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2.6 PFAS Releases to the Environment

This section summarizes sources of PFAS releases to the environment that have the potential for significant environmental impact, based on the type and magnitude of the release, and the types and concentrations of PFAS associated with that release. These sources are sites where PFAS could be, or are known to have been, released to the environment, even if the site is not the location where the PFAS were generated or used. Refer to Section 2.1 for a discussion of the relative significance of releases and source control, as not all of these facilities will have, or have been documented to have, PFAS releases, and not all releases are of the same magnitude. "In the absence of high-quality testing data, PFAS contamination could be presumed" (Salvatore et al. 2022) at major sources and certain facilities as mentioned below. In addition, USEPA has developed an online database called the PFAS Analytic Tools that provides access to different sources of information about potential PFAS sources, drinking water sampling data, occurrence in environmental media, among others (USEPA 2023).

These major sources are located both in the United States and abroad, and include:

- industrial facilities that produce PFAS or process PFAS, or facilities that use PFAS chemicals or products in manufacturing or other activities (Section 2.6.1)
- areas where fluorine-containing Class B firefighting foams are stored, used, or released (Section 2.6.2)
- waste management facilities, such as landfills (Section 2.6.3)
- wastewater treatment residuals and areas of biosolids production and application, with more significant impacts associated with industrial wastewater discharges (<u>Section 2.6.4</u>).

The fate and transport processes and distribution of PFAS in the environment are discussed in <u>Section 5</u>. Media-specific occurrence data are discussed in <u>Section 6</u>. Information about risk assessment, and human and ecological receptors is included in <u>Section 9</u>. Discussion of conceptual site model (CSM) components for each of the PFAS release categories listed above are included in <u>Section 10.2.1</u>.

2.6.1 Major Manufacturing and Industry Sources

Industrial source sites include primary and secondary manufacturing facilities. Primary manufacturing facilities are those where PFAS-containing products are synthesized and made into products or chemical feedstocks, or where PFAS are used as processing aids in fluoropolymer production. PFAS processing aids are not intended to be in the final product, but may be present at trace quantities (<u>3M Company 2003</u>; <u>Buck et al. 2011</u>).

Secondary manufacturing facilities may use fluoropolymers and PFAS-based materials produced at primary manufacturing facilities as part of industrial processes, such as the application of coatings to finished products. In some industrial settings, PFAS have been used for worker safety purposes, such as using 6:2 FTS or PFOS-based materials to suppress harmful mists during electroplating activities (Section 2.6.1.3).

PFAS composition and release mechanisms will vary for each facility. The composition of PFAS released from industrial facilities depends on the type of PFAS produced or used by the facility.

The general PFAS release mechanisms and pathways at industrial facilities are illustrated in CSM Figure 2-18 and include wastewater and stormwater discharges; on- and off-site disposal of solid wastes; accidental releases such as leaks and spills; and stack and fugitive emissions. Stack emissions may result in aerial deposition of PFAS to soil and surface water (with subsequent leaching and infiltration to groundwater) related to the facility (Davis et al. 2007; Shin et al. 2011), as well as short- and long-range air transport of PFAS. Industrial facilities may also contain areas where fire training or fire response using AFFF has occurred, AFFF storage areas, and AFFF fire suppression systems inside buildings. Like many AFFF release sites, industrial sites may also have releases of co-contaminants (solvents, petroleum products, etc.) that could potentially influence fate and transport of PFAS.

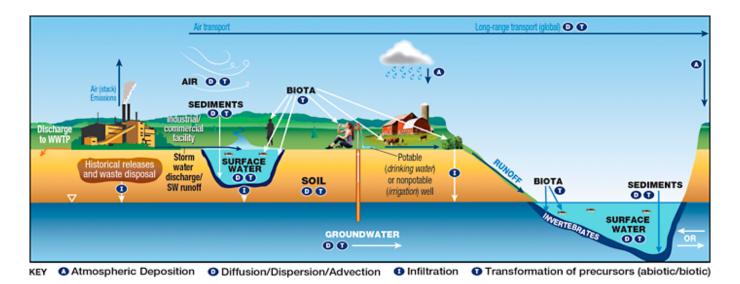


Figure 2-18. CSM for industrial sites.

Source: Adapted from figure by L. Trozzolo, TRC, used with permission.

The following subsections provide further details regarding potential sources of PFAS releases to the environment from PFAS use in manufacturing or industrial processes; these are not presented in order of the potential for significance of a release.

