylene bags, and transported on ice to Ecology headquarters where they were stored frozen at –20° C. Rainbow trout collected from lakes averaged 270 mm total length and 211 gm in weight.

When ready for processing, fish were partially thawed then scales were removed for aging by WDFW. Composite samples of homogenate tissue were prepared by methods described by EPA and the Washington State Toxics Monitoring Program (EPA, 2000; Seiders, 2003). Briefly, fish were scaled, skin-on fillets removed, and equal mass aliquots of tissue were homogenized with three passes through a Kitchen-Aid food processor for each composite. Homogenates were placed in a 4-oz organics-free glass jar with Teflon lid liner and certificate of analysis and stored frozen.

All resection was done with non-corrosive stainless steel implements on a clean aluminum foil surface. Persons preparing samples wore non-talc polyethylene or nitrile gloves changed between samples. Resection and homogenizing equipment was cleaned using Liquinox® detergent and hot tap water, followed by rinses with deionized water, pesticide grade acetone, and pesticide grade hexane, then air-dried in a fume hood before use.

Laboratory Analysis and Data Quality

Sample Preparation

Feed

Feed samples were first Soxhlet extracted using 1:1 methylene chloride/hexane, then solvent exchanged into hexane and adjusted to 10 ml. Extracts were split, half for PCB/chlorinated pesticide and half for PBDE and lipid analysis.

Extracts for PCB and chlorinated pesticide analysis were eluted through 2 gm micro Florisil® columns first with 100% hexane and collected as the “0% Florisil fraction”, followed by elution with 1:1 hexane/preserved diethyl ether, collected as the “50% Florisil fraction”. When the

"0% Florisil fractions" were solvent-reduced, the remaining extracts were as much as 50% lipids, unsuitable for gas chromatography (GC) analysis, and were therefore added to the "50% Florisil fraction". The combined extracts were then back-extracted with acetonitrile to remove lipids and re-eluted through 2 gm micro Florisil® columns with 100% hexane ("0% Florisil fraction") and 1:1 hexane/preserved diethyl ether ("50% Florisil fraction"). Each fraction was solvent-exchanged to iso-octane and concentrated to 1 ml. One-half of the "50% Florisil fraction" and the "0% Florisil fraction" were treated with concentrated sulfuric acid prior to analysis. The remainder of the "50% fraction" was analyzed without acid treatment.

Fish Tissue

Tissue samples were first Soxhlet extracted using 1:1 methylene chloride/hexane, then solvent exchanged into hexane and adjusted to 10 ml. Extracts were split, half for PCB/chlorinated pesticide and half for PBDE and lipid analysis.

Extracts for some PCB analyses (sample nos. 05248100 – 05248109) were eluted through 2 gm micro Florisil® columns with 100% hexane, solvent-exchanged to iso-octane, and concentrated to 1 ml. Extracts were treated with concentrated sulfuric acid prior to analysis.

For chlorinated pesticide analysis and some PCB analyses (sample nos. 05144080 – 05144090), extracts were eluted through 2 gm micro Florisil® columns with 100% hexane and collected as the "0% Florisil fractions", followed by elution with 1:1 hexane/preserved diethyl ether. The hexane/ether fractions were adjusted to 5 ml and back-extracted with acetonitrile to remove lipids and re-eluted through 2 gm micro Florisil® columns with 1:1 hexane/preserved diethyl ether, and collected as the "50% Florisil fraction". Each fraction was solvent-exchanged to iso-octane and concentrated to 1 ml. One-half of the "50% Florisil fraction" and the "0% Florisil fraction" were treated with concentrated sulfuric acid prior to analysis. The remainder of the "50% fraction" was analyzed without acid treatment.

Analyses for PCBs, chlorinated pesticides, and PBDEs were conducted at the Manchester Environmental Laboratory (MEL) using dual column GC/ECD. Sample preparation and analysis methods were modifications of EPA SW-846 Methods 3540, 3620, and 8081/8082.

Samples for PCDDs/PCDFs were analyzed at Pacific Rim Laboratories, Inc. (Surrey, B.C.) using high resolution GC/MS isotope dilution methodology of EPA Method 1613B. Percent lipid was analyzed gravimetrically at MEL. The complete list of analytes is in Appendix B.

Data Quality

Overall quality of the data was fair. Precision for Aroclor analysis was 11% relative percent difference. Analysis was also performed with a high degree of precision for DDT compounds (14%), other chlorinated pesticides (17%), PBDEs (17%), and PCDD/Fs (13%).

One standard reference material was analyzed along with feed and tissue; NIST 1974b – Organics in Frozen Mussel Tissue (https://srmors.nist.gov/tables/view_table.cfm?table=109-2.htm). Chlorinated pesticide analytes, including DDT compounds, were only 67% of certified concentrations on average. Total PCBs were 82% of the reference concentration. These results indicate a possible low bias for these analyte groups.

Data Analysis

PCB, DDT, PBDE, and lipid concentrations were compared between hatchery and lake rainbow trout using the Wilcoxon signed-rank test, a non-parametric equivalent of the paired t-test (Zar, 1984). Spearmann ranked correlations among contaminant groups and sample types were done using SYSTAT 9.01 software program (SPSS, 1998). Non-detected values were treated as zero for statistical tests to avoid misinterpretation of comparisons between hatchery tissue samples and the lake tissue samples, which had different detection limits for the same analytes.

Non-detects were also treated as zero for samples analyzed in duplicate. Therefore, values presented as the mean of duplicate analyses may be biased low. The complete set of chemistry data is in Appendix C.

Results

Field Observations

Physical observations of rainbow trout collected from lakes indicated that they originated from hatcheries sampled earlier in the year. Most specimens had gnawed pectoral fins, or dorsal and caudal fin erosion. Scale annuli patterns also indicated fish were from the year's stock of catchables (John Sneva, WDFW, written communication, 8/10/2005). This physical evidence, coupled with local knowledge from WDFW District Biologists and WDFW plant records, supports the conclusion that the trout had been raised as catchables from known hatcheries.

Rainbow trout collected from lakes in June were larger on average than just prior to their release from hatcheries (Table 3). Increases in total length averaged 15%, and weight gain was 39% on average. Mean condition factors in hatchery and planted trout were 1.11 and 1.02, respectively. Condition factors fell below 1.0 in half the lakes, suggesting food supply was limited. Gut contents were not examined, but aside from the fin erosion mentioned previously, the fish collected from lakes appeared healthy and took bait and lures readily in most cases.

Contaminants in Feed and Fish

PCBs, DDT, and PBDEs

Most feed and fish tissue samples contained measurable concentrations of PCBs (Table 4). Aroclor-1254 was the most commonly detected, followed by 1260, 1242, and 1248; none of the other Aroclors were detected.

All samples contained DDT compounds, with 4,4'-DDE comprising 74% of the ΣDDT on average. All but one of the tissue samples contained low levels of PBDEs; none of the feed samples had detectable PBDEs. PBDE-47 was the most common congener detected, followed by 99, 71/100, and 138/209.

Mean concentrations of lipids in feed were high (16.8%) compared to tissue. This high fat diet resulted in high lipid levels in hatchery rainbow trout fillet tissue (mean of 3.2%). Although the catchable rainbow trout increased in size following planting, it appears that muscle lipid was depleted to meet their energy requirements, with an average 60% decrease in lipid content (1.2%).

Table 3. Length and Weight of Rainbow Trout Collected from Hatcheries and Lakes.

Hatchery/Lake	2005 Collection Date	N	Total Length (mm, mean ± SD)	Weight (gm, mean ± SD)	Condition Factor (mean ± SD)
Arlington Hatchery	29-Mar	10	245 ± 16	165 ± 34	1.11 ± 0.09
Lone Lake	16-Jun	10	306 ± 12	334 ± 37	1.17 ± 0.06
Chelan Hatchery	5-Apr	10	253 ± 16	178 ± 37	1.08 ± 0.11
Molson Lake	13-Jun	10	296 ± 20	303 ± 59	1.16 ± 0.08
Columbia Basin Hatchery	5-Apr	10	230 ± 15	140 ± 31	1.15 ± 0.08
Warden Lake	9-Jun	10	251 ± 10	147 ± 23	0.93 ± 0.08
Eells Springs Hatchery	1-Apr	10	230 ± 8	142 ± 19	1.16 ± 0.07
Summit Lake	13-Jun	7	259 ± 13	160 ± 16	0.92 ± 0.12
Ford Hatchery	4-Apr	10	197 ± 15	83 ± 15	1.09 ± 0.06
Fan Lake	14-Jun	8	290 ± 14	271 ± 47	1.11 ± 0.15
Mossyrock Hatchery	5-Apr	10	260 ± 14	190 ± 31	1.08 ± 0.06
S. Lewis Co. Park Pond	14-Jun	8	259 ± 10	176 ± 24	1.01 ± 0.07
Puyallup Hatchery	1-Apr	10	218 ± 20	111 ± 31	1.04 ± 0.11
North Lake	13-Jun	10	245 ± 12	141 ± 21	0.96 ± 0.08
Spokane Hatchery	4-Apr	10	210 ± 13	98 ± 21	1.04 ± 0.08
Chapman Lake	15-Jun	4	243 ± 10	125 ± 13	0.87 ± 0.04
Tucannon Hatchery	4-Apr	10	206 ± 18	108 ± 30	1.21 ± 0.06
Donnie Lake	16-Jun	10	254 ± 19	145 ± 33	0.87 ± 0.07
Vancouver Hatchery	5-Apr	10	298 ± 24	303 ± 91	1.12 ± 0.10
Lacamas Lake	17-Jun	9	285 ± 14	249 ± 37	1.07 ± 0.06
Troutlodge Hatchery	4-Apr	10	374 ± 22	678 ± 133	1.29 ± 0.19

Condition Factor = (W[g] x 100/L[cm]^3)
N = number
SD = standard deviation

Table 4. Lipid, ΣPCB, ΣDDT, and ΣPBDE Concentrations in Feed and Rainbow Trout Fillet Tissue (ng/g, ww).

Sample Type/Location	% Lipid	ΣPCB	ΣDDT	ΣPBDE
Hatchery Feed				
Arlington	18.19	**13.8**	**6.3**	U(0.25-1.2)
Chelan	13.75	**34.8**	**9.4**	U(0.25-1.2)
Columbia Basin	14.47	**11.6**	**6.3**	U(0.24-1.2)
Eells Spring	12.70	**12.5**	**5.9**	U(0.24-1.2)
Ford*	25.85	U(2.5)	**3.7**	U(0.25-1.2)
Mossyrock	19.64	**27.6**	**11.0**	U(0.25-1.2)
Puyallup*	16.14	U(2.5)	**6.6**	U(0.25-1.2)
Spokane	15.79	**16.4**	**5.9**	U(0.25-1.2)
Tucannon	15.01	**8.2**	**21**	U(0.25-1.2)
Vancouver	16.08	**13.3**	**5.8**	U(0.25-1.2)
Hatchery Rainbows				
Arlington	3.97	**12.1**	**4.8**	**0.64**
Chelan	3.05	**67**	**4.1**	**1.09 J**
Columbia Basin	4.10	**18.5**	**6.5**	**0.90 J**
Eells Spring*	2.42	U(2.4)	**2.7**	**0.52**
Ford	2.35	U(2.5)	**2.5**	**0.24 J**
Mossyrock	2.69	**15.8**	**3.9**	**0.89 J**
Puyallup	3.07	U(2.3)	**2.4**	**0.24**
Spokane	2.48	**11.7**	**2.9**	**1.10 J**
Tucannon	3.69	U(2.4)	**5.3**	**0.27**
Vancouver*	4.00	**4.8**	**4.0**	**0.71 J**
Troutlodge	5.39	**14.4**	**5.7**	**0.84 J**
Planted Rainbows				
Lone Lake*	1.67	U(4.8)	**1.9**	**0.96 J**
Molson Lake	2.05	**8.6**	**5.8**	U(0.49-2.4)
Warden Lake	0.61	U(4.9)	**3.7**	**0.46 J**
Summit Lake*	0.40	**5.0**	**3.2**	**0.56**
Fan Lake	2.66	U(5.0)	**57**	**0.40 J**
South Lewis Co. Park Pond	0.77	**5.9**	**2.4**	**1.33 J**
North Lake	0.65	U(4.9)	**2.9**	**1.23 J**
Chapman Lake	0.44	**11.8**	**5.0**	**1.01 NJ**
Donnie Lake	1.29	U(5.0)	**3.6**	**0.25 J**
Lacamas Lake	1.50	U(5.0)	**3.0**	**0.42 J**

ΣPCB = the sum of detected Aroclors
ΣDDT = the sum of detected 4,4' and 2,4' homologues of DDD, DDE, and DDT
ΣPBDE = the sum of detected PBDE congeners analyzed
Detected concentrations in **bold**
* Samples analyzed in duplicate. Results shown are mean of laboratory analyses.
U - The analyte was not detected at or above the reported result
J - The analyte was positively identified. The associated numerical value is an estimate.
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate.

Lipid catabolism in muscle and consequent contaminant mobilization may explain higher mean ΣPCB in hatchery fish compared to fish from lakes (13 and 3.1 ng/g, respectively; Figure 2). Differences between contaminant concentrations in hatchery and planted rainbow trout appear to be regulated by more than lipid decreases, however. Mean ΣDDT concentrations in tissue were higher in lakes (8.8 ng/g) compared to tissues from hatchery fish (3.9 ng/g), although concentrations were nearly identical when the outlier from Fan Lake was removed.

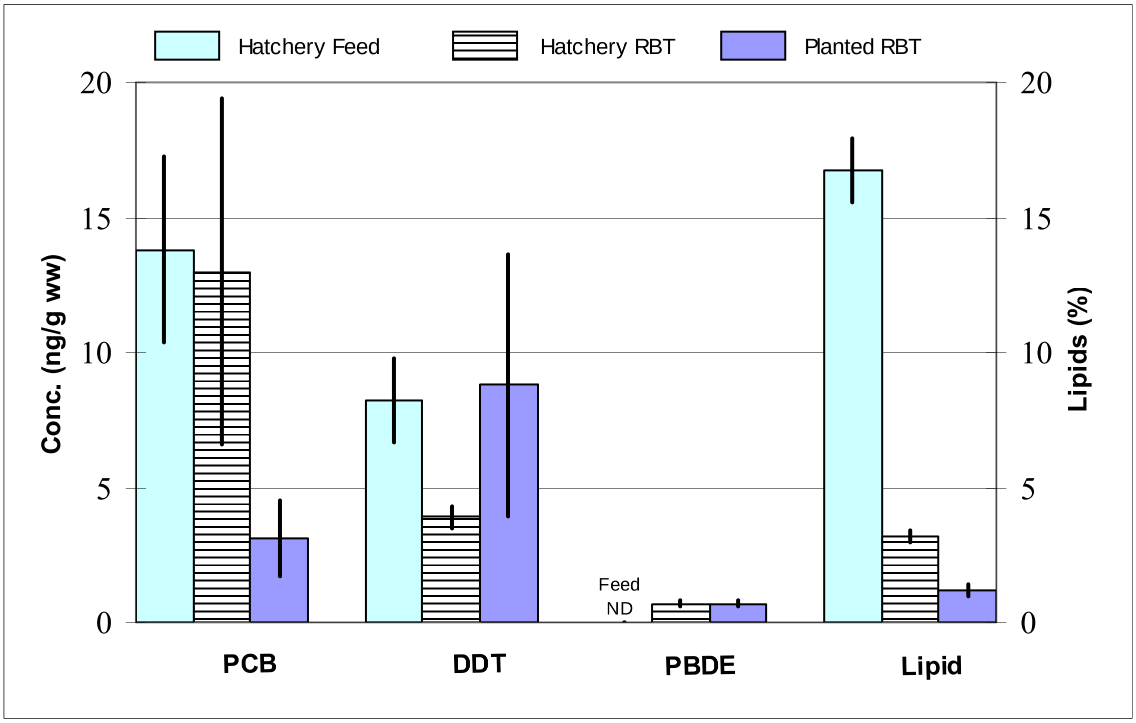

Figure 2. Mean (SE) Concentrations of ΣPCBs, ΣDDT, ΣPBDE, and Percent Lipid in Hatchery Feed, Hatchery Rainbow Trout, and Planted Rainbow Trout.

