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Scrubbers are an increasing threat to marine ecosystems globally, as more ships install these systems to comply with the IMO 2020 sulfur fuel standard. The risk is so significant that the science advisory body to the IMO, the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP), stated in its report to the IMO that these systems may actually violate the UNCLOS prohibition on against converting one type of pollution to another. The continuous discharges from open loop scrubbers and bleedoff from closed loop systems contribute to toxic burdens in already strained ecosystems, with thermal, heavy metal, and endocrine disrupting contaminants. A study published last month based on three decades of UK data found that scrubbers may be a significant 'hidden' source of toxic heavy metal sediment contamination. At this moment, when the Southern Resident killer whales and the Chinook salmon on which they depend are fight for survival, we cannot afford more toxic inputs into their habitats. The IMO regulations are weak, developed long before there was any significant uptake of scrubber technology and are not based on scientific data (which is why the IMO is trying to revisit this issue now). In order to protect sensitive marine and coastal ecosystems, we must ban the discharges of all scrubber wastes in Puget Sound. This is a unique opportunity for the cruise sector to demonstrate their purported commitment to healthy seas by voluntarily agreeing to stop dumping of scrubber washwater and bleedoff in the Sound - without the reservation that there should be some reconciliation with scientifically unsound and pitifully weak IMO standards. If they refuse to do this voluntarily, then the WA Dept of Ecology must take action to ban the use of scrubbers as a compliance mechanism for the ECA, as the state of California has already done, in order to not only protect the health of Puget Sound but also the health of the communities along its shores.

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**EVALUATION AND HARMONIZATION OF RULES AND GUIDANCE ON THE  
DISCHARGE OF LIQUID EFFLUENTS FROM EGCS INTO WATERS,  
INCLUDING CONDITIONS AND AREAS**

**Report of the GESAMP Task Team on exhaust gas cleaning systems**

**Note by the Secretariat**

**SUMMARY**

*Executive summary:* This document provides the report of the GESAMP Task Team on exhaust gas cleaning systems

*Strategic direction,  
if applicable:* 1

*Output:* 1.23

*Action to be taken:* Paragraph 2

*Related documents:* MEPC 74/18 and PPR 7/12/5

**Introduction**

1 The final report of the GESAMP Task Team on exhaust gas cleaning systems, as approved by GESAMP, is set out in the annex.

**Action requested of the Sub-Committee**

2 The Sub-Committee is invited to note the annexed report.

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ANNEX  
REPORT OF THE GESAMP TASK TEAM ON EXHAUST GAS CLEANING SYSTEMS



**GESAMP**  
Joint Group of Experts on the  
Scientific Aspects of Marine  
Environmental Protection

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# EXHAUST GAS CLEANING SYSTEMS

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## A Roadmap to Risk Assessment

Advice to the Marine Environment Protection Committee and its

Sub-Committee on Pollution Prevention and Response

Report of the GESAMP Task Team on EGCS

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## Table of Contents

List of Acronyms and Abbreviations	4
List of Figures	8
List of Tables	9
ACKNOWLEDGEMENT	10
EXECUTIVE SUMMARY	11
Short Summary	13
1.    INTRODUCTION TO THE REPORT	15
2.    PROBLEM DEFINITION	17
3.    TERMS OF REFERENCE	17
4.    DOCUMENTS AVAILABLE TO THE TASK TEAM ON EGCS	18
4.1    IMO Submissions	19
4.2    Relevant Documents	21
5.    INVENTORY OF EXHAUST GAS CLEANING SYSTEMS	26
5.1.    Introduction to EGCS	26
5.2.    Wet EGC Systems	26
5.2.1.    Open Loop Systems (OL)	26
5.2.2.    Closed Loop Systems (CL)	29
5.2.3.    Hybrid Systems	30
5.3    Environmental aspects of EGCS washwater	30
6.    INVENTORY OF CHEMICALS	31
6.1.    Introduction to Relevant Chemicals	31
6.2    General Concerns regarding Chemicals in EGCS washwater	31
6.3.    Chemicals in EGCS Washwater: Background Considerations	32
6.4.    Main Contaminants	32
6.5.    General Concerns of Environmental Impacts of EGCS Washwater Discharges	33
6.6.    Further Evaluation of Washwater Chemicals	34
6.7.    Background Documents	35
6.8    Inorganic Chemicals	36
6.8.1.    SO <sub>x</sub>	36
6.8.2.    NO <sub>x</sub>	39
6.8.3    Trace Metals	42
6.9.    Particulate Matter	47
6.10   Organic Contaminants	48
6.11   Conclusions and Recommendations on Chemicals	58
7.    POSSIBILITIES FOR RISK ASSESSMENT, INCLUDING ERA AND HHRA	59
7.1.    Exposure Assessment	59
7.1.1.    Introduction	59
7.1.2.    Initial Mixing Models	59
7.1.3    Near Field Models	60
7.1.4    Far Field Models	61
7.1.5    Available Models	62
7.1.5.1.    MAMPEC-BW	62

	7.1.5.2. STEAM3	64
	7.1.5.3. DREAM	65
	7.1.6 Conclusions and Recommendations on Exposure Assessment	65
7.2.	Hazard Assessment, including PBT and CMR	66
	7.2.1 Human Health Hazard Assessment for CMR Properties	66
	7.2.1.1 Extrapolation and Derivation of Reference Values	67
	7.2.2 Environmental Hazard Assessment for PBT Properties	74
	7.2.2.1 Availability of Data	74
	7.2.3. Environmental hazard assessment for aquatic toxicity	76
	7.2.3.1 Availability of Data	76
	7.2.3.2 Extrapolation and Derivation of Reference Values	80
	7.2.4 Whole Effluent Toxicity (WET) Testing	83
	7.2.4.1 Conclusions and Recommendations for WET	86
	7.2.5 Conclusions and Recommendations on Hazard Assessment	87
7.3.	Potential for Risk Assessment	88
	7.3.1 Human Health Risk Assessment Approaches	88
	7.3.2 Exposure via Different Sources and Background Concentrations	89
	7.3.3 Possibilities for Environmental Risk Assessment	89
7.4	Recommendation on Risk Assessment	90
8.	UNCERTAINTY	91
9.	FURTHER RESEARCH	91
	9.1 Conclusions and Recommendations on Future Research	92
10.	ROLE OF GESAMP	93
11.	CONCLUSIONS AND RECOMMENDATIONS	94
	11.1 General	94
	11.2 With respect to Chapter 6	96
	11.3 With respect to Chapter 7	97
	11.3.1 Exposure Assessment	97
	11.3.2 Hazard Assessment	97
	11.3.3 Whole Effluent Toxicity	98
	11.3.4 Risk Assessment	99
	11.4 Recommendations for future Work	99
12.	REFERENCES	102
13.	APPENDIX	118
	Table A1	119
	Table A2	121

## List of Acronyms and Abbreviations

Acronyms/ Abbreviation	Explanation
AA-EQS	Annual Averaged Environmental Quality Standard
ADI	Acceptable Daily Intake
AI	Adequate Intake
AIS	Automatic Identification System
AR	Average Requirement
BAT	Best Available Techniques
BATNEEC	Best Available Technology Not Entailing Excessive Costs
BMD	Benchmark Dose
BMDL	Lower confidence limit of the benchmark dose
BSH	Bundesamt für Seeschifffahrt und Hydrographie (Bundesamt für Seeschifffahrt und Hydrographie in Germany)
BWMS	Ballast Water Management Systems
BWWG	Ballast Water Working Group
BOTU	Bleed-Off Treatment Unit
BTEX	Benzene, Toluene, Ethylbenzene, Xylene
CAPEX	Capital expenditure
CAS	Chemical Abstracts Service
CESA	Community of European Shipyards' Associations
CFD	Computational Fluid Dynamics modelling
CL	Closed Loop
CLIA	Cruise Lines International Association
CLP	Classification, Labelling and Packaging of Chemicals
CRM	Certified Reference Materials
CMR	Carcinogenicity, Mutagenicity and Reproductive toxicity
COD	Chemical Oxygen Demand
CONTAM	Panel on Contaminants in the Food Chain
CRM	Certified Reference Material
CSS	Current Status Scenario
DBP	Disinfection By-Products
DELWAQ	Delft Water Quality model
DMEL	Derived No Effect Level
DNEL	Derived Minimal Effect Level
DOC	Dissolved Organic Carbon
DREAM	Dose-related Risk and Effect Assessment Model
DU	Decanting Unit
DW	Drinking Water
ECA	Emission Controlled Areas
ECHA	European Chemicals Agency
EEA	European Environmental Agency
EF	Emission Factor
EFSA	European Food Safety Authority
EC50	Effect Concentration causing effects for 50% of the population
EG	Exhaust Gas
EGCS	Exhaust Gas Cleaning Systems
EGCSA	Exhaust Gas Cleaning Systems Association

EGSE	Exhaust Gas Scrubber Effluent
EGR	Exhaust Gas Recirculation
ELV	Emission Limit Values
EQS	Environmental Quality Standards
ERA	Environmental Risk Assessment
EU	European Union
FNB	Food and Nutrition Board
FNU	Formazine Nephelometric Unit
FW	Flow rate of washwater
GC-MS(-MS)	Gas Chromatography Mass Spectrometry (could be double MS)
GC-TQ-MS	Gas Chromatography Triple Quad Mass Spectrometry
GESAMP	IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UN Environment/UNDP/ISA Joint Group of Experts on the Scientific Aspects of Marine environmental Protection
GHG	Green House Gas
GHS	Globally Harmonized System of Classification and Labelling of Chemicals
HC	Hazardous Concentration
HFO	Heavy Fuel Oil
HHRA	Human Health Risk Assessment
HMW	High molecular weight
IARC	International Agency for Research on Cancer
IED	Industrial Emissions Directive
IKSR-CIPR-ICBR	Internationale Kommission zum Schutz des Rheins (same in French and Dutch)
IMO	International Maritime Organization
INF	Information document used in meetings at IMO
IPPC	Integrated Pollution Prevention and Control
IQ	Intelligence Quotient
IRIS	Integrated Risk Information System
ISO	International Standardization Organization
ITMS	Ion Trap Mass Spectrometry
JECFA	Joint FAO/WHO Expert Committee on Food Additives
kWh	Kilowatt hour
LMW	Low Molecular Weight
LNG	Liquefied / Liquid Natural Gas
LOEC	Lowest Effect Concentration
LOQ	Level of Quantification
LSFO	Low Sulfur Fuel Oil
MAC-EQS	Maximum Allowable Concentration Environmental Quality Standard
MAMPEC(-BW)	Marine Anti-foulant Model for the Prediction of Environmental Concentrations (as amended for Ballast Water)
MARPOL	International Convention for the Prevention of Pollution from Ships
MCR	maximum Continuous Rating
MDO	Marine Diesel Oil
MEPC	Marine Environment Protection Committee
MGO	Marine Gas Oil
MIS	Maximum Installation Scenario
ML	Maximum Limits
MRL	Maximum Residue Limit
MPC	Maximum Permissible Concentrations
MSFD	Marine Strategy Framework Directive
MV	Motor Vessel
MWh	Megawatt hour

NA	Not Available or Not Applicable
NGO	Non-Governmental Organization
NOEC	No-effect concentration
NOx	Nitric Oxides, generally a combination of NO and NO <sub>2</sub>
NRC	National Research Council
NRWQC	National Recommended Water Quality Criteria
NTU	Nephelometric turbidity unit
NMVOC	Non-Methane Volatile Organic Compounds
OECD	Organization for Economic Cooperation and Development
OL	Open Loop
OPEX	Operational expenditure
OSPAR	Oslo and Paris commissions
PAH	Polycyclic Aromatic Hydrocarbons
PBT	Persistence, Bioconcentration and Toxicity
PCB	polychlorinated biphenyls
PCDD	polychlorinated dibenzodioxins
PCDF	polychlorinated dibenzofurans
PEC	Predicted Environmental Concentration
PHS	priority hazardous substances
PM	Particulate Matter
PMTDI	Provisional Maximum Tolerable Daily Intake
PNEC	Predicted No-Effect Concentration
PNED	Predicted No-Effect Dilution
POC	Particulate Organic Carbon
PON	Particulate Organic Nitrogen
POP	Persistent Organic Pollutant or Particulate Organic Phosphorous
PPR	Sub-Committee on Pollution Prevention and Response
PRI	Population Reference Intake
PTWI	Provisional Tolerable Weekly Intake
REACH	Registration, Evaluation Authorization of Chemicals
RIVM	Rijksinstituut voor Volksgezondheid en Milieu (National Institute for Public Health and the Environment in The Netherlands)
RNA	Ribonucleic acid
RoRo	Roll-on-Roll-off
SCHER	Former Scientific Committee on Health and Environmental Risks of the EU
SCR	Selective Catalytic Reduction
SECA	SOx Emission Controlled Areas
SFOC	Specific Fuel Oil Consumption
SINTEF	Selskapet for INdustriell og TEknisk Forskning ved norges tekniske hoegskole (The Foundation for Scientific and Industrial Research at the Norwegian Institute of Technology)
SOLAS	International Convention for the Safety Of Life At Sea
SOx	combination of SO <sub>2</sub> and SO <sub>3</sub>
SPM	Suspended Particulate Matter
STEAM3	Ship Traffic Emission Assessment Model, version 3
TDI	Tolerable Daily Intake
TEF	Toxic Equivalence Factor
TG	Test Guideline
ToF-MS	Time of Flight Mass Spectrometry
ToR	Terms of Reference
TRS	Technical Report Series (WHO)
TWI	Tolerable Weekly Intake

UBA	Umwelt Bundesamt, German Environmental Department
UNECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Programme
USA	United States of America
USDT	United States Department of Transportation
USEPA	United States Environmental Protection Agency
UV	Ultra Violet light
vPvB	very Persistent and very Bioaccumulating
WET	Whole Effluent Toxicity
WFD	Water Framework Directive
WHO	World Health Organization

## List of Figures

#	Description
Figure 1	MARPOL Annex VI fuel sulfur limits
Figure 2	Open loop exhaust gas cleaning system
Figure 3	Closed loop exhaust gas cleaning system
Figure 4	Ship smoke stack and EGCS washwater emissions
Figure 5	Calculated values of $EF_{m\ SO_2}$ ( $g\ SO_2\ kg_{HFO}^{-1}$ ) and $EF_{p\ SO_2}$ ( $g\ SO_2\ (kWh)^{-1}$ ) for $SO_2$ as a function of S concentration in HFO
Figure 6	Concentration of sulfate ion in washwater ( $g\ L^{-1}$ ) as a function of S concentration in HFO and for $SFOC = 0.16\ kg\ (kWh)^{-1}$
Figure 7	Volume of buffer-depleted normal seawater ( $m^3\ kg_{HFO}^{-1}$ and $m^3\ (kWh)^{-1}$ ) as a function of S concentration in HFO
Figure 8	Relative contribution of $NO_x$ and $SO_x$ emissions on water acidification
Figure 9	Minimum and maximum concentrations were used to calculate a range of EF values for each metal
Figure 10	Relative 16 EPA PAH based on all measurement campaigns
Figure 11	Temporal trend for global PAH shipping emissions in metric tons since 1960
Figure 12	Schematics of (left) the near-field and (right) the far-field model in top-view
Figure 13	Layout of the commercial harbour and the shipping lane in MAMPEC
Figure 14	Steps in the risk assessment approach (modified from GESAMP (2019))

## List of Tables

#	Description
Table 1	MARPOL Annex VI fuel sulfur limits
Table 2	Overview of submissions to IMO bodies, MEPC and PPR, on EGCS
Table 3	Overview of otherwise relevant documentation considered by the Task Team
Table 4	H <sup>+</sup> and nitrate ion emission factors for EGCS
Table 5	Total Trace Metal Concentrations ( $\mu\text{g L}^{-1}$ ) reported in different area
Table 6	Average concentrations and ratios of heavy metals in the dissolved versus the particulate phase in EGCS washwater on the basis of measured data from PPR 6/INF.20
Table 7	Mass-standardized emission factor ( $\text{EF}_m$ ) values for V, Fe, Ni, Pb, Zn, Cd, Hg, As, Cr and Cu and power-standardized emission factors ( $\text{EF}_p$ ) calculated for a SFOC of $0.19 \text{ kg HFO (kWh)}^{-1}$
Table 8	Dissolved PAHs environmental concentrations ( $\text{ng L}^{-1}$ )
Table 9	Ranges of summed concentrations of parent $\Sigma$ PAHEPA-16 and phenanthrene in EGCS washwater (in open loop operation); concentrations adjusted to $45 \text{ m}^3 (\text{MWh})^{-1}$ washwater flow rate only for EGCSA (2018)
Table 10	Concentrations of alkylated dibenzothiophenes, phenanthrenes and naphthalenes in closed loop EGCS in the BOTU feed water in comparison to total 16 EPA PAH ( $\mu\text{g L}^{-1}$ )
Table 11	Total PAH discharged per MW and year, based on the evaluated measurement campaigns
Table 12	CMR properties for trace metals
Table 13	CMR properties for 16 EPA PAHs
Table 14	Guideline values for selected CMR chemicals
Table 15	Guideline values for selected chemicals with threshold effects
Table 16	Legally established PBT properties of 16 EPA PAHs in the US and EU
Table 17	Overview of the derived risk limits for 16 EPA PAHs individually
Table 18	Overview of the derived PNECs or similar reference values for individual dissolved trace metals contained in EGCS washwater
Table 19	Summary results of WET testing using the actual discharge water from EGCS
Appendix	
Table A1	Log $K_{ow}$ , freshwater solubility, and estimated acute and chronic toxicity of PAH frequently found in crude and refined petroleum
Table A2	Trace metal and PAH concentrations ( $\mu\text{g L}^{-1}$ ) in open loop EGCS washwaters measured in the provided measurement campaigns



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## EXECUTIVE SUMMARY

The Task Team on Exhaust Gas Cleaning Systems (EGCS) was convened on a request of the Marine Environmental Protection Committee (MEPC) to the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) to give an opinion on the potential environmental and public health effects of EGCS effluents. According to its Terms of Reference (ToR), the Task Team assessed the available evidence relating to the environmental impact of EGCS taking into account the information provided in several background documents and other information made available or accessed by the Task Team. It concerned in the first place, this information concerned the submissions on EGCS to the Sub-committee on Pollution Prevention and Response (PPR) and MEPC, and in the second place, other relevant scientific publications and other and publicly available reports that were accessible to the members of the Task Team based on the personal knowledge of the members. The Task Team attributed specific weight to the PPR and MEPC papers as this information could be considered as belonging to the knowledge of the stakeholders in International Maritime Organization (IMO) fora and were therefore specifically relevant in the scope of the ToR provided to the Task Team. The other data was considered as valuable additional information that may be confirmative or non-confirmative to the information presented in the PPR and MEPC papers. The Task Team applied a similar level of scientific scrutiny to all information available and where deemed appropriate this included a critical review focusing on applicability and uncertainties.

An important part of the assessment of the available information was the establishment of concentrations of chemicals involved in the application of EGCS. This related not only to the assessment of actually performed measurements of EGCS washwater content, but also to an analysis of specific processes that were involved in a more generalized way. This would be necessary if a risk assessment had been the aim. EGCS normally applied on ships were of the open or closed loop type and of a hybrid type of these two systems. Open Loop EGCS applied generally non-treated seawater as source for the washwater and closed loop EGCS were more prone to use freshwater made up on board. The hybrid type EGCS was able to operate in both modes. A major difference of the two types was the discharge of the washwater. **In open systems, the washwater was generally directly discharged to the receiving waters with basically only a minimum of post-treatment.** Closed loop EGCS used a separate tank to store and reuse the washwater and therefore it had the possibility of post-treatment of the washwater by chemicals, to recover the original pH of the washwater and to apply a sedimentation step to collect and separate the sediment in the washwater. **The final volume of washwater discharge to the environment was, therefore, substantially lower although the concentrations of contaminants in this discharge volume may be higher than for open loop EGCS.**

As there was (per definition) combustion involved in the propulsion of ships, the normal combustion chemicals were present, like different kinds and amounts of polycyclic aromatic hydrocarbons, ash and soot. Also many trace metals were normally present in these exhaust gases in relation to the sources of the fuel. Fuels may have a wide variety in natural content of trace metals. Finally, a third group of products being present in the washwater of EGCS was the components in the lubricants of the ships. Also here, a wide variety of concentrations and amounts was present. **However, the Task Team identified several flaws where the sampling, analysis and identification of the contaminants were concerned.** Recognizing the possible variations in the composition of washwater the Task Team

was able to achieve a sufficiently clear view on the potential concentrations of chemicals in the wastewater. These also implied organic contaminants, like alkylated polycyclic aromatic hydrocarbons, for which no discharge limits exist, but which were normally present in the exhaust gas and partially end up in the EGCS wastewater.

The Task Team also assessed the results of available simulations for predicting the environmental concentrations of target substances. Several documents informed about the application of models to describe the fate and behavior of substances in aquatic systems taking also into account the specific circumstances of the aquatic environment for shipping. In this respect, power output of the engines may have an important influence on the functioning of EGCS as well as the alkalinity of the source water and the receiving waters. The Group assessed the modelling activities in the different documents available. Generally, the model MAMPEC was used by the authors, probably because it was applied already in several jurisdictions in a regulatory way, like the assessment of antifouling paints in the European Union (EU) and the United States of America (USA) as it was recommended by the Organization for Economic Cooperation and Development (OECD). It was also applied in the risk assessment of disinfection by-products of ballast water management systems, where a specific emission scenario and geographic conditions of the receiving port was developed, MAMPEC-BW. However, the Task Team on EGCS became aware of two other models that with some effort could be made applicable for the situation of the evaluation of EGCS: STEAM3 and DREAM. Both models were developed for completely different applications, air emissions from ships and drilling wastes from oil platforms. Some features of both models are appealing, like the automatic identification system for ships (AIS) for STEAM3 and the multicomponent analysis for DREAM. The Task Team concluded after its assessment that the first model to be investigated further would be the MAMPEC(-BW) model as its development status would be more appropriate for the evaluation of EGCS. The use of a model would present a useful building block in the risk assessment process.