2.6.1.1 Building and Construction

Similar to other products, the chemical attributes of PFAS have led to advancements in building and construction materials. One particular application has been in composite wood and oriented strand board (OSB). Over the last 50 years, wood-based materials have used numerous additives for product strength and durability. A recent study performed on wood samples and OSB found primarily short-chain PFCAs and PFOA at concentrations ranging from 1.38 to 13.9 micrograms per kilogram (µg/kg) (Bečanová et al. 2016). Furthermore, wood fiber insulation has been shown to contain high amounts of PFHpA and other 5- to 8-carbon chain PFCAs (Bečanová et al. 2016). Many manufacturers use urea- or phenol-formaldehyde due to their performance and low cost; however, the composition of the resins used by many manufacturers is proprietary.

Other materials, including certain types of building insulation (phenolic foam) have shown high amounts of PFOS. Additionally, PFAS (predominantly C8–C20 gamma-omega-perfluorotelomer thiols with acrylamide) have been used in the production of lightweight concrete, concrete sandwich panels, and lightweight concrete blocks (Bečanová et al. 2016; Posner et al. 2013). The prevalence of these building materials in the construction of fire training areas, AFFF storage facilities, and other areas potentially exposed to PFAS led to potential issues with demolition waste. The porous nature of these materials (for example, concrete, brick) could lead to PFAS adsorption/absorption, representing a potential source of PFAS when disposed in landfills or recycling facilities (Australia Government DOD 2018).

PFAS, including fluoropolymers such as PTFE, are used in the manufacture of architectural fabrics, such as those used in the construction of roof domes, including large stadiums and transportation facilities (Performance Fluoropolymer Partnership 2021; Glüge et al. 2020).

PFOS-related chemicals have several uses in paint and varnishes. They can be used as wetting, leveling, and dispersing agents, and have also been used to improve gloss and antistatic properties. Additionally, they can be used as additives in dyestuff and ink. Furthermore, they can be used as pigment grinding aids or as agents to combat pigment flotation problems (KEMI 2004; RPA 2004). Fluorosurfactants are commonly used in coatings application for substrate wetting, leveling, reduction of surface tension, oil repellency, and dirt pickup resistance (Danish EPA 2015; Posner et al. 2013).

Information received from different suppliers within the paint and varnish industry suggests that fluorinated surfactants in general are much more expensive alternatives compared to other surfactants. Therefore, fluorosurfactants are used only for special purposes in paint and varnishes, where it is necessary to gain such a low surface tension that no other (nonfluorinated) alternatives can achieve (Danish EPA 2015).

Studies pertaining to PFAS in building materials continue to be published. The Green Science Policy Institute recently published a report that summarizes currently available studies for the purposes of informing those in the building and construction industry of the presence of PFAS and eliminating unnecessary uses (Fernandez, Kwiatkowski, and Bruton 2021).

2.6.1.2 Cable and Wiring

In the 1950s the wire and cable industry began to use extruded grades of PTFE. This is a suspension polymerization process, which does not require surfactants, unlike dispersion polymerizations (for example, Teflon-coated pans). Melt extrusion is the process by which most fluoropolymers are applied to wires. For instance, FEP, PFA, and PVDF are heated to 260°C and then melt extruded over wire to continuous lengths. The equipment used for melt-processable fluoropolymers requires temperature sensitivity of 427°F. PTFE is processed via paste extrusion for coating PTFE over wires due to its high melting point (ASTSWMO 2015; Kotthoff et al. 2015; Lau et al. 2007; Lindstrom, Strynar, and Libelo 2011; Oliaei et al. 2013; Renner 2001; Trudel et al. 2008). For more information on the safe handling of fluoropolymer resins during processing, see the Plastics Industry Association (2019) guidance document.

2.6.1.3 Metal Finishing and Plating

Electroplating is a process that uses electric current to apply a metal coating to the surface of an object. Metallic ions in an acidic electrolyte solution are used in the electrochemical deposition of metal coatings to the surface of the cathode (<u>USEPA</u> 1996).