Neither ΣPCB nor ΣDDT were significantly different in hatchery fish compared to planted fish (Wilcoxon signed-rank test, α=0.05). There was also no significant difference in PBDEs between groups, although this was not surprising since mean concentrations were identical (0.66 ng/g, respectively).

PCBs tended to be highest in hatchery fish whose food had comparatively high PCB concentrations. For example, the Chelan Hatchery had high ΣPCB in both feed and fish, while Ford and Puyallup Hatcheries had no detectable PCBs in either feed or fish. However, this pattern was not true for ΣDDT and ΣPBDE in hatchery samples.

To examine possible relationships among sample types and contaminants, a ranked correlation analysis was performed on major variables in hatchery and lake samples (Table 5). There were no strong correlations between contaminants in hatchery fish and their lake counterparts. With the exception of lipids and ΣDDT in hatchery fish, lipids were not highly correlated with ΣPCB, ΣDDT, or ΣPBDE, a somewhat surprising finding but lending support to the notion that factors other than lipid may be the primary determinants in contaminant residue levels, particularly for DDT compounds and PBDEs.

Table 5. Spearman Ranked Correlation Matrix of Major Variables in Feed and Tissue Samples.

	Hatch. Feed Lipid									
Hatch. Feed ΣPCB	-0.182	Hatch. Feed ΣPCB								
Hatch. Feed ΣDDT	-0.159	0.226	Hatch. Feed ΣDDT							
Hatch. Fish Lipid	-0.139	-0.012	0.256	Hatch. Fish Lipid						
Hatch. Fish ΣPCB	-0.156	**0.753**	0.280	0.356	Hatch. Fish ΣPCB					
Hatch. Fish ΣDDT	-0.333	0.280	0.402	**0.721**	**0.563**	Hatch. Fish ΣDDT				
Hatch. Fish ΣPBDE	-0.365	**0.817**	0.086	0.170	**0.822**	0.426	Hatch. Fish ΣPBDE			
Planted Fish Lipid	0.467	0.030	-0.030	0.042	0.125	0.127	-0.213	Planted Fish Lipid		
Planted Fish ΣPCB	-0.307	**0.753**	0.131	**-0.519**	0.405	-0.171	**0.685**	-0.294	Planted Fish ΣPCB	
Planted Fish ΣDDT	-0.285	-0.109	-0.293	-0.382	0.019	-0.042	0.182	0.176	0.246	Planted Fish ΣDDT
Planted Fish ΣPBDE	0.382	0.103	0.055	-0.103	-0.006	-0.394	0.043	**-0.539**	0.184	**-0.636**

Spearman correlation coefficients ≥|0.500| in **bold**

Dioxins and Furans

Four samples each of hatchery feed and rainbow trout tissue were analyzed for 2,3,7,8-substituted PCDDs and PCDFs (Table 6). Toxic equivalents (TEQs) were calculated using toxic equivalency factors (TEFs) proposed by Van den Berg et al., 1998.

Table 6. TEQ Concentrations (pg/g, ww) in Four Feed and Rainbow Trout Samples and Percent TEQ Contribution by Congener.

	Vancouver	Mossyrock	Ford*	Spokane
Hatchery Feed				
TEQ	0.562	1.226	0.028	1.194
2,3,7,8-TCDD	33%	30%	0%	16%
1,2,3,7,8-PeCDD	38%	44%	0%	53%
1,2,3,4,7,8-HxCDD	0%	0%	0%	0%
1,2,3,6,7,8-HxCDD	4%	4%	0%	4%
1,2,3,7,8,9-HxCDD	0%	3%	0%	3%
1,2,3,4,6,7,8-HpCDD	2%	1%	18%	1%
OCDD	0%	0%	1%	0%
2,3,7,8-TCDF	9%	7%	71%	12%
1,2,3,7,8-PeCDF	2%	2%	10%	1%
2,3,4,7,8-PeCDF	11%	9%	0%	10%
1,2,3,4,7,8-HxCDF	0%	0%	0%	0%
1,2,3,6,7,8-HxCDF	0%	0%	0%	0%
2,3,4,6,7,8-HxCDF	0%	0%	0%	0%
1,2,3,7,8,9-HxCDF	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDF	0%	0%	0%	0%
1,2,3,4,7,8,9-HpCDF	0%	0%	0%	0%
OCDF	0%	0%	0%	0%
Hatchery Rainbows				
TEQ	0.053	0.041	0.012	0.024
2,3,7,8-TCDD	0%	0%	0%	0%
1,2,3,7,8-PeCDD	0%	0%	0%	0%
1,2,3,4,7,8-HxCDD	0%	0%	0%	0%
1,2,3,6,7,8-HxCDD	0%	0%	0%	0%
1,2,3,7,8,9-HxCDD	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDD	0%	0%	0%	0%
OCDD	0%	0%	0%	0%
2,3,7,8-TCDF	35%	68%	35%	68%
1,2,3,7,8-PeCDF	11%	17%	0%	0%
2,3,4,7,8-PeCDF	39%	0%	0%	0%
1,2,3,4,7,8-HxCDF	13%	12%	56%	28%
1,2,3,6,7,8-HxCDF	0%	0%	0%	0%
2,3,4,6,7,8-HxCDF	0%	0%	0%	0%
1,2,3,7,8,9-HxCDF	0%	0%	0%	0%
1,2,3,4,6,7,8-HpCDF	2%	2%	9%	3%
1,2,3,4,7,8,9-HpCDF	0%	0%	0%	0%
OCDF	0%	0%	0%	0%

* Samples analyzed in duplicate. Results shown are mean of laboratory analyses.

TEQs were much higher in hatchery feed compared to rainbow trout, with the exception of Ford Hatchery feed which had a TEQ up to 40 times lower than the other hatcheries. Like other contaminants, it appears that TEQ concentrations were not determined primarily by lipid content. Congeners contributing to TEQ were markedly different between high and low TEQ samples, with tetra- and penta-substituted dioxin congeners providing approximately 70% of the toxicity. In contrast, the Ford feed sample had none of these congeners detected.

Congener patterns were even more distinct when feed and fish tissue samples were compared. None of the tissue samples contained detectable concentrations of PCDDs. Most of the toxicity in tissue samples was derived from tetra-, penta-, and hexa-substituted furan congeners.

Other Chlorinated Pesticides

In addition to DDT compounds, nine chlorinated pesticides or breakdown products were detected in feed samples, six were detected in hatchery rainbow trout, and four were found in trout collected from lakes (Figure 3). DDMU, like its parent DDT, was detected in all hatchery feed samples. Although DDMU was much higher in Fan Lake trout (4.4 ng/g) compared to other lakes, feed from the originating hatchery (Ford) had the lowest concentration (0.2 ng/g), and Ford Hatchery rainbow trout did not have a detectable DDMU concentration.

Dieldrin had an unusual detection pattern among samples; it was detected in only 10% (Mossyrock, one of ten) feed samples, and 20% of lake fish tissue samples, but was found in 70% of the hatchery tissue samples. It should be noted, however, that detection limits for dieldrin in feed samples were approximately three-to-five times higher than in tissue samples (see Appendix C). Hexachlorobenzene was found in only one sample from planted rainbow trout (Summit Lake, 0.7 ng/g) but not in fish from the original population at Eells Springs Hatchery. The comparatively high concentration and lack of apparent link to the hatchery suggests these fish may have accumulated hexachlorobenzene from a local source in Summit Lake.

Pentachloroanisole, a degradation product of pentachlorophenol, was the only additional compound detected in planted rainbow tissue. Concentration of pentachloroanisole doubled in Lacamas Lake fish compared with the original hatchery population, suggesting a possible local source. Tucannon hatchery rainbows also apparently accumulated pentachloroanisole from the hatchery feed, although residues in the hatchery fish did not persist while the fish were in a (Donnie) lake environment.

Nonachlor and cis-chlordane, two components of commercial-grade chlordane, were found in at least half of the hatchery feed and fish samples. Another chlordane component, trans-chlordane, was found in 40% of feed samples. Two other chlorinated pesticides, toxaphene and methoxychlor, were found in 30% and 20% of feed samples, respectively.

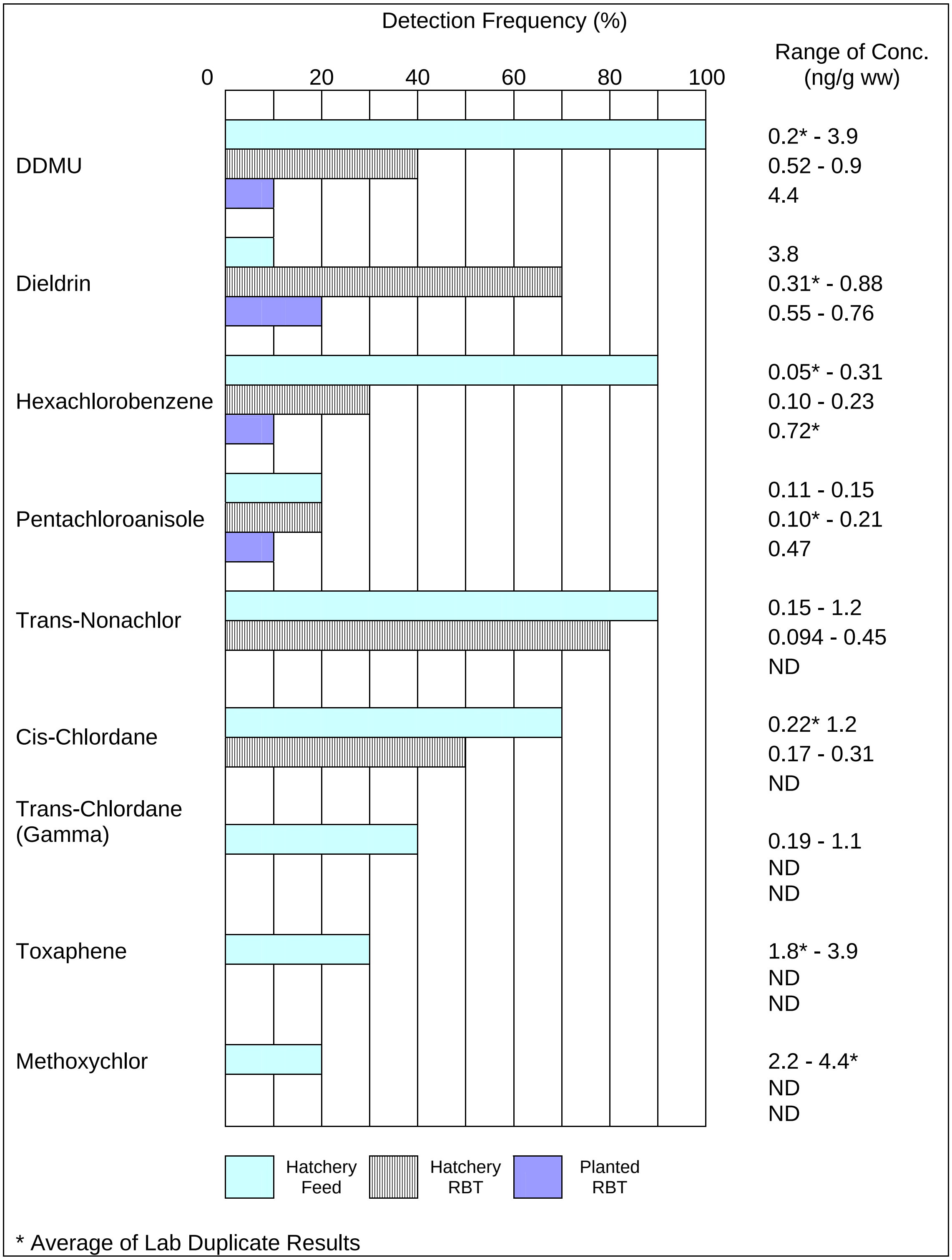

Figure 3. Detection Frequency of Chlorinated Pesticides (Excluding DDT Compounds) and Range of Detected Concentrations.

Discussion

Feed Ingredients and Residue Tolerances

The four brands of feed analyzed during the present 2005 survey have a wide range of contaminant levels. Rainbow trout collected at hatcheries gained approximately 50-80% of their final weight on these feeds, and may have gained most of their initial weight on starter feeds with similar ingredients.

Feed sack labels obtained at the time of sampling indicated that minimum amounts of crude protein (40-45%) were similar among feeds, as were maximum amounts of crude fiber (1.5-5%) and ash (9-12%). Crude fat was the other major component, constituting 10-24% of the weight which generally showed good agreement with lipid analysis in the present study (r^2=0.64).

In terms of contaminant residues, the origin of the lipids in the feed is probably a much greater concern than their percent by weight. All of the feeds sampled advertise fish oil as a major ingredient which is likely to be the major source of PCBs, chlorinated pesticides, PBDEs, PCDD/Fs, and other lipophilic contaminants (Jacobs et al., 2002).

The exact source of the fish oil and fish meal, as well as other major ingredients in hatchery feed (wheat flour, soybean oil, blood meal, feather meal, and poultry by-product), may change from batch to batch, depending on the availability and cost of the raw products. Fishmeal may derive largely from anchovy, menhaden, capelin, and herring, and fish oil is a by-product of the fishmeal manufacturing industry.

The negative correlation between lipids and major contaminants in feed is another indication that the source of fish oil is probably a more important determinant in contaminant residues than lipid content by weight. However, contaminant concentrations in feed apparently play a large role in accumulation by fish, probably due to the high rate of weight conversion from feed to fish (>80%). This is demonstrated by the strong correlation between ΣPCB in hatchery feed and hatchery fish (r=0.75), and a weaker yet positive correlation for ΣDDT (r=0.40).

In the case of the Tucannon hatchery, where the fish are raised in water from the Tucannon River for six months prior to stocking, fish weight gain actually exceeded the weight of feed provided (conversion of 163%) due to availability of natural prey items.