The Task Team identified many gaps in the IMO submissions and the scientific literature where toxicological and ecotoxicological data was concerned. For most trace metals, sufficient information was available to determine predicted no effect concentrations in environmental aquatic systems for an acute and chronic environmental risk assessment. However, for the sediment phase of the aquatic environment insufficient data were available and the use of equilibrium partitioning or quantitative structure activity relations maybe used. The Task Team did not have sufficient time to analyze this further.

With respect to polycyclic aromatic hydrocarbons (PAH), the Task Team on EGCS identified the same kind of missing (eco-)toxicity data for the sediment. In addition, there was a lack of data on the acute and chronic ecotoxicity for aquatic organisms most generally used, like the relevant OECD Test Guidelines for three different aquatic organisms, algae, crustaceans and fish. For the human health risk assessment, the Task Team recommended that the most suitable indicators of PAH in food are PAH4 and PAH8. Also in this area, the Task Team did not have sufficient time to search for more data that could fill the (eco-)toxicity gaps identified. The Task Team was aware of the hesitations in the scientific arena, but also the public opinion in general, on the further testing of substances using test animals. A perspective may be available in the possibilities of *in vitro* or *in silico* developments. For *e.g.* the ecotoxicological and toxicological data this is already partly available.

To the Task Team on EGCS, it was quite clear from the beginning of its work that carrying out a preliminary risk assessment was not possible considering the available information and despite the

examples provided on risk assessments already carried out. In this regard, the Task Team noted that secondary poisoning (via consumption of seafood) had been mentioned as a likely exposure route for humans. The report, however, showed several shortcomings and insufficient scientific evidence to support the way forward in performing risk assessments according to the methods presented. The Task Team was also clear in its recommendation to support a risk assessment to be carried out by GESAMP or some other entity. GESAMP would be available in this further process if the recommendation would be endorsed by the Committee. **Contributory to a risk assessment, the Task Team considered it would be advisable in parallel to generate more information on the direct ecotoxicological effects of EGCS washwaters through well-established whole effluent toxicity (WET) testing.** Such testing should be carried out in consultation with GESAMP to avoid inconsistent testing. An internationally accepted guideline for WET tests using seawater was not available but may be developed in this setting. Also, for this recommendation the Task Team was aware of the time constraints that may be accompanied with this development of a WET test guideline.

The Task Team recognized also that current available evidence on chemicals in EGCS washwater effluents and its importance for the environment, should call for an increased and broad focus on this topic from both the science community and from policymakers. This would provide a framework for future work allowing a more accurate estimation and prediction of current and future EGCS chemicals releases, and their spatial and temporal representation including further modeling efforts combining ship traffic data with ship emission factors and in-situ measurements of contaminants. The revision and improvement of existing guideline limits and development of a new guideline for certain chemicals and should take into account future updated chemical concentration levels, **by using harmonized procedures in terms of washwaters sampling and analysis to ensure better comparability in different data sets.**

Finally, this document presented the report of the findings of the Task Team on EGCS to PPR 7 and proposed actions as appropriate.

### **Short Summary**

With respect to the ToR the following conclusions and recommendations were established:

- The Task Team on EGCS concluded that many useful documents were already available in the IMO submissions and the in the scientific literature but the Task Team on EGCS also identified data gaps in mainly basic (eco-)toxicological information of EGCS effluent contaminants, especially PAHs. The Task Team on EGCS recommended filling these data gaps that were essential for a future risk assessment.
- The Task Team also identified several useful tools for the simulation of predicted environmental concentrations of target substances. Several available documents presented their tools and their approach to an environmental risk assessment. In this respect the Task Team identified a missing harmonized approach that could be considered suitable for more areas around the globe than currently available. Therefore, the Task Team recommended the development of such a harmonized approach. In the view of the Task Team on EGCS this specifically means the development of a widely accepted mathematical calculation model, based on MAMPEC-BW, as a start. **In addition, a database containing all physico-chemical characteristics, (eco-)toxicological effect data and fate and behavior data of all relevant**

contaminants in exhaust gas effluents, using the GESAMP-BWWG database as a starting point.

- The Task Team on EGCS, in addition to the recommendation in the second bullet, recommended to gather and probably develop further knowledge on data with respect to the socio-economic data to be able to perform an environmental impact assessment in a global sense as the risk assessment approach recommended by the Task Team did not include risk management options, like cost-benefit analysis, risk mitigation measures and global policies. For the Task Team on EGCS, it was clear that an approach taking into account potential global effects with respect to acidification and eutrophication would put the risk assessment of exhaust gas effluent contaminants into the correct context.
- The Task Team was not able to perform a preliminary risk assessment due to the missing data as defined in the earlier bullet points.
- The Task Team recommended a clear way forward to the development of a risk assessment related to exhaust gas effluent contaminants using MAMPEC-BW as an environmental exposure assessment tool, developing a database with data on physico-chemical characteristics, (eco-) toxicological effects and fate and behavior of all relevant contaminants using the model of the database of GESAMP-BWWG.
- With respect to the entity to carry out the proposed future approach, the Task Team on EGCS recommended to position this work again under the supervision of GESAMP and was available to take on-board any successive work, whether or not in the same composition as additional expertise in the area of database development and adjusting the proposed model to comply with the identified needs were considered necessary.
- The Task Team on EGCS identified data gaps in the (eco-)toxicological effects area of the relevant exhaust gas effluents contaminants. The Task Team on EGCS was also aware of international resistance to effect testing using test animals, therefore, other approaches may have to be developed, such as in vitro and in silico methods, quantitative structure activity relations (QSAR) or read-across methods. In addition to these methods, the Task Team recommended the development of whole effluent toxicity (WET) testing methods that could be internationally accepted. The Task Team noted that these WET tests were applied successfully in the area of ballast water management systems, although these methods were not fully harmonized yet.
- The Task Team is of the opinion that the current report fulfils its ToR.

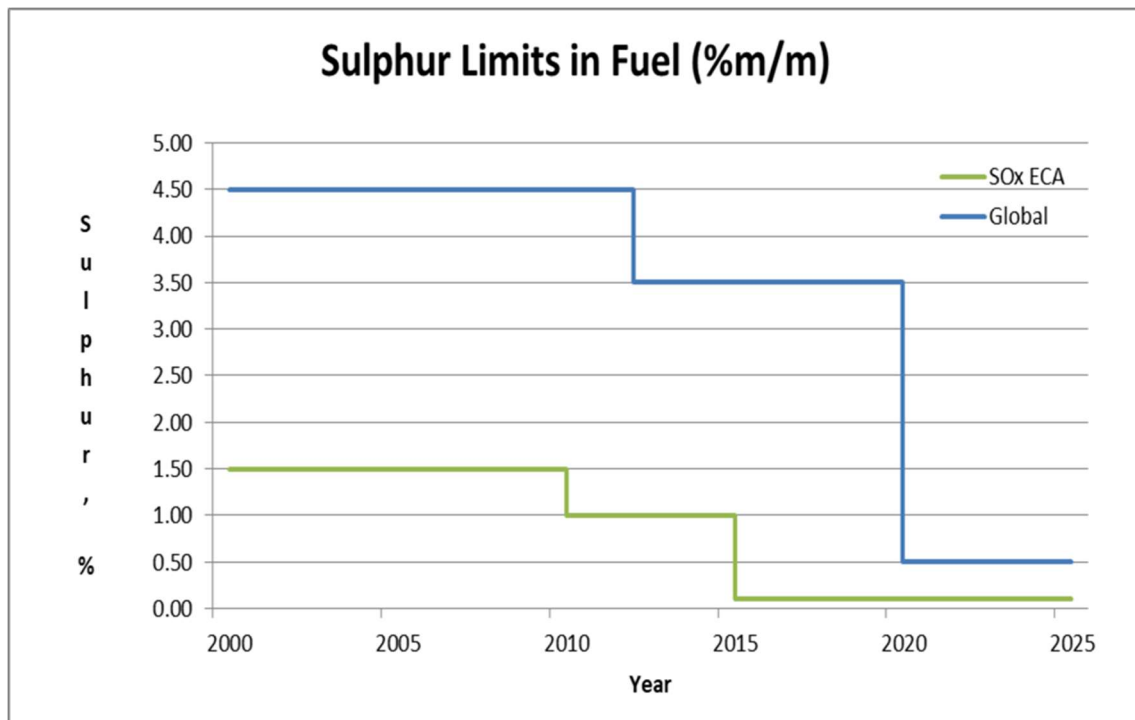
## 1. INTRODUCTION TO THE REPORT

At the International Maritime Organization (IMO), in its Marine Environment Protection Committee (MEPC) and its Sub-Committee, Pollution Prevention and Response (PPR), the 2015 Guidelines for Exhaust Gas Cleaning Systems (EGCS) (resolution MEPC 259(68) (MEPC, 2015)) are under review in relation to Regulation 14.4 of the International Convention for the Prevention of Pollution from Ships (MARPOL) Annex VI that sets out a sulfur limit for fuel oil in SOx emission controlled areas (SECA). The sulfur limit has been defined at 0.10% in SECAs and 3.50% m m<sup>-1</sup> (mass by mass) outside SECAs. A new Regulation coming into effect on 1 January 2020 limits the sulfur content of fuels outside SECAs at 0.50% m m<sup>-1</sup>. Table 1 and Figure 1 show the development of the sulfur limits, regulated by MARPOL Annex VI since 2005.

**Table 1. MARPOL Annex VI fuel sulfur limits**

Date	Sulfur Limit in Fuel (% m m <sup>-1</sup> )	
	SOx ECA	Global
2005 May 19 <sup>th</sup>	1.5	4.5
2010 June 1 <sup>st</sup>	1.0	
2012 January 1 <sup>st</sup>		3.5
2015 January 1 <sup>st</sup>	0.1	
2020 January 1 <sup>st</sup>		0.5

The established ECAs are Baltic Sea area, North Sea area, North American area, and United States Caribbean Sea area.



**Figure 1. MARPOL Annex VI fuel sulfur limits**

As an equivalent method to the use of a compliant fuel, whose sulfur content was less than 0.50%, technologies that are at least as effective in term of reducing SOx emissions are allowed. Since 22 July 2005 when the first EGCS Guideline was adopted, the number of ships equipped with EGCS has grown from 300 to a projected 3,800 on 1 January 2020 (MEPC, 2016). This increased use of EGCSs has triggered a discussion on the costs and benefits of this equivalent technology and specifically on the direct environmental risks and impacts of increasing operational discharges of EGCS washwater by ships equipped with such systems. The PPR Sub-Committee agreed that the currently available information at MEPC and PPR, as contained in the documents PPR 6/INF.20 (PPR, 2018), PPR 6/20 (PPR, 2019), MEPC 74/INF.10 (MEPC, 2019b), MEPC 74/INF.24 (MEPC, 2019c) and MEPC 74/INF.27 (MEPC, 2019d) sufficiently warranted the undertaking of further scientific research and to subsequently submit the results to future sessions to facilitate the work on the revision of the 2015 EGCS Guidelines.

Based on the discussions during the meetings at IMO, it was suggested that an independent study would be helpful and the PPR Sub-Committee requested the Secretariat to explore the possibility of GESAMP carrying out a review of the relevant scientific literature and also overseeing a modelling study of the impacts of discharge washwater from exhaust gas cleaning systems and to inform the Sub-Committee at its next session. GESAMP, in its meeting from 9 to 12 September 2019, approved the terms of reference, that are outlined in Chapter 3 of this document, and including a timeline for the preparation of a report on this topic. Subsequently, GESAMP endorsed the establishment of a Task Team on EGCS.

The Task Team on EGCS consisted of 8 scientists with different expertise. The following persons were members of the Task Team on EGCS:

- Eric Adams (USA) with expertise in fluid dynamics, mathematical modelling;
- Brigitte Behrends (Germany) with expertise in marine and environmental chemistry;
- Annette Dock (Sweden) with expertise in human toxicology and human health risk assessment;
- Shinichi Hanayama (Japan) with expertise on ecotoxicology, ocean chemistry;
- Jan Linders (The Netherlands, Chair) with expertise in general risk assessment, mathematical modelling;
- Richard Luit (The Netherlands) with expertise in environmental risk assessment;
- Claude Rouleau (Canada) with expertise in chemistry;
- Jacek Tronczynski (France) with expertise in environmental marine biogeochemistry, oceanography.

The Task Team on EGCS was asked to report its findings, as an information document, to the seventh meeting of the Sub-Committee on Pollution Prevention and Response (PPR 7), that was scheduled at IMO in London from 17 to 21 February 2020. Factually, this meant that the Task Team had to finalize its report in the last week of November as two additional weeks were needed for the peer review of GESAMP.

When considering the scope of the work, the Task Team strongly pointed out that the time to produce a report was considered too short to address all requirements of the ToR in sufficient detail and sufficient scientific depth. The Task Team understood the time pressure of the work in relation

to the entry into effect of the 2020 standards. The Task Team was of the opinion that much more could have been achieved if more time had been available to reach a thorough scientific report. The approaches and methods used in the report by the Task Team were not the subject of in-depth scientific debate and therefore opinions expressed may not be necessarily fully shared. The Task Team would like to request some consideration of the readers in this respect. PPR and MEPC would now have the task to decide on a report that had several uncertainties that may limit the usefulness of the report. In fact, the Task Team recommended that the report would need further elaboration in future.

## **2. PROBLEM DEFINITION**

As stated above, the new Regulation 14.4 of MARPOL Annex VI (MEPC, 2019h) limited the exhaustible SO<sub>x</sub> gas to 0.50% m m<sup>-1</sup> in so-called outside SECAs by 1 January 2020, whereas it was already 0.10 % m m<sup>-1</sup> inside SECAs. Until 1 January 2020 still a limit of 3.50% was required outside SECAs. The use of EGCS, also called scrubbers, were applied as an alternative for the low sulfur limits to be achieved under the new Regulation, provided an equivalent level of SO<sub>x</sub> emission reduction was achieved (Winnes, *et al.*, 2018a). It was recognized that to achieve the new limits a considerable amount of SO<sub>x</sub> would have to be washed out of the exhaust gas to be able to meet the newly required limit. As the contaminants washed out of the exhaust gas, they would successively be introduced in the marine aquatic environment questions were raised at MEPC (MEPC, 2019b; MEPC, 2019c; and MEPC, 2019d) whether or not the chemicals in the washwater could present unintended consequences to the marine aquatic environment and could have environmental effects on aquatic ecosystems and toxicological effects on humans. The Task Team noted that, in this respect, it could even be argued that EGCS are potentially in conflict with Article 195 of UNCLOS “Duty not to transfer damage or hazards or transform one type of pollution into another”. Together with the nearby date of entering in effect of the Regulation 14.4, the need for quick scientific advice to MEPC through PPR was obvious.

## **3. TERMS OF REFERENCE**

The Terms of Reference (ToR) to the Task Team on EGCS agreed at the annual meeting of GESAMP held in New York from 9 to 12 September 2019 were as follows:

- To assess the available evidence relating to the environmental impact of exhaust gas cleaning system effluents taking into account the information provided in several background documents and other information made available or accessed by the Task Team;
- To assess the results of available simulations for predicting the environmental concentrations of target substances;
- To provide information on whether there was a need for more scientific research with respect to the environmental impacts of EGCS washwater discharges; and
- To report its findings, as an information document, to PPR 7.



It should be noted that the Task Team had not been requested to carry out a risk assessment following its review of the available evidence. With respect to the possibilities of a risk assessment the Task Team could decide to consider three options:

1. to carry out a preliminary risk assessment;
2. to recommend that a risk assessment should be carried out (by GESAMP or some other entity);
3. to request that further studies are undertaken to provide sufficient evidence to carry out a risk assessment.

Based on the ToR, the Task Team on EGCS set the scope of the work to be performed as follows:

- The Task Team is limiting its assessment to relevant effects of washwater releases. The potential effects of the still remaining emissions to air are not assessed in this report;
- The Task Team included environmental pollution (emissions), (eco-)toxicological, physico-chemical (pH, thermal), and biogeochemical (acidification and eutrophication) effects in its assessment;
- The Task Team recognized that the term “impact” in its ToR is ambiguous as it may be interpreted in a wider or a narrower meaning. For the purpose of this report taking into account the time limitations, the Task Team decided to restrict its work to the scope of the second bullet in the ToR, which is more in the context of the word “risk” than to make an economical and sociological assessment that is generally more related to the term “impact”.
- The Task Team also took into account different spatial (geographical) scales, open seas, harbours, special areas etc.;
- The Task Team assessed the possibilities of risk assessments for several environmental compartments normally considered in risk assessments (water, sediment dwelling organisms, secondary poisoning). The chemical and physical properties of the contaminants are considered;
- To identify the potential human health risks the Task Team examined the potential exposure pathways for humans via the environmental and coastal amenities.

#### **4. DOCUMENTS AVAILABLE TO THE TASK TEAM ON EGCS**

In recent years several Members States and Non-Governmental Organization (NGOs) have submitted documents to IMO on the topic of EGCS. Next to these documents, numerous scientific articles have been published in the literature. The Task Team was requested to assess the available information. The Task Team did not carry out an in depth literature search as most of the information was already available through several IMO submissions, especially [MEPC 74/INF.10 \(MEPC, 2019b\)](#), submitted by Panama with the particular purpose of such a search and the members of the Task Team brought in

their own knowledge and in addition, also because the time needed for an additional search activity was not available.

In this Chapter, the IMO submissions and relevant documents that reached the Task Team on EGCS during the course of its work were presented accompanied by a short evaluation.

#### 4.1 IMO Submissions

In Table 2 an overview was given of the submissions to IMO on EGCS. It concerned submissions to MEPC and PPR.

**Table 2. Overview of submissions to IMO bodies, MEPC and PPR, on EGCS**

IMO-id	Member State /NGO	Title
MEPC 70/INF.6	Secretariat	Assessment of fuel oil availability – final report
MEPC 73/INF.5	CESA	Study report on analyses of water samples from exhaust gas cleaning systems
MEPC 74/14/1	Austria, <i>et al.</i>	Proposal for evaluation and developing harmonized rules and guidance on the discharge of liquid effluents from exhaust gas cleaning systems
MEPC 74/14/7	CLIA	Comments on the proposal for evaluation and developing harmonized rules and guidance on the discharge of liquid effluents from exhaust gas cleaning systems
MEPC 74/14/8	CESA	Environmental impact assessment of EGCS discharges for generic risk-based, requirements adequately addressing all available technologies
MEPC 74/14/9	China	Comments on document MEPC 74/14/1
MEPC 74/INF.10	Panama	Scrubber Environmental Impact Literature Review
MEPC 74/INF.24	Japan	Report on the environmental impact assessment of discharge water from exhaust gas cleaning systems
MEPC 74/INF.27	CLIA	Compilation and assessment of 281 cruise ship EGCS washwater samples
MEPC.170(57)	Secretariat	2008 Guidelines For Exhaust Gas Cleaning Systems
MEPC.184(59)	Secretariat	2009 Guidelines For Exhaust Gas Cleaning Systems
MEPC.259(68)	Secretariat	2015 Guidelines for Exhaust Gas Cleaning Systems
MEPC.307(73)	Secretariat	2018 Guidelines for the Discharge of Exhaust Gas Recirculation (EGR) Bleed-Off Water
PPR 6/20	Secretariat	Report to the Marine Environment Protection Committee
PPR 6/INF.20	Germany	Results from a German project on washwater from exhaust gas cleaning systems

Below a short description and evaluation was provided. Several of these documents will be dealt with in more detail in the relevant sections of the report of the Task Team.

- MEPC 70/INF.6: the submission by the Secretariat contained the final report of CE Delft (The Netherlands) on the availability of fuel oil. It gave an overview of the different types of fuel used in shipping, the origin of the fuels, the capacity and the refinery actions needed to produce the fuel. The study also defined three scenarios for future fuel developments.
- MEPC 73/INF.5: the submission by CESA to PPR contained the report on analyses of water samples from EGCS used on ships. The submission reported on the analysis of 238 samples of EGCS washwater discharges from 20 different vessels in the Baltic and North Sea ECAs. Substances analyzed were the USEPA 16 PAHs, several metals, BTEX (benzene, toluene, ethylbenzene and xylene) and nitrates/nitrites.
- MEPC 74/14/1: this submission of the EU dealt with a proposal for the evaluation and developing harmonized rules and guidance on the discharge of liquid effluents from EGCS. The authors foresaw a deterioration of the marine environment by the extensive use of EGCS and proposed to further develop guidance to industry with regard to new technological developments.
- MEPC 74/14/7: the submission of CLIA commented on submission MEPC 74/14/1 and dealt with its willingness to cooperate with the proposal by sharing details on a science-based modelling analysis that was currently performed. It was the understanding of the Task Team that a draft report of this study was presented in the paper of Faber, *et al.* (2019).
- MEPC 74/14/8: the submission by CESA was also a comment on document MEPC 74/14/1 and proposed carrying out an environmental impact assessment of EGCS discharges in which all available technologies were included. The potential of these technologies to avoid regulatory action to forbid certain technologies should be investigated as well. An independent body was advised to be installed to perform the study.
- MEPC 74/14/9: also this submission by China was a comment on MEPC 74/14/1 and supported in general that paper. In addition, China proposed a four-stage approach to develop a future marine environmental risk assessment methodology. The Task Team considered that the proposal by China was fully suitable to achieve this goal, however, the Task Team was of the opinion that pursuing the proposal would require several years of work (order probably >10y) and would cost several millions of dollars. The scientific challenge of such a project would be great.
- MEPC 74/INF.10: this document was submitted by Panama and gives an overview of the scientific literature on the environmental impacts of EGCS. The review was carried out by prof. John Heywood and Dr. Emmanuel Kasseris, both of Massachusetts's Institute for Technology (MIT) in the US, and they presented two areas of concern related to EGCS: 1) potential effects of EGCS discharge on marine life and biogeochemical processes and 2) doubts on the true equivalence between ships using EGCS and ships using low sulfur fuel with regards to air emissions. As air emissions were outside the ToR of the Task Team the last issue was not discussed in the report of the Task Team. Several of the identified reports in this submission were also discussed in the current report.
- MEPC 74/INF.24: Japan submitted this document and it dealt with an approach to the environmental impact of the discharge of EGCS. The analysis included a short- and a long-term assessment of the potential effects of using EGCS. In the short-term assessment a dilution model was applied to the EGCS discharge based on propeller influenced mixing and indicating no effects and sufficient dilution to meet national Japanese water quality criteria standards. In addition, the study applied the MAMPEC-model in three Japanese harbours and

in the study WET testing was also reported. As a general conclusion the report identified no unacceptable effects to the marine environment on short- and long-term perspectives. These results were further discussed in the report of the Task Team on EGCS.