PFAS, particularly PFOS, have been used as mist suppressants that are added to metal plating and finishing baths to prevent air emissions of toxic metal fumes. Glüge et al. (2020) identified PFAS use in chrome, nickel, copper, tin, and zinc plating for lowering surface tension. In the United States, amendments to the National Emissions Standards for Hazardous Air Pollutants (NESHAP) under the Clean Air Act included a requirement to phase out the use of PFOS-based fume suppressants (a fume suppressant that contains 1% or greater PFOS by weight) in chromium electroplating by 2015 (USEPA 2012). Some countries have phased out the use of PFOS in some electroplating operations, adopting the use of other fluorotelomers (for example, 6:2 FTS) as a substitute in hard chrome plating operations (Danish EPA 2015; KEMI 2015) or changing decorative chrome plating operations to employ the less toxic trivalent chromium. PFAS known by the trade name F-53B (11-Chloroeicosafluoro-3-oxaundecane-1-sulfonic acid [11Cl-PF3OUdS or F-53B Minor], 9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid (9Cl-PF3ONS or F-53B Major]) have been used as metal plating mist suppressants in China (USEPA 2021; Bao et al. 2019). The toxicology of F-53B is reviewed in Section 17.2.6.1 (note that toxicological research may use different nomenclature for F-53B, namely 6:2 chlorinated polyfluoroether sulfonate (6:2 CIPFESA) and 8:2 CIPFESA (Munoz et al. 2019).

Many different types of electroplating solutions can be used in plating activities, including hard and decorative chrome plating; chromic acid anodizing; nickel, cadmium, or lead plating; metal plating on plastics; and alkaline zinc plating. Chrome electroplating is the most significant contributor as it relates to PFAS use. In this process, PFAS are used as surfactants to reduce the surface tension of the electrolyte solution. Historically, PFOS was commonly used at a concentration of 5–10% to limit the development of bubbles and the emission of hexavalent chromium aerosols to workplace air, thereby reducing the potential hazard to workers posed by hexavalent chromium (<u>USEPA 2009</u>) (<u>OSHA 2013</u>) (<u>Danish EPA 2015</u>).

Studies show use of PFAS in these settings can result in high concentration wastewater discharges (<u>USEPA 2009</u>) and air emissions. Once the electrolyte solution can no longer be used, it may be treated to remove chromium and other metals, but PFOS and other PFAS may be present in effluent and deposited in sewage sludge (<u>Danish EPA 2015</u>). Investigations in Minnesota traced PFOS releases from one chrome plating operation to a wastewater treatment plant (WWTP) where elevated levels of PFOS were detected in the biosolids, effluent water, and fish in the receiving surface water (<u>ATSDR 2008</u>). Air emissions from another Minnesota chrome plater were found to have accumulated on the roof of the facility and from there contaminated stormwater and snow melting from the roof, which in turn contaminated the groundwater, a nearby surface water system, and fish (<u>MPCA 2016</u>).

According to a study in Michigan by the Michigan Department of Environment, Great Lakes, and Energy (EGLE), 320 metal finishers that had a history of using fume suppressants were found to have PFOS in wastewater effluent. The report noted that 15% of metal finishers were discharging to WWTPs at concentrations greater than screening criteria (12 ppt PFOS) and 5% were discharging greater than 1,000 ppt PFOS (MI EGLE 2020). Of the metal finishers discharging PFOS above screening criteria, 89% used hexavalent and/or trivalent chromium in their current or past processes. Chrome platers in Michigan were determined to be in compliance with the NESHAP and many replaced PFOS with a fume suppressant containing 6:2 FTS. Some chrome platers did not use PFOS-containing chemicals to control fumes and have not been found to be sources of PFOS to WWTPs. Nearly half of the chrome platers regulated under the NESHAP used mechanisms other than chemical fume suppression. It was concluded that current effluent containing PFOS from facilities that have complied with NESHAP originates from historical use of PFOS-containing fume suppressants.

2.6.1.4 Industrial Surfactants and Fluoropolymer Production

PFAS have been, and currently are, instrumental as surfactants in industrial and commercial production. In the recent past, some information pertaining to specific surfactant uses of PFAS has become publicly available, though much information still remains unavailable. Most well documented is the historical use of PFOA as a processing aid in the manufacturing of PTFE, where APFO is used to help mix together the chemicals needed to combine units of tetrafluoroethylene (TFE) to make PTFE. Similarly, APFN, the ammonium salt of perfluorononanoic acid (PFNA), has also been used in the production of PVDF. PVDF polymers that are produced with the aid of APFN are sold in solid phase, with notable residual APFN concentrations (100–200 ppm) (Prevedouros et al. 2006).

Since the voluntary phaseout of PFOA and related PFAS chemistries, replacement chemistries such as ADONA and the GenX process chemicals are now used in the production of fluoropolymers.

The PFAAs used as polymerization aids may occur as impurities/residuals in some fluoropolymer products, as discussed in detail in Section 2.2.2.1

PFAS are also used in the manufacturing of plastics and fluoropolymers, rubber, and compression mold release coatings. These have applications in tubing, piping, drums, molds, and resins (<u>Poulsen et al. 2005</u>; <u>Prevedouros et al. 2006</u>).