The only domestic regulation concerning POP contaminants is a federal PCB residue tolerance of 2,000 ng/g for feed components of animal origin (21 CFR 109.30). The Canadian Food Inspection Agency has an “actionable level” of 2,000 ng/g for PCBs, but it only applies to fish oil destined for animal feed (CFIA, 2003). In 2001, the European Union (EU) established maximum tolerable levels of PCDD/F TEQs in fish in various fish products and feeds. The maximum levels for fish feed and fish oil for use other than direct human consumption is 2.25 and 6.0 pg TEQ/g, respectively (Directive 2001/102/EC).

Table 7 shows a summary of major contaminants in hatchery feeds and commercial aquaculture feeds for salmonids. Care should be used interpreting the available data since information on the intended species is often not included. For instance, feed for commercial salmon aquaculture typically contains higher fish oil content than trout feed, and some species, such as catfish and tilapia, may be fed entirely vegetable-based feeds.

Table 7. Contaminant Concentrations in Various Hatchery and Commercial Aquaculture Feeds and Applicable Regulations (mean concentrations unless otherwise noted).

Source	ΣPCB (ng/g ww)	ΣDDT (ng/g ww)	ΣPBDE (ng/g ww)	Dioxin TEQ (pg/g ww)	Lipid (%)	Ref.
Contaminant Levels in Feed						
Feeds from 6 manufacturers used in 11 USF&WS National Fish Hatcheries	1.94*	11.33	nr	0.227	16.7	Maule et al., 2006
Mean of 8 commercial salmon aquaculture feeds from Scottish sources	105	12.1	5.1	nr	28.8	Jacobs et al., 2002
Mean of fish feed/fishmeal samples originating from Canada (n=14), U.S. (n=7), Iceland (n=1), Peru (n=1), and Russia (n=1)	Can.–30.7† U.S.–16.5† Ice.–12.1† Peru–0.6† Rus.–12.7†	Can.–21.1 U.S.–23.3 Ice.– <7 Peru–nr Rus.–nr	nr	Can.–1.0 U.S.–1.1 Ice.–0.23 Peru–ND Rus.–0.22	nr	CFIA, 2006
Feed from the 2 largest global suppliers for commercial salmon aquaculture; 9 samples from North and South America and 4 samples from Europe	15 (approx. Amer. median) 60 (approx. Eur. median)	nr	nr	1 (approx. Amer. median) 4 (approx. Eur. median)	nr	Hites et al., 2004
Confidential	nr	nr	nr	1.21	nr	Hermann et al., 2004
WDFW trout hatchery feed	*13.8*	*8.2*	*ND (<0.25)*	*0.75*	*16.8*	*present study, 2005*
Regulatory Levels in Feed						
U.S. Food and Drug Admin.	2,000	ne	ne	ne	ne	21 CFR 109.30(a)(6)
European Union	ne	ne	ne	2.25	ne	Directive 2001/102/EC 21

*sum of 14 dioxin-like congeners
†sum of 72 congeners
nr - not reported
ND – not detected
ne - not established

Maule et al. (2006) analyzed numerous batches of feeds used at eleven U.S. Fish and Wildlife Service (USF&WS) National Fish Hatcheries. They found a lower mean dioxin TEQ and ΣPCB than reported here, although the ΣPCB was derived from only 14 dioxin-like PCB congeners. Interestingly, the bulk of the dioxin TEQ was derived from 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD similar to the feed samples analyzed for the present survey. ΣDDT and percent lipid contents were similar between the two studies.

Comparison of contaminants in feed and salmon fillet from commercial aquaculture operations showed ΣPCB, ΣDDT, and ΣPBDE higher in feed compared to fish tissue at similar proportions to those reported here (Jacobs et al., 2002). Although the mean ΣDDT was similar to results for the present study, ΣPCB was an order of magnitude higher. The feeds, which were from Scottish sources, had much higher lipids (mean of 28.8%) than found here (mean of 16.8%). Hites et al. (2004) reported ΣPCB and dioxin TEQ levels approximately four-fold higher in commercial salmon aquaculture feeds from Europe compared to North and South America. Like other results, the Hites et al. (2004) study found POP concentrations slightly higher in feed compared to fish, on average, but lipid concentrations were not given.

The Canadian Food Inspection Agency (CFIA) analyzed 24 samples originating primarily from Canada and the U.S., but also from Iceland, Peru, and Russia, although little additional information is provided about the samples (CFIA, 2006). ΣPCB for U.S., Icelandic, and Russian samples were similar to those reported here, while Canadian feed had about twice the concentration on average. DDT levels were about twice the levels reported in other samples, while the dioxin TEQs tended to be within ranges reported by other investigators.

The only reported PBDE analysis of feed was done in the Jacobs et al. (2002) study which found ΣPBDE averaging 20 times the analytical reporting limits for the present study. PBDE-47 was the predominant congener found and, although not found in WDFW hatchery feeds, it was the major congener in rainbow trout from hatcheries and lakes and is one of the most abundant congeners found in Washington freshwater fish (Johnson and Olson, 2001).

None of the average POP concentrations reported in these studies violated residue tolerances from the applicable regulations, with the exception of the European median dioxin TEQ reported by Hites et al. (2004). A study by Herrmann et al. (2004), designed specifically to look at compliance with EU standards, found a mean concentration of dioxin TEQ about one-half the EU residue tolerance. However, 95th percentile values for both fish feed (2.71 pg/g) and fish oil (6.30 pg/g) exceeded the dioxin TEQ standards.

Depuration/Uptake of Contaminants in Lakes

One of the underlying goals of this study was to determine if lakes act as purifying environments or if hatchery fish stocked in lakes accumulate additional contaminants. Cursory inspection of the data suggests depuration occurs for PCBs and chlorinated pesticides other than DDT compounds, and no differences are seen for ΣDDT (with the Fan Lake outlier removed) and ΣPBDE. Although pairwise comparisons show no statistical differences between ΣPCB in hatchery and lake rainbow trout, mean ΣPCB in hatchery fish are substantially higher, even when the outlier from Chelan Hatchery (67 ng/g) is removed.

Part of the decrease in ΣPCB may be the significant decrease in lipid content of the planted fish. It is a well-known fact that PCB concentrations are often positively correlated with lipid in tissue. Lipid metabolism, and in particular lipid catabolysis, to meet energy requirements may mobilize lipids from muscle to high lipid organs such as the liver, kidney, and brain, thus decreasing muscle PCB concentrations (Jørgensen et al., 2002). Gamete production is another mechanism for transfer of PCBs due to loss of muscle lipids, although it is unlikely that the fish examined had reached sexual maturity.

As mentioned previously, the absence of a strong correlation between lipids and ΣPCB suggests PCB concentrations are controlled by factors other than the percentage of lipid. One possible explanation for altered PCB concentrations is that some lakes allow PCBs to be shed from muscle tissue while others increase the PCB accumulated by fish. Molson, Warden, South Lewis County Park, and Lone lakes received fish with the highest ΣPCB, and all four lakes had the greatest depuration. Increases in ΣPCB only occurred where concentrations were low-to-moderate in hatchery fish (with the exception of the very small increase in Spokane Hatchery-Chapman Lake fish), suggesting net accumulation only occurs when PCB concentrations in tissue are initially low.

ΣDDT concentrations were low in muscle tissue of hatchery rainbow trout precluding an opportunity to observe substantial depuration. As many lakes had increased levels of ΣDDT as had decreases, all small changes with the exception of Fan Lake.

Fan Lake is a medium-sized (32 hectare) lake approximately 20 miles north of Spokane, situated in a small drainage basin (1,600 hectares). It is one of the lowermost in a chain of lakes along the West Branch of the Little Spokane River. Since the Fan Lake basin is relatively undeveloped and the lake is connected to other lakes only through its outlet, it appears unlikely that high ΣDDT accumulating in Fan Lake rainbow trout is a result of basin-wide or area-wide contamination.

Comparison of Tissue Concentrations to Applicable Criteria

Criteria to protect human health from harmful pollutants in ingested water and fish were issued to Washington State in EPA's 1992 National Toxics Rule (NTR, 40 CFR 131.36). The human health-based criteria, if met, will generally ensure that public health concerns do not arise, and that fish advisories are not needed. Sampling of either water or edible fish tissue may be conducted to assess compliance with the NTR criteria (Ecology, 1992), but tissue is generally preferred because POPs are often found at concentrations in water below reasonably available laboratory detection limits.

It should be recognized that POPs exceeding the NTR criteria in fish tissue do not necessarily signal the need for a fish consumption advisory, nor does it imply the existence of a public health concern. Assessment of risks to the fishing public and consumption advice is carried out by the Washington State Department of Health (DOH), often based on data collected by Ecology and WDFW. While DOH supports Ecology's use of the NTR criteria for identifying potentially contaminated waters and for developing source controls to keep water quality at or below criteria, it does not use the NTR criteria to establish fish advisories. Instead, DOH evaluates contaminants in fish tissue using established risk assessment paradigms. These include tools for:

1. Analysis of risks – calculating allowable meal limits based on known contaminant concentrations, estimates of exposure in specific groups or populations.
2. Risk management – e.g., reduction in contaminants through preparation and cooking techniques, known health benefits from fish consumption, contaminant concentrations or health risks associated with replacement foods, and cultural importance of fish.
3. Risk communication – the outreach component of a fish advisory or a conclusion that an advisory is unnecessary.

Table 8 shows NTR criteria for chemicals analyzed in the present 2005 survey compared to concentrations in rainbow trout. NTR criteria have not been established either for PBDEs or for some of the chlorinated pesticides analyzed here.

There are 15 instances where contaminants in rainbow trout exceed NTR criteria. Most of the exceedances are for ΣPCB (three lakes and six hatcheries), followed by dieldrin (one lake and four hatcheries) and 4,4'-DDE (one lake). In all, seven of the eleven hatchery samples (including Troutlodge) exceeded NTR criteria for at least one chemical, and five of the ten planted fish samples exceeded the criteria.

When criteria are not met, the waterbody is considered impaired and placed on the federal Clean Water Act section 303(d) list and may require a Total Maximum Daily Load (TMDL) study to address the impairment. TMDLs identify the sources of a pollutant and allocate pollutant loads among sources in order to bring the waterbody in compliance with standards.

One of the questions this study sought to answer is whether contaminants accumulated by fish in hatcheries could contribute, in whole or in part, to the water quality impairment of a waterbody and its consequent addition to the 303(d) list. This question was not intended to be directed at the lakes analyzed in the present survey. Instead, at issue is whether hatchery-derived contamination is worthy of inquiry by investigators who are conducting surveys on individual lakes and streams where the source of contamination is uncertain and where waterbodies may be considered for 303(d) listing.

Table 8. National Toxics Rule Criteria Compared to Contaminant Residues in Hatchery and Planted Rainbow Trout Fillet (ng/g ww except pg/g ww for 2,3,7,8-TCDD).

Contaminant	Criterion	Concentrations in present study	No. of Criterion Exceedances	Location(s) of Criterion Exceedance
ΣPCB	5.3	4.85 - 67	9	Chapman Lake Molson Lake South Lewis Co. Park Pond Chelan Hatchery Columbia Basin Hatchery Mossyrock Hatchery Troutlodge Arlington Hatchery Spokane Hatchery
4,4'-DDT	32	0.14 - 1.9		
4,4'-DDE	32	1.9 - 45	1	Fan Lake
4,4'-DDD	45	0.29 - 9.6		
2,3,7,8-TCDD	0.07	0.011 - 0.053		
Aldrin	0.65	ND		
Dieldrin	0.65	0.32 - 0.88	5	Warden Lake Mossyrock Hatchery Spokane Hatchery Arlington Hatchery Eells Springs Hatchery
Endrin	3,216	ND		
Endrin Aldehyde	3,216	ND		
alpha-BHC	1.7	ND		
beta-BHC	1.6	ND		
gamma-BHC (Lindane)	8.2	ND		
Chlordane (total)	8.3	ND		
Endosulfan I	540	ND		
Endosulfan II	540	ND		
Endosulfan Sulfate	540	ND		
Heptachlor	2.4	ND		
Heptachlor Epoxide	1.2	ND		
Hexachlorobenzene	6.7	0.10 - 0.73		
Toxaphene	9.8	ND		

ND - not detected

To examine this question on a statewide basis, results of the present survey were compared to all of the data on contaminants in tissue in Washington. In order to provide a suitably comparable data set, data that met the following conditions were extracted from Ecology's Environmental Information Management (EIM) database; 1) samples analyzed since 1998, 2) results only for rainbow trout, brown trout, and cutthroat trout since these are the primary resident species raised to catchable size at WDFW hatcheries, and 3) fillet data only. Only results for ΣPCB, 4,4'-DDE, and dieldrin were selected since these were the contaminants found here that exceeded NTR criteria. Waterbodies known to have large sources of these chemicals (e.g., Spokane River for PCBs, Yakima River for 4,4'-DDE and dieldrin) were removed.

Figure 4 shows the cumulative distribution of all ΣPCB data for fish in Washington using the selection process previously described. Sixty percent of the 46 samples exceed the NTR criterion, and approximately 50% of samples statewide had concentrations that fall into the range of detectable ΣPCB concentrations in hatchery rainbow trout, excluding the Chelan Hatchery sample. One hundred percent of samples fall into the hatchery ΣPCB range when all of the hatchery rainbows are considered.

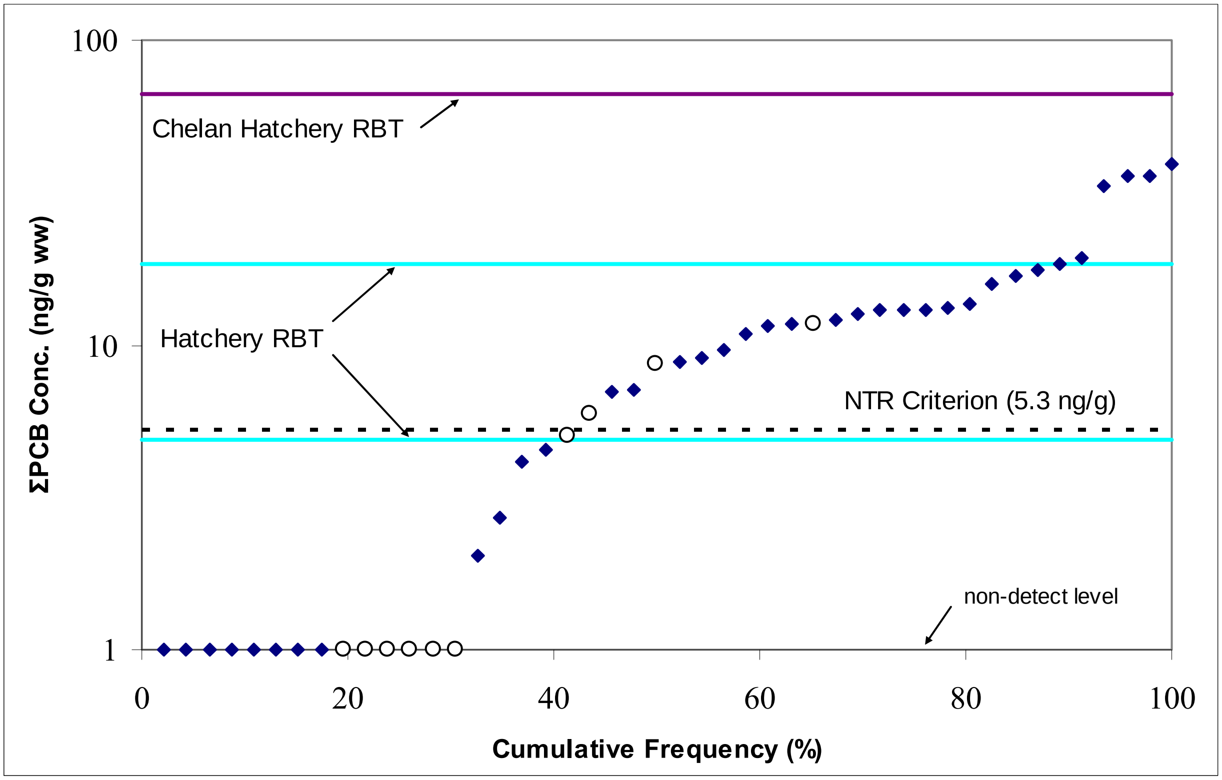

Figure 4. Cumulative Frequency Distribution of ΣPCB Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected ΣPCB concentrations in hatchery fish. Dashed line is the NTR criterion for ΣPCBs.)