- MEPC 74/INF.27: this CLIA submission dealt with the analytical results of 281 EGCS washwater samples collected from cruise ships and included 54 test parameters, PAHs and heavy metals. The submission concluded that internationally accepted guidelines, *e.g.* EU surface water standards and the World Health Organization (WHO) drinking water guidelines were not breached by the EGCS discharges.
- MEPC.170(57), MEPC.184(59), MEPC.259(68) and MEPC.307(73): these documents contained the MEPC resolutions on the guidelines for exhaust gas cleaning systems as they had developed during the years, 2008, 2009, 2015 and 2018, respectively.
- PPR 6/20: the document contained the report of PPR 6 to MEPC 74 and in section 11 dealt with the discussion on EGCS in relation to several submissions of member states and NGOs to PPR 6 and presented the wish of several delegations, as suggested by GESAMP, that a generalized marine environmental risk assessment was to be developed at least for some model harbours. The work of the Task Team on EGCS should be viewed in this respect.
- PPR 6/INF.20: the submission by Germany highlighted some preliminary results of a German project on washwater from EGCS. Samples taken from open loop (OL) and closed loop (CL) EGCS were compared and assessed in relation to the potential environmental impact of EGCS. Upon a request of the Task Team for the final version of this study, the Task Team was informed that the final report was not yet ready.

#### 4.2 *Relevant Documents*

The Task Team on EGCS considered the scientific papers and some background documents as especially valuable for its task. An overview of these documents was given in Table 3. For further details the reader was referred to the list of references.

**Table 3. Overview of otherwise relevant documentation considered by the Task Team**

First author	Year of publication	Title
Reed, M.	2002	DREAM: a Dose-Related Exposure Assessment Model Technical Description of Physical-Chemical Fates Components
Hufnagl, M.	2005	Effects of Sea Water Scrubbing – Final Report, rev.
Rye, H.	2007	Development of a Numerical Model for Calculating Exposure to Toxic and Nontoxic Stressors in the Water Column and Sediment from Drilling Discharges
USDT	2011	Exhaust-Gas-Cleaning-Systems-Guide-Rev. A---Final-Report
USEPA	2011	Exhaust Gas Scrubber Washwater Effluent
Hansen, P.	2012	Exhaust Gas scrubber Installed Onboard MV Ficaria Seaways
Kjølholt, J.	2012	Assessment of possible impacts of EGCS water discharges on the marine environment
Hassellöv, I.-M.	2013	Shipping contributes to ocean acidification
Tripp, L.	2014	The effects of seawater-scrubber-masters-report
Boer, den	2015	Scrubbers – An Economic and Ecological Assessment
Lange, B.	2015	Impacts of EGCSs on the Environmental Situation in Ports and Coastal Waters
Stips, A.	2016	Scoping report on the potential impact of on-board desulphurization on water quality in SOx Emission Control Areas
Johansson	2017	Global assessment of shipping emissions in 2015 on a high spatial and temporal resolution
Karlberg, M.	2017	The future for micro-plankton in the Baltic Sea – Effects of SWS and climate change
Koski, M.	2017	Ecological effects of EGCS water discharge on coastal plankton: potential synergistic effects of contaminants reduce survival and feeding of the copepod <i>Acartia tonsa</i>
Turner, D.R.	2017	Shipping and the environment: Smokestack emissions, scrubbers and unregulated oceanic consequences
Endres, S.	2018	A New Perspective at the Ship-Air-Sea-Interface: The Environmental Impacts of Exhaust Gas EGCS Discharge
Winnes, H.	2018	Environmental analysis of marine exhaust gas EGCSs on two Stena Line ships, Summary Activity 3, EGCSs closing the loop
Magnusson, K.	2018	Risk Assessment of marine exhaust gas EGCS water, Task 2, Activity 3, EGCSs closing the loop
ABS	2018	ABS Advisory On Exhaust Gas EGCS Systems
Carnival	2019	Compilation and Assessment of Lab Samples from EGCS Washwater Discharge on Carnival ships
Ushakov, S.	2019	Meeting future emission regulation at sea by combining low-pressure EGR and seawater scrubbing
Ytreberg	2019	Effects of scrubber washwater discharge on microplankton in the Baltic Sea
Faber, J.	2019	The Impact of EGCS Washwater Discharges on Port Water and Sediment

As for the official submission to IMO, a short characterization was given of the most relevant articles in the scientific literature and grey literature that the Task Team on EGCS was aware of. It concerned in chronological order:

- Reed, M. & Hetland, B., 2002: The authors described the development of the DREAM, a Dose Related Exposure Assessment Model. The model was designed to support management decisions on the environmental risks of complex mixtures. The model had been further developed since, see also Rye, *et al.*, 2007 and Brønner, 2019, personal communication.
- Hufnagl, M., Liebezeit, G., Behrends, B., 2005: the report described the results of five sampling campaigns in the harbours of Calais and Dover and on board of a ferry that was equipped with an EGCS. PAHs, pH, nutrients, temperature, trace metals and nutrients were measured. No negative effects were detected on the port environment nor on eutrophication effects.
- Rye, H.; Reed, M.; Frost, T.K.; Smit, M.G.D.; Durgut, I.; Johansen, Ø.; Ditlevsen, M.K., 2008. A successive paper on DREAM that presented an application to model stressors in the water column and the sediment from drilling discharges.
- USDT, 2011: the document guided members of the US Ship Operations Cooperative Program in the selection of equipment including EGCS.
- USEPA, 2011: The document contained information on the use of EGCS onboard vessels. It reported on scavenging pollutants from the EGCS, like combustion products, fuel and lubricants, the source water used and the EGCS itself.
- Hansen, P., 2012: the report discussed the results of measurements at the EGCS on board the MV Ficaria Seaways. The EGCS was of a hybrid type and provided information on the advantages and disadvantages of both EGCS types. The system was analyzed in relation to the energy consumption of the vessel.
- Kjølholt, J.; Aakre, S.; Jürgensen, C. and Lauridsen, J., 2012: this study was initiated by the Danish Environmental Protection Agency and provided additional information on the potential environmental effects to the marine environment. As an overall result the study concluded that the pollutant concentrations were far below levels of concern and in some local conditions the levels determined could slightly exceed the environmental quality guidelines of the European Union.
- Hassellöv, I.-M., Turner, D.R., Lauer, A. and Corbett. J.J., 2013: discussed introduction of scrubbers in the context of modelling the shipping-derived pH decreases worldwide indicating that sea areas with heavy shipping traffic and seasonal stratification could be subject to larger pH decreases on a seasonal basis.
- Tripp, L., 2014: The report was mainly focusing on the Baltic Sea and discussed results from research based on laboratory measurements, experimental mesocosms and natural Baltic Sea microbial communities. It was shown that 10% effluent concentrations had effects on nutrient availability, primary production, photosynthetic activity and bacterial populations. Further research was recommended to the impact of shipping pollutants in the Baltic Sea.
- Boer, E. den and Hoen, M. 't, 2015: this was an often cited paper and dealt with an analysis of environmental and economic aspects of EGCS application in comparison to the use of Marine Gas Oil (MGO). It concluded also that EGCS may have a negative impact on the marine environment due to acidification, eutrophication and the accumulation of hazardous

hydrocarbons and heavy metals in case the dilution was limited. The authors of the study further advocated a cost benefit analysis between the potentially harmful impacts of EGCS and the benefits for the society as a whole and the ship owners in particular.

- Lange, B., Markus, T. and Helfst, L. P., 2015: the report analyzed several earlier reports published on EGCS, especially in the Baltic and North Sea. Due to the high pressure on the environment due to shipping, construction material extraction, energy, tourism and discharges of industry and agriculture, a further deterioration of the environment due the discharge of EGCS was considered unwanted and proposals were formulated to prohibit the use of EGCS unless a multilateral regulation would be established.
- Stips, A., Bolding, K., Macias, D., Bruggeman, J., and Coughlan, C., 2016: have used a spatially-resolved model to examine the potential impact of scrubber operation on acidification of the basin scale of the North Sea over a 1-year period. Authors conclude that the largest effects are confined to near-coastal areas, most particularly in the vicinity of major ports, where the acidifying effect due to SO<sub>x</sub> can equal or exceed that due to CO<sub>2</sub>.
- Johansson, L., Jalkanen, J.-P., Kukkonen, J., 2017: the authors developed a model for the global assessment of shipping emissions. The model STEAM made use of the Automated Identification System for ships to identify their route and emissions. The authors claimed that application to EGCS emissions and ballast water should be possible.
- Karlberg, M., 2017: the report dealt with a study on the micro-plankton population in the Baltic and potential threats in the future. The discharge of EGCS may have deteriorating effects on the aquatic organisms. The author applied different concentrations of diluted EGCS effluent to algal species and found no direct effects to the micro-plankton species composition but caution was recommended as long term effects were still unknown.
- Koski, M.; Stedmon, C. and Trapp, S., 2017: The paper reported on three items: 1) threshold concentrations of the copepod *Acartia tonsa* to exposure to EGCS discharge water, 2) whether the effects depended on the exposure route and 3) whether the exposure was detectable in organisms collected in the field. The authors were of the opinion that synergistic effects could play a role on the plankton productivity and bioaccumulation of metals.
- Turner, D.R., Hassellöv, I.-M., Ytreberg E. and Rutgersson A., 2017: Through modelling projections on the basin scale of the Baltic Sea, have shown that ships would become the major source of strong acid addition to surface marine waters if there was widespread use of wet EGCS.
- Endres, S.; Maes, F.; Hopkins, F.; Houghton, K.; Mårtensson, E.M.; Oeffner, J.; Quack, B.; Singh, P. and Turner, D., 2018: the paper presented the results of an international workshop on the impacts of ship emissions on biogeochemical processes at the air-sea interface and their socio-economic relevance. It was concluded that despite existing guidelines there was a risk for acidification, eutrophication and accumulation of pollutants in the marine environment due to EGCS.
- Winnes, H.; Granberg, M.; Magnusson, K.; Malmaeus, M.; Mellin, A.; Stripple, H.; Yaramenka; Zhang, Y., 2018: This report was a summary report of a much wider project of which the environmental part was dealt with in the next publication by Magnusson, *et al.* (2018). See below. There were projects on air emission measurements, risk assessment of marine exhaust gas scrubber water, cost benefit analysis and evaluation of exhaust gas scrubber systems for ship applications in a system perspective.

- Magnusson, K.; Thor, P.; and Granberg, M., 2018: the report was a part of a larger project on the effects of EGCS (see above) and this part dealt with the results of measurements on two ships where all the relevant parameters were analyzed. In addition, the discharge was tested for toxicity on field collected species, a zooplankton species and a sediment dwelling blue mussel. Effects were found with 0.04% of closed loop effluent and 1% of open loop effluent. Although there were found effects on organisms in some tests, the actual concentrations of individual compounds were always about 3 orders of magnitude lower than the concentrations that showed effects.
- ABS, 2019: the report was especially intended for ship designers, owners and operators to provide them with the latest information concerning EGCS. It gave information about regulatory requirements as well as the newest available technologies and was regularly updated.
- Carnival, 2019: the report summarized the results of 79 washwater samples on 23 ships using USEPA protocols and training taken in 2016. In the samples 54 parameters were identified and analyzed by certified laboratories. The results were compared to national and international point source discharge limits. Now an update was available with 281 samples on 53 ships. Although the report mentioned gaps in suitable standards, especially for PAHs, no exceedance of any set of limits was determined.
- Ushakov, S.; Stenersen, D.; Einang, P.M.; Ask, T.Ø., 2019: the paper reported on the environmental performance of full-scale combined low-pressure exhaust gas recirculation and seawater scrubbing system. The standard parameters were measured, pH, turbidity and nitrates, which values were below the IMO requirements, but the values for certain metals (vanadium, nickel and zinc) and the PAHs in the washwater indicated certainly of lesser quality and would need sufficient dilution. Considering the measurements of arsenic, copper and molybdenum, the substances should give cause to some concern.
- Ytreberg, E., Hassellöv, I.-M., Nylund, A.T., Hedblom, M., Al-Handal, A. Y. and Wulff, A., 2019: in the paper scrubber washwater was tested on a Baltic micro-plankton community. A significant increase in chlorophyll-a, particulate phosphorous, carbon and nitrogen was observed when the community was exposed to 10% scrubber washwater for 13 days. Also in the laboratory effects were determined: a 10% effect concentration (EC10) = 8.6% and EC10 = 5.5% for two different primary producers.
- Faber, J., Hattum, B. v., and Kleissen, F., 2019: the report was still a draft report and a follow-up of a presentation by the authors during MEPC 74. It was not clear to the Task Team in what detail it may be used as the information may be subject to changes. The report dealt with an approach to a risk assessment procedure by estimating the amount of chemical substances discharged based on energy related parameters and a set of scenarios, the application of the MAMPEC-model and regionally accepted water quality standards (the EU Maximum Allowable Concentration of the Environmental Quality Standard (MAC-EQS) and the Annual Average of the Environmental Quality Standard (AA-EQS) that will enter into force in the EU in 2021). Four different harbour scenarios had been defined as well as different background concentration assumptions. For sediments, the report identified several gaps in the available standards and also used national standards from the UK, The Netherlands and Canada. The authors did not find any significant breaching of the standards as far as available, although they noted that the results of the report were dependent on reference scenario (the number off ships in a port, and their fuel consumption and the port itself).



In the appropriate sections of this report of the Task Team on EGCS, these publications will be dealt with further.

## 5. INVENTORY OF EXHAUST GAS CLEANING SYSTEMS

### 5.1 Introduction to EGCS

This chapter gives an overview of current EGCS. EGCS could be subdivided into wet and dry EGCS. The category of the wet systems may be divided in three categories: the open loop (OL) systems, the closed loop (CL) systems and the hybrid systems. As a synonym or a short description often the name scrubbers was used. In this report consistently the abbreviation EGCS will be used. Generally, the OL systems were used with seawater whilst the CL systems worked with freshwater as washwater. Hybrid systems could use either system depending where they were sailing (Japan, 2019).

In wet EGCS, water was used to dissolve the SO<sub>2</sub> from the exhaust gas. The washwater was either discharged directly back into the sea or was re-circulated under the addition of neutralizing agents (see section 5.2). In a dry EGCS calcium hydroxide (Ca(OH)<sub>2</sub>) granules or sodium bicarbonate powder formed with the SO<sub>2</sub> calcium sulfate (CaSO<sub>4</sub>; gypsum) or sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>). Both dry EGCS were generating solid waste streams. This assessment concentrated on the investigation of the environmental impact of EGCS washwater and possible chemicals used in the process. Dry EGCS do not produce a solid waste stream to the environment and were therefore not considered further in this report. For more technical details, please refer to the numerous publications in the area of EGCS.

### 5.2 Wet EGC Systems

Wet EGCS used seawater or freshwater, some in combination with chemical additives (*e.g.* NaOH). OL EGCS (section 5.2.1) used seawater like a flow-through system, whereas CL EGCS (section 5.2.2) could use fresh- or seawater, which was re-circulated, plus neutralization chemicals (NaOH, Na<sub>2</sub>CO<sub>3</sub> or MgO) for the accumulating sulfuric acid. The hybrid type EGCS (section 5.2.3) could operate in OL or CL modes. Further, the inert gas cleaning EGCSs of oil tankers belonged into the category of wet EGCS.

#### 5.2.1. Open Loop Systems (OL)

An open loop EGCS would take water from the marine or freshwater environment where the vessel was sailing. The system would bring this water in contact with the exhaust gas. After processing, the gaseous and particulate contaminants in the exhaust gas were partly washed-out and discharged to the marine or freshwater surroundings of the ship. **This meant that the contaminants that were washed-out and removed from the exhaust gas were discharged into the water phase as dissolved or particulate chemical species.** A typical layout of an OL EGCS with its basic components was given in Figure 2. It should be noted that the washwater treatment and dilution by using clean seawater as indicated in this example were optional.

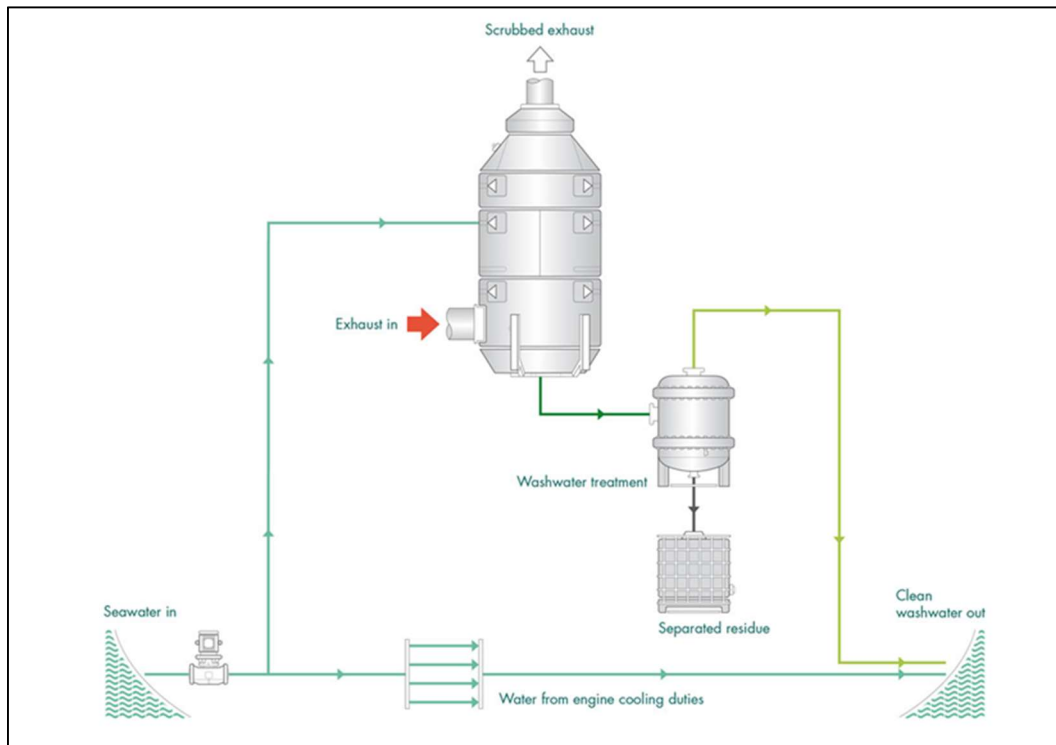
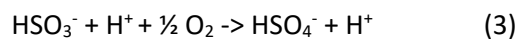
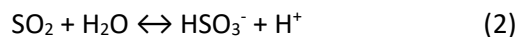
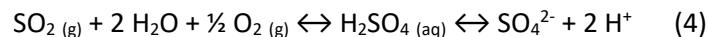


Figure 2. Open loop exhaust gas cleaning system (Courtesy EGCSA)

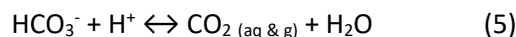
In OL EGCS, seawater was pumped and sprayed in a counter current against exhaust gas flow in the scrubbing tower, where it dissolved the sulfurous gases  $\text{SO}_2$  (equation 1) and  $\text{SO}_3$  to form sulfurous acid (equation 2), which was oxidized to sulfuric acid (equation 3). In summary, the dissolved  $\text{SO}_2$  was transformed by chemical reaction to sulfate ions and two hydrogen ions, which generated the acidity (equation 4). As follows:



Overall reaction:



Seawater was slightly alkaline (pH 8.2-8.4) due to carbonate and other ions present in seawater and therefore had a natural buffer capacity. This alkalinity of seawater was used to neutralize the acids, which are formed in the scrubbing process (equation 5, overall reaction).



Equation 5 further showed that the washwater releases two moles  $\text{CO}_2$  per one mole  $\text{SO}_2$  dissolved in the water. **When the natural alkalinity was depleted, the washwater turned acidic. The actual washwater could have a pH of 3 or even lower depending on the ratio of water flow sprayed and exhaust gas flow. The lower the pH, the less  $\text{SO}_2$  would be dissolved.**

The water consumption depended on the sulfur content of the fuel, the engine load and the intended efficiency of the scrubbing process. As a rule of thumb, usually up to  $90 \text{ m}^3 (\text{MWh})^{-1}$  was

required, based on Resolution MEPC.259(68) (MEPC, 2015). However, in a measurement campaign on OL EGCS by the German BSH, the water consumption was 75 – 140 (on average 100) m<sup>3</sup> (MWh)<sup>-1</sup> (PPR, 2018). After reacting with the exhaust gas, the washwater was either discharged directly back to the sea or was treated in a washwater treatment plant and/or diluted before discharge.

Other substances occurring in the exhaust gas would also be taken up by the washwater. These were unburnt oil (both fuel and lubricant), polycyclic aromatic hydrocarbons (PAH), trace metals, nitrate (from NO<sub>x</sub> adsorption) and particles such as black carbon, soot and ash.

The 2015 Guidelines for Exhaust Gas Cleaning Systems (MEPC, 2015) had set limits for pH, phenanthrene equivalence for PAH (PAH<sub>phe</sub>), turbidity measured as formazine nephelometric units (FNU) or nephelometric turbidity units (NTU) and nitrates. These parameters, except pH and nitrates, must be monitored continuously at the inlet of the EGCS and at the outlet of the EGCS, but before any kind of dilution. Washwater pH was allowed to be monitored only at the point of discharge, if it achieved the condition of 1) shown below. On the other hand, as no online and onboard monitoring methods for trace metals existed, these must be measured in the lab after discrete sampling. Relevant criteria were formulated in MEPC (2015) and may be summarized as follows (Magnusson, *et al.*, 2018):

- 1) Criteria for pH: The discharged EGSE should have a pH of no less than 6.5 at 4 m from the overboard discharge point with the ship stationary. Computed model may be used to simulate the pH at 4 m distances together with the reference of receiving seawater condition (*i.e.* alkalinity of 2,200 μmol L<sup>-1</sup> and pH 8.2).
- 2) Criteria for PAH concentration: The criterion for EGSE was that the maximum continuous PAH concentrations must not exceed 50 μg-PAH<sub>phe</sub> L<sup>-1</sup> above the PAH concentration in the inlet water and was normalized for a washwater flow rate through the EGC unit of 45 t (MWh)<sup>-1</sup> where the MW refers to the maximum continuous rating (MCR) or 80% of the power rating of the fuel oil combustion unit.
- 3) Criteria for turbidity: The maximum continuous turbidity in Exhaust Gas Scrubber Effluent (EGSE) should not be greater than 25 FNU (formazine nephelometric units) or 25 NTU (nephelometric turbidity units) or equivalent units, above the inlet water turbidity.
- 4) Criteria for nitrates: The EGCS water treatment system should prevent the discharge of nitrates beyond that associated with a 12% removal of NO<sub>x</sub> from the exhaust, or beyond 60 mg·L<sup>-1</sup> normalized for EGSE discharge rate of 45 tons (MWh)<sup>-1</sup>, whichever was greater.