2.6.1.5 Paper Products and Packaging

Since the 1960s, PFAS have been used as grease-proofing agents on food contact materials (FCM) to prevent oil, grease, and moisture from foods from leaking through the packaging. This includes coated paper and cardboard such as pizza boxes, microwavable popcorn bags, parchment paper, fast food wrappers, paper cups, pet food bags, and other items (Rao and Baker 1994; Hekster, Laane, and De Voogt 2003; Poulsen et al. 2005; Trudel et al. 2008; Buck et al. 2011).

The U.S. Food and Drug Administration (FDA) currently approves more than 90 unique monomer and polymer PFAS in FCMs (USFDA 2016). In January 2016, the FDA rescinded approval for three families of long-chain PFAS used in FCMs, but these had been voluntarily removed from the market in 2011. N-MeFOSE and NEtFOSE were historically used to produce surface coatings for textiles and paper products (Zaggia and Ameduri 2012). PFAS currently used in FCM include polyfluorinated polyether-based polymers and shorter chain PFAAs (Wang, et al. 2015; Schaider et al. 2017). See Section 8.2 for additional information regarding a voluntary phaseout of 6:2 FTOH.

The most common PFAS detected in U.S. fast food wrappers include PFCAs (for example, PFOA and PFHxA), PFSAs (for example, PFBS), and fluorotelomer sulfonates (for example, 6:2 FTS) (Schaider et al. 2017). Six of 20 FCM tested were found to contain detectable levels of PFOA even though in 2011 U.S. manufacturers had voluntarily agreed to stop distributing FCM that were manufactured using PFOA via an FDA initiative. The methodology was not sensitive enough to detect if the PFAS were intentionally added to the packaging material or if they were attributed to unintentional background levels (Schaider et al. 2017). Refer also to Section 2.4.3 on the USEPA 2010/2015 PFOA Stewardship Program, which discusses the phaseout of PFOA and potential sources of PFOA that may remain in commercial and consumer products.

2.6.1.6 Photolithography/Semiconductor Industry

The semiconductor industry historically has used PFOS for their surface-active properties in the fabrication of imaging devices such as digital cameras, cell phones, printers, and scanners (Poulsen et al. 2005). Studies have shown semiconductor waste streams containing the PFAAs PFBS, PFHxS, PFOS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, and PFDOA (Lin, Panchangam, and Lo 2009). Similarly, in photolithography processes, PFOS has been used predominantly in applying top-layer antireflective coatings (TARCs), bottom antireflective coatings (BARCs), and etchants. Smaller quantities of PFOS and longer-chain PFAS have been used in wet etchants, film developers, cleaners, protective coatings, and color filters (SIA 2008), with ongoing uses permitted (Section 2.4).

2.6.1.7 Textiles, Leather, and Apparel (Including Carpet and Furniture)

Surface treatment of textiles, leather, carpet, and furniture upholstery with PFAS to make them stain, oil, and water repellent occurs both before (that is, at the factory) and after consumer acquisition for ongoing stain, oil, and water repellency (Prevedouros et al. 2006; Ahrens 2011; Herzke, Olsson, and Posner 2012). Aftermarket PFAS-containing stain-repellent products for carpets allow consumers to treat carpets and textiles at home (Renner 2001; Hekster, Laane, and De Voogt 2003). Losses to the environment can be related to dry cleaning and laundering activities (Poulsen et al. 2005; 3M Company 2000).

Home textiles, including furniture and carpeting, as well as aftermarket PFAS surface treatment products, are also sources of

long-chain perfluorinated chemical exposure (<u>Guo et al. 2009</u>). Textile coating operations may use water-emulsion or powdered feedstocks that contain greater proportions of PFCAs compared to PFSAs (<u>Lassen et al. 2015</u>; <u>Gremmel, Frömel, and Knepper 2016</u>). According to California EPA (CalEPA) <u>CalEPA (2018)</u>, pg. 12, "The PFAS polymers used in carpets, rugs, and other textiles can contain various amounts of mobile residual raw materials, impurities, or degradation products, including PFAAs and other PFAA precursors such as fluorotelomer alcohols (FTOHs) and perfluoroalkyl sulfonamide alcohols." Releases to the environment could occur from disposal of carpet cleaning wastewater (<u>CalEPA 2018</u>). Physical degradation of some consumer products (such as PFAS-treated textiles and carpets, as well as paper) may be a source of PFAS in house dust (<u>Björklund</u>, <u>Thuresson</u>, and de <u>Wit 2009</u>).