Only two of the 38 trout samples (5%) exceed the NTR criterion for 4,4'-DDE, including the Fan Lake result found in the present survey (Figure 5). Approximately 40% of the samples are within the hatchery fish range, with more than 70% falling below the maximum 4,4'-DDE level in hatchery rainbows.

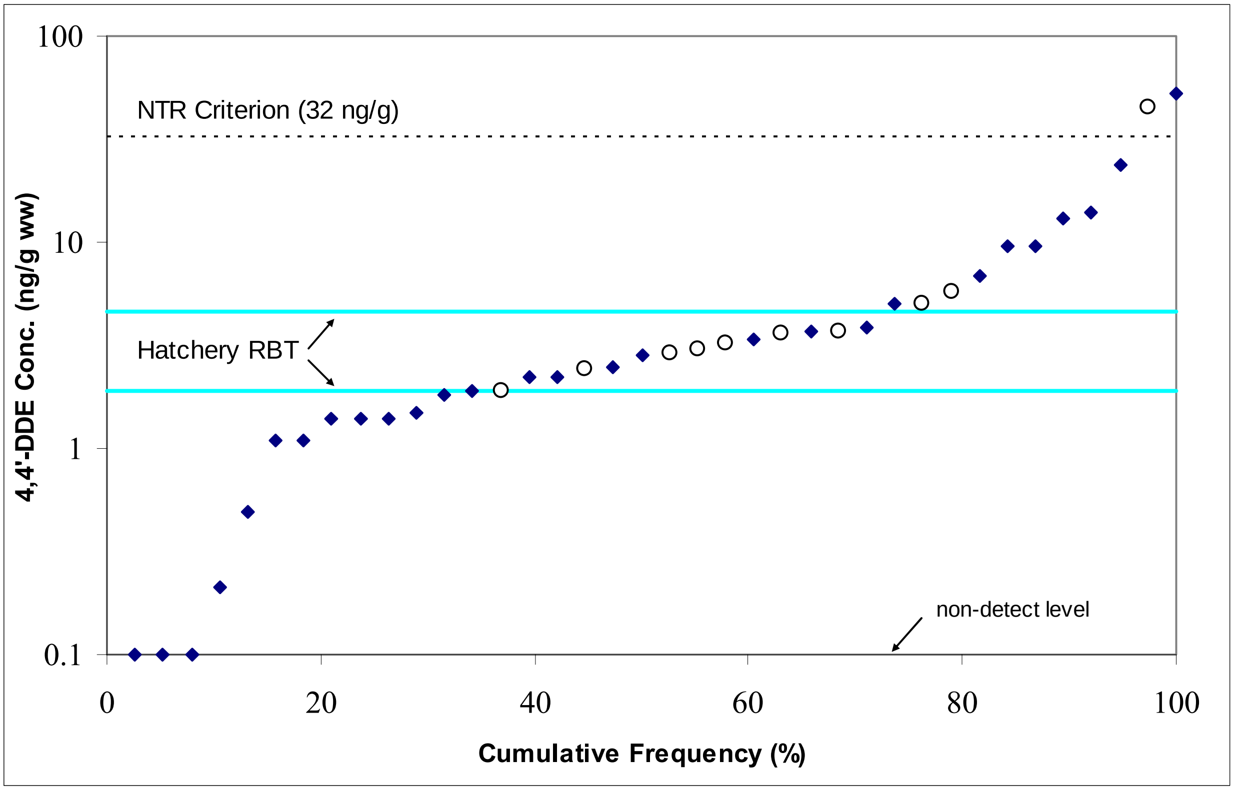

Figure 5. Cumulative Frequency Distribution of 4,4'-DDE Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected 4,4'-DDE concentrations in hatchery fish. Dashed line is the NTR criterion for 4,4'-DDE.)

None of the 38 samples screened from the EIM database had detectable levels of dieldrin; North Lake and Warden Lake are the only "unpolluted" sites where dieldrin has been detected in common trout species (Figure 6). Dieldrin in Warden Lake rainbow trout (0.76 ng/g) exceeds the NTR criterion (0.65 ng/g).

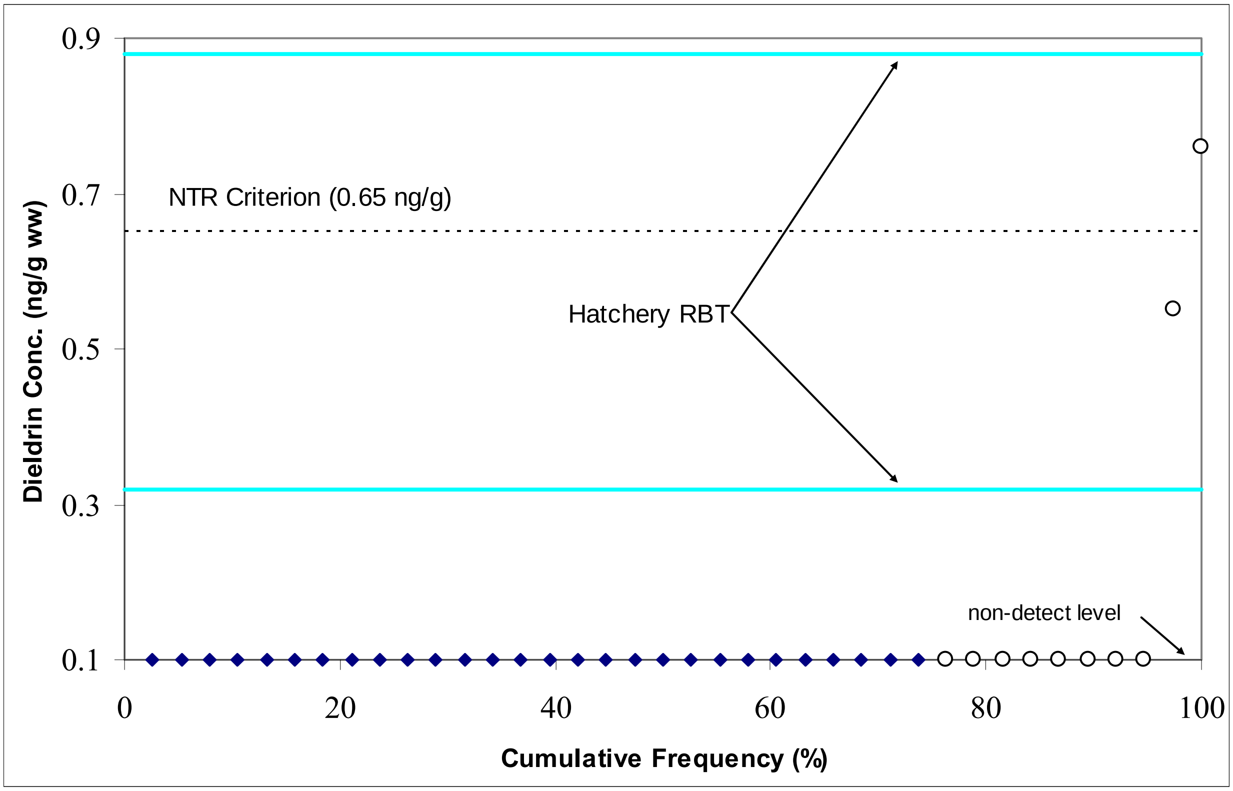

Figure 6. Cumulative Frequency Distribution of Dieldrin Concentrations in Fillet Tissue of Rainbow Trout (RBT), Cutthroat Trout, and Brown Trout from Washington Lakes and Streams. (Open circles represent data from lakes sampled in the present survey. Solid gray lines bound the range of detected dieldrin concentrations in hatchery fish. Dashed line is the NTR criterion for dieldrin.)

There are several inferences that may be drawn from these comparisons to screened data. One supposition is that some portion of POPs found in trout is derived from burdens obtained at a hatchery, although this presumes that the fish were planted. However, there is no concomitant information on the origin of the fish used for these comparisons.

The extent of hatchery-derived POPs remaining in planted fish is variable and appears to depend largely on the amount of time lapsed since stocking. Trout caught around the time of opening day – typically mid-to-late April – will have contaminant concentrations nearly identical to concentrations found in fish just prior to their removal from hatcheries. It appears that as the fishing season progresses, the concentrations in fillet tissue generally decrease, particularly for PCBs and some chlorinated pesticides, although this is a pattern which is less consistent for DDT compounds and PBDEs. Unfortunately, the planted cohorts that were sampled for each hatchery-planted pair could not be sampled further to track fillet concentrations during subsequent periods.

One of the implications of these results, particularly from the practical standpoint of a regulatory agency, is that waterbodies may be included on the 303(d) list due to contamination stemming from hatcheries. Taken further, 303(d) listed waters often require a TMDL to assess contaminant sources. Sources considered for TMDLs are typically point sources (e.g., piped effluent) and nonpoint sources (e.g., agricultural and urban runoff, atmospheric deposition) which normally occur in the vicinity of the impaired waterbody. However, no known TMDLs in Washington have included hatchery fish as a contaminant source. For PCBs, and to a lesser extent dieldrin, hatchery fish may contribute to impairment and, in some cases, may cause the bulk of impairment. Therefore, TMDL investigators may want to consider including hatchery fish as contaminant sources among other sources.

Conclusions

Rainbow trout acquire low-to moderate concentrations of persistent organic pollutants (POPs) while residing at Washington Department of Fish and Wildlife (WDFW) hatcheries. Feed used to raise rainbow trout to catchable size (≥6”) contains concentrations of PCBs, DDT compounds, PCDD/Fs, and several additional chlorinated pesticides at higher wet-weight concentrations than in fillet tissue of the fish specimens analyzed, suggesting that the POP accumulation pathway is primarily through the feed. This is consistent with findings of other aquacultural studies, although other possible pathways of contaminant accumulation were not examined for this study. Low levels of PBDEs were also present in rainbow trout tissue, but were not detected in trout feed.

Fish feed is high in lipids and shows variable amounts of contaminants, but POP concentrations were not correlated with the percent lipid in feed samples. This suggests the source of lipids, largely derived from marine oil, is an important determinant in POP concentrations. The positive correlation between feed and fish ΣPCB and between feed and fish ΣDDT supports the conclusion that feed is the primary contaminant source to hatchery fish. WDFW trout hatchery feed has POP concentrations similar or lower than feeds analyzed in other studies, and appears to have much lower PCB concentrations than feeds used in commercial salmon aquaculture.

It appears that fillet tissue concentrations of some POPs, particularly PCBs, decrease following stocking in lakes, although this finding is inconclusive. ΣDDT concentrations may increase in the lake environment even as fillet lipid concentrations decrease significantly. Fish from only one location – Fan Lake in Pend Oreille County – showed a substantial increase in contaminants (DDT compounds) following residence in the wild.

In the 21 rainbow trout fillet samples analyzed (11 from hatcheries including Troutlodge and 10 from lakes), there are 15 instances where contaminants exceed (do not meet) regulatory criteria. Most of the exceedances are for ΣPCB (three lakes and six hatcheries), followed by dieldrin (one lake and four hatcheries) and 4,4’-DDE (one lake). Considering the POP levels in catchable rainbow trout just prior to planting, it appears likely that at least part of the contaminant burden is hatchery-derived, with the notable exception of DDT compounds in Fan Lake as described previously.

Based on comparisons between waterbodies in Washington State and POP data reported here, it is possible that trout caught in “unpolluted” lakes and streams contain contaminants originating from WDFW hatcheries. It is also possible that some listings for impaired waters, particularly listings for PCBs, may be due to hatchery-contaminated fish. Therefore, Total Maximum Daily Load (TMDL) project managers may want to consider hatchery fish as a source of contaminant loads.

Recommendations

Based on results of this 2005 study, it is recommended that fish feed and trout fillet tissue sampling be expanded to include all 26 WDFW hatcheries raising catchable trout. Samples should be analyzed for the persistent organic pollutants (POPs) in the present study, with PCDD/F analysis included for all samples. Water in hatcheries should also be sampled where contaminant levels in fish are exceptionally high. Any water sampling should be performed using semi-permeable membrane devices or other methods to achieve low detection limits for POPs.

More data are needed to assess depuration or accumulation of contaminants in catchable trout following planting in lakes. Ideally, fish could be sampled during several periods to better track trends in contaminant levels over time. Whole fish analysis should also be considered along with fillet sampling, to determine if contaminant burdens are conserved in fish following mobilization of lipids in muscle tissue.

A review of the current 303(d) list should be conducted to identify cases where tissue data used to assess impairment may have come from WDFW catchable trout plants. TMDL project managers should consider the implications of hatchery fish as a possible source of contaminants to waterbodies being assessed.