For nitrates/nitrites, an online method was available and was published by Azzarro (2013). Further impacts on the washwater included reduced pH, temperature rise, increased chemical oxygen demand (COD), reduced oxygen concentration and decreased alkalinity.

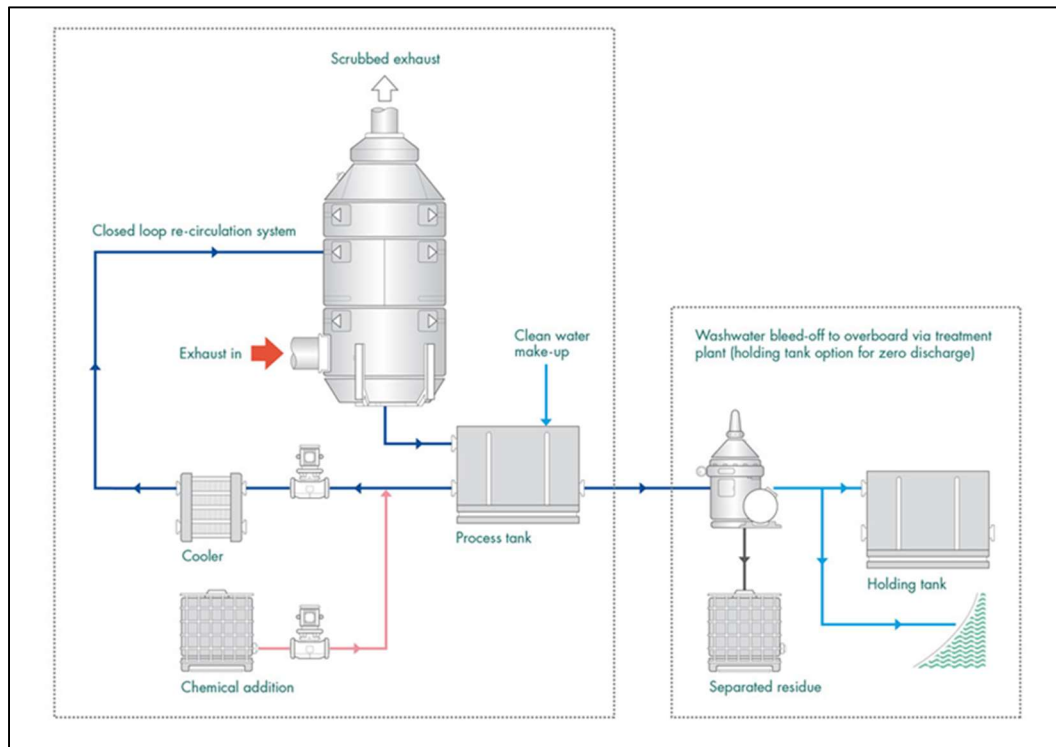


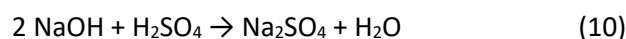
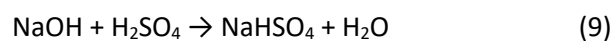
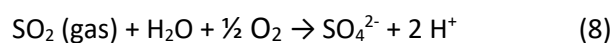
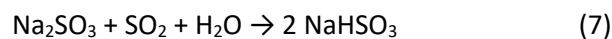
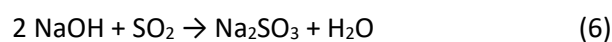
Figure 3. Closed loop exhaust gas cleaning system (Courtesy EGCSA)

### 5.2.2. Closed Loop Systems (CL)

In a closed loop exhaust gas cleaning system, the washwater was not discharged to the surrounding waters but was fed to a clean-up process tank to separate the sedimentable particles from the water phase and held stored on-board until it could be disposed of in a harbour. **Only a small amount of water was directly discharged, so-called bleed-off, see below.** A typical layout of a CL EGCS that could operate for some time without discharging any water was presented in Figure 3.

The effluent discharge of CL systems was substantially less than for the open loop system:  $0.1 - 0.3 \text{ m}^3 (\text{MWh})^{-1}$  (EGCSA, 2019) and was generally called “bleed-off”.

In CL EGCS, the reaction equations are as follows:



In CL EGCS the fresh- or seawater was re-circulated and was therefore suitable for ship operation in areas with low alkalinity. The reactions of the gaseous sulfur compounds were similar to the OL systems, but to compensate the lack and the consumption of alkalinity, chemical additives must be added to the water to neutralize the generated sulfuric acid. The simplest chemical was sodium hydroxide (NaOH), which was used as 50% solution in water. The consumption varied, depending on the fuel oil sulfur content, from  $1$  to  $11 \text{ L} (\text{MWh})^{-1}$  for 0.9% and 2.9% S in the fuel, respectively (Kjølholt, *et al.*, 2012). The higher the NaOH dosage, the better was the efficiency of  $\text{SO}_2$  reduction.

The usage of NaOH as neutralizing chemical generated sodium salts like sulfate ( $\text{Na}_2\text{SO}_4$ ), bisulfite ( $\text{NaHSO}_3$ ) and sulfite ( $\text{Na}_2\text{SO}_3$ ). The hydroxide ion reacted with the hydroxonium ions to form water. To prevent re-circulating water to be saturated with ions and above-mentioned pollutants the washwater was treated in a water treatment plant. The washwater treatment plant included chemical additives (e.g. aluminium(III)sulfate (Ochoa-González, *et al.*, 2012)) and/or mechanical treatment to remove particles and other pollutants (PPR, 2018). Following the water treatment, the water was returned to the EGCS. **The water treatment plant generated sludge, which was stored in dedicated sludge tanks for later disposal in port reception facilities.**

Further, some bleed-off water was discharged after treatment in the “bleed-off treatment unit” (BOTU). The discharge rate was typically in the range of  $0.1 - 0.3 \text{ m}^3 (\text{MWh})^{-1}$  under a re-circulation rate of  $20 \text{ m}^3 (\text{MWh})^{-1}$  (MEPC, 2008). Freshwater was added with the same flow rate to top up the process water. The bleed-off water could be stored in a holding tank for later discharge or disposal. The sludge of the BOTU was pumped into a decanter unit to allow settlement. The supernatant water was fed back into the BOTU and the sludge was stored for later disposal ashore.

### 5.2.3. Hybrid Systems

The hybrid EGCS could be operated in OL and CL modes using either seawater (OL) or freshwater (CL). An advantage of hybrid EGCS was that they could be operated in areas where the discharge of EGCS effluents was prohibited or in areas with low alkalinity (estuaries, harbours, rivers, the Baltic and other sensitive areas) (Aalborg, 2010). The system combined features of the OL and the CL system and was, therefore, more complicated. In the CL mode the effluent had to be stored on-board for the duration that the tank volume would permit.

## 5.3 Environmental aspects of EGCS washwater

In the preceding sections the existing EGCS were presented with their main features. MEPC (2019b) concluded that there was no doubt in the literature that the use of EGCS in marine applications was effective in removing SOx ( $\text{SO}_2$  and  $\text{SO}_3$ ). However, a closer examination revealed that there may be significant issues regarding exhaust particulate emissions.

The Task Team tried to explore the effects of EGCS use in the environmental and human health effects contexts, with the ToR as the guiding principle. **Both types of systems discharged water containing several chemical substances in varying concentrations to the aquatic environment.** The emission of these systems to the atmosphere was considered outside the ToR of the Task Team. The intention was to give an overview of the environmental and human health effects for the aquatic environment of the oceans and coastal areas caused by the lower emission standards to the atmosphere based on the use of EGCS on ships. The Task Team on EGCS, therefore, focused on the emission route of EGCS to marine surface water. Exposure routes were limited to the discharge of the direct effluent and the bleed-off water.

**The environmental aspects of EGCS washwater also determined the effects to humans because of the secondary poisoning of aquatic species that may find their way to human food via bioconcentration and biomagnification.** Bioconcentration was considered to be the first step in the

food chain while biomagnification was more related to the following levels in the food chain. In the way forward, these limitations had to be kept in mind in the risk assessment process proposal.

## 6. INVENTORY OF CHEMICALS

### 6.1. Introduction to Relevant Chemicals

The main aim of using EGCS on the sea-going ships was the reduction of SO<sub>x</sub> emissions if heavy fuel oil (HSFO, High Sulfur Heavy Fuel Oil) was used for the propulsion of the ship to a level that met the requirements of MARPOL Annex VI, Regulation 14.4.

Alternatively, distillate fuels were used for compliance with sulfur regulations. The distillate fuels such as marine gas oil (MGO) and marine diesel oil (MDO) were primarily used to comply also with the IMO regulation under MARPOL Annex VI. Exhaust gas cleaning systems operating in open loop mode, were low-cost and may be preferred alternatives to high-cost, low-sulfur fuel. Other technological solutions for sulfur abatements were also developed and implemented such as novel engine technologies, exhaust gas recirculation and fuel emulsifiers (Endres, *et al.*, 2018).

The evidence on how these different options enabled ships to operate within sulfur emission limits and how alternative solutions compared in the reduction of relevant chemical emissions and discharges had been yet poorly studied (Lehtoranta, *et al.*, 2019 and references there in, Zhou *et al.*, 2017, Endres *et al.*, 2018). This topic was seen as important by the Task Team on EGCS but it remained out of the scope of the present information report. **It was recognized by the Task Team on EGCS that there was a need for scientific assessments comparing alternative emission reduction techniques in terms of their overall environmental performance.**

### 6.2. General Concerns regarding Chemicals in EGCS washwater

The principle of the EGCS operation must result in the efficient transfer of sulfur oxides from the ships' exhaust gases to the washwaters of their EGCS. Through the operation of EGCS, other chemical contaminants present in the exhaust gas of the ships were also transferred, in different proportions, into the washwaters. The washwaters and chemical substances from open loop EGCS were discharged into the marine environments where the ships operate (offshore traffic lines, berth bays, estuaries, harbours and ports). **The main concern was how the shipping sector could address sulfur regulations with EGCS and tackle the environmental impacts related to the transfer of polluting chemicals from air to marine water and their aquatic marine ecosystems.**

The recognition of ships washwater as a new waste category from EGCS also triggered policy processes. This included introduction, enforcements and necessary revisions of emission limits, as for instance already adopted regulations in the scope of Annex VI to the MARPOL Convention and other national and international regulations (*e.g.* European Directives – EU 2019/883 of 17 April 2019 on port reception facilities for the delivery of waste from ships (EU, 2019)). Such policy efforts aimed at the environmental protection from chemical pollution and at the achievement and maintenance of clean, healthy and productive aquatic environments. The current regulations for on-going marine transport were, however, less strict than those applied to land-based transport within the European Union where in several Union Regulation the NO<sub>x</sub> emission was reduced, for example (Turner, *et al.*, 2017 and references therein).

### 6.3. Chemicals in EGCS Washwater: Background Considerations

The washwater from OL EGCS may basically contain most of the chemical species present in the exhaust gas of the marine diesel engines. The comprehensive chemical characterization of heavy petroleum fuels and their combustion exhaust gases from diesel engines were largely characterized in scientific literature (examples of recent publications: Celo, *et al.*, 2015, Käfer, *et al.*, 2019, Jiang, *et al.*, 2019, Corbin, *et al.*, 2018 and references therein). The exhaust gases form complex chemical mixtures and their full characterization demanded highly elaborate analytical techniques and methods (Käfer *et al.*, 2019). These efforts were augmented because exhaust gases of diesel engines were classified as carcinogenic to humans – Group 1 (IARC/WHO, 2012, Benbrahim-Tallaa, *et al.*, 2012). In the routine emissions assessments, many chemicals, often present at low trace levels in the exhaust gases and in EGCS washwaters, were not surveyed, even if these compounds were recognized as toxic and contributing to genotoxic (mutagenic and carcinogenic) character of EG. The species of concern are suites of metals, PAHs, volatile organic hydrocarbons and chlorinated hydrocarbons (Attfield, *et al.*, 2012, IARC/WHO, 2012).

Furthermore, in the appreciation of chemicals of concern in the EGCS washwaters, it should be taken into account that the composition of chemical mixtures in the washwater effluent would vary and would mainly depend on the HSHFO specification, its composition, engine characteristics, combustion temperature, ship and EGCS operation conditions. The EGCS washwater dissolved gaseous and soluble contaminants, emulsified non-combusted fuels oil and lubricants, and partly removed suspended particulate matter with adsorbed contaminants. The chemical composition of the mixture would also depend on the efficiency of transfer processes from exhaust gas to washwater, directly influenced by compounds' properties like solubility in water and their sorption capacity to particulate matter.

The revision of the IMO 2015 EGCS Guidelines contained a new Appendix 3 to facilitate and improve washwater data collection. PPR 7 was expected to complete this revision of the IMO 2015 EGCS Guidelines. This revision focused on the structure, clarification of principles and terminology as well as on the improvement of the certification framework principles and requirements. Section 10 of the revised guidelines on EGCS discharge water criteria had not been changed, due to the lack of scientific evidence.

### 6.4. Main Contaminants

The main contaminants in the ship exhaust gases were largely characterized as nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO<sub>x</sub>), particulate matter (PM) with a range of aerodynamic diameters between micrometers to nanometers (Mastral and Calleán, 2000, Moldanová, *et al.*, 2009, Celo, *et al.*, 2015, Käfer, *et al.*, 2019, Jiang, *et al.*, 2019). The emitted particles and gases contained also black carbon (BC), elemental and organic carbon (EC and OC) and other toxic contaminants such as trace metals (TMs) and polycyclic aromatic hydrocarbons (PAHs). Ships also emitted a large number and significant amounts of non-methane volatile organic compounds (NMVOC) and multiple other chemical species. In addition, roughly, for every ton of SO<sub>2</sub> input from EGCS washwaters, the ocean would not absorb about half a ton of CO<sub>2</sub> (Stips, *et al.*, 2016). It was, however, reported that the



introduction of EGCS on the ships would not strongly affect ship GHG emissions to the atmosphere, because CO<sub>2</sub> removal by washwater was generally low (< 5 %) (Køcks, *et al.*, 2013).

Contrary to the exhaust gases, the available evidence on the main contaminants identified and quantified in the EGCS washwaters from peer reviewed scientific literature was limited. Therefore, it could be appreciated indirectly via studies based on the measurements of the chemicals removal efficiency from the exhaust gases (Celo, *et al.*, 2015, Lehtoranta, *et al.*, 2019). However, more direct determinations of chemical concentrations in the washwaters were also needed for a better assessment of their potential environmental impacts.

In the following sections of the report in this chapter the Task Team on EGCS summarized estimated and measured concentrations of chemicals, grouped in inorganic SO<sub>x</sub>, NO<sub>x</sub> and trace metals (TM), particulate matter (PM) containing both inorganic and organic pools, and organic compounds, mainly PAH in the EGCS washwaters.

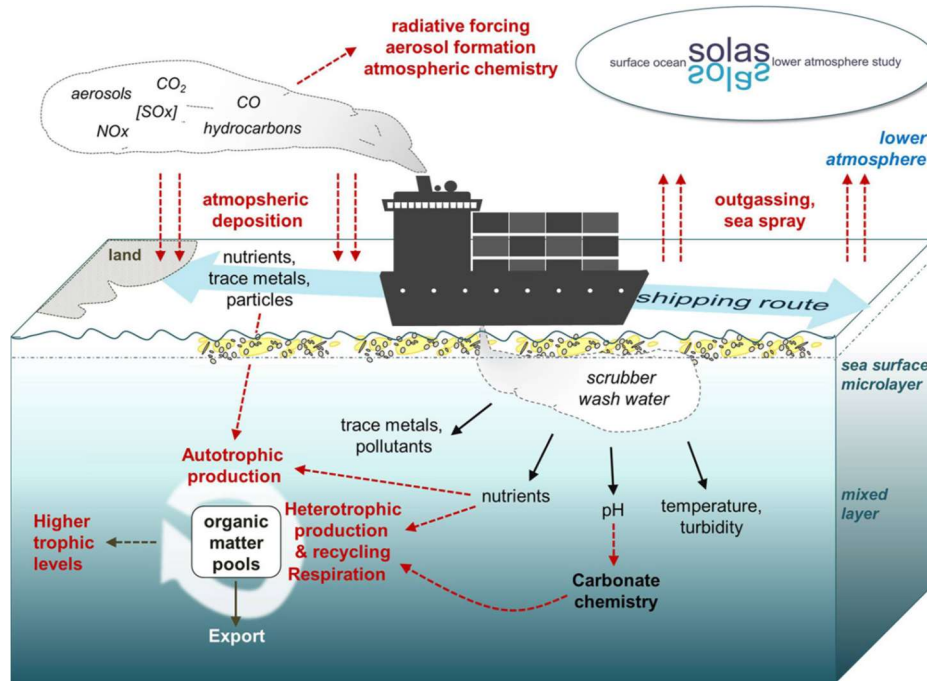
#### 6.5. General Concerns of Environmental Impacts of EGCS Washwater Discharges

A number of studies concluded that the EGCS discharge water may impact biogeochemical processes (such as acidification, eutrophication) and marine life through accumulation of pollutants in the marine environment, especially in the coastal regions, with often already higher concentrations of contaminants and less dilution and dispersal potential compared to the open sea (Hufnagl, *et al.*, 2005, Køcks, *et al.*, 2013, Kjølholt, *et al.*, 2012, Boer and Hoen, 2015, BSH, 2019, Endres, *et al.*, 2018, Hassellöv, *et al.*, 2013, Koski, *et al.*, 2015, Turner, *et al.*, 2017, Turner, *et al.*, 2018, Ytreberg, *et al.*, 2019). This was considered a developing area of research and new European projects were now initiated. In the appreciation of environmental impacts the relevance of spatially non-uniform addition of acidifying, eutrophying and polluting chemicals should be carefully considered in the assessment efforts, with the main pressures occurring in the areas of intense shipping operations (Stips, *et al.*, 2016). There was no full understanding of the EGCS washwater discharges on ecological functioning of coastal marine ecosystems. In particular, the biological effects of increasing operation of EGCS in shipping transport had been addressed in only a few studies (Koski, *et al.*, 2017; Ytreberg, *et al.*, 2019). **Additional accumulation of persistent, bioconcentrating and toxic (PBT) contaminants such as metals and PAH, especially in the coastal areas and semi-enclosed seas, may lead to exceedance of environmental quality standards** (Endres, *et al.*, 2018, Kjølholt, *et al.*, 2012, Turner, *et al.*, 2017). **Furthermore, there was a risk of cumulative and interactive effects of different pollutants, which should be considered** (Payтана, *et al.*, 2009, Chouvelon, *et al.*, 2019, Lange, *et al.*, 2015). **Metals and PAHs interacted and bioconcentrated in the first trophic levels of plankton and were transferred to higher trophic levels through food webs, especially to filter feeding marine molluscs** (Valavanidis, *et al.*, 2008, Chouvelon, *et al.*, 2019, Echeveste, *et al.*, 2011, Echeveste, *et al.*, 2012).

Scientific evidence allowing careful appreciation of the short- and long-term potential environmental impacts of the large-scale uses of exhaust gas cleaning systems, was poor (Endres, *et al.*, 2018). This was further challenging because the nature of the environmental responses was wide and complex, ranging from effects on marine ecosystems biogeochemistry, threats to marine life, climate change and feedbacks, to human health hazards (Endres, *et al.*, 2018, Fuglestedt, *et al.*, 2009, Corbett, *et al.*, 2007). Such potential, anticipated, modelled, and evidenced environmental impacts were recently summarized by **Endres, *et al.* (2018)** and are depicted in the Figure 4. This later publication was the most comprehensive available scientific summary of the environmental aspects of EGCS



discharge in the marine environment, given also in the wider perspective of the global shipping emissions, international and national global and local regulations and legal aspects of EGCS waste discharge, including alternative technologies for emission reduction (Endres, *et al.*, 2018).



**Figure 4 Ship smoke stack and EGCS water emissions at the ship-air interface and their potential effects on atmospheric chemistry and marine ecosystems in the surface ocean (reproduced from Endres, *et al.*, 2018)**

#### 6.6. Further Evaluation of Washwater Chemicals

The Task Team on EGCS, in its further evaluation of relevant chemicals in the EGCS washwaters, will take into account several background documents. The additional bibliographic review was limited in the scope of the Task Team, and the main considerations were limited to the marine environment and open loop systems.

In the section below the selected contaminant groups were presented along the following general lines:

- i) major chemical species and their ambient environmental “background” concentrations of contaminants;
- ii) measured and estimated contaminant concentrations in the washwater of the open loop systems;
- iii) examples of estimation of chemical discharge by washwater effluents;
- iv) further consideration of environmental risks of specific chemicals;
- v) explanation on the discharge limits in the current EGCS Guideline.

Several rationales and limitations for this presentation should be pointed out, namely:

- i) Well-documented and established ranges of ambient concentrations of contaminants in the open sea, coastal waters and specific areas of bays, harbours and ports were

- established in practice and recommended for their use in the risk assessment efforts in the areas where pollution from OL EGCS was added;
- ii) The data on the measured contaminant concentrations in the EGCS washwater, in the peer review studies available, was limited; therefore, based on the available emission factors, different transfer and dilution assumptions, estimated contaminant concentrations in the EGCS washwaters were proposed; this was reported as a useful exercise for instance for the discharge dilution modelling studies;
  - iii) On the basis of the measured and estimated contaminant concentrations in the outlet of EGCS washwater, several contaminant discharge loads were provided; their significance could not be fully discussed in the scope of the present information report, but the level of washwater contaminant inputs should be further assessed and taken into account in the risk assessment efforts;
  - iv) The present report also could not cover all aspects of possible environmental impacts related to EGCS contaminant discharges into the marine environment, however, more realistic approaches of environmental risk assessments in the field were highlighted.