It should be noted that many treated home textiles and carpets are now manufactured with alternatives to long-chain PFAS; however, these products can have a long useful life, making it possible that items previously treated with long-chain PFAS are still in use (<u>Brooke et al. 2004</u>). A 2009 study of over 100 consumer products conducted by the USEPA and Arcadis indicated that pretreated carpet, treated upholstery and textiles, as well as other floor treatments, are likely the largest source of PFAS receptor exposure in American homes (<u>Guo et al. 2009</u>).

Other studies have since shown nonpolymeric PFAS in leather samples and outdoor textiles to impart water, oil and stain resistance; applications include protective clothing, outerwear, footwear, umbrellas, tents, and sails (OECD 2013; Walters and Santillo 2006; Kotthoff et al. 2015). Durable water repellent (DWR) is a fabric surface finish that creates a protective barrier. It is typically added at the factory, but is also available to consumers for apparel maintenance (Brooke et al. 2004). The finishes/treatments are applied to materials in mills/tanneries and as aftermarket applications by professionals or do-it-yourself consumers as aqueous dispersions. In some aftermarket applications, they are applied as solutions in hydrocarbon-based or halogenated solvents (OECD 2013).

2.6.1.8 Other Potential Commercial or Domestic Sources of PFAS Releases to the Environment

There is the potential for everyday uses of PFAS to result in relatively smaller releases of PFAS to the environment. Of note, these may include, but are not limited to leaching from materials to media (for example, well construction and plumbing materials), discharges to on-site wastewater disposal systems from use of household products and cosmetics, discharges from car washing and waxing, and use of ski waxes (professional ski wax technicians may have significant inhalation exposures to PFAS (Nilsson et al. 2013). Snowmelt and surface waters (Kwok et al. 2013), as well as snow, soil, and groundwater (Carlson and Tupper), near ski areas may have measurable PFAS impacts.

2.6.2 Class B Fluorine-Containing Firefighting Foams

Some Class B firefighting foams designed for extinguishing flammable liquid hydrocarbon fires and vapor suppression may contain fluorine. These foams can be a major source of local PFAS release to the environment, with the CSM included in Figure 2-19.

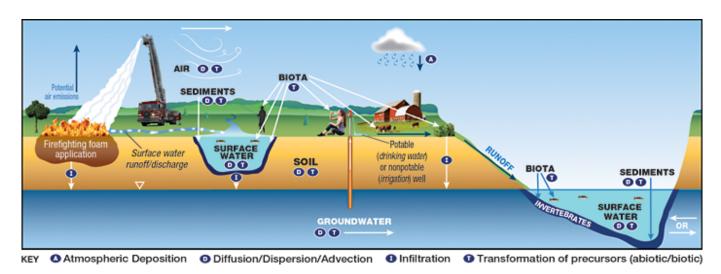


Figure 2-19. CSM for fire training areas.

Source: Adapted from figure by L. Trozzolo, TRC, used with permission.

Class B firefighting foams are commercial surfactant solutions that have been (and continue to be) stored and used for fire suppression, fire training, and flammable vapor suppression at military installations and civilian facilities and airports (<u>Hu et</u>

<u>al. 2016</u>), as well as at petroleum refineries and bulk storage facilities, and chemical manufacturing plants and storage facilities (<u>CONCAWE 2016</u>). Additionally, local fire departments in communities have used and may maintain quantities of firefighting foam in their inventories for use in training and emergency response. Facilities that manufactured firefighting foams and landfills that received firefighting waste are also potential sources. Refer to <u>Section 3</u> for more detailed information about firefighting foams.

2.6.3 Solid Waste Management Facilities

Environmental releases associated with the use of PFAS-containing products are primarily related to management of solid waste (for example, disposal of used items in a municipal solid waste (MSW) landfill or other legacy disposal areas). Other solid waste facilities, such as scrap yards and metal salvage facilities, might also be a potential source of release to the environment. Some PFAS are considered hazardous waste by some states (Section 8). Additional information pertaining to disposal of PFAS and PFAS-containing materials at MSW landfills can be found in the 2020 USEPA Interim Guidance on the Destruction and Disposal of Perfluoroalkyl and Polyfluoroalkyl Substances and Materials Containing Perfluoroalkyl and Polyfluoroalkyl Substances (USEPA 2020).