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Appendices

Appendix A – Glossary of Acronyms and Units

303(d) – Section 303(d) of the federal Clean Water Act
CFIA – Canadian Food Inspection Agency
DDD – 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane
DDE – 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene
DDT – 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane
DOH – Washington State Department of Health
ECD – electron capture detector
Ecology – Washington State Department of Ecology
EIM – Environmental Information Management
EPA – U.S. Environmental Protection Agency
EU – European Union
GC – gas chromatography
MEL – Manchester Environmental Laboratory
MS – mass spectrometry
NIST – National Institute of Standards and Technology
NTR – National Toxics Rule
PBDE – polybrominated diphenyl ether
PCB – polychlorinated biphenyl
PCDD – polychlorinated dibenzo-*p*-dioxin
PCDF – polychlorinated dibenzofuran
POP – persistent organic pollutant
SRM – standard reference material
TEF – toxic equivalency factor
TEQ – toxic equivalent
TMDL – total maximum daily load (water cleanup plan)
USF&WS – U.S. Fish and Wildlife Service
WDFW – Washington Department of Fish and Wildlife
ww – wet weight
Σ – sum of

Units of measurement

ng/g – nanograms per gram (parts per billion)
pg/g – picograms per gram (parts per trillion)

Appendix B – Target Analytes and Reporting Limits

Table B. Target Analytes and Reporting Limits

Analyte	Reporting Limit (ng/g ww)	Analyte	Reporting Limit (ng/g ww)	Analyte	Reporting Limit (pg/g ww)
PCBs		**Chlorinated Pesticides**		**PCDDs/PCDFs**	
Aroclor-1016	2.3 – 5.0	2,4'-DDE	0.47 – 1.0	2,3,7,8-TCDD	0.03 – 0.05
Aroclor-1221	2.3 – 5.0	2,4'-DDD	0.47 – 1.0	1,2,3,7,8-PeCDD	0.07 – 0.11
Aroclor-1232	2.3 – 5.0	2,4'-DDT	0.47 – 1.0	1,2,3,4,7,8-HxCDD	0.05 – 0.2
Aroclor-1242	2.3 – 5.0	4,4'-DDT	0.47 – 1.0	1,2,3,6,7,8-HxCDD	0.05 – 0.2
Aroclor-1248	2.3 – 5.0	4,4'-DDE	0.47 – 1.0	1,2,3,7,8,9-HxCDD	0.05 – 0.2
Aroclor-1254	2.3 – 5.0	4,4'-DDD	0.47 – 1.0	1,2,3,4,6,7,8-HpCDD	0.05 – 0.2
Aroclor-1260	2.3 – 5.0	DDMU	0.47 – 1.0	OCDD	0.36
PBDEs		Aldrin	0.47 – 1.0	2,3,7,8-TCDF	0.03 – 0.05
		Dieldrin	0.47 – 10	1,2,3,7,8-PeCDF	0.06 – 0.1
PBDE-47	0.23 – 0.65	Endrin	0.96 – 10	2,3,4,7,8-PeCDF	0.04 – 0.12
PBDE-66	0.25 – 0.50	Endrin Aldehyde	0.96 – 10	1,2,3,4,7,8-HxCDF	0.2
PBDE-71	0.25 – 0.50	Endrin Ketone	0.96 – 10	1,2,3,6,7,8-HxCDF	0.05 – 0.1
PBDE-99	0.25 – 0.50	alpha-BHC	0.47 – 1.0	2,3,4,6,7,8-HxCDF	0.05 – 0.2
PBDE-100	0.25 – 0.50	beta-BHC	0.47 – 1.0	1,2,3,7,8,9-HxCDF	0.05 – 0.2
PBDE-138	0.25 – 0.50	gamma-BHC (Lindane)	0.47 – 1.0	1,2,3,4,6,7,8-HpCDF	0.05 – 0.2
PBDE-153	0.25 – 0.50	delta-BHC	0.47 – 1.0	1,2,3,4,7,8,9-HpCDF	0.05 – 0.2
PBDE-154	0.25 – 0.50	cis-Chlordane (alpha)	0.47 – 1.0	OCDF	0.21 – 0.23
PBDE-183	0.25 – 0.50	trans-Chlordane (gamma)	0.47 – 1.0		
PBDE-190	0.25 – 0.50	Oxychlordane	0.47 – 1.0		
PBDE-209	1.2 – 6.2	Chlordane (technical)	0.98 – 5.0		
		Chlorpyriphos	0.96 – 10		
		Dacthal (DCPA)	0.96 – 10		
		Endosulfan I	0.96 – 10		
		Endosulfan II	0.96 – 10		
		Endosulfan Sulfate	0.96 – 10		
		Heptachlor	0.47 – 1.0		
		Heptachlor Epoxide	0.96 – 10		
		Hexachlorobenzene	0.47 – 1.0		
		Methoxychlor	0.47 – 1.0		
		Mirex	0.47 – 1.0		
		cis-Nonachlor	0.47 – 1.0		
		trans-Nonachlor	0.47 – 1.0		
		Pentachloroanisole	0.47 – 1.0		
		Toxaphene	0.98 – 10		

Appendix C – Complete Results of Lipid and Contaminant Analysis

Table C-1. Complete Results of Percent Lipids and PCB Aroclor Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Lipids (%)	PCB-aroclor 1016		PCB-aroclor 1221		PCB-aroclor 1232		PCB-aroclor 1242		PCB-aroclor 1248		PCB-aroclor 1254		PCB-aroclor 1260		PCB-aroclor 1262		PCB-aroclor 1268	
Hatchery Feed																				
Arlington	5144102	18.19	2.5	UJ	2.5	UJ	2.5	UJ	2.7	J	5.5	UJ	8.1	J	3	J	2.5	UJ	2.5	UJ
Chelan	5144096	13.75	2.5	U	2.5	U	2.5	U	5	UJ	8	NJ	18		8.8		2.5	U	2.5	U
Columbia Basin	5144098	14.47	2.4	UJ	2.4	UJ	2.4	UJ	2.4	UJ	4.7	UJ	7.6	J	4	J	2.4	UJ	2.4	UJ
Eells Spring	5144103	12.7	2.4	UJ	2.4	UJ	2.4	UJ	2.6	J	4.6	UJ	6.9	J	3	J	2.4	UJ	2.4	UJ
Ford	5144099	25.75	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ
Ford-Dup	5144099-Dup	25.95	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Mossyrock	5144097	19.64	2.5	UJ	2.5	UJ	2.5	UJ	3.8	J	10	UJ	16	J	7.8	J	2.5	UJ	2.5	UJ
Puyallup	5144104	15.75	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ
Puyallup-Dup	5144104-Dup	16.52	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Spokane	5144100	15.79	2.5	UJ	2.5	UJ	2.5	UJ	2.8	J	6.1	UJ	9.3	J	4.3	J	2.5	UJ	2.5	UJ
Tucannon	5144101	15.01	2.5	UJ	2.5	UJ	2.5	UJ	2.5	UJ	2.8	J	5.4	NJ	2.5	UJ	2.5	UJ	2.5	UJ
Vancouver	5144095	16.08	2.5	UJ	2.5	UJ	2.5	UJ	2.6	J	5.5	UJ	7.7	NJ	3	J	2.5	UJ	2.5	UJ
Hatchery Rainbows																				
Arlington	5144087	3.97	2.5	U	2.5	U	2.5	U	2.1	J	3.5	UJ	6.5	NJ	3.5	J	2.5	U	2.5	U
Chelan	5144081	3.05	2.4	U	2.4	U	2.4	U	8.5	UJ	20	NJ	47		3.4	UJ	2.4	U	2.4	U
Columbia Basin	5144083	4.1	2.5	U	2.5	U	2.5	U	2.5	U	3.5	J	9		6		3.6	UJ	2.5	U
Eells Spring	5144088	2.7	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Eells Spring-Dup	5144088-Dup	2.13	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U
Ford	5144084	2.35	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U
Mossyrock	5144082	2.69	2.4	U	2.4	U	2.4	U	2.4	U	4.8	UJ	9	J	6.8	J	2.4	U	2.4	U
Puyallup	5144089	3.07	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U	2.3	U
Spokane	5144085	2.48	2.5	U	2.5	U	2.5	U	2.5	U	2.6	J	5.2	NJ	3.9	J	2.5	U	2.5	U
Troutlodge	5144090	5.39	2.4	U	2.4	U	2.4	U	2.6	J	5.6	UJ	7.9		3.9	J	2.4	U	2.4	U
Tucannon	5144086	3.69	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U	2.4	U
Vancouver	5144080	4.86	2.5	U	2.5	U	2.5	U	2.5	U	3.2	NJ	3.6	NJ	2.9	NJ	2.5	U	2.5	U
Vancouver-Dup	5144080-Dup	3.14	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U	2.5	U

Table C-1 (cont'd). Complete Results of Percent Lipids and PCB Aroclor Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Lipids (%)	PCB-aroclor 1016		PCB-aroclor 1221		PCB-aroclor 1232		PCB-aroclor 1242		PCB-aroclor 1248		PCB-aroclor 1254		PCB-aroclor 1260		PCB-aroclor 1262		PCB-aroclor 1268	
Planted Rainbows																				
Chapman Lake	5248102	0.44	5	U	5	U	5	U	5	U	5	U	6.7	J	5.1	J	5	U	5	U
Donnie Lake	5248103	1.29	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Fan Lake	5248104	2.66	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Lacamas Lake	5248100	1.5	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U	5	U
Lone Lake	5248108	1.63	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U
Lone Lake-Dup	5248108-Dup	1.71	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U
Molson Lake	5248101	2.05	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	8.6	J	4.9	U	4.9	U	4.9	U
North Lake	5248106	0.65	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U
South Lewis Co. Park Pond	5248105	0.77	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	5.9	J	4.9	U	4.9	U	4.9	U
Summit Lake	5248109	0.35	4.8	U	4.8	U	4.8	U	4.8	U	4.8	U	4.7	J	4.8	U	4.8	U	4.8	U
Summit Lake-Dup	5248109-Dup	0.45	5.0	U	5.0	U	5.0	U	5.0	U	5.0	U	**5.2**	J	5.0	U	5.0	U	5.0	U
Warden Lake	5248107	0.61	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U	4.9	U

U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate
Dup - Duplicate

Table C-2. Complete Results of DDT Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	2,4'-DDD		2,4'-DDE		2,4'-DDT		4,4'-DDD		4,4'-DDE		4,4'-DDT	
Hatchery Feed													
Arlington	5144102	0.37	J	0.49	U	0.49	U	2.3		2.8		0.78	NJ
Chelan	5144096	0.57	UJ	0.5	U	0.5	U	3.1		4.8		1.5	
Columbia Basin	5144098	0.28	J	0.49	U	0.49	U	1.4		4.4		0.21	J
Eells Spring	5144103	0.38	J	0.48	U	0.48	U	2		2.8		0.72	J
Ford	5144099	0.12	J	0.5	U	0.12	J	0.61		2.3		0.31	J
Ford- Dup	5144099-Dup	0.17	J	0.5	U	0.13	J	0.86		2.4		0.38	J
Mossyrock	5144097	0.43	J	0.5	U	0.5	U	3.4		6		1.2	NJ
Puyallup	5144104	0.32	J	0.5	U	0.15	J	1.2		4.3		0.49	J
Puyallup-Dup	5144104-Dup	0.3	J	0.49	U	0.15	J	1.4		4.3	NJ	0.55	
Spokane	5144100	0.2	J	0.5	U	0.5	U	0.77		4.2		0.7	NJ
Tucannon	5144101	0.85		0.5	U	0.16	J	4.4		15		0.7	J
Vancouver	5144095	0.43	J	0.5	U	0.5	U	2.1		2.6		0.69	J
Hatchery Rainbows													
Arlington	5144087	0.12	J	0.49	U	0.49	U	0.84		3.8		0.49	U
Chelan	5144081	0.49	U	0.49	U	0.49	U	0.7		2.8		0.6	NJ
Columbia Basin	5144083	0.5	U	0.61	UJ	0.5	U	1.8		3.9		0.77	NJ
Eells Spring	5144088	0.1	J	0.5	U	0.5	U	0.55		1.8		0.25	J
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	0.49	U	0.5		1.9		0.24	J
Ford	5144084	0.49	U	0.49	U	0.49	U	0.33	J	2.2		0.49	U
Mossyrock	5144082	0.48	U	0.48	U	0.48	U	0.91		2.7		0.24	NJ
Puyallup	5144089	0.47	U	0.47	U	0.47	U	0.29	J	2		0.14	J
Spokane	5144085	0.49	U	0.49	U	0.49	U	0.52		2.2		0.2	J
Troutlodge	5144090	0.15	J	0.49	U	0.49	U	1.5		3.4		0.65	J
Tucannon	5144086	0.49	U	0.49	U	0.49	U	0.52		4.6		0.18	J
Vancouver	5144080	0.5	U	0.5	U	0.5	U	0.7		4		0.26	J
Vancouver-Dup	5144080-Dup	0.12	J	0.5	U	0.5	U	0.59		2.1		0.27	J
Planted Rainbows													
Chapman Lake	5248102	1	U	1	U	1	U	1	U	5.1		1	U
Donnie Lake	5248103	1	U	1	U	1	U	1	U	3.6		1	U
Fan Lake	5248104	1	U	1	U	1	U	9.6	J	45		1.9	J
Lacamas Lake	5248100	1	U	1	U	1	U	1	U	3.0		1	U
Lone Lake	5248108	0.98	U	0.98	U	0.98	U	0.98	U	1.9	J	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.97	U	0.97	U	0.97	U	1.9	J	0.97	U
Molson Lake	5248101	0.98	U	0.98	U	0.98	U	0.98	U	5.8		0.98	U
North Lake	5248106	0.97	U	0.97	U	0.97	U	0.97	U	2.9		0.97	U
South Lewis Co. Park Pond	5248105	0.96	U	0.98	U	0.98	U	0.96	U	2.4		0.96	UJ
Summit Lake	5248109	0.96	U	0.96	U	0.96	U	0.96	U	3.0		0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	1.0	U	1.0	U	1.0	U	3.3		1.0	U
Warden Lake	5248107	0.97	U	0.97	U	0.97	U	0.97	U	3.7		0.97	U

U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate

Table C-3. Complete Results of PBDE Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	PBDE-047		PBDE-066		PBDE-071		PBDE-099		PBDE-100		PBDE-138	
Hatchery Feed													
Arlington	5144102	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Chelan	5144096	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Columbia Basin	5144098	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ
Eells Spring	5144103	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ
Ford	5144099	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Ford-Dup	5144099-Dup	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Mossyrock	5144097	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Puyallup	5144104	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Puyallup-Dup	5144104-Dup	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Spokane	5144100	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Tucannon	5144101	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Vancouver	5144095	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ
Hatchery Rainbows													
Arlington	5144087	0.64		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Chelan	5144081	0.95		0.24	U	0.14	J	0.24	U	0.24	U	0.24	U
Columbia Basin	5144083	0.65	J	0.25	U	0.16	J	0.089	NJ	0.25	U	0.25	U
Eells Spring	5144088	0.55		0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Eells Spring-Dup	5144088-Dup	0.49		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Ford	5144084	0.24	J	0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Mossyrock	5144082	0.73		0.24	U	0.16	J	0.24	U	0.24	U	0.24	U
Puyallup	5144089	0.24		0.23	U	0.23	U	0.23	U	0.23	U	0.23	U
Spokane	5144085	0.55		0.25	U	0.25	U	0.25	U	0.11	J	0.1	J
Troutlodge	5144090	0.62		0.24	U	0.15	J	0.07	J	0.24	U	0.24	U
Tucannon	5144086	0.27		0.24	U	0.24	U	0.24	U	0.24	U	0.24	U
Vancouver	5144080	0.72		0.25	U	0.13	J	0.25	U	0.25	U	0.25	U
Vancouver-Dup	5144080-Dup	0.56		0.25	U	0.25	U	0.25	U	0.25	U	0.25	U
Planted Rainbows													
Chapman Lake	5248102	0.91		0.5	U	0.5	U	0.1	NJ	0.5	U	0.5	U
Donnie Lake	5248103	0.25	J	0.5	U	0.5	U	0.5	U	0.5	U	0.5	U
Fan Lake	5248104	0.4	J	0.5	U	0.5	U	0.5	U	0.5	U	0.5	U
Lacamas Lake	5248100	0.29	J	0.5	U	0.5	U	0.5	U	0.13	J	0.5	U
Lone Lake	5248108	0.65		0.49	U	0.49	U	0.24	J	0.15	J	0.49	U
Lone Lake-Dup	5248108-Dup	0.54		0.48	U	0.48	U	0.13	J	0.20	J	0.48	U
Molson Lake	5248101	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ
North Lake	5248106	0.78		0.48	U	0.48	U	0.29	J	0.16	J	0.48	U
South Lewis Co. Park Pond	5248105	1		0.49	U	0.49	U	0.33	J	0.49	U	0.49	U
Summit Lake	5248109	0.87		0.48	U	0.48	U	0.12	J	0.14	J	0.48	U
Summit Lake-Dup	5248109-Dup	0.65	U	0.50	U	0.50	U	0.50	U	0.50	U	0.50	U
Warden Lake	5248107	0.46	J	0.48	U	0.48	U	0.48	U	0.48	U	0.48	U