#### 6.7. Background Documents

A number of reports submitted by IMO/MEPC to GESAMP EGCS Task Team as “background documents” were listed above. More reports were submitted to the group during its meeting in London on October 29 to November 1 and a few additional documents were downloaded through the internet.

Nevertheless, the Task Team would like to draw attention to the need for more assured and scientifically valid and independently peer reviewed literature data, in contrast to the measurement and environmental impact assessment reports presented in selected background documents. In this section, concerns were raised especially on the quality of chemical analysis data. In a number of reports, no basic information was provided either on the sampling techniques, or on the quality assurance and control of the analytical methods used. Even if certain methods used were claimed to be according to certified analytical procedures, the information was needed on the analytical performance of the specific trials. Information was missing on the blank controls, the recovery levels of contaminants extraction, the precision, accuracy and reproducibility of the analytical methods, based on the certified reference materials (CRMs) analysis. The methods quantification limit (LOQs) were often not communicated. In a few reports, the detection limits were very high, resulting in no detection of contaminants. Lack of all this information put a great uncertainty on a number of presented results and the quality of the provided data could not be fully judged and trusted. The use of these data in the appreciation of environmental impacts and/or in the risk assessment models may result in inconsistent interpretations.

Another concern was that a number of these reports presented the data on the concentrations of contaminants in the seawater – qualified as “inlet seawater”. The concentrations in the samples of the “inlet seawater” were considered as “ambient – background concentration” and frequently were used for the assessments of an added pollution and related ecological risks. However, the inlet seawater was often collected after the passage through the onboard pump system and may be contaminated mostly by metals originating from the ship itself (copper containing antifouling paints in the sea chests, cathodic pipe protection systems, also releasing copper). This portion of

contaminants, even if not directly related to the exhaust gas wash-out process, would be discharged into seawater if EGCS would not be used (cf. in Kjølholt, *et al.*, 2012) and therefore should not be regarded as ambient background concentrations.

Furthermore, in most of the reported data on contaminant concentrations in the inlet and the outlet washwaters, there was no information on the separation of dissolved, particulate or total contaminants. The extraction efficiency of the “total”, in one extraction step, that was both dissolved and particulate contaminants, was uncertain. The quantification of the contaminants’ discharge and mass budgets was biased. The so-called ambient pollution existing in the areas where ships operate may indeed come from different sources, but also from current EGCS contaminant discharges in washwater. The subtraction of contaminant concentrations in inlet water from those of outlet washwaters was basically questionable for the exposure assessments.

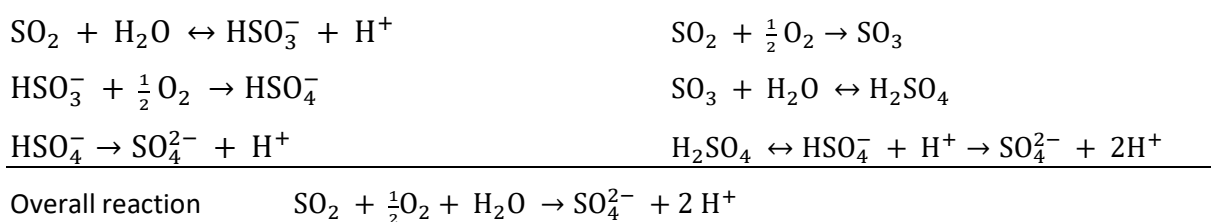
## 6.8. Inorganic Chemicals

The inorganic contaminants, like metals and SO<sub>x</sub> present in the exhaust gases were directly related to the fuel oil quality, whereas NO<sub>x</sub> were formed in the combustion process itself. Internationally, shipping accounted for about 17.0 Mt y<sup>-1</sup> for nitrogen oxides (NO<sub>x</sub>) emissions and 9.7 Mt y<sup>-1</sup> of sulfur oxides (SO<sub>x</sub>) emissions in 2012. In contrast to this, Celo, *et al.* (2015) estimated that 15% of the world’s nitrogen oxides (NO<sub>x</sub>) emissions and 5 – 8% of sulfur oxides (SO<sub>x</sub>) emissions was shipping related. The question was which part of these contaminant loads would be transferred from air to the sea and what were the potential long-term environmental consequences.

For all inorganic chemicals, Emission Factors (EF) were calculated. In the case of SO<sub>x</sub> and trace metals, EFs were calculated from the concentrations measured in fuel, as it was assumed they were transferred quantitatively to exhaust gas following combustion. It was also assumed that the removal efficiency of the EGCS was 100%, which represented a worst-case discharge scenario. In the case of NO<sub>x</sub>, EF available from the literature were used and it was assumed that 12% of NO<sub>x</sub> was transferred from the exhaust gas to the washwater (MEPC, 2015, Resolution MEPC.259(68)).

### 6.8.1. SO<sub>x</sub>

The removal of sulphur oxides (SO<sub>2</sub>, SO<sub>3</sub>) to reduce acid-rain-causing emissions to the atmosphere resulting from shipping activities, in compliance with MARPOL Annex VI (MEPC, 2019h), while using HSHFO (S > 0.5 % m m<sup>-1</sup>) was an important driver behind the installation of EGCS systems onboard ships. The chemistry of SO<sub>x</sub> removal by EGCS was simple, but the process results in direct and localized emissions of large washwater volumes characterized by a low pH (≈ 3) and high sulfate concentrations. As chemical reactions governing the removal of both SO<sub>2</sub> and SO<sub>3</sub> from exhaust gas results in the same overall reaction, only the former was used in the calculation below:



HSHFO mass-efficiency factor ( $EF_m$ , units of  $g SO_2 kg_{HFO}^{-1}$ ) and power-normalized ( $EF_p$ , units of  $g SO_2 kWh^{-1}$ ) EFs for  $SO_2$  were calculated as a function of S concentration in HFO (% S by mass in HSHFO, or  $g_S g_{HFO}^{-1} \times 100$ ), assuming that 100 % of sulfur in fuel was converted to  $SO_2$ . Molar masses of S ( $MM_S$ ) and  $SO_2$  ( $MM_{SO_2}$ ) are 32.06 and 64.07  $g mol^{-1}$ , respectively. As 1 mole S reacts with 1 mole  $O_2$  to yield 1 mole  $SO_2$ , it could be written that:

$$[1] \quad \left(\frac{\% S}{100}\right) \times \left(\frac{1000 g_{HFO}}{kg_{HFO}}\right) \times \left(\frac{1}{MM_S}\right) = \left(\frac{mol S}{kg_{HFO}}\right) = \left(\frac{mol SO_2}{kg_{HFO}}\right)$$

Since  $EF_m$  for  $SO_2$  ( $EF_{m SO_2}$ ) could be calculated as:

$$[2] \quad EF_{m SO_2} = \left(\frac{mol SO_2}{kg_{HFO}}\right) \times MM_{SO_2}$$

Combination of equations [1] and [2] yields:

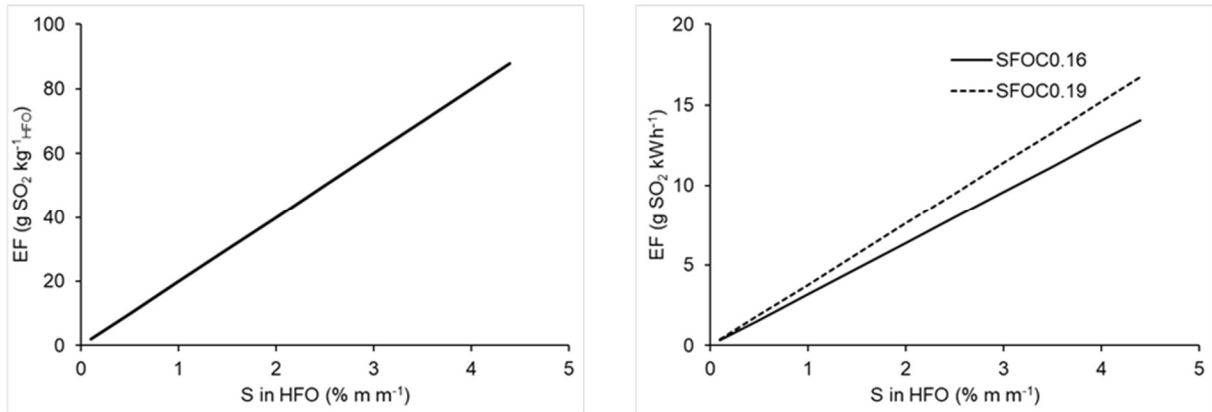
$$[3] \quad EF_{m SO_2} = \left(\frac{\% S}{100}\right) \times 1000 \times \left(\frac{MM_{SO_2}}{MM_S}\right)$$

or

$$[4] \quad EF_{m SO_2} = \% S \times 19.98$$

As the power-normalized EF for  $SO_2$  ( $EF_{p SO_2}$ ) was the product of  $EF_{m SO_2}$  and specific fuel oil consumption (SFOC, units of  $kg_{HFO} (kWh)^{-1}$ ), from equation [4] one could write:

$$[5] \quad EF_{p SO_2} = EF_{m SO_2} \times SFOC = \% S \times 19.98 \times SFOC$$



**Figure 5.** Calculated values of  $EF_{m SO_2}$  ( $g SO_2 kg_{HFO}^{-1}$ ) and  $EF_{p SO_2}$  ( $g SO_2 (kWh)^{-1}$ ) for  $SO_2$  as a function of S concentration in HFO. Minimum (solid line) and maximum (dotted line) values of SFOC used to calculate  $EF_{p SO_2}$  were 180 and 225  $g HFO (kWh)^{-1}$ , respectively

Depending on S concentration in the fuel,  $EF_{m SO_2}$  could be expected to vary from 2 to 88  $g SO_2 kg_{HFO}^{-1}$  and  $EF_{p SO_2}$  from 0.4 to 20  $g SO_2 kWh^{-1}$ . As sulfur oxides are ultimately converted to  $SO_4^{2-}$  ions ( $MM_{SO_4^{2-}} = 96.06 g mol^{-1}$ ), the concentration of the latter ( $[SO_4^{2-}]$ , in  $g L^{-1}$ ) in washwater could be calculated as a function of the flow rate in EGCS (FW, in  $m^3 (MWh)^{-1}$ ) following:

$$[6] \quad [\text{SO}_4^{2-}] = \frac{\text{EF}_{\text{pSO}_2} \times 1000 \text{ (kWh MWh}^{-1}) \times \left( \frac{\text{MM}_{\text{SO}_4^{2-}}}{\text{MM}_{\text{SO}_2}} \right)}{\text{FW} \times 1000 \text{ (L m}^{-3})} = \frac{\text{EF}_{\text{pSO}_2} \times \left( \frac{\text{MM}_{\text{SO}_4^{2-}}}{\text{MM}_{\text{SO}_2}} \right)}{\text{FW}}$$

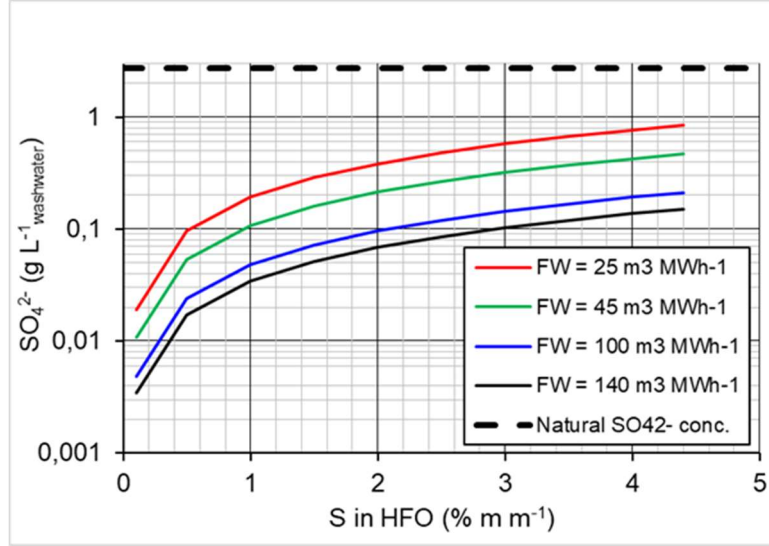


Figure 6. Concentration of  $\text{SO}_4^{2-}$  ion in washwater ( $\text{g L}^{-1}$ ) as a function of S concentration in HFO and for  $\text{SFOC} = 180 \text{ g (kWh)}^{-1}$ . Values calculated for  $\text{SFOC} = 225 \text{ g (kWh)}^{-1}$  are 18.8 % higher

Again, as a worst-case scenario and for the sake of simplicity, a complete conversion of  $\text{SO}_2$  to  $\text{SO}_4^{2-}$  within the EGCS was assumed. The concentration of  $\text{SO}_4^{2-}$  ions in EGCS effluent was calculated for FW values of 50, 100 and 150  $\text{m}^3 \text{ (kWh)}^{-1}$  (Figure 6). It could be seen that  $\text{SO}_4^{2-}$  concentrations in washwater were expected to remain below the average concentration found in seawater.

The worst effect of  $\text{SO}_x$  removal by EGCS was the production and disposal of large volumes of acidic washwater ( $\text{pH} \approx 3$ ). Corrosion may result if non-corrosion resistant material for EGCS construction and associated piping were selected. Nevertheless, a discussion on corrosion was out of the scope of the present document. At sea, EGCS usually were run at a flow rate high enough, so that the limited buffer capacity of marine waters was enough to neutralize the  $\text{H}^+$  ions produced and comply with requirement of pH of at least 6.5 at 4 meters from the point of discharge. However, disposal of acidic washwater may adversely affect the pH of water bodies of lower alkalinity that may have a lower buffering capacity, particularly if maritime traffic was congested. To evaluate the impact of EGCS acidic washwater on seawater buffering capacity, the volume of seawater completely depleted of its buffering capacity was calculated, considering that 2 moles of  $\text{H}^+$  ions were produced for each mole of  $\text{SO}_2$  removed by EGCS, the mass-normalized EF of  $\text{H}^+$  ions ( $\text{EF}_{\text{mH}^+}$ ) it could be written from equation [1]:

$$[7] \quad \left( \frac{\text{mol H}^+}{\text{kg}_{\text{HFO}}} \right) = 2 \left( \frac{\text{mol SO}_2}{\text{kg}_{\text{HFO}}} \right) = 2 \times \left( \frac{\% \text{ S} \times 10}{32.06} \right)$$

The mass-normalized EF of  $\text{H}^+$  ions ( $\text{EF}_{\text{mH}^+}$ ) was then calculated with:

$$[8] \quad \text{EF}_{\text{mH}^+} = \left( \frac{\text{mol H}^+}{\text{kg}_{\text{HFO}}} \right) \times \text{MM}_{\text{H}^+} = 2 \times \left( \frac{\% \text{ S} \times 10}{32.06} \right) \times \text{MM}_{\text{H}^+} = \% \text{ S} \times 0.629$$

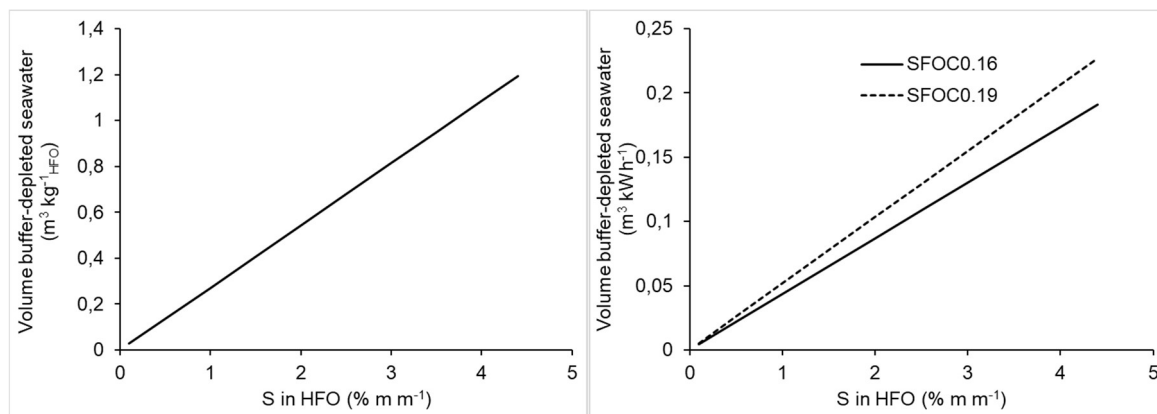
where  $MM_{H^+}$  was the molar mass of  $H^+$  ions ( $1.008 \text{ g mol}^{-1}$ ). Power-normalized EF for  $H^+$  ions ( $EF_{pH^+}$ ) was given by

$$[9] \quad EF_{pH^+} = EF_{mH^+} \times SFOC = \%S \times 0.629 \times SFOC.$$

The volume of buffer-depleted seawater (in  $\text{m}^3 \text{ kg}_{\text{HFO}}^{-1}$  or  $\text{m}^3 (\text{kWh})^{-1}$ ) as a function of Alkalinity (in  $\text{mol } H^+ \text{ L}^{-1}$ ) and EFs was then:

$$[10] \quad V_{\text{buffer-deplete}} = \left( \frac{EF_{mH^+} \text{ or } EF_{pH^+}}{\text{Alkalinity} \times MM_{H^+}} \right) \times \left( \frac{\text{m}^3}{1000 \text{ L}} \right)$$

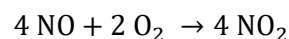
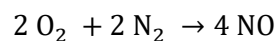
The volume of buffer-depleted normal seawater as a function of S concentration in fuel was calculated as an example (Figure 7). In the case of high-sulfur fuels, the volume of buffer-depleted seawater could reach up to a cubic meter for each kg of fuel burned. These figures were inversely proportional to alkalinity. This meant that the impact of EGCS washwater could be more important in water bodies with low salinity and low alkalinity. The Central Baltic Sea had an alkalinity of ca.  $1.5 \text{ meq L}^{-1}$  and it could be as low as  $0.7 - 1.0 \text{ meq L}^{-1}$  in the Gulf of Bothnia and the Gulf of Finland (Muller, *et al.*, 2016).  $V_{\text{buffer-depleted}}$  being inversely proportional to alkalinity, its value for a given S concentration in fuel oil would be 1.53, 2.30 and 3.28 times higher at 1.5, 1.0 and  $0.7 \text{ meq L}^{-1}$ , compared to  $2.3 \text{ meq L}^{-1}$ .

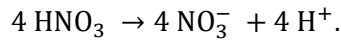
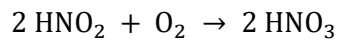
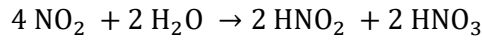


**Figure 7. Volume of buffer-depleted normal seawater ( $\text{m}^3 \text{ kg}_{\text{HFO}}^{-1}$  and  $\text{m}^3 (\text{kWh})^{-1}$ ) as a function of S concentration in HFO (left panel). Alkalinity was  $2.3 \text{ meq L}^{-1}$ . Minimum (solid line) and maximum (dotted line) values of SFOC (right panel) used to calculate  $EF_{pH^+}$  were 180 and  $225 \text{ g HFO (kWh)}^{-1}$ , respectively**

### 6.8.2 NOx

Production of nitrogen oxides resulted from the reaction of atmospheric  $N_2$  with  $O_2$  under high pressures and high temperatures typical of marine diesel engines. Briefly, nitrogen gas first formed NO with oxygen, which would further be oxidized to  $NO_2$ . Water solubility of NO was negligible whereas  $NO_2$  readily would produce nitrous and nitric acids with water. Ultimately, nitrous acid would be oxidized to nitric acid, which dissociated to  $H^+$  and  $NO_3^-$  ions.





Emission factors for NO<sub>x</sub> were not easily predictable as their value fluctuated with the type of fuel (e.g., residual fuel oil versus marine distillate), engines and actual load. Measured values of EF<sub>NO<sub>x</sub></sub> from Tier 1 engines found in the literature (Agrawal, *et al.*, 2010, Cooper and Andreasson, 1999, Cooper and Gustaffson, 2004, Zetterdahl, *et al.*, 2016) were on average 12.5 ± 3.2 g NO<sub>x</sub> (kWh)<sup>-1</sup> (range 7.5 – 18.7) and 64 ± 18 g NO<sub>x</sub> kg<sub>HFO</sub><sup>-1</sup> (range 36 – 93) for ship engines of at least 4.5 MW and burning heavy fuel oil (or residual oil) or marine distillate. Considering that 12% of NO<sub>x</sub> was assumed to be transferred from the exhaust gas to the EGCS washwater (MEPC, 2015, Resolution MEPC.259(68)), that NO<sub>x</sub> values are usually reported as NO<sub>2</sub>, that 1 mole NO<sub>2</sub> transferred to EGCS washwater will be ultimately converted to 1 mole HNO<sub>3</sub>, EF values for NO<sub>2</sub>, H<sup>+</sup> and nitrate ion were calculated and are shown in Table 4.