Landfills can be sources of PFAS because they are the ultimate repositories for PFAS-contaminated industrial waste, sewage sludge from wastewater treatment facilities, and waste from site mitigation, as well as for PFAS-bearing consumer wastes, such as goods treated with hydrophobic, stain-resistant coatings (Busch et al. 2010; Eggen, Moeder, and Arukwe 2010). But the type and concentration of PFAS vary greatly among landfills, due to variations in the waste streams. Industrial waste can be a significant source of PFAS in landfills (as well as in wastewater and biosolids), particularly those that accept waste from facilities involved in the production or application of PFAS (Oliaei et al. 2013). Although MSW will contain PFAS due to its presence in so many consumer products, it generally is expected to have lower concentrations than landfills that accept industrial waste. Given the production timeline of PFAS, industrial, commercial, and consumer products and waste disposed since the 1950s are potential sources of PFAS release to the environment. As PFAS manufacturing processes change with time, the resulting type and composition of waste streams also change. PFAS production and use began several decades before the enactment of federal and state regulations governing waste disposal; as a consequence, environmental and drinking water impacts from disposal of legacy PFAS industrial and consumer waste have been documented (Oliaei, Kriens, and Weber 2010; Shin et al. 2011; MPCA 2017).

<u>Figure 2-20</u> illustrates common elements of CSMs associated with the potential release scenarios at waste management facilities.

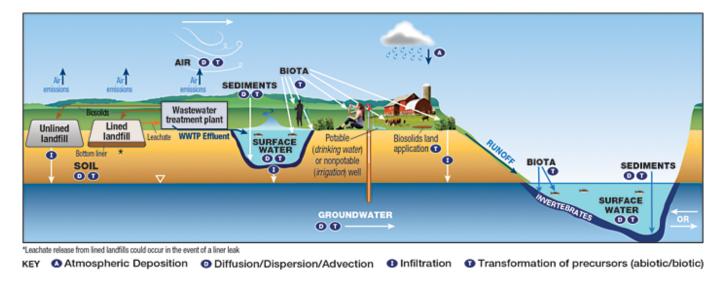


Figure 2-20. CSM for landfills and WWTPs.

Source: Adapted from figure by L. Trozzolo, TRC, used with permission.

2.6.3.1 Landfill Construction

Landfills are either lined or unlined (Figure 2-20). MSW landfills constructed since the 1990s are required by federal or state regulations to install a composite liner, a layer of compacted soil, and a leachate collection system (40 CFR 258.40). Although some states may have implemented construction standards at an earlier date, most landfills constructed before the 1990s were not required to have synthetic flexible membrane liners, compacted soil liners, or leachate collection

systems, causing waste to be in direct contact with underlying soil or groundwater. Construction and demolition (C&D) landfills or nonmunicipal solid waste landfills are subject to the requirements specified in 40 CFR 257 Part A (and if they intend to accept very small quantity generator waste, they are also subject to 40 CFR 257 Part B). Minimum design criteria for landfill liners are not specified in 40 CFR 257. Therefore, new C&D and nonmunicipal solid waste landfills may be permitted and constructed (or new cells added to existing facilities) without synthetic liners. Some states may have more restrictive requirements. Therefore, unlined landfills (and legacy disposal areas not classified as landfills) have a higher potential of contributing PFAS to groundwater (Oliaei et al. 2013). Properly constructed and operated modern landfills provide one of the few available disposal/management options for PFAS-containing waste, including wastewater solids, remedial/treatment waste, and consumer products. The USEPA Interim Guidance on the Destruction and Disposal of Perfluoroalkyl and Polyfluoroalkyl Substances and Materials Containing Perfluoroalkyl and Polyfluoroalkyl Substances provides further discussion about the use of landfills for management of PFAS-containing wastes, the potential for PFAS to be released to the environment from landfills, and the additional research and data that are needed to further assess the effectiveness of managing PFAS discharges and emissions from all landfills (USEPA 2020).

Landfills are currently required to use a daily cover or alternate daily cover. It is acceptable for alternative daily cover to include materials such as sludge, sludge-derived products, shredded automotive parts, spray-on foams, and other materials (Pohland and Graven 1993) that are possible sources of PFAS. Landfill caps reduce infiltration of water to waste and may reduce the overall mass of PFAS entering the environment from a landfill, but more research on their effectiveness is needed (Hamid, Li, and Grace 2018).

Leachate from some MSW landfills has been shown to be a source of PFAS release to the environment (Busch et al. 2010; Eggen, Moeder, and Arukwe 2010), although the fate and transport processes for PFAS through landfills into leachate are not well understood at this time. The processes for managing leachate have implications on the ultimate fate and transport of PFAS. Leachate collected from landfills is typically treated on site or transported to either a WWTP or evaporation ponds. If liners or leachate collection systems fail, PFAS may directly enter the environment. Modern landfills with properly constructed and operated liner and leachate collection systems should generally protect the underlying groundwater from PFAS releases. Leachate treatment by WWTPs is common prior to discharge to surface water or distribution for agricultural or commercial use (Lang 2016). However, standard WWTP technologies are generally ineffective at reducing or eliminating PFAS (Hamid and Li 2016; Ahrens et al. 2016; CRC CARE 2017). As a result, the discharge of landfill leachate, even if treated at WWTPs, can be a significant source of release of some PFAS to the environment (Ahrens et al. 2016; CRC Care 2017).