Table C-3 (cont'd). Complete Results of PBDE Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	PBDE-153		PBDE-154		PBDE-183		PBDE-190		PBDE-209	
Hatchery Feed											
Arlington	5144102	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Chelan	5144096	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Columbia Basin	5144098	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	1.2	UJ
Eells Spring	5144103	0.24	UJ	0.24	UJ	0.24	UJ	0.24	UJ	1.2	UJ
Ford	5144099	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Ford-Dup	5144099-Dup	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Mossyrock	5144097	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Puyallup	5144104	0.25	U	0.25	U	0.25	U	0.25	U	1.2	U
Puyallup-Dup	5144104-Dup	0.25	U	0.25	U	0.25	U	0.25	U	1.2	U
Spokane	5144100	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Tucannon	5144101	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Vancouver	5144095	0.25	UJ	0.25	UJ	0.25	UJ	0.25	UJ	1.2	UJ
Hatchery Rainbows											
Arlington	5144087	0.24	U	0.24	U	0.24	U	0.24	U	3.1	U
Chelan	5144081	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Columbia Basin	5144083	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Eells Spring	5144088	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Eells Spring-Dup	5144088-Dup	0.24	U	0.24	U	0.24	U	0.24	U	3.0	U
Ford	5144084	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Mossyrock	5144082	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Puyallup	5144089	0.23	U	0.23	U	0.23	U	0.23	U	2.9	U
Spokane	5144085	0.25	U	0.25	U	0.25	U	0.25	U	0.34	J
Troutlodge	5144090	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Tucannon	5144086	0.24	U	0.24	U	0.24	U	0.24	U	3	U
Vancouver	5144080	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Vancouver-Dup	5144080-Dup	0.25	U	0.25	U	0.25	U	0.25	U	3.1	U
Planted Rainbows											
Chapman Lake	5248102	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Donnie Lake	5248103	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Fan Lake	5248104	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Lacamas Lake	5248100	0.5	U	0.5	U	0.5	U	0.5	U	6.2	U
Lone Lake	5248108	0.49	U	0.49	U	0.49	U	0.49	U	6.1	U
Lone Lake-Dup	5248108-Dup	0.48	U	0.48	U	0.48	U	0.48	U	6.0	U
Molson Lake	5248101	0.49	UJ	0.49	UJ	0.49	UJ	0.49	UJ	2.4	UJ
North Lake	5248106	0.48	U	0.48	U	0.48	U	0.48	U	6.1	U
South Lewis Co. Park Pond	5248105	0.49	U	0.49	U	0.49	U	0.49	U	6.1	U
Summit Lake	5248109	0.48	U	0.48	U	0.48	U	0.48	U	6	U
Summit Lake-Dup	5248109-Dup	0.50	U	0.50	U	0.50	U	0.50	U	6.2	U
Warden Lake	5248107	0.48	U	0.48	U	0.48	U	0.48	U	6.1	U

U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate

Table C-4. Complete Results of PCDD/F Analysis of Feed Samples (pg/g ww)

Sample Number	TEF	Vancouver Hatchery			Mossyrock Hatchery			Ford Hatchery			Ford Hatchery			Spokane Hatchery		
		Sample 05144105		TEQ	Sample 05144106		TEQ	Sample 05144107		TEQ	Sample 05144107-Dup		TEQ	Sample 05144108		TEQ
2,3,7,8-TCDF	0.1	0.525		0.0525	0.798		0.0798	0.182		0.0182	0.222		0.0222	1.4		0.14
1,2,3,7,8-PeCDF	0.05	0.251	J	0.01255	0.591		0.02955	0.1	UJ	0	0.115	J	0.00575	0.218	J	0.0109
2,3,4,7,8-PeCDF	0.5	0.126	J	0.063	0.226	J	0.113	0.12	UJ	0	0.12	UJ	0	0.246	J	0.123
1,2,3,4,7,8-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,6,7,8-HxCDF	0.1	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0	0.1	UJ	0
2,3,4,6,7,8-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,7,8,9-HxCDF	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,4,6,7,8-HpCDF	0.01	0.2	UJ	0	0.207	J	0.00207	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,4,7,8,9-HpCDF	0.01	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
OCDF	0.0001	0.404	J	0.00004	0.403	J	0.00004	0.32	J	0.00003	0.344	J	0.00003	0.377	J	0.00004
2,3,7,8-TCDD	1	0.186		0.186	0.367		0.367	0.05	UJ	0	0.05	UJ	0	0.192		0.192
1,2,3,7,8-PeCDD	1	0.215	J	0.215	0.534		0.534	0.11	UJ	0	0.11	UJ	0	0.637		0.637
1,2,3,4,7,8-HxCDD	0.1	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0	0.2	UJ	0
1,2,3,6,7,8-HxCDD	0.1	0.235	J	0.0235	0.494		0.0494	0.2	UJ	0	0.2	UJ	0	0.437		0.0437
1,2,3,7,8,9-HxCDD	0.1	0.2	UJ	0	0.333		0.0333	0.2	UJ	0	0.2	UJ	0	0.339		0.0339
1,2,3,4,6,7,8-HpCDD	0.01	0.857		0.00857	1.6		0.016	0.524		0.00524	0.474		0.00474	1.25		0.0125
OCDD	0.0001	8.14		0.00081	16.5		0.00165	3.38		0.00034	3.3		0.00033	11.5		0.00115
TEQ total				0.56197			1.22581			0.02381			0.03305			1.19419

TEF - Toxicity Equivalence Factor from Van den Berg et al., 1998
TEQ - Toxic Equivalent
UJ - The analyte was not detected at or above the reported estimated result
J - The analyte was positively identified. The associated numerical value is an estimate

Table C-5. Complete Results of PCDD/F Analysis of Rainbow Trout Tissue Samples (pg/g ww)

Sample Number	TEF	Vancouver Hatchery			Mossyrock Hatchery			Ford Hatchery			Ford Hatchery			Spokane Hatchery		
		Sample 05144092		TEQ	Sample 05144093		TEQ	Sample 05144091		TEQ	Sample 05144091-Dup		TEQ	Sample 05144094		TEQ
2,3,7,8-TCDF	0.1	0.187		0.0187	0.282		0.0282	0.037	J	0.0037	0.042	J	0.0042	0.163		0.0163
1,2,3,7,8-PeCDF	0.05	0.111	J	0.00555	0.143	J	0.00715	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
2,3,4,7,8-PeCDF	0.5	0.041	J	0.0205	0.04	UJ	0	0.04	UJ	0	0.04	UJ	0	0.04	UJ	0
1,2,3,4,7,8-HxCDF	0.1	0.07	J	0.007	0.051	J	0.0051	0.075	J	0.0075	0.053	J	0.0053	0.067	J	0.0067
1,2,3,6,7,8-HxCDF	0.1	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
2,3,4,6,7,8-HxCDF	0.1	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
1,2,3,7,8,9-HxCDF	0.1	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0	0.06	UJ	0
1,2,3,4,6,7,8-HpCDF	0.01	0.095	J	0.00095	0.078	J	0.00078	0.094	J	0.00094	0.105	J	0.00105	0.082	J	0.00082
1,2,3,4,7,8,9-HpCDF	0.01	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
OCDF	0.0001	0.31	J	0.00003	0.225	J	0.00002	0.313	J	0.00003	0.289	J	0.00003	0.336	J	0.00003
2,3,7,8-TCDD	1	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0	0.03	UJ	0
1,2,3,7,8-PeCDD	1	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0	0.07	UJ	0
1,2,3,4,7,8-HxCDD	0.1	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0	0.10000	UJ	0
1,2,3,6,7,8-HxCDD	0.1	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0
1,2,3,7,8,9-HxCDD	0.1	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0	0.05	UJ	0
1,2,3,4,6,7,8-HpCDD	0.01	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0	0.08	UJ	0
OCDD	0.0001	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0	0.36	UJ	0
TEQ total				0.05273			0.04125			0.01217			0.01058			0.02385

TEF - Toxicity Equivalence Factor from Van den Berg et al., 1998
TEQ - Toxic Equivalent
UJ - The analyte was not detected at or above the reported estimated result
J - The analyte was positively identified. The associated numerical value is an estimate

Table C-5. Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Aldrin		Alpha-BHC		Beta-BHC		Chlordane		Chlor-pyriphos		cis-Chlordane	
Hatchery Feed													
Arlington	5144102	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	UJ	0.52	
Chelan	5144096	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.5	U
Columbia Basin	5144098	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.68	NJ
Eells Spring	5144103	0.48	UJ	0.48	U	0.48	U	2.4	U	2.4	UJ	0.47	NJ
Ford	5144099	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.19	J
Ford-Dup	5144099-Dup	0.50	UJ	0.50	U	0.50	U	2.5	U	2.5	UJ	0.25	J
Mossyrock	5144097	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	1.2	NJ
Puyallup	5144104	0.5	UJ	0.5	U	0.5	U	2.5	U	10	UJ	0.5	U
Puyallup-Dup	5144104-Dup	0.49	UJ	0.49	U	0.49	U	2.5	U	9.9	UJ	0.49	U
Spokane	5144100	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	UJ	0.39	NJ
Tucannon	5144101	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	UJ	0.5	U
Vancouver	5144095	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.52	
Hatchery Rainbows													
Arlington	5144087	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.24	NJ
Chelan	5144081	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.49	U
Columbia Basin	5144083	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.25	NJ
Eells Spring	5144088	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.5	U
Eells Spring-Dup	5144088-Dup	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.49	U
Ford	5144084	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.49	U
Mossyrock	5144082	0.48	UJ	0.48	U	0.48	U	2.4	U	2.4	U	0.31	NJ
Puyallup	5144089	0.47	UJ	0.47	U	0.47	U	2.3	U	2.3	U	0.47	U
Spokane	5144085	0.49	UJ	0.49	U	0.49	U	2.5	U	2.5	U	0.49	U
Troutlodge	5144090	0.49	UJ	0.49	U	0.49	U	2.4	U	1.2	J	0.44	NJ
Tucannon	5144086	0.49	UJ	0.49	U	0.49	U	2.4	U	2.4	U	0.17	J
Vancouver	5144080	0.5	UJ	0.5	U	0.5	U	2.5	U	2.5	U	0.2	NJ
Vancouver-Dup	5144080-Dup	0.50	UJ	0.50	U	0.50	U	2.5	U	2.5	U	0.17	NJ
Planted Rainbows													
Chapman Lake	5248102	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Donnie Lake	5248103	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Fan Lake	5248104	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Lacamas Lake	5248100	1	UJ	1	UJ	1	U	5	U	4	U	1	U
Lone Lake	5248108	0.98	UJ	0.98	UJ	0.98	U	0.98	U	3.9	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	UJ	0.97	UJ	0.97	U	4.8	U	3.9	U	0.97	U
Molson Lake	5248101	0.98	UJ	0.98	UJ	0.98	U	4.9	U	3.9	U	0.98	U
North Lake	5248106	0.97	UJ	0.97	UJ	0.97	U	4.9	U	3.9	U	0.97	U
South Lewis Co. Park Pond	5248105	0.98	UJ	0.96	UJ	0.96	U	2.4	U	0.96	U	0.96	UJ
Summit Lake	5248109	0.96	UJ	0.96	UJ	0.96	U	4.8	U	3.8	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	UJ	1.0	UJ	1.0	U	5.0	U	4.0	U	1.0	U
Warden Lake	5248107	0.97	UJ	0.97	UJ	0.97	U	4.9	U	3.9	U	0.97	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	cis-Nonachlor		Dacthal (DCPA)		DDMU		Delta-BHC		Dieldrin	
Hatchery Feed											
Arlington	5144102	0.49	U	2.5	UJ	0.69	NJ	0.49	UJ	2.5	UJ
Chelan	5144096	0.5	U	2.5	U	1.5	NJ	0.5	UJ	2.5	U
Columbia Basin	5144098	0.49	U	2.4	UJ	0.73	NJ	0.49	UJ	2.4	UJ
Eells Spring	5144103	0.48	U	2.4	UJ	0.58	NJ	0.48	UJ	2.4	UJ
Ford	5144099	0.5	U	2.5	UJ	0.4	NJ	0.5	UJ	2.5	UJ
Ford-Dup	5144099-Dup	0.50	U	2.5	UJ	0.50	U	0.50	UJ	2.5	UJ
Mossyrock	5144097	0.5	U	2.5	U	1.2	NJ	0.5	UJ	3.8	NJ
Puyallup	5144104	0.5	U	10	UJ	1.1	NJ	0.5	UJ	10	UJ
Puyallup-Dup	5144104-Dup	0.49	U	0.49	UJ	1.0	NJ	0.49	UJ	9.9	UJ
Spokane	5144100	0.5	U	2.5	UJ	0.85	NJ	0.5	UJ	2.5	UJ
Tucannon	5144101	0.5	U	2.5	UJ	3.9	NJ	0.5	UJ	2.5	UJ
Vancouver	5144095	0.5	U	2.5	U	0.6	NJ	0.5	UJ	2.5	U
Hatchery Rainbows											
Arlington	5144087	0.49	U	2.5	U	0.9	NJ	0.49	UJ	0.77	J
Chelan	5144081	0.49	U	2.4	U	0.52	NJ	0.49	UJ	0.58	NJ
Columbia Basin	5144083	0.5	U	2.5	U	0.63	NJ	0.5	UJ	0.5	J
Eells Spring	5144088	0.5	U	2.5	U	0.5	U	0.5	UJ	0.75	J
Eells Spring-Dup	5144088-Dup	0.49	U	2.4	U	0.49	U	0.49	UJ	0.63	NJ
Ford	5144084	0.49	U	2.5	U	0.49	U	0.49	UJ	0.49	UJ
Mossyrock	5144082	0.48	U	2.4	U	0.48	U	0.48	UJ	0.88	NJ
Puyallup	5144089	0.47	U	2.3	U	0.47	U	0.47	UJ	0.47	UJ
Spokane	5144085	0.49	U	2.5	U	0.49	U	0.49	UJ	0.78	J
Troutlodge	5144090	0.49	U	2.4	U	0.49	NJ	0.49	UJ	0.75	J
Tucannon	5144086	0.49	U	2.4	U	0.62		0.49	UJ	0.49	UJ
Vancouver	5144080	0.5	U	2.5	U	0.5	U	0.5	UJ	0.33	NJ
Vancouver-Dup	5144080-Dup	0.50	U	2.5	U	0.50	U	0.50	UJ	0.30	J
Planted Rainbows											
Chapman Lake	5248102	1.0	U	4.0	U	1.0	U	1.0	UJ	0.79	UJ
Donnie Lake	5248103	1.0	U	4.0	U	1.0	U	1.0	UJ	0.79	UJ
Fan Lake	5248104	1.0	U	4.0	U	4.4		1.0	UJ	0.80	UJ
Lacamas Lake	5248100	1.0	U	4.0	U	1.0	U	1.0	UJ	0.80	UJ
Lone Lake	5248108	0.98	U	3.9	U	0.98	U	0.98	UJ	0.78	UJ
Lone Lake-Dup	5248108-Dup	0.97	U	3.9	U	0.97	U	0.97	UJ	0.78	UJ
Molson Lake	5248101	0.98	U	3.9	U	0.98	U	0.98	UJ	0.78	UJ
North Lake	5248106	0.97	U	3.9	U	0.97	U	0.97	UJ	0.55	NJ
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	0.98	U	0.96	U	0.96	U
Summit Lake	5248109	0.96	U	3.8	U	0.96	U	0.96	UJ	0.77	UJ
Summit Lake-Dup	5248109-Dup	1.0	U	4.0	U	1.0	U	1.0	UJ	0.80	UJ
Warden Lake	5248107	0.97	U	3.9	U	0.97	U	0.97	UJ	0.76	NJ