**Table 4. H<sup>+</sup> and nitrate ion emission factors for EGCS**

	EF <sub>m</sub> (g kg <sub>HFO</sub> <sup>-1</sup> )		EF <sub>p</sub> (g (kWh) <sup>-1</sup> )	
	X ± SD	range	X ± SD	range
<b>NO<sub>2</sub></b>	7.7 ± 2.2	4.3 – 11.2	1.5 ± 0.4	0.9 – 2.2
<b>H<sup>+</sup></b>	0.26 ± 0.07	0.14 – 0.37	0.051 ± 0.013	0.030 – 0.075
	V <sub>buffer-depleted</sub> (m <sup>3</sup> kg <sub>HFO</sub> <sup>-1</sup> ) (alkalinity = 2.3 meq H <sup>+</sup> L <sup>-1</sup> ) 0.014 ± 0.004 (range 0.008 – 0.021)			
	EF <sub>m</sub> (g kg <sub>HFO</sub> <sup>-1</sup> )		EF <sub>p</sub> (g (kWh) <sup>-1</sup> )	
	X ± SD	range	X ± SD	range
<b>NO<sub>3</sub><sup>-</sup></b>	15.9 ± 4.4	8.9 – 23.0	3.1 ± 0.8	1.9 – 4.6
	Load (kg kton <sub>HFO</sub> <sup>-1</sup> ) 15 900 ± 4400 (range 8900 – 23,000)			

### Environmental impacts of SO<sub>x</sub> and NO<sub>x</sub>

The acidification process in the ocean was enhanced by the deposition of anthropogenic SO<sub>x</sub> and NO<sub>x</sub> (Doney, *et al.*, 2007). Through modelling projections on the basin scale of the Baltic Sea, it had been shown that ships would become the major source of strong acid addition to surface marine waters (Turner, *et al.*, 2017). This acid burden from shipping may also be significant in the open ocean. It had been estimated that emissions of SO<sub>x</sub> and NO<sub>x</sub> in the heavily trafficked waters of the open ocean could lead to significant regional pH reductions of the same order of magnitude as anthropogenic CO<sub>2</sub>-driven acidification (Hassellöv, *et al.*, 2013; Stips, *et al.*, 2016). This later publication found critical regions with high ship traffic intensity, for example along shipping lanes and



in the Rotterdam port area (Stips, *et al.*, 2016). Here the contribution from SO<sub>2</sub> injection could be as much as 20 times larger than the North Sea mean (decrease of SO<sub>x</sub> and NO<sub>x</sub> = 0.0025 y<sup>-1</sup>) and would therefore be about double the impact from increasing CO<sub>2</sub> concentrations.

The relative contribution of NO<sub>x</sub> and SO<sub>x</sub> to ocean acidification was dependent upon the concentration of S in fuel (Figure 8): with low sulfur fuels, NO<sub>x</sub> emissions were responsible for most of the water acidification whereas their contribution would become negligible compared to SO<sub>x</sub> in high-sulfur fuels.

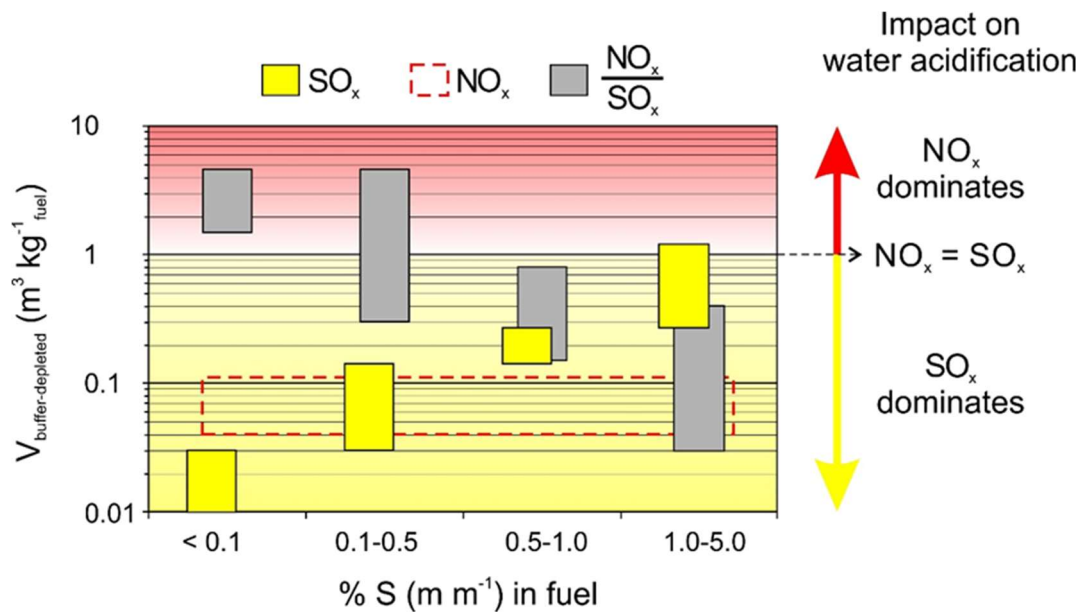


Figure 8. Relative contribution of NO<sub>x</sub> and SO<sub>x</sub> emissions on water acidification

## Eutrophication

Eutrophication was considered a relevant environmental problem for different areas of the world oceans (Gauss, *et al.*, 2013). Shipping contributed to nitrate emission and therefore, to the eutrophication processes especially in specific areas such as coastal semi-enclosed seas where nitrogen may be the limiting nutrient. (EEA, 2015, HELCOM, 2015; Jonson, *et al.*, 2015 and references therein). In more offshore and oligotrophic regions, away from direct terrestrial agricultural runoff, ship derived nitrogen, but also limiting iron, may have effects on the productivity of pelagic phytoplankton and its community structure (EEA, 2015, HELCOM, 2015, Endres, *et al.*, 2018).

IMO MARPOL guidelines limited the discharge of nitrates in washwater effluent stating that EGCS NO<sub>x</sub> removal from the exhaust gases should not be more than 12% and / or below 60 mg L<sup>-1</sup> normalized concentration in washwater discharge rate at 45 m<sup>3</sup> (MWh)<sup>-1</sup> (IMO, 2008b).

Most of the surveyed studies showed that the nitrogen amount washed out by EGCS was relatively limited and its concentrations in the washwaters were well below IMO guidelines (Hufnagl, *et al.* 2005, BSH, 2019, Boer den and Hoen, 2015). Hufnagl, *et al.* (2005), indicated that EGCS nitrogen from washwater appeared unlikely to cause a concern, because the quantities compared to other sources were very low and should be quickly assimilated in marine ecosystems. The BSH (2019) study showed that in the maximum EGCS installation scenario (in North Sea, English Channel, and Baltic Sea), EGCS



wastewater discharge of nitrates was significantly lower than river Rhine inputs in the North Sea. Therefore, the potential for significant increase of primary production and eutrophication appeared to be low.

Finally, it should be also noted that part of the exhaust NO<sub>x</sub> emissions would end up in the sea, independent of the use of an EGCS. This was mainly because, as noted above, most of NO<sub>x</sub> was not well soluble and was not removed in any significant quantities from exhaust gasses, whereas it was a major nitrogen chemical species in diesel engine exhausts.

### 6.8.3. Trace metals

#### Major and trace elements in HFO

The most abundant metal element in the heavy fuel oils was vanadium (V), which was also particularly toxic (Thompson and Orvig, 2003). It was followed by other metals such as Ni, Fe, Ca, Na present in significant amounts and other less-abundant species: Si, Al, Zn, Cr, Cu, K, Ba, Mo, Mn, Mg, Sn and trace elements As, Cd and Hg. Mainly V, Ni and Fe were known as the indigenous elements of crude oils, but their relative composition varied from oil to oil (Celo, *et al.*, 2015). It was recognized that almost all content of metals present in HFO may be transferred into exhaust gas and ultimately into washwaters (Celo, *et al.*, 2015). The EFs of these elements were correlated with their concentrations in the fuel (Agrawal, *et al.*, 2008a and b, Stippula, *et al.*, 2014, Moldanová, *et al.*, 2009). Therefore, the composition of metals in the washwaters would vary depending on the fuel oil used. Other parameters, such as type and technology of the engine, speed of operation and engine load would also affect the chemical composition of ship exhausts and washwaters. For instance, the prevalence of V and Ni in ship plumes was also reported in a number of studies and was used as an indicator of the contribution of shipping to the emissions in the regions of heavy maritime traffic (Celo, *et al.*, 2015 and references therein). The minor trace elements were also determined in the lanthanoid series, where the major elements were La and Ce (Celo, *et al.*, 2015). These elements together with V and Ni were considered as possible tracers of primary PM emissions by marine shipping (Viana, *et al.*, 2009, Celo, *et al.*, 2015, Viana, *et al.*, 2014; and references therein). The majority of studies on trace metals coming from EGCS focus on: As, Cd, Co, Cr, Cu, Fe, Mn, V, Ni, Pb and Zn as being of potential environmental concern.

#### Environmental Trace Metal Concentrations

The rationale for the presentation of contaminants' ambient environmental concentrations was given under the PAH paragraph (see below). The fundamentals of this discussion are also relevant for trace metals.

Total trace element concentrations in the open sea and seawaters (Atlantic, Pacific and Indian oceans), and available coastal regional waters were given in the Table 5. Metal data for harbour seawater was scarce and was expected to be highly variable. (Broeker and Peng, 1982, Bruland and Lohan, 2004, Mason, 2013, Chakraborty and Owens, 2014, Jahan and Strezov, 2017, Martínez-Soto, *et al.*, 2016). In harbours, most of the available data reported is on the sediment compartment. Therefore, the data presented in Table 5 should not be taken as representative values for harbour zones but was reported to show the variability of metal concentrations. This showed that ECGS

washwaters may impact the marine environment to various extents, mostly depending on local anthropogenic activities. That also meant that pristine marine environments like many “paradise islands” and other touristic spots may be significantly impacted by the presence of cruise ships.

As a rule of thumb, metal concentrations in coastal seas and harbours were 2 to 3 orders of magnitude higher than in open sea respectively.

**Table 5. Total Trace Metal Concentrations ( $\mu\text{g L}^{-1}$ ) reported in different areas; < indicates below detection limit**

[ $\mu\text{g L}^{-1}$ ]	World Open Ocean			World Coastal waters		Australia Harbour 1		Australia Harbour 2		Mao Minorca Harbour	
	min	max	average	min	max	min	max	min	max	min	max
V	1.5	2.0	1.5	-	-			3	8		
Fe	0.001	0.1	0.03	-	-	180	350	8	530	0.07	1.9
Ni	0.1	0.7	0.5	0.9	2.1	0.3	1.9	BDL	9	0.16	0.35
Pb	<0.002	0.03	0.02	1.0	1.5	0.4	55	1	7	0.04	0.5
Zn	0.003	0.6	0.3	12	22	14	67	1	35	0.1	3.9
Cd	<0.001	0.1	0.06	-	-	0.1	0.8	1	7	0.01	0.04
Hg	<0.002	-	0.0004	-	-	-	-	BDL	BDL		
As	1.5	1.9	1.7	0.8	1.5	3.2	8.2	2	8		
Cr	0.2	0.3	0.2	-	-	0.4	1.3	1	2		
Cu	0.03	0.3	0.3	0.9	1.9	0.9	350	1	40	0.1	3.4
Mn	0.004	0.27	0.02	1.1	6.0	6.5	160	1	51		
Al	0.01	1.1	0.5	-	-	-	-	1	1200		
Co	<0.0006	0.001	0.001	-	-	2.2	5	1	2	0.01	0.04
Mo	-	-	10	-	-	-	-	6	13	9.1	14
Ag	<0.001	0.004	0.002	-	-	-	-	BDL	1		
Se	0.04	0.2	0.1	-	-	-	-	1	5		

At seawater pH conditions, metals were mostly adsorbed onto particulates (ca. 80 to 99%), the dissolved part represented a small fraction of the total metal concentration (ca. 20 to 1%). As described in Hatje, *et al.* (2003) metal desorption depends on pH, SPM loading and salinity. Desorption of particulate metals was correlated to pH change towards acidic conditions.

### Washwater concentrations of metals

The desorption of metals increased sharply within narrow changes of pH (1 to 2 units) for Cu, Zn, Cd and Ni. Increasing release of metals to the dissolved phase was expected in the EGCS washwater effluents, because of their high acidity, thus enhancing metals bioavailability in the receiving ambient seawaters. This in turn would make these metals releases of greater concern for the health of coastal communities.

Although only limited data were currently available on monitoring of metals in EGCS washwaters, the reviewed surveyed studies on open-loop scrubber washwater indicates high to very high metal

concentrations. The summary table of the examined results was given in the Appendix, Table A2. The wide ranges of metals concentrations were determined in EGCS washwaters.

Vanadium had been found to be the dominating dissolved metal in several EGCS effluent studies with concentrations ranging from 10 – 300  $\mu\text{g L}^{-1}$  in OL and 3 – 9  $\text{ng L}^{-1}$  in CL EGCS study (PPR 6/INF.20). This compared well with a measurement campaign carried out by EGCSA and Carnival, where on average 210 and 140  $\mu\text{g V L}^{-1}$  in OL washwater were found, respectively. However, the average concentrations may be influenced by non-detected or below detection limit results. The overall range found for V was from 10 to 860  $\mu\text{g L}^{-1}$ .

The solubility of  $\text{V}_2\text{O}_5$  was 0.92  $\text{g L}^{-1}$  at a temperature of 20 °C. The predicted no effect concentration (PNEC) of  $\text{V}_2\text{O}_5$  for marine water was 2.5  $\mu\text{g L}^{-1}$  and 240  $\text{mg kg}^{-1}$  for sediments (ECHA, 2019b). All of the measured  $\text{V}_2\text{O}_5$  concentrations are higher than this PNEC value for seawater. A recent study to determine levels for water quality guidelines in the case of bitumen extraction in Canada (Schiffer and Liber, *et al.*, 2017) had also shown the acute and chronic toxicity of V on marine zoo- and phytoplankton. V concentrations starting at 0.1  $\text{mg L}^{-1}$  seemed to impact adversely the zooplankton. ISO 8217 specifies that maximum concentration of V should be limited to <150  $\text{mg kg}^{-1}$ . The estimated concentration of V in washwater, using the ISO specification, SFOC ( $\text{g-fuel (kWh)}^{-1}$ ) and average flow rate of EGCS ( $\text{m}^3 (\text{MWh})^{-1}$ ), was about 3 times higher than the above mentioned concentration of 0.1  $\text{mg L}^{-1}$ , therefore, further assessment will be needed for potential risk by V.

**Table 6. Average concentrations and ratios of heavy metals in the dissolved versus the particulate phase in EGCS washwater on the basis of measured data from PPR 6/INF.20**

[ $\mu\text{g L}^{-1}$ ]	Average	Average	Average	Average	Ratio	Ratio
	diss. conc. OL discharge	diss. conc. CL discharge	part. conc. OL discharge	part. conc. CL discharge	diss./part. OL discharges	diss./part. CL discharges
V	78	5700	11	620	7.4	9.3
Ni	16	1200	3.8	280	4.1	4.4
Cu	6.4	18	0.7	4.4	9.1	4.1
Zn	4.7	150	0.3	53	16	2.9
As	3.3	15	<LOD	1		15
Cd	0.03	0.14	<LOD	0.01		14
Pb	0.08	0.01	0.08	1.2	1.0	0.01

As stated above, due to the high acidity of the washwater, the solubility of metals was increased. This was supported by a measurement campaign of the BSH (PPR 6/INF.20), where both particulate and dissolved heavy metals were analyzed. The average dissolved vanadium concentration had been found to be ca. 7 times (OL) to 9 times (CL) higher than the average particulate vanadium analyzed in the suspended matter (Table 6). The same trend holds true for all the other metals.

The environmental potential impacts of copper and zinc discharged by EGCS were recently presented by Turner, *et al.*, 2017. The average concentrations of both copper and zinc in washwaters were 6 and 340  $\mu\text{g L}^{-1}$ , respectively. The highest total copper and zinc concentrations reported in discharge

water were 260 and 537  $\mu\text{g L}^{-1}$ , respectively (Appendix, Table A2). In total, 18 discharge waters have been analyzed for metal concentrations and the average concentrations of copper and zinc are 60 and 136  $\mu\text{g L}^{-1}$ , respectively (Table 6). Thus, the average daily load of copper and zinc from a medium-sized RoRo vessel equipped with a 12 MW main engine would be 780 g Cu and 1770 g Zn.

### Estimated concentrations and discharges of metals

Mass-standardized emission factor ( $EF_m$ ) values for V, Fe, Ni, Pb, Zn, Cd, Hg, As, Cr and Cu were taken as their concentrations measured in HSHFO (Moldanova, *et al.*, 2009; Agrawal, *et al.*, 2010; Cooper and Gustafsson, 2004; Linak and Miller, 2000; Corbin, *et al.*, 2018; Huffman, *et al.*, 2000; Table 7). Power-standardized emission factors ( $EF_p$ ) were calculated in turn for a SFOC of 0.19  $\text{kg}_{\text{HFO}} (\text{kWh})^{-1}$  with:

$$EF_p = EF_m \times SFOC = g (\text{kWh})^{-1}.$$

**Table 7.** Mass-standardized emission factor ( $EF_m$ ) values for V, Fe, Ni, Pb, Zn, Cd, Hg, As, Cr and Cu and power-standardized emission factors ( $EF_p$ ) calculated for a SFOC of 0.19 kg HFO ( $\text{kWh})^{-1}$

Element	Concentration in HFO ( $\text{g kg}^{-1}$ ) = $EF_m$				$EF_p$ ( $\text{g (kWh)}^{-1}$ , SFOC = 0.19 ( $\text{kg}_{\text{HFO}} (\text{kWh})^{-1}$ ))			
	Mean	SD	min	max	Mean	SD	min	max
V	1.2E-01	6.9E-02	3.5E-02	2.2E-01	2.3E-02	1.3E-02	6.7E-03	4.2E-02
Fe	1.9E-02	4.4E-03	1.3E-02	2.5E-02	3.7E-03	8.4E-04	2.5E-03	4.8E-03
Ni	2.9E-02	6.1E-03	1.7E-02	3.5E-02	5.4E-03	1.2E-03	3.2E-03	6.7E-03
Pb	1.7E-03	2.0E-03	1.0E-04	4.5E-03	3.2E-04	3.7E-04	1.9E-05	8.6E-04
Zn	2.3E-02	3.5E-02	1.0E-03	7.4E-02	4.3E-03	6.6E-03	1.9E-04	1.4E-02
Cd	4.6E-04	2.6E-04	1.3E-05	6.0E-04	8.8E-05	4.8E-05	2.5E-06	1.1E-04
Hg	7.7E-05	4.7E-05	3.0E-06	1.2E-04	1.5E-05	8.8E-06	5.7E-07	2.3E-05
As	3.1E-04	3.0E-04	1.0E-04	8.5E-04	5.9E-05	5.8E-05	1.9E-05	1.6E-04
Cr	1.1E-03	9.3E-05	9.6E-04	1.2E-03	2.0E-04	1.8E-05	1.8E-04	2.3E-04
Cu	1.8E-03	1.4E-03	5.6E-04	3.5E-03	3.5E-04	2.6E-04	1.1E-04	6.7E-04

A range of trace metals concentration in discharge water was calculated using minimum and maximum values of  $EF_p$  and a flow rate FW of 100  $\text{m}^3 \text{MWh}^{-1}$ . This figure was compared with concentration data for trace metals in OL EGCS at discharge published by Koski, *et al.* (2017), PPR (2018, PPR 6/INF.20 in the figure), MEPC (2019f, CESA, open loop in the figure) and BSH, (2019, Germany in the figure). It may be concluded from this figure that the concentration ranges predicted (white boxes in figure 9) from the calculated metals EFs correspond quite well to the concentrations actually measured. This may be used as a tool for environmental risk assessment and to detect anomalies, such as too high Cu, Zn and Cr concentrations that may be indicative of EGCS corrosion.

The BSH preliminary report (2019) provided an assessment of metals (and for other contaminants, see below for PAHs) discharge-loads from open loop systems (and also close loop) to the Baltic Sea, North Sea and English Channel. The estimations were performed for EGCS “current status” - CSS and “maximum installation” MIS scenarios, taking into account shipping activities, washwater water flow

rates, and actual contaminant concentrations in washwater effluents. In the current scenario situation, the total (dissolved and particulate fractions) annual discharge of studied metals in the area of the North Sea, Baltic Sea and the English Channel were in the ranges: V (4 – 8 tons), Ni (2 – 26 t), Cu (0.6 – 7 t), Zn (0.8 – 46 t), As (0.4 – 2 t), Cd (0.003 – 0.02 t) and Pb (0.03 – 0.8 t). In the maximum installation scenario, these discharges for metals with washwaters effluents were: V (41 – 1010 t), Ni (18 – 240 t), Cu (5 – 63 t), Zn (7 – 430 t), As (3 – 22 t), Cd (0.02 – 0.25 t) and Pb (0.3 – 7 t). These ranges indicated significant inputs of metals accumulating in the environment. This situation presented a problem for some coastal areas with high shipping density and potential exceedance of environmental quality standards. The inputs of these elements coming from EGCS washwaters may, at local scales, in such areas, impair achieving good environmental status, failing the objectives of international agreements and regulations such as European Directives (MSFD, WFD and others).

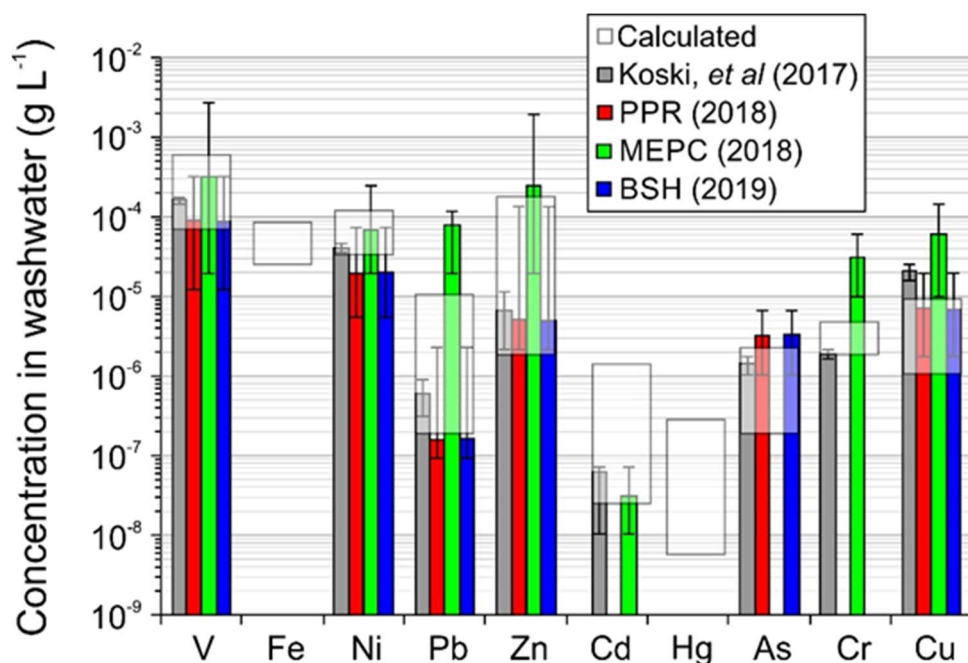


Figure 9. Minimum and maximum concentrations were used to calculate a range of EF values for each metal

### Metal environmental impacts

Certain trace metals had essential biological functions within a narrow range of optimal concentrations (essential elements), while others had no known biological role (non-essential elements) and were recognized for their toxic effects on aquatic organisms, even at low concentrations (Mason, 2013). The addition of both essential and non-essential elements into marine environment led to effects and impacts. Taxa- and species-specific metal regulation mechanisms (*i.e.* uptake, storage and/or elimination) had been described for both essential and non-essential elements (Wang and Rainbow, 2010). Their transfer between biogeochemical compartments, bioconcentration in organisms and biomagnification in food webs depended on their concentrations and speciation in both abiotic (habitat) and biotic (food sources) environments (Neff, 2002; Rainbow, 2002). Marine organisms were hence exposed to and accumulate contaminants via dissolved and

trophic pathways; the latter being the main route for trace metal intake by medium to high trophic level consumers such as fish (Mathews and Fisher, 2009; Pouil, *et al.*, 2016).