2.6.3.2 Waste Age

Landfills containing sources of PFAS may continue to release PFAS to leachate at slow but relatively steady rates for decades following initial placement. In modeled anaerobic landfill reactors, most of the release is attributed to biological, not physical, mechanisms, indicating that the low solubility of the compounds is not solely responsible for slow release rates from landfills (Allred et al. 2015; Lang et al. 2016). Although landfill leachate PFAS concentrations can be relatively high, landfill leachate discharged to WWTPs for treatment generally is considered a relatively minor source to the environment because the volume of leachate generated annually and sent to a WWTP for treatment is low compared to the flow volume in most WWTPs (Busch et al. 2010). However, legacy industrial waste landfills may constitute a major source of PFAS release to the environment (ATSDR 2008, 2012).

2.6.3.3 PFAS Composition

PFAS composition and concentration in leachates vary depending on waste age, climate, and waste composition (Allred et al. 2015; Lang et al. 2017). Relative concentrations of PFAS in leachate and groundwater from landfills are different from those at WWTPs and AFFF-contaminated sites. PFAS with fewer than eight carbons tend to dominate landfill leachate because they are less hydrophobic and therefore more likely to partition to the aqueous phase (Huset et al. 2011; Higgins and Luthy 2007). In particular, 5:3 fluorotelomer carboxylic acid (FTCA) is a common and often dominant constituent of PFAS found in landfills and is released from carpet in model anaerobic landfill reactors. This compound could prove to be an indicator of PFAS in the environment originating from landfills (Lang et al. 2017, 2016).

PFAS may also be released to the air from landfills, predominantly as fluorotelomer alcohols (FTOHs) and perfluorobutanoate (PFBA). In one study, total PFAS concentrations were 5–30 times greater at landfills than at background reference sites (Ahrens et al. 2011). PFAS release rates vary with time for a given waste mass, with climate (for example, rainfall) as the apparent driving factor for the variations (Lang et al. 2017; Benskin, Li, et al. 2012). Gas collection systems commonly employed at modern landfills should reduce possible PFAS emissions to air.

2.6.4 Wastewater Treatment and Wastewater Treatment Residuals and Biosolids

Consumer and industrial use of PFAS-containing materials, including disposal of landfill leachate and firefighting foam, may discharge PFAS-containing wastewater to municipal and industrial WWTPs (<u>Lin, Panchangam, and Lo 2009</u>; <u>Ahrens et al. 2009</u>) private septic systems, or other wastewater disposal systems.

2.6.4.1 Wastewater Treatment

WWTPs can provide the following pathways for PFAS to the environment (Figure 2-20):

- point source discharges of effluent
- leakage or unintended releases from surface impoundments and structures
- air emissions
- management and disposal of biosolids (https://www.epa.gov/biosolids) and other byproducts generated during the treatment process (Section 2.6.4.2).

The composition of PFAS in these media is a function of the different sources to the WWTP influent and the WWTP processes (Chen, Lo, and Lee 2012; Oliaei, D. Kriens, and Kessler 2006; Frömel et al. 2016; Schultz et al. 2006), including:

- type and concentration of PFAS received by the WWTP, particularly those that receive industrial wastewater discharges from industrial facilities manufacturing or using PFAS
- biological and chemical transformation of polyfluorinated substances (that is, precursor PFAS) to intermediate and terminal degradation products, such as PFAAs
- physical or chemical partitioning, or both.

Conventional sewage treatment methods used in WWTPs do not efficiently remove PFAAs (<u>Ahrens et al. 2011</u>; <u>Schultz et al. 2006</u>). Even WWTPs with advanced treatment technologies (such as granular activated carbon (GAC), powdered activated carbon (PAC), or reverse osmosis (RO)) may not fully remove all PFAS if these systems were not designed with the intent to remove PFAS in addition to other targeted contaminants. Some PFAAs are frequently detected in WWTP effluent (for example, PFOA and PFBS), with concentrations of some PFAS ranging up to hundreds of ng/L. <u>Ahrens et al. (2011)</u> and <u>Hamid and Li (2016)</u> suggested that WWTP effluent is a major source of PFAAs to surface waters.