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Endo-sulfan I		Endo-sulfan II		Endo-sulfan Sulfate		Endrin		Endrin Aldehyde		Endrin Ketone	
Hatchery Feed													
Arlington	5144102	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Chelan	5144096	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Columbia Basin	5144098	2.4	UJ	2.4	UJ	REJ		2.4	UJ	REJ		REJ	
Eells Spring	5144103	2.4	UJ	2.4	UJ	REJ		2.4	UJ	REJ		REJ	
Ford	5144099	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Ford-Dup	5144099-Dup	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Mossyrock	5144097	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Puyallup	5144104	10	UJ	10	UJ	10	UJ	10	UJ	10	UJ	10	UJ
Puyallup-Dup	5144104-Dup	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ	9.9	UJ
Spokane	5144100	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Tucannon	5144101	2.5	UJ	2.5	UJ	REJ		2.5	UJ	REJ		REJ	
Vancouver	5144095	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Hatchery Rainbows													
Arlington	5144087	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Chelan	5144081	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Columbia Basin	5144083	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Eells Spring	5144088	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Eells Spring-Dup	5144088-Dup	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Ford	5144084	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Mossyrock	5144082	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Puyallup	5144089	2.3	U	2.3	UJ	REJ		2.3	U	REJ		REJ	
Spokane	5144085	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Troutlodge	5144090	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Tucannon	5144086	2.4	U	2.4	UJ	REJ		2.4	U	REJ		REJ	
Vancouver	5144080	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Vancouver-Dup	5144080-Dup	2.5	U	2.5	UJ	REJ		2.5	U	REJ		REJ	
Planted Rainbows													
Chapman Lake	5248102	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Donnie Lake	5248103	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Fan Lake	5248104	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Lacamas Lake	5248100	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Lone Lake	5248108	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
Lone Lake-Dup	5248108-Dup	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
Molson Lake	5248101	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
North Lake	5248106	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	0.96	UJ	0.96	U	0.96	UJ	0.96	U
Summit Lake	5248109	3.8	U	3.8	U	3.8	UJ	3.8	U	3.8	UJ	3.8	U
Summit Lake-Dup	5248109-Dup	4.0	U	4.0	U	4.0	UJ	4.0	U	4.0	UJ	4.0	U
Warden Lake	5248107	3.9	U	3.9	U	3.9	UJ	3.9	U	3.9	UJ	3.9	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Heptachlor		Heptachlor Epoxide		Hexachloro -benzene		Lindane		Methoxy-chlor		Mirex	
Hatchery Feed													
Arlington	5144102	0.49	U	2.5	UJ	0.16	J	0.49	U	2.5	UJ	0.49	UJ
Chelan	5144096	0.5	U	2.5	U	0.31	J	0.5	U	2.5	UJ	0.5	UJ
Columbia Basin	5144098	0.49	U	2.4	UJ	0.49	UJ	0.49	U	2.4	UJ	0.49	UJ
Eells Spring	5144103	0.48	U	2.4	UJ	0.15	J	0.48	U	2.4	UJ	0.48	UJ
Ford	5144099	0.5	U	2.5	UJ	0.16	J	0.5	U	2.5	UJ	0.5	UJ
Ford-Dup	5144099-Dup	0.50	U	2.5	UJ	0.19	J	0.50	U	2.5	UJ	0.50	UJ
Mossyrock	5144097	0.5	U	2.5	U	0.3	J	0.5	U	2.2	J	0.5	UJ
Puyallup	5144104	0.5	U	10	UJ	0.5	UJ	0.5	U	4.9	J	0.5	UJ
Puyallup-Dup	5144104-Dup	0.49	U	9.9	UJ	0.10	J	0.49	U	4.0	J	0.49	UJ
Spokane	5144100	0.5	U	2.5	UJ	0.12	J	0.5	U	2.5	UJ	0.5	UJ
Tucannon	5144101	0.5	U	2.5	UJ	0.16	J	0.5	U	2.5	UJ	0.5	UJ
Vancouver	5144095	0.5	U	2.5	U	0.17	J	0.5	U	2.5	UJ	0.5	UJ
Hatchery Rainbows													
Arlington	5144087	0.49	U	0.49	U	0.23	J	0.49	U	2.5	U	0.49	U
Chelan	5144081	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Columbia Basin	5144083	0.5	U	0.5	U	0.5	U	0.5	U	2.5	U	0.5	U
Eells Spring	5144088	0.5	U	0.5	U	0.5	U	0.5	U	2.5	U	0.5	U
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Ford	5144084	0.49	U	0.49	U	0.49	U	0.49	U	2.5	U	0.49	U
Mossyrock	5144082	0.48	U	0.48	U	0.48	U	0.48	U	2.4	U	0.48	U
Puyallup	5144089	0.47	U	0.47	U	0.47	U	0.47	U	2.3	U	0.47	U
Spokane	5144085	0.49	U	0.49	U	0.1	J	0.49	U	2.5	U	0.49	U
Troutlodge	5144090	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Tucannon	5144086	0.49	U	0.49	U	0.49	U	0.49	U	2.4	U	0.49	U
Vancouver	5144080	0.5	U	0.5	U	0.3	NJ	0.5	U	2.5	U	0.5	U
Vancouver-Dup	5144080-Dup	0.50	U	0.50	U	0.50	U	0.50	U	2.5	U	0.50	U
Planted Rainbows													
Chapman Lake	5248102	1	U	0.79	UJ	1	U	1	U	4	U	1	U
Donnie Lake	5248103	1	U	0.79	UJ	1	U	1	U	4	U	1	U
Fan Lake	5248104	1	U	0.8	UJ	1	U	1	U	4	U	1	U
Lacamas Lake	5248100	1	U	0.8	UJ	1	U	1	U	4	U	1	U
Lone Lake	5248108	0.98	U	0.78	UJ	0.98	U	0.98	U	3.9	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U
Molson Lake	5248101	0.98	U	0.78	UJ	0.98	U	0.98	U	3.9	U	0.98	U
North Lake	5248106	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U
South Lewis Co. Park Pond	5248105	0.98	U	0.96	U	0.98	U	0.96	U	0.96	U	0.98	U
Summit Lake	5248109	0.96	U	0.77	UJ	0.69	J	0.96	U	3.8	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	0.80	UJ	0.76	J	1.0	U	4.0	U	1.0	U
Warden Lake	5248107	0.97	U	0.78	UJ	0.97	U	0.97	U	3.9	U	0.97	U

Table C-5 (cont'd). Complete Results of Chlorinated Pesticide (Excluding DDT Compounds) Analysis of Feed and Fish Tissue Samples (ng/g ww)

	Sample Number	Oxychlordane		Pentachloro-anisole		Toxaphene		trans-Chlordane		trans-Nonachlor	
Hatchery Feed											
Arlington	5144102	0.49	U	0.49	UJ	4.9	U	0.49	U	0.68	
Chelan	5144096	0.5	U	0.5	UJ	5	U	1.1	NJ	1.1	NJ
Columbia Basin	5144098	0.49	U	0.49	UJ	3.9	NJ	0.2	J	0.7	
Eells Spring	5144103	0.48	U	0.48	UJ	4.8	U	0.19	J	0.44	J
Ford	5144099	0.5	U	0.5	UJ	5	U	0.5	U	0.15	J
Ford-Dup	5144099-Dup	0.50	U	0.50	UJ	5.0	U	0.50	U	0.16	J
Mossyrock	5144097	0.5	U	0.15	J	5	U	0.2	NJ	1.2	
Puyallup	5144104	0.5	U	0.5	UJ	3.6	J	0.5	U	0.5	U
Puyallup-Dup	5144104-Dup	0.49	U	0.49	UJ	4.9	U	0.49	U	0.49	U
Spokane	5144100	0.5	U	0.5	UJ	2.6	J	0.5	U	0.8	
Tucannon	5144101	0.5	U	0.11	J	5	U	0.5	U	0.3	J
Vancouver	5144095	0.5	U	0.5	UJ	5	U	0.5	U	0.77	J
Hatchery Rainbows											
Arlington	5144087	0.49	U	0.49	U	4.9	U	0.49	U	0.27	J
Chelan	5144081	0.49	U	0.49	U	4.9	U	0.49	U	0.42	J
Columbia Basin	5144083	0.5	U	0.5	U	5	U	0.5	U	0.45	J
Eells Spring	5144088	0.5	U	0.5	U	5	U	0.5	U	0.2	J
Eells Spring-Dup	5144088-Dup	0.49	U	0.49	U	4.9	U	0.49	U	0.17	J
Ford	5144084	0.49	U	0.49	U	4.9	U	0.49	U	0.49	U
Mossyrock	5144082	0.48	U	0.48	U	4.8	U	0.48	U	0.3	NJ
Puyallup	5144089	0.47	U	0.47	U	4.7	U	0.47	U	0.094	J
Spokane	5144085	0.49	U	0.49	U	4.9	U	0.49	U	0.49	U
Troutlodge	5144090	0.49	U	0.49	U	4.9	U	0.49	U	0.57	J
Tucannon	5144086	0.49	U	0.18	J	4.9	U	0.49	U	0.12	NJ
Vancouver	5144080	0.5	U	0.21	J	5	U	0.5	U	0.3	J
Vancouver-Dup	5144080-Dup	0.50	U	0.50	U	5.0	U	0.50	U	0.20	J
Planted Rainbows											
Chapman Lake	5248102	1	U	1	U	9.9	U	1	U	1	U
Donnie Lake	5248103	1	U	1	U	9.9	U	1	U	1	U
Fan Lake	5248104	1	U	1	U	10	U	1	U	1	U
Lacamas Lake	5248100	1	U	0.47	J	10	U	1	U	1	U
Lone Lake	5248108	0.98	U	0.98	U	0.98	U	0.98	U	0.98	U
Lone Lake-Dup	5248108-Dup	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U
Molson Lake	5248101	0.98	U	0.98	U	9.8	U	0.98	U	0.98	U
North Lake	5248106	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U
South Lewis Co. Park Pond	5248105	0.96	U	0.96	U	9.6	U	0.96	U	0.98	U
Summit Lake	5248109	0.96	U	0.96	U	9.6	U	0.96	U	0.96	U
Summit Lake-Dup	5248109-Dup	1.0	U	1.0	U	10	U	1.0	U	1.0	U
Warden Lake	5248107	0.97	U	0.97	U	9.7	U	0.97	U	0.97	U

Dup – Duplicate
U - Analyte was not detected at or above the reported result
UJ - Analyte was not detected at or above the reported estimated result
J - Analyte was positively identified. The associated numerical result is an estimate
NJ - There is evidence that the analyte is present. The associated numerical result is an estimate
REJ - Data are unusable for all purposes

JANET T. MILLS
ATTORNEY GENERAL

TEL: (207) 626-8800
TTY USERS CALL MAINE RELAY 711

STATE OF MAINE
OFFICE OF THE ATTORNEY GENERAL
6 STATE HOUSE STATION
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FAX: (207) 496-3291

March 17, 2015

By certified mail

Gina McCarthy, Administrator
U.S. Environmental Protection Agency
Office of the Administrator, Mail Code 1101A
1200 Pennsylvania Avenue, NW
Washington DC 20460

Eric Holder, Attorney General
U.S. Department of Justice
950 Pennsylvania Avenue, NW
Washington DC 20530

Re: **60-Day Notice of Maine's Intent to Sue EPA Over EPA's Action Dated February 2, 2015**

Dear Administrator McCarthy and Attorney General Holder:

By this letter, the State of Maine ("Maine") hereby provides notice of its intent to sue the United States Environmental Protection Agency ("EPA") and/or the United States over EPA's recent action dated February 2, 2015, which, among other things, unlawfully disapproves many of Maine's water quality standards. EPA's action also unlawfully usurps Maine's role in promulgating such standards under the Clean Water Act, and creates a two-tiered water quality regulatory system in violation of the *Maine Indian Land Claims Settlement Act*, 25 U.S.C. § 1721 *et seq.* ("MICSA") and the *Act to Implement the Maine Indian Land Claims Settlement Act,* set forth in Maine law at 30 M.R.S. §§ 6201 *et seq.* ("MIA") (collectively the "Settlement Acts").

In 1980 the Federal Government, Maine, and several Maine Indian Tribes negotiated a comprehensive settlement of Indian land claims to an area consisting of approximately two-thirds of Maine's land mass. Congress approved that settlement in MICSA, which also ratified and confirmed MIA. Together, these Settlement Acts create and define a nationally unique state-tribal relationship, and unambiguously confirm Maine's regulatory authority over Indian lands and natural resources. 30 M.R.S. § 6204; 25 U.S.C. § 1725(b)(1). Under the Settlement Acts, Maine's authority to regulate environmental matters applies uniformly throughout the State, without distinction as to tribal and non-tribal lands and natural resources, and this premise is foundational to the Settlement Acts. When the First Circuit Court of Appeals was called upon to interpret and apply these provisions in a case involving EPA's refusal to recognize Maine's authority to implement the Clean Water Act in Indian territory, the Court held that the Settlement

Acts are "about as explicit … as is possible" in conferring environmental regulatory authority on the State over Indian lands and natural resources. *Maine v. Johnson*, 498 F.3d 37, 43 (1st Cir. 2007).