In a recent study, Koski, *et al.* (2017) investigated the threshold concentrations of metals in the EGCS discharge washwaters for survival, feeding and reproduction for the copepod, *Acartia tonsa*, and how the determined responses depended on the exposure routes. They also looked at whether the effects of discharge washwaters could be detected in field-collected organisms. Their findings indicated that a direct exposure to discharge EGCS washwaters increased adult copepod mortality and reduced feeding. The metals concentrations were orders of magnitude lower than the lethal concentrations suggesting synergistic effects on plankton productivity and bioaccumulation of metals. Ytreberg, *et al.* (2019) investigated how microplankton species and communities may be affected by EGCS washwater discharges in the Baltic Sea. The main result showed scrubber washwater to stimulate the microplankton community; shown in increased chlorophyll *a* concentrations as well as POP, POC and PON concentrations in the 10% scrubber washwater treatment. Cd and Pb toxicity thresholds had also been studied for marine phytoplankton (Echeveste, *et al.*, 2012). Values of  $0.2 \cdot 10^{-6} \text{ g L}^{-1}$  and  $20 \cdot 10^{-6} \text{ g L}^{-1}$  of Cd and Pb respectively were found to be lethal (LC50). Considering that HFO Cd and Pb concentrations are 2000 and 85 times higher than these thresholds respectively, acute toxic effects arising from the discharge by EGCS of these elements might also be considered.

### Trace metals discharge limits

Up to this date, there were no regulations or guidelines for specific trace metals discharge concentrations in EGCS washwaters. However, the MEPC 2015 Guidelines stated that the environmental criteria for EGCS residues needed updating and that ship owners and scrubber manufacturers were requested to additionally monitor heavy metals such as cadmium, copper, nickel, lead, zinc, arsenic, chromium, and vanadium in the washwater. The Task Team on EGCS recommended the development of harmonized guideline procedures for selected trace metal concentrations in EGCS washwater effluents.

### 6.9 Particulate Matter

Particulate matter (PM), along with the gas-phase, was one of most studied fractions emitted in exhaust gases from the marine diesel engines. The emitted particulate matter (PM) properties, mass, size distribution, chemical composition and structures were extensively investigated (Moldanovà, *et al.*, 2009 and references therein). A number of recent studies linked PM of ship emissions to both negative health effects for exposed populations, and to significant changes in climate patterns (Corbett, *et al.*, 2007, Tronstad, *et al.*, 2012, Fuglestvedt, *et al.*, 2009). With the actual growth trend in utilization of EGCS, new studies were carried out in order to clarify the effects of EGCS introduction on particle mass, number and size, and related chemicals emissions, in the wide context of health hazards and global environmental impacts (Lehtoranta, *et al.*, 2019 and references therein).

In the present report, the Task Team did not review the large number of scientific publications on emissions and impacts of PM, which would be out of the scope of the ToR. The surveyed studies in the background documents did not provide much information on how EGCS affect particulate matter. However, considering the large spectrum of PM environmental impacts and its crucial role as

the main carrier of different chemicals this topic should be further examined in the context of EGCS washwater effluents discharge.

### 6.10 Organic Contaminants

A great number of organic contaminants were emitted by ships diesel engines through either incomplete combustion of the fuel or via formation during the combustion processes. All of these chemicals may be found in EGCS washwater. The amounts and chemical composition of emitted compounds was related to the type of fuel used and the engine operations. The numerous organic compounds determined in the exhaust gas of marine engines were mainly hydrocarbons like linear and cyclic hydrocarbons (e.g. alkanes, alkenes, terpenes, etc.), mono-aromatics (benzene, toluene, ethylbenzene, xylene, together often named BTEX) and a great number of polycyclic aromatic hydrocarbons (PAHs). A large group of organic compounds was also more operationally defined as non-methane volatile organic compounds (NMVOC), covering a variety of volatile hydrocarbons, aldehydes, ketones and others.

The emission factors for many of the individual chemicals and their groups for marine engines were reported by Agrawal, *et al.*, 2010, Cooper and Peterson, 1996, Cooper and Andreasson, 1999, Cooper, 2001, Cooper and Gustaffson, 2004, and Zetterdahl, *et al.*, 2016. Methane, together with black carbon and N<sub>2</sub>O were quantified as shipping non-CO<sub>2</sub> climate pollutants, whereas NMVOC and other contaminants belonged to air pollutants because of their volatility (Smith, *et al.*, 2015, Olmer, *et al.*, 2017). The amounts of NMVOC emitted by international ships were estimated to be globally 609 kt (kilotons) in 2012, whilst Olmer, *et al.* (2017) estimated an amount of 795 kt in 2015. Toxic halogenated organic compounds were also identified in the marine combustion exhaust gases and particles. The groups of compounds such as polychlorinated dibenzodioxins (PCDDs) and dibenzofurans (PCDFs), polychlorinated biphenyls (PCB) were found at low trace levels and should also be accounted for in their environmental budgets.

#### **Polycyclic aromatic hydrocarbons (PAHs)**

IMO had already regulated the maximum PAH concentration in the discharged EGCS washwaters from the viewpoint of environmental protection. Furthermore, PAHs were the largest known group of carcinogenic substances recognized as global contaminants and being on the lists of priority pollutants in regional frameworks (EPA, 2014; EU, 2013). They were persistent compounds and accumulate in the marine environment. As potential mutagens and/or carcinogens (Allen, *et al.*, 1998; Durant, *et al.*, 1998) PAHs represented a significant public health concern and threats to ecosystems. The total PAH emissions from shipping operations were seen as a significant contributor to their global budgets (Shen, *et al.*, 2013). Environmental quality standards (OSPAR, 2004, EU, 2013 and US EPA, 2014) existed for certain PAH compounds in these regional frameworks.

#### **PAH environmental concentration**

The concentrations of PAHs in marine waters were relatively well documented in scientific literature (González-Gaya *et al.*, 2016, Lohmann, *et al.*, 2013 and references there in). An in-depth and critical review of bibliographic data was not attempted in this report. The intention was to assess the ranges



for environmental concentrations of PAHs to be regarded as their reference levels for environmental assessments of EGCS discharge. The concentrations of PAHs in marine waters are generally in the wide range from parts per quadrillion (ppq – pg L<sup>-1</sup>) to parts per billion (ppb - µg L<sup>-1</sup>). Such wide ranges and low levels of concentrations required suitable analytical techniques and often large volumes of water samples to perform the analysis. Gas chromatography-mass spectrometry (GC-MS) has become the leading reference technique for the analysis of PAHs. The highest environmental concentrations of PAHs were reported in specific areas: ports, harbours, after operational and accidental oil spills and in certain environments such as for instance in macro-tidal estuaries with maximum turbidity zones (Zhou, *et al.*, 2000, Reddy and Quinn, 2001, Tronczynski, *et al.*, 2004, González, *et al.*, 2006). The developments of complementary analytical techniques (Time of Flight Mass Spectrometry (ToF-MS), Gas Chromatography Mass Spectrometry (could be double MS) (GC-MS-MS), Ion Trap Mass Spectrometry (ITMS) and Gas Chromatography Triple Quad Mass Spectrometry (GC-TQ-MS)) had also allowed the detection of PAHs at very low levels including those in open ocean waters. Whereas the relevance of being able to detect very low concentrations of PAHs should be now taken into account and linked to risk assessment which in turn should result in a sensible policy as far as monitoring was concerned.

As an example, the Task Team had assembled data on PAH concentrations in open oceanic waters, coastal waters and for oil spills in Table 8. PAH was a loosely defined analyte, because it was a mixture of many compounds. Therefore, their summarized concentrations varied and depended on which and how many individual compounds were quantified and summarized. For example, Guigue, *et al.* (2011, 2014) reported that when taking into account alkylated derivatives (*i.e.*, mono-, di-, tri- or tetra-methyl PAHs) the concentration of dissolved total PAHs may increase by a factor 1.5 – 3 in coastal and harbour waters.

**Table 8. Dissolved PAHs environmental concentrations (ng L<sup>-1</sup>)**

Ocean area	Mean	Range	Σ PAH Sum range	References
Open ocean		0.1 – 10	Σ13 – 19	Witt and Matthäus, 2001; Stortini, <i>et al.</i> , 2009; Berrojalbiz, <i>et al.</i> , 2011
Coastal areas		1.6 – 500	Σ13 – 19	Guitart, <i>et al.</i> , 2007; Valavanidis, <i>et al.</i> , 2008; Qiu, <i>et al.</i> , 2009 Tronczynski, <i>et al.</i> , 2004
North Atlantic	1.8		Σ16	González-Gaya, <i>et al.</i> , 2016
South Atlantic	2.0		Σ16	González-Gaya, <i>et al.</i> , 2016
North Pacific	1.4		Σ16	González-Gaya, <i>et al.</i> , 2016
South Pacific	1.5		Σ16	González-Gaya, <i>et al.</i> , 2016
Indian	1.1		Σ16	González-Gaya, <i>et al.</i> , 2016
West Atlantic		0.8 – 2.8	Σ16	Tronczynski, <i>et al.</i> , 2004
East Atlantic	0.4	0.2 – 0.7	phenanthrene	Nizzetto, <i>et al.</i> , 2008
Atlantic	0.2	0.03 – 1.4	phenanthrene	Lohmann, <i>et al.</i> , 2013
Oil spills		3.9 – 100 000	Different ranges	Reddy and Quinn, 2001; Tronczynski, <i>et al.</i> , 2004; González, <i>et al.</i> , 2006; Reddy, <i>et al.</i> , 2012; Wade, <i>et al.</i> , 2016



The concentrations of particulate PAHs (associated with suspended matter in the water column and sediments) were not reported here. The ranges of concentrations varied widely – from open sea to coastal and estuarine environments. In the water column particulate PAH concentrations often depended on the levels of suspended particulate matter (SPM). In shallow waters, estuaries and ports particulate PAH concentrations may vary with natural (tides, currents) and mechanical (e.g. dragging, ship propellers) bottom sediment resuspension.

### PAH concentrations in EGCS washwaters

The Task Team took firstly into consideration available data on the PAH analysis carried out using gas chromatography coupled to mass spectrometry (GC-MS). The optical method with UV/fluorescence sensor continuous determination of phenanthrene will be discussed later.

The sources of PAH in scrubber washwater were three-fold: PAH could constitute a large fraction of the fuel oils (petrogenic PAH), which by incomplete combustion could end up in the exhaust gas (Czech, *et al.*, 2018) and finally in the EGCS washwater. The same would hold true for the lubrication oil. The third source was the formation of PAH during combustion (pyrogenic PAH). Marine fuels like heavy fuel oils (HFO) were composed of tens of thousands of compounds with c. 75% hydrocarbons, including PAHs (OSPAR, 2004).

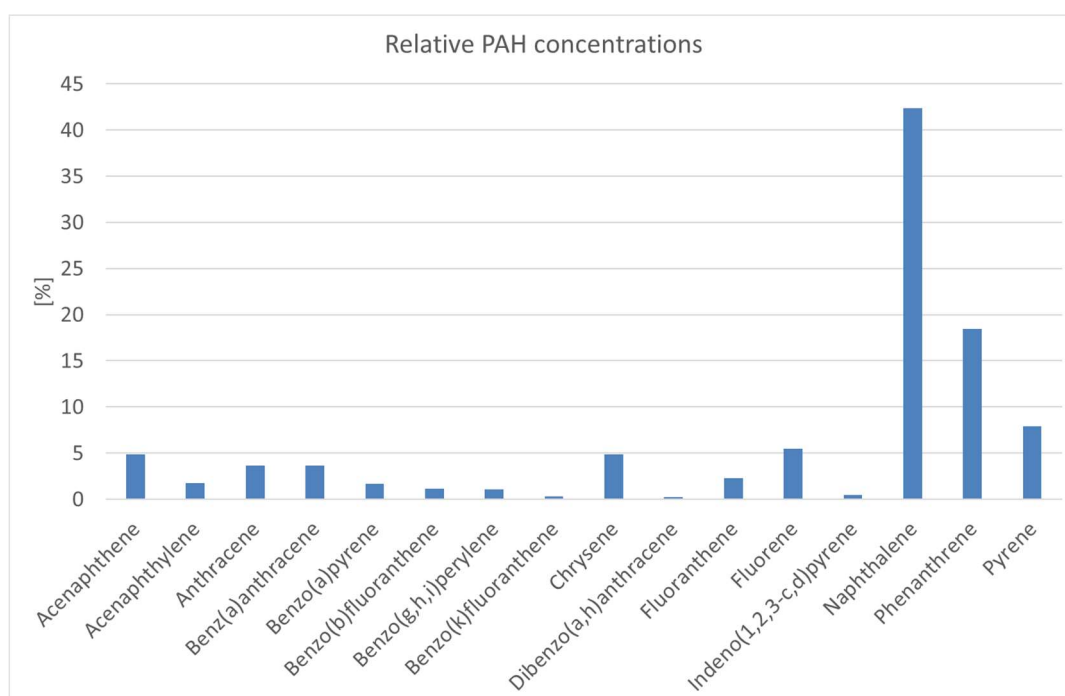
Pyrogenic PAH either occurred in the gas phase (2-3 ring PAH) or attached to particles like soot (4-6 ring PAH) (Neff, 2002). There were many studies specifying the 16 EPA PAH, but there was little information on their derivatives like alkylated PAH, although they were more abundant in the environmental compartments, persistent, and sometimes more toxic (Casala, *et al.*, 2014). The results on PAH concentrations in EGCS washwaters published as surveyed reports of EGCS trials had not been peer reviewed. A number of these reports were submitted to the Task Team on EGCS in the series of background documents, whereas available evidence on PAH concentrations and their thorough chemical characterization in EGCS washwaters, published in peer reviewed scientific literature was very limited or not found.

The concentrations of PAHs in washwater for open loop EGCS were at the range of ppb levels ( $\mu\text{g L}^{-1}$ ) as summarized from selected number of surveyed studies (Table 9). The summary table of all examined results was given in Appendix, Table A2. These reported concentrations were not normalized on washwater flow rate (except for EGCSA, 2018), but were only for non-substituted parent PAHs and might be 2 to 3 times higher if alkylated homologues would be added (Guigue, *et al.*, 2014, BSH, 2019).

Based on all evaluated background documents, an average relative distribution of the (dissolved) parent 16 PAH in the EGCS washwater was calculated. The data were presented in the Appendix, Table A2, references were in the footnote of the table. Figure 10 showed the averaged PAH concentrations of all measurement campaigns. The dominant PAH were naphthalene (42%), phenanthrene (18%) and pyrene (8%) in EGCS effluents. Their average (maximum) concentrations were 1.8 (14), 1.2 (6.1) and 0.17 (2.6)  $\mu\text{g L}^{-1}$ , respectively.

**Table 9. Ranges of summed concentrations of parent  $\Sigma$  PAH<sub>EPA-16</sub> and phenanthrene in EGCS washwater (in open loop operation); concentrations adjusted to 45 m<sup>3</sup> (MWh)<sup>-1</sup> washwater flow rate only for EGCSA (2018). BSH data was only from a preliminary report, the full report was expected in 2019**

Source	PPR 6/INF.20, BSH, 2019	MEPC 73/INF5, EGCSA, 2018	Magnusson, <i>et al.</i> , 2018	Hufnagl, <i>et al.</i> , 2005
$\Sigma$ PAH <sub>EPA-16</sub> ( $\mu\text{g L}^{-1}$ )	1.6 – 19	0.5 – 24	22	12. – 20
Phenanthrene ( $\mu\text{g L}^{-1}$ )	0.7 – 2.9	0.08 – 6.1	10	5.1 – 8.2



**Figure 10. Relative 16 EPA PAH based on all measurement campaigns**

Almost none of the PAH determinations in washwaters was done on separate particulate and dissolved fractions, whereas a great portion of the total PAHs emitted by diesel engines were bound to soot particles (Hufnagl, *et al.*, 2005; Buhaug, *et al.*, 2006). A significant fraction of particulate PAHs in washwaters was thus not determined by many of the surveyed studies. This was a gap to be taken into account in the further appreciation of EGCS PAH risks. Most of the results systematically showed prevalence of lower molecular weight PAHs (2-3 rings) in the EGCS washwaters, indicating predominance of dissolved PAH compounds. These results suggested again that the particulate PAH fraction (with higher molecular weight) may be underestimated.

The low molecular weight (LMW) PAHs with 2 to 3 rings ( $\text{Log } K_{ow} < 4$ ) were usually dispersed in the water column and were less persistent in the environment because of their high volatility. High molecular weight (HMW) PAHs (e.g., 3 rings and more) were more hydrophobic ( $\text{Log } K_{ow} > 4$ ) and therefore, less water soluble and more persistent (Rice, *et al.*, 2001). HMW PAH tended to adsorb to particles and soot that were dispersed in the scrubber washwater and would be discharged, unless filtered.

PAH solubility in freshwater decreased with increasing PAH molecular weight (Appendix, Table A1). The solubility of some alkyl-PAHs was greater than that of the parent compounds and decreased with an increase of alkylation. With increasing seawater salinity, the solubility of PAH tended to decrease with decreasing water temperature (Neff, 2002). At all salinities and temperatures, anthracene was less soluble than its isomer, phenanthrene. These physical properties of PAH affected their bioavailability and toxicity to freshwater and marine organisms (Neff, *et al.*, 2004).

As already stated above, the review of the Task Team revealed significant issues with methodological protocols and on how the measurements were actually organized and performed. This was important with analysis of PAH concentrations in EGCS washwaters and especially when their potential environmental impacts should be assessed (and not merely examination of EGCS compliance performance). Not normalized conditions of test studies, for instance, engine power and operating loads, washwater flows, resulted in different PAH emissions in exhaust gases and consequently in discharged EGCS effluent washwaters.

### Other PAH

The 16 EPA PAH compounds encompassed only parent PAHs. However, there were many other potentially toxic polyaromatic compounds (PACs). In environmental toxicity studies where fossil material was involved, adherence to the EPA list underestimated the toxic potential of the analyzed material (Andersson and Achten, 2015). Three groups of polycyclic aromatic compounds (PAC) were missing: larger PAHs, alkylated PACs, and compounds containing heteroatoms (mainly N, O, S). All of the parent PAH occur in single- or multi-alkylated forms. The NOS-heterocyclic compounds contained a heteroatom in their internal structure, e.g., like dibenzofurans including a central oxygen atom.

Table A1 in the Appendix clearly showed the increasing toxicity with increasing number of alkylation. For naphthalene the acute and chronic toxicities increased by a factor of 116 and 237 times, respectively from the parent naphthalene to the 4-fold methylated naphthalene (C4-naphthalene). The same trend could be found for phenanthrene, where the toxicity of C4-phenanthrene was even higher than that of benzo[a]pyrene. The high aromatic content of HFO also caused a high concentration of PAH derivatives. For almost all PAHs, the highest concentrations of alkylated PAH were present, e.g. the C3-alkylated naphthalene derivatives.

In the BSH measurement campaign (PPR 6 INF.20) also two methylated naphthalenes were analyzed and found in significant concentrations ( $2 - 19 \mu\text{g L}^{-1}$ ) for 1-methyl and 2-methyl naphthalene, respectively, in OL scrubber discharge water. The total dissolved 16 EPA PAH were  $4.7 \mu\text{g L}^{-1}$  on average (min.  $1.2$ ; max  $13 \mu\text{g L}^{-1}$ ). This result showed that the detection of only two methylated PAH already exceeded the concentration of the parent PAH by a factor of 4.5. Magnussen, *et al.* (2018) measured a variety of alkylated PACs in scrubber washwater, but only in closed loop systems. These included alkylated naphthalenes, alkylated phenanthrenes and alkylated dibenzothiophenes. The

concentrations of alkylated dibenzothiophenes ( $3,000 \mu\text{g L}^{-1}$ ), phenanthrenes and naphthalenes ( $9,200 \mu\text{g L}^{-1}$ ) in CL EGCS in sum exceeded the measured total 16 EPA PAH ( $780 \mu\text{g L}^{-1}$ ) by a factor of more than 15 (Table 10) and these PACs were only a small selection of alkylated PAH. It was not possible to extrapolate these results to OL EGCS, but due to the solubilities and occurrence of these compounds in the exhaust gas, one could only assume a similar ratio.

**Table 10** Concentrations of alkylated dibenzothiophenes, phenanthrenes and naphthalenes in closed loop EGCS in the BOTU feed water in comparison to total 16 EPA PAH ( $\mu\text{g L}^{-1}$ ). (Magnussen, *et al.*, 2018)

Compounds	Concentration in BOTU feed water ( $\mu\text{g L}^{-1}$ ) CL
Total 16 EPA PAH	780
Dibenzothiophene	150
Methyldibenzothiophene	700
2,3-Methyldibenzothiophene	330
Trimethyldibenzothiophene	790
4-Methyldibenzothiophene	280
Total dibenzothiophenes	3000
Methylphenanthrene	3300
Dimethylphenanthrene	4000
Trimethylphenanthrene	1500
Dimethylnaphthalene	320
Trimethylnaphthalene	120

In many of the measurement campaigns, there was a lack of information regarding the EGCS operating conditions during washwater sampling, especially those concerning removal of particulate matter (open or closed loop, use of decantation tank and multi-cyclone treatment, etc.), which would have a direct influence on particulate PAH levels in washwaters. In addition, the extraction recovery for the analysis of particulate PAH from the bulk seawater, that was not a filtered water sample (often used in the trials), would be lower or minimal – *i.e.* significantly less efficient than extraction of lower molecular weight dissolved compounds. All these factors may lead to the apparently higher level of lower molecular weight PAH in EGCS washwaters. The question was also what was the fate of higher molecular weight PAH produced in large quantities (mainly adsorbed on soot particles) by heavy fuel oil combustion? Indeed, a high concentration of PAH in the hundreds of  $\text{mg kg}^{-1}$  in EGCS sludge wastes was determined (Kjølholt, *et al.*, 2012). These findings are in contrast to  $\mu\text{g L}^{-1}$  level concentrations of PAH determined by surveyed studies in washwater effluents from open loop EGCS. It was also possible that a significant fraction of particulate PAH was not removed by EGCS and remained emitted to the atmosphere. Data on a better mass balance budget of PAH emissions from ships with EGCS were needed. Such data was also required for the careful assessment of environmental threats and health hazards related with discharge of contaminants by EGCS.