Evaluation of full-scale WWTPs has indicated that conventional primary (sedimentation and clarification) and secondary (aerobic biodegradation of organic matter) treatment processes can change PFAS concentrations and subgroups. For example, studies have shown increased concentrations of PFAAs in effluent, presumably from degradation of precursor PFAS (Schultz et al. 2006), and the possible creation of PFAAs from the oxidation of polyfluorinated precursors during the treatment process (Oliaei, D. Kriens, and Kessler 2006; Frömel et al. 2016; Houtz et al. 2018).

PFAS may be concentrated in wastewater solids (for example, sewage sludge) generated throughout the wastewater treatment process (Schultz et al. 2006). PFAS may also be present in septage (solids removed from septic systems). Depending on waste management and disposal practices, land application or landfill disposal of wastewater solids, biosolids, or septage could potentially contaminate the environment.

Hu et al. (2016) suggested that the presence of WWTPs in an area could be predictive of the presence of PFOS and PFOA in drinking water. PFOS and PFOA are two of the most frequently detected PFAS in wastewater (Hamid and Li 2016). Using WWTP effluent-impacted surface water as a source of drinking water can, in turn, recycle the PFAS back to the WWTP, recirculating PFAS in the water cycle (Hamid and Li 2016).

At some WWTPs, studies have shown concentrations of PFAS in ambient air at WWTPs to be 1.5–15 times greater than background reference sites (Hamid and Li 2016). Hamid and Li (2016) noted that these elevated air concentrations of total PFAS include *poly*fluoroalkyls and that this has important implications considering the potential for their long-range transport and subsequent degradation to recalcitrant PFAAs. PFAS distribution (primarily PFAAs and FTOH, with higher concentrations of FTOH) changes based on the specific PFAS sources in the effluent and the type of treatment methods employed at the WWTP. Lagoon systems contain a greater fraction of PFAAs.

2.6.4.2 Biosolids Production and Application

PFAS (measured as PFCAs and PFSAs) have been found in domestic sewage sludge (<u>Higgins et al. 2005</u>; <u>Yoo et al. 2009</u>), and PFAS occurrence in biosolids is reported to be prevalent and nationwide (<u>Venkatesan and Halsden 2013</u>). Given that more than half of the sewage sludge produced in the United States is applied to agricultural land as biosolids (<u>USEPA 2017</u>), there

is the potential for release of PFAS to the environment associated with biosolids production and application. PFAS are not known to be added to biosolids during processing or application.

PFAS may be introduced to the environment through the land application of biosolids as a beneficial soil amendment, potentially allowing PFAS to enter surface water through runoff or to infiltrate to groundwater (<u>Lindstrom et al. 2011</u>). The potential effects on groundwater or surface water depend on the amount and composition of PFAS present in biosolids, soil properties, infiltration rate, and land application practices. PFAS concentrations can be elevated in surface and groundwater in the vicinity of agricultural fields that received PFAS-contaminated biosolids over an extended period of time (<u>Washington et al. 2010</u>). The Washington et al. study was completed in an area that received industrial wastewater discharges from several PFAS-related industrial dischargers. Other studies indicate that the potential PFAS releases from municipal biosolids (for example, those generated from facilities that do not receive PFAS-related industrial discharges), may still impact water quality, but at an apparent lower relative impact than at the industrial-influenced biosolids application sites (<u>Gottschall et al. 2017</u>).

The most abundant PFAS found in biosolids (PFOS and PFOA) are the same as those found in WWTP effluent, although biosolids may also contain other long-chain PFAS (Hamid and Li 2016; Washington et al. 2010). Although multiple studies have reported statistically significant data showing transformation of polyfluorinated substances to PFAAs in land-applied biosolids (Yoo et al. 2010; Sepulvado et al. 2011; Washington et al. 2010), other evidence indicates that some polyfluorinated substances remain in biosolids-amended soils for many years to decades (Yoo et al. 2010; Rich et al. 2015; Washington et al. 2018).

Application of municipal biosolids as a soil amendment can result in a transfer of PFAS to soil (Sepulvado et al. 2011). These PFAS can then be available for uptake by some plants and soil organisms (Yoo et al. 2011). There are indications that PFAAs can enter the food chain through the use of biosolids-amended soil (Lindstrom et al. 2011; Blaine et al. 2013; Blaine et al. 2014; Navarro et al. 2017). It is noted, however, that PFAAs present at one municipal biosolids application site were not found in grain grown in the application plot (Gottschall et al. 2017). Hamid and Li (2016) suggested that short-chain (< C7) PFAAs in biosolids subsequently used in land applications can lead to contamination of food (Section 5.6).

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