Under the Clean Water Act, Maine also has the responsibility to set water quality standards for all of its intrastate waters, including all waters in Maine's Indian lands. In order to ensure that these standards are consistent with the Clean Water Act, Maine periodically submits its new and revised water quality standards to EPA for review and approval. 33 U.S.C. § 1313. For many years, EPA approved Maine's standards without distinction as to Indian lands and waters, as the Clean Water Act and the Settlement Acts each require. However, beginning around 2004, EPA for the first time began inserting limiting language into its approval letters stating that those decisions did "not extend to waters that are within Indian territories or lands." Despite the First Circuit's emphatic ruling against EPA in the *Johnson* case, and despite Maine's repeated requests that EPA approve Maine's water quality standards throughout the State, EPA continued to refuse to approve these standards for waters in Indian lands based on EPA's unfounded jurisdictional concerns. At roughly the same time, and without Maine's knowledge, EPA also provided ongoing assistance to Maine's tribes in pursuit of separate tribal water quality goals – in some cases pursuant to the terms of a confidentiality agreement obligating EPA to use its best efforts to avoid freedom of access disclosures, which ensured that Maine was kept in the dark.

As a consequence, and in order to force EPA to fulfill its mandatory duties under the Clean Water Act, Maine filed a lawsuit against EPA in July 2014 asking for, among other things, a court order requiring EPA to 1) honor Maine's environmental regulatory jurisdiction to set water quality standards for all intrastate waters, including waters in Indian lands, and 2) act on all of Maine's outstanding standards for such Indian waters. During the briefing on Maine's lawsuit, and perhaps in recognition of the weaknesses of EPA's historical jurisdictional position, EPA finally conceded what *Johnson* had already decided years earlier – that Maine does indeed have the environmental regulatory authority under the Settlement Acts to set water quality standards for all Maine waters, including those in Indian lands. Also during briefing, and presumably in order to avoid the embarrassment of a court ordering the relief formally requested by Maine, EPA committed to finally acting on Maine's outstanding water quality standards by the end of January 2015. Based on these developments, and in light of the Settlement Acts and the *Johnson* decision, Maine expected that EPA would finally end its decade-long delay and approve all of Maine's outstanding water quality standards for waters in Indian lands, as they had already been approved for non-tribal waters. As we now know, this was never EPA's intention.

EPA's action, which was by letter dated February 2, 2015, consists of various EPA approvals, disapprovals, and ongoing inaction with respect to Maine's water quality standards. As telegraphed by EPA during its briefing of Maine's July 2014 lawsuit, EPA's action correctly concludes that Maine has statewide environmental regulatory jurisdiction under the Settlement Acts to set water quality standards for all Maine waters, including Maine's waters in Indian lands. However, EPA's action then takes a surprisingly convoluted and unlawful approach in order to reach what appears to be a pre-determined result. This portion of EPA's February 2, 2015 action is carefully built upon a series of unlawful determinations designed to get around Maine's statewide environmental regulatory authority under the Settlement Acts. These unlawful determinations collectively form the shaky underpinnings of the result that EPA appears to have sought all along – the disapproval of all of Maine's human health criteria for Indian waters, even though those same standards were previously approved by EPA for the same waters immediately outside of Maine's tribal areas. The unlawful aspects of EPA's February 2, 2015 action that contribute to EPA's ultimate decision to unlawfully disapprove Maine's human health criteria standards for Indian waters include, without limitation, the following:

1. EPA unlawfully asserts that, prior to its February 2, 2015 action, none of Maine's water quality standards were ever approved for waters in Indian lands, including those standards that were fully approved by EPA before 2004 without any qualification as to their effect on waters in Indian lands;

2. EPA unlawfully asserts that it must make a formal finding of Maine's environmental regulatory jurisdiction over waters in Indian lands before EPA can approve any of Maine's water quality standards for such waters;

3. EPA unlawfully asserts that the Settlement Acts must be construed in favor of the tribes and against Maine on jurisdictional issues;

4. EPA unlawfully asserts that its duties under the Clean Water Act must be informed by EPA's interpretation of the purpose, as opposed to the actual terms, of the Settlement Acts;

5. EPA unlawfully interprets the narrow portions of MIA that permit certain Maine Indian tribes to take fish without restriction within their reservations provided that such fish takings are for individual sustenance only, as more broadly constituting a designated use of "tribal sustenance fishing" with respect to waters in Indian lands;

6. EPA unlawfully interprets Maine's existing designated use of "fishing," as used throughout Maine's water classification system, as meaning "tribal sustenance fishing" with respect to waters in Indian lands;

7. EPA unlawfully usurps Maine's role as a "State" under the Clean Water Act by establishing its own newly-created designated use of "tribal sustenance fishing" in Maine without the benefit of any public input or other required process;

8. EPA unlawfully interprets its newly-created designated use of "tribal sustenance fishing" as requiring heightened human health water quality criteria for Maine waters in Indian lands, which EPA claims will also affect discharges into other surrounding Maine waters;

9. EPA unlawfully analyzes its newly-created designated use of "tribal sustenance fishing" in the context of tribal-only population data as opposed to general Maine population data, which EPA uses for all other Maine waters; and

10. EPA unlawfully disapproves Maine's human health water quality criteria with respect to waters in Indian lands.

In effect, EPA's February 2, 2015 action, by disapproving Maine's otherwise approved human health criteria for only those waters in Maine's Indian lands, creates a two-tiered water regulatory system that elevates the water regulatory goals of Maine's Indian tribes over the rest of Maine in violation of the Clean Water Act, the Settlement Acts, and the *Johnson* decision. To add insult to injury, EPA's February 2, 2015 action also indicates that any heightened standards ultimately implemented in Indian waters will have a regulatory reach beyond Maine's Indian waters to non-tribal waters within the same watersheds. To make matters still worse, EPA does not define the scope of what it considers to be the affected Maine waters in Indian lands. In this way, EPA's February 2, 2015 action, in addition to being unlawful, irresponsibly disrupts settled regulatory expectations and causes uncertainty with respect to Maine's long-established water classification system.

As a consequence, Maine intends to sue EPA in order to challenge EPA's disapproval of Maine's human health water quality criteria with respect to the unspecified waters in Indian lands, as well as other aspects of EPA's February 2, 2015 action, including, without limitation, the unlawful EPA determinations described above. Maine believes that all of its forthcoming legal challenge to EPA's February 2, 2015 action may properly be brought as an appeal of EPA's final February 2, 2015 action pursuant to the Administrative Procedures Act, 5 U.S.C. § 701 *et seq.* ("APA").

However, as an alternative to proceeding with an appeal under the APA, and in an abundance of caution, Maine also intends to assert as part of its forthcoming lawsuit against EPA, mandatory duty claims pursuant to 33 U.S.C. § 1365(a)(2). Pursuant to 33 U.S.C. §§ 1313(a) and 1313(c)(3), EPA has the non-discretionary duty to approve Maine's human health water quality criteria for all of Maine's intrastate waters, including Maine's Indian waters, when, like here, those standards were determined by EPA to meet the requirements of the Clean Water Act 1) without qualification as to their effect in Indian waters, as was the case with all of Maine's standards before 2004; and 2) for the same waters immediately outside of tribal areas. EPA violated this non-discretionary duty by, among other things, 1) attempting to revisit its pre-2004 unqualified approvals of Maine's water quality standards in order to retroactively reconsider their effect in Indian waters; and 2) disapproving Maine's human health water quality criteria with respect to waters in Indian lands after having approved those same standards for waters immediately outside of tribal areas in violation of the Settlement Acts.

With respect to these and any other mandatory duties alleged or determined to be at issue in EPA's February 2, 2015 action, Maine hereby provides this notice of its intent to sue EPA pursuant to 33 U.S.C. § 1365(b)(2). The entities giving this notice are the State of Maine, which is a sovereign state, and the Maine Department of Environmental Protection, which is an agency of the State of Maine, both of which are represented in this matter by Maine's Attorney General, Janet T. Mills. The address and contact information of the Maine Attorney General and the Commissioner of the Maine Department of Environmental Protection are as follows:

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Patricia W. Aho
Commissioner
Maine Department of Environmental Protection
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Counsel of record in this matter include the following:

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Sincerely,

Janet T. Mills
Maine Attorney General

Sincerely,

Patricia W Aho

Patricia W. Aho
Maine Department of
Environmental Protection

cc: The Honorable Paul LePage
The Honorable Susan Collins
The Honorable Angus King
The Honorable Bruce Poliquin
The Honorable Chellie Pingree
Kirk Francis, Chief
Frederick Moore, III, Chief
William Nicholas, Sr., Chief
Brenda Commander, Chief
Edward Peter-Paul, Chief
H. Curtis Spalding, EPA Region I Administrator

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
REGION 10
1200 Sixth Avenue, Suite 900
Seattle, WA 98101-3140

OFFICE OF THE
REGIONAL
ADMINISTRATOR

APR 0 8 2014

Maia Bellon, Director
Department of Ecology
P.O. Box 47600
Olympia, Washington 98504-7600

Dear Director Bellon:

I am writing in regards to your February 14, 2014 letter to Michael Grayum, Executive Director of the Northwest Indian Fisheries Commission and more recent conversations we have had with you and your staff. The U.S. Environmental Protection Agency was strongly encouraged by the Department of Ecology's timeline to adopt human health criteria with a draft rule by the end of March 2014 and a final rule submitted to the EPA by December 31, 2014. Although the EPA was disappointed to hear that Ecology will no longer be able to meet the commitment to issue a draft rule in March, we understand that Ecology remains committed to adopting a final rule by the end of 2014.

The EPA supports Ecology's use of the scientifically sound regional and local fish consumption data available in Washington as it develops its human health criteria. As I have previously stated, the best available science includes evidence of fish consumption rates well above 6.5 grams per day among high fish consumers in Washington, which raises concerns that the human health criteria currently in effect for Clean Water Act purposes in Washington are not sufficiently protective. In addition, as we have also discussed, another important element of a final rule is choosing a cancer risk level that provides risk protection for all Washington citizens, including communities that eat higher amounts of fish. The EPA remains committed to supporting Ecology as it moves forward with a rulemaking based on sound scientific data. This will help facilitate the EPA's review of Ecology's new or revised water quality standards, which requires EPA to ensure that criteria are protective of applicable designated uses and based on a sound scientific rationale, consistent with 40 CFR § 131.11(a).

If Ecology does not follow through with its stated timeframe for final rule adoption, the EPA intends to take the steps necessary to allow for a proposal of federally revised human health criteria for Washington, via amendment of the National Toxics Rule human health criteria for Washington, by May 31, 2015. For the sake of clarity, I note that this letter does not constitute, and is not intended as, an Administrator determination under CWA § 303(c)(4)(B).

The EPA is very appreciative of the challenging work that Ecology has undertaken thus far to adopt human health water quality criteria. I look forward to continuing our partnership to support state adoption of human health criteria to better ensure health protection for all Washington citizens. If you would like to discuss these topics further, please contact me directly or Dan Opalski, our Director for the Office of Water and Watersheds, at (206) 553-1855.

Sincerely,

Dennis J. McLerran
Regional Administrator

cc: Michael Grayum, Executive Director
Northwest Indian Fisheries Commission

Brian Cladoosby, Tribal Chairman
Swinomish Tribe

From:	Opalski, Dan
Sent:	Monday, March 17, 2014 10:35 AM
To:	ksus461@ecy.wa.gov
Subject:	FW: Listing and EJ Discussion

Kelly –

A slight addendum…

Consistent with our previous discussions, I wanted to confirm that Oregon DEQ has relatively recently proposed 303d listings for mercury based upon fish tissue concentrations (not relying on an OHA advisory).

I am understanding that some communication is now out on the end of March date slipping. I'd appreciate a chance to check in soon on this.

Thx. DanO.

From: Opalski, Dan
Sent: Saturday, March 15, 2014 12:26 AM
To: 'Susewind, Kelly (ECY)'
Subject: RE: Listing and EJ Discussion

Kelly –
Sorry I didn't get more back to you on the listing sooner. Here is what was reported to me:

DEQ has only listed impaired waters for fish tissue contamination when the Oregon Health Authority (OHA, formerly DoH) has issued fish advisories, i.e., DEQ has not done any independent review of fish tissue data for listing purposes, even for mercury, which is shown as a fish tissue criterion in their WQS.

OHA calculates their fish consumption advisories for a maximum of four, eight-ounce, meals per month which translates into 30.25 grams of fish per day, much lower than the 175 grams per day upon which the Oregon toxics criteria for human health are based**.**

So best I can tell this tracks pretty well with your understanding. However, I would say that because OHA's advisories, which trigger 303d listings, are based on fish tissue data, it is accurate to say that these listings are based upon tissue data. But it remains notable that DEQ does not independently review the tissue data for these purposes, nor does there appear to be any explicit translation of the tissue concentrations to their water column standards, and, finally, the fish consumption rate used by OHA is quite a ways off from the rate underlying their standards.

DanO.

From: Susewind, Kelly (ECY) [mailto:KSUS461@ECY.WA.GOV]
Sent: Tuesday, March 11, 2014 1:41 PM
To: Opalski, Dan
Cc: Bellon, Maia (ECY)
Subject: Listing and EJ Discussion

Hey Dan

Did a little looking on my own following today's discussion.

Listing:

The 2012 Oregon assessment states that:

- New and revised human health criteria apply to pollutants in the water column except for methyl mercury…..
- Category 5 listings require two or more samples not meeting the most stringent applicable criterion of a specific substance in the water, or
- A fish consumption advisory issued for a specific water body based on pollutants in fish tissue

We acknowledged that Oregon lists based on fish advisories, but that is far different than saying they do listing based on tissue. A quick perusal of Oregon's fish advisories only shows a few advisories generally based on mercury and PCBs.

We've also been contacted by DEQ staff regarding our listing policy because they are getting pressure to list based on tissue "like Washington."

Is there more information that I am missing?

EJ

I have a copy of the document: "EPA Policy on Environmental Justice for Tribes and Indigenous Peoples." It's a pre-decisional working draft dated November 14,2012.

Is that the document Dennis referred to?

The only real pertinent language I could find in that document was:

4. THE EPA ASSESSES THE POTENTIAL FOR DISPROPORTIONATELY HIGH AND ADVERSE HUMAN HEALTH OR ENVIRONMENTAL EFFECTS ON TRIBES OR INDIGENOUS COMMUNITIES.
a. The EPA considers both quantitative and qualitative information about the potential disproportionately high and adverse human health or environmental effects pertaining to, and/or provided by, tribes or indigenous stakeholders.
b. The EPA works to understand Traditional Ecological Knowledge and its role in protecting public health and the environment, and to understand community definitions of health and the environment.

As we discussed, tribal members, and anyone eating high amounts of fish, are at higher risk. They are at a risk exactly proportionate to the consumption rate and will be at the same ratio (proportion) regardless of where the rule lands. Interpreting this section of the policy to mean that they can't be at a higher risk would frustrate the entire system the HHC equations are based on and make it impossible to comply.

Is there a statement somewhere that one in a million risk rate is the baseline to establish environmental justice? Or that a higher risk rate is inherent in the approach, but establishes some criteria to define "disproportionately high and adverse effects?

I’m not trying to be argumentative, but we are getting to the end of a very contentious process, and I really need to understand these concepts in order to advise decision makers.

Thanks

Kelly