In the present discussion the Task Team on EGCS would draw attention to the factual difficulties related to the determination of PAH concentrations in the EGCS washwater effluents. Determining chemicals in marine matrices posed quite a challenge and an entire chain of processes, from sampling at sea through the conservation, transport and preparation steps that led to the final analysis, was considerably demanding and required trained personnel. Such standardized controlled protocols for sampling and PAH analysis could not be routinely performed in real-world ship activities.

In short, the Task Team on EGCS concluded that given the PAH determination in EGCS washwaters, together with the lack of compulsory information (test conditions, design of EGCS, sampling protocols etc.) and the quality of obtained data, limit the comparability of the results and the representativeness of the surveyed studies. These factors accounted for uncertain results in this area.

#### PAH discharge-loads with EGCS washwaters

The appraisals of contaminant loads from the discharge of EGCS washwater effluents were probably even more relevant than their concentrations for their environmental risk assessments. This was raised in several surveyed studies (Endres, *et al.*, 2018, BSH, 2018), especially for persistent and long-term accumulating substances. The accumulation in certain areas with high shipping activities may potentially lead to the exceedance of contaminant environmental quality standards. The significance of the contaminant loads may be also appreciated through comparison with other contributing contaminant sources.

**Table 11. Total PAH discharged per MW and year, based on the evaluated measurement campaigns**

PAH g (MWy) <sup>-1</sup>	Min	Max	Average
Acenaphthene	0.0	630	61
Acenaphthylene	0.0	230	24
Anthracene	0.0	470	24
Benzo(a)anthracene	2.4	470	27
Benzo(a)pyrene	3.9	220	21
Benzo(b)fluoranthene	3.9	150	18
Benzo(g,h,i)perylene	3.9	140	14
Benzo(k)fluoranthene	0.0	36	7.3
Chrysene	0.0	630	35
Dibenzo(a,h)anthracene	2.4	32	6.7
Fluoranthene	0.0	300	35
Fluorene	0.0	710	130
Indeno(1.2.3-c.d)pyrene	0.0	55	8.3
Naphthalene	2.4	5500	720
Phenanthrene	2.4	2400	450
Pyrene	2.8	1000	66

The BSH preliminary report (2019) provided an assessment of PAH (and other contaminants) discharge-loads from open loop systems (and closed loop) to the Baltic Sea, North Sea and English Channel. The estimations were performed for EGCS “current status” (CSS) and “maximum installation” (MIS) scenarios, taking into account shipping activities, washwater water flow rates, and actual contaminant concentrations in washwater effluents. The range of total annual loads of  $\sum\text{PAH}_{\text{EPA16}}$  to these regional Seas was estimated to be between 0.5 and 6.0 tons for current state situation and between 5.0 to 60 tons for the maximum installation scenario. These inputs were compared to the whole river Rhine basin PAH annual loads to the North Sea, that were estimated to be between 6.5 – 7 tons. Therefore, the current and potential loads of PAH from EGCS were recognized to be a significant cause of concern regarding chemical pollution of these regional Seas (Baltic, North Sea and English Channel).

To calculate the mass load per installed MW of the ship’s engine, the minimum, maximum and average concentrations of the PAH had been calculated as loads in  $\text{g MW}^{-1}$  per year, based on a washwater flowrate of  $45 \text{ m}^3 (\text{MWh})^{-1}$ . It should be noted that some installations used a higher flowrate of up to  $140 \text{ m}^3 (\text{MWh})^{-1}$  (PPR 6/INF.20). Therefore, the total masses discharged per year could be a factor of 2-3 times higher. These values had to be multiplied with installed power (MW) of an individual ship to calculate the total emissions per ship. For some PAH the total mass load emitted by one ship per year will then easily be in the kg range as shown in Table 11.

The impact of EGCS installation on shipping emissions of PAHs should also be recognized in the global perspective. The global annual PAH shipping emissions were estimated to be at 10700 tons in 2014 (Shen, *et al.*, 2013 and personal communication with prof. S. Tao). These global emissions had increased by a factor of over 2.5 times since 1960 (Figure 11). Comparable PAH emissions were here estimated to be in the range of 6500 – 15,500  $\text{t y}^{-1}$  on the basis of PAH emission factors and global shipping fuel consumption in 2015. These values indicated also that in 2014 – 2015 shipping would thus account for 1.3 to 3.1% of the global (all sources) of PAH emissions (Shen, *et al.*, 2013. González-Gaya, *et al.*, 2016). The consistent estimates by different methods and models suggested that PAH emission factors could be used for the determination of different scenarios for PAH loads discharged with EGCS washwaters, providing useful data for their environmental risk assessments.

On the other hand, it should also be noted that the absolute amounts of emitted PAH by the shipping sector would not change with the introduction of EGCS, unless other factors would change, such as use of modern engines, different fuels (distillates, LNG), lower speed, etc. The large-scale introduction of EGCS on ships would change the spatial distribution of contaminant loads as well as the routes of their entry to the environment. Higher amounts of PAH (and other contaminants) would be directly introduced by discharge of EGCS washwater effluents into shallow coastal waters, regional semi-enclosed seas and in bays and ports. There was a rather wide consensus in the literature that discharge of contaminants by ship EGCS was a main cause of concern with respect to possible short and long term impacts on the environment, especially in the areas of high shipping activities, with low degree of flushing and under high existing pollution levels and anthropic pressures (Endres, *et al.*, 2018). This was the justification for further scientific investigations and it warranted also careful revisions of policy regulations and decisions.

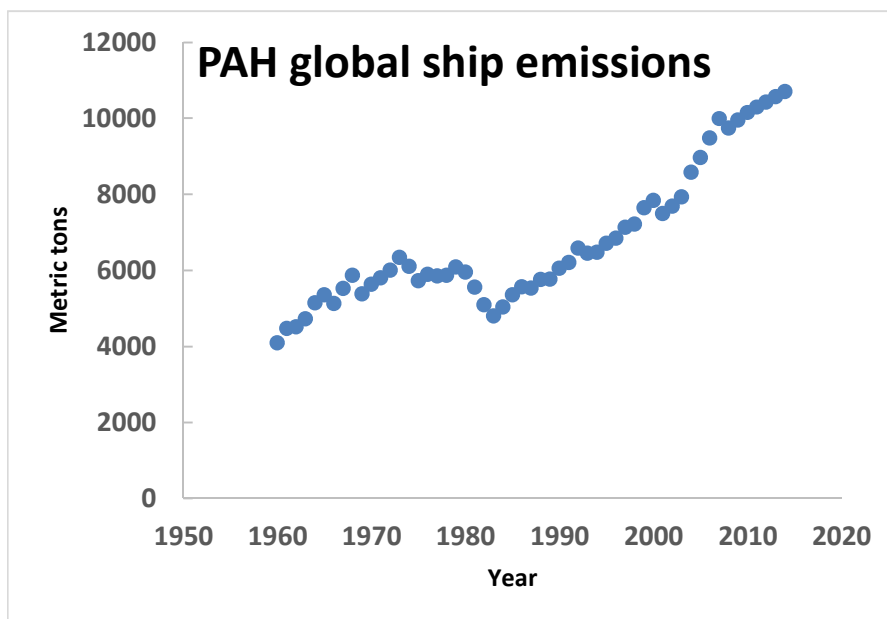


Figure 11. Temporal trend for global PAH shipping emissions in metric tons since 1960 (Shen *et al.*, 2013 and personal communication with prof. S. Tao)

The Task Team concluded that in different papers different values were reported on the trend of future PAH emissions from ships. The Task Team would prefer to have information available from established sources based on sound data. This was a clear case that the Task Team would have a preference for the data presented in PPR 6/INF.20, but these values may also be prone to uncertainties.



#### PAH guideline limit discharge concentration

IMO Resolution MEPC.259(68) provided a guideline limit defined as phenanthrene equivalence ( $PAH_{phe}$ ) for PAHs concentrations in EGCS washwater effluents. The limit concentration determined before any dilution for open loop operation, at the outlet of the EGCS, was related to EGCS washwater flow rates. The method for  $PAH_{phe}$  determination was defined as an optical measurement with UV or fluorescence detection by means of an online-sensor installed onboard, allowing continuous monitoring of the dissolved PAH discharge. This  $PAH_{phe}$  limit should be revised because it was not well defined and its level of protection was questionable. The optical measurement of PAH was introduced to have a permanent control of PAH and indirectly oil discharges during operation of an EGCS. similar to an oil in water monitoring device for bilge water alarms. However, its operational application was frequently failing whereas GC-MS analysis of PAH was demanding and required trained technicians and could only be done in a laboratory after discrete samplings. Generally, the examined results determined by GC-MS met the IMO criteria for PAH discharge in OL-EGCS washwater, whereas the rarely presented optically determined data showed frequent detection failures or not comparable results with the GC-MS determination (BSH, 2019).

The historical development of phenanthrene equivalence ( $PAH_{phe}$ ) criterion for PAH might have been well intended, but the rationale for this was not well established. There was no clear definition stated and in practice, its application could introduce many flaws and misunderstandings. Historically, the PAH limit was derived relative to several oil discharge limits and its concentration of



PAH in oil. To relate this limit to PAH toxicity, the toxic equivalence factors (Nisbet and LaGoy, 1992, Fisher, *et al.*, 2011) of individual compounds should be used to express the results. Such a concept was mainly proposed for PAH health hazard quantification. This required also the use of analytical techniques allowing individual PAH compounds determination. On the other hand, if one would like to express the concentration of a “total” PAH (*i.e.* for a defined number of individual compounds, like 16 EPA-PAHs) as an equivalent of one compound determination, like phenanthrene, a conversion factor should be established and used. Such a concept was barely proposed and developed in the scientific literature – mainly because of impracticalities, such as, for instance, changing relative composition of PAH deriving from different PAH sources. For the IMO guideline, such conversion would be even further complicated because phenanthrene concentrations measured by an optical technique should be eventually converted to equivalent 16 EPA PAHs concentrations determined by GC-MS or other analytical techniques. This would be very unreliable.

In practice, almost all surveyed studies compared PAH<sub>phe</sub>-equivalency value limit to summed concentrations of PAH determined by GC-MS analysis. This was not correct. Thus, the terminology should be revised – until any PAH “equivalency” was defined and appropriate conversion factor was determined.

The onboard installation of EGCS systems was accompanied now by optical sensors installations for PAH continuous monitoring. Certain PAH fluorescence sensor manufacturers claimed that a conversion factor of 6 should be applied (multiplication, causing underestimation) to convert from optical phenanthrene determination to 16 EPA-PAHs. This was somewhat confusing with scientific literature data concerning the development and application of optical sensors, like MiniFluo-UV, installed on the ocean on-going underwater gliders for continuous detection of selected PAH (Cyr, *et al.*, 2019). The authors showed that the MiniFluo sensor used in this study agrees within one order of magnitude with the concentration determined by GC-MS (overestimation by a factor 7 on average (Cyr, *et al.*, 2019). These apparently contradictory results for the conversion of optical phenanthrene concentrations to 16 EPA PAH would require further investigation, because the online measurement of PAH<sub>phe</sub>-equivalents could lead to imminent exceedances or underestimations of PAH discharges from EGCS.

Furthermore, the optical measurement was subject to the strong interferences (quenching, scattering of emitted light, etc.), which may be related, for instance, to changing suspended particulate matter and organic matter concentrations. This was particularly important in industrial applications, such as in EGCS. Additionally, one of the important disadvantages was also the fact that optical measurement would totally overlook particulate PAH present in washwaters, as this was technically not feasible. This could only be done by frequent sampling followed by laboratory GC-MS analysis.

A simple calculation showed that in a maximized scenario of all ships equipped with OL-EGCS, taking in account the 2015 HFO world fuel consumption by the shipping sector (Smith, *et al.*, 2015) and known emission factors for PAH (Cooper and Peterson, 1996, Cooper and Gustaffson, 2004) and the 50 µg L<sup>-1</sup> PAH limit and assuming a washwater discharge flow rate of 45m<sup>3</sup> (MWh)<sup>-1</sup>, the global emission of PAH would be 5.9 Mt. This amount would be about 10 times higher than the world-wide PAHs emissions from all sources (all biomass, all fossil fuel combustion (Shen, *et al.*, 2013, González-Gaya, *et al.*, 2016). That was considered unrealistic, but clearly indicating that, in fact, the actual IMO PAH guideline limit did not provide any restriction for PAH discharges.



### 6.11. Conclusions and Recommendations on Chemicals

The Task Team on EGCS formulated the following conclusions and recommendations with respect to the Chapter on chemicals:

1. Well-documented and established ranges of contaminant ambient concentrations in the open sea, coastal waters and specific areas of bays, estuaries, harbours and ports were recommended for their use in the risk assessment efforts in areas where effluents of OL EGCS was discharged into the ambient environment;
2. Extensive chemical characterization of substances in EGCS washwater effluents, including toxic and genotoxic species (such as heterocycle and alkyl-PAH) was recommended and needed for better appraisal of contaminants emissions through EGCS and their environmental threats for marine life. **In this context, EGCS washwater measurements should also include monitoring of particulate and dissolved contaminant concentrations, especially for PAHs, trace metals and all other relevant chemicals.** The Task Team recognized also the necessary improvements and need for harmonized procedures in terms of washwaters sampling and analysis to ensure better comparability in different data sets. With respect to these points the Task Team recognized also that the **current available data and practice was insufficient;**
3. Besides improving chemical emissions characterization, the development of spatial and temporal in situ measurements of contaminants along shipping lanes and in all areas of high volume of ship activities was considered important to gain a more comprehensive understanding of EGCS related pollution distribution and the environmental consequences of large-scale uses of EGCS. In respect to this, the ship emissions monitoring network may be further developed. Such developments were recommended by the Task Team, considering also an enhancement of cooperation with independent academic institutions;
4. **In terms of total amounts of contaminant discharges through EGCS, it appeared that large scale uses of these systems may lead the deterioration of environmental status, especially in the ecologically vulnerable and sensitive areas such as coastal waters, semi-enclosed seas and also in ports and harbours.** In respect of this, the Task Team recommended further assessment and appraisal of contaminant loads discharged with EGCS washwater effluents. The development of alternative effective methods and protocols based on combining ship traffic data with contaminants emission factors were fundamental for the improvement of EGCS contaminants loads monitoring and assessments. The spatial representation of related discharges of pollutants would require further modeling efforts combining ship traffic data with ship emission factors and in-situ measurements of contaminants. The amounts of EGCS contaminants should also be further taken into account in the appreciation of their environmental risk assessment approaches and methods;
5. The Task Team recommended the revision of the existing PAH<sub>phe</sub> guideline limit, including revision of its definition, methodology of determination and its limit level. The development of a new limit guideline for selected trace metal concentrations in EGCS washwater effluents was also recommended;
6. Considering the complexity of the topic and its importance for the marine environment, the Task Team recognized a great need for more interdisciplinary research development to

better predict ship emission and the use of EGCS and their chemicals impacts on acidification, eutrophication and related climate feed backs.

## 7. POSSIBILITIES FOR RISK ASSESSMENT, INCLUDING ERA AND HHRA

### 7.1. Exposure Assessment

#### 7.1.1 Introduction

The exposure assessment was considered an essential part of a risk assessment (see section 7.3). It identified the Predicted Environmental Concentration (PEC) of the relevant chemical contaminants for which the risk assessment was carried out. The PEC could be identified with different methods, varying from rather simple, and easy to perform calculation based on worst case assumptions and conservative fate to complicated 3-D simulation models with more realistic assumptions and natural behavior of substances with respect to degradation and sorption. Some features of the different types of modelling will be dealt with in the next sections. Finally, a short overview was presented of the available models with similar possibilities as the Task Team considered relevant for the evaluation of the potential risk assessment for EGCS.

Contaminants in the washwater discharge from EGCS were of potential environmental concern in open waterbodies with heavy ship traffic such as straits, shipping channels and canals, and/or enclosed or semi-enclosed waterbodies with weak flushing such as bays and harbours. Different types of models were appropriate for each. These two domains were typically analyzed using near field and far field models respectively. There was also interest in predicting concentrations at very short distances from the ship for which initial mixing models could be used. In each case the goal was to predict concentrations that could be compared against threshold concentrations determined from (marine) ecotoxicological studies. The models could be used to predict the concentrations of specific contaminants (*e.g.*, pH, a particular trace metal or a PAH). Models had also been used to predict the dilution of the discharge which could be used in connection with whole effluent toxicity testing.

#### 7.1.2 Initial Mixing Models

Washwater was discharged through the ship's hull, below the waterline, a few meters from the propeller. The region within a few meters of the discharge received rapid mixing because of the washwater discharge momentum, the ship's wake and propeller wash. These same factors contributed to making the flow complex and 3-dimensional, such that analysis required use of either direct measurements of a tracer, or computational fluid dynamics modeling (CFD). An example of CFD modeling, applied to EGCS, was provided by MEPC 74/INF.24 (MEPC, 2019c) where, in addition to near source concentrations, the distributions of turbulent viscosity and diffusivity were generated. Because initial mixing was a function mainly of ship and discharge properties, such mixing was independent of the site (waterbody) so a single CFD analysis could be applied to all ships and discharge scenarios of a given type.

The Task Team noted that such initial mixing models had been used to predict near source pH and in particular to confirm compliance with the regulation of pH exceeding 6.5 at a distance of 4 m from the source. Over this distance, mixing alone did not generate sufficient dilution to achieve a pH of 6.5; hence washwater treatment (buffering) must be employed.

### 7.1.3 Near Field Models

The Task Team considered the near field to extend from the initial mixing region described above to a distance of several kilometers behind the ship. MEPC III/7 (MEPC, 1975) used results of earlier field campaigns in the US, Netherlands and Norway and a large scale lab study to derive an empirical formula for the dilution of conservative contaminants in the wake of a moving ship. Such contaminants were found to be directly proportional to the contaminant mass loading, and proportional to the ship length and velocity and the distance behind the ship, to powers of -1.6, -1.0, and -0.4. Thus, the most influential independent variable was the ship length. Mixing was also shown to depend on the location, orientation and the number of orifices employed (*e.g.*, orifices on either side of the stern). Later experimental studies and analyses conducted during the 1980s (Lewis, 1985; Byrne, *et al.*, 1988; Lewis and Riddle, 1989, and Delvigne, 1987) found that the MEPC III.7 formula underestimated observed dilutions (overestimated peak concentrations) by a factor of three or more, rendering its use quite conservative.

Rather than relying directly on field measurements, a near field mathematical model, calibrated to field observations could also be used. Applied to an open waterbody, such a model would start with a description of the steady-state 3-D concentration distribution  $c(x,y,z)$  resulting from a continuous effluent discharge from a single ship. Important variables here were the lateral and vertical turbulent diffusivities which should be calibrated to observed concentrations in the field and would reflect characteristics such as ship length, speed and position  $x$ . For relatively short distances from the discharge, the results of an initial mixing analysis could be used to generate “starting” conditions, though the influence of initial mixing would die off at longer distances (order of a kilometer or more) from the ship. In the frame of reference of a moving ship,  $c(x,y,z)$  could be determined by solving an advection-diffusion equation. In congested waterways, there were usually multiple vessels, often of different type (tanker, cargo, etc.), each contributing overlapping contaminant plumes. See Figure 12.

In general, near field concentrations in the  $x$ ,  $y$ , and  $z$  directions would depend on the frequency of vessel arrivals (often obtainable by AIS), their spacing, type, and washwater volumetric flow rate, initial mixing (as described above), ambient current speed and lateral and vertical diffusivities, and waterway dimensions (width, depth and length). Contaminants may also be lost due to processes such as biodegradation, evaporation or sorption / settling, but the associated time constants for these processes may be larger than the time scales in the near field (hours), in which case the contaminants could be treated as conservative.

An example of a near field model application was presented in MEPC 74/INF.24 (MEPC, 2019c) which analyzed concentrations aft of a line of vessels traveling in a shipping lane entering Tokyo Bay. The study concluded that a minimum dilution of at least 5000 in 60 s would be achievable, and independently noted that the dilution ratio of > 5000 would exceed the predicted no-effects dilution (PNED) of 5000 calculated from their results of whole effluent toxicity tests. The study included several conservative (worst case) assumptions including peak rates of vessel daily peak congestion, and the assumption that vessel orientation was precisely “line astern”. The latter assumption would produce maximum concentrations when the vessel wakes are superimposed (see Figure 12); a slight off-set in lateral vessel orientation, or a mild lateral current, would cause the individual wakes to separate, leading to smaller peak concentrations (higher dilutions), albeit over larger areas. Also, the

predicted no-effects dilution of 5000 was quite uncertain and possibly conservative, as it included an assessment factor of 1000 relating the minimum dilution of 5 (20% of discharge concentration) in which acute effects were observed in the laboratory, extrapolated down to a predicted no-effects dilution (see 7.2.4). On the non-conservative side, the model used spatially uniform, isotropic turbulent diffusivities derived from the initial mixing model described above. The initial mixing zone was quite turbulent and, more likely, the diffusivity would decrease with distance as predicted for the wake behind a moving ship (Lewis, 1985; Byrne, *et al.*, 1988; Lewis and Riddle, 1989, Delvigne, 1987 and Tennekes and Lumley, 1972). Overall, however, the analysis was conservative and through judicious choice of input parameters, the methodology could be applied to other waterbodies.

An important consideration would be the exposure time experienced by an organism subject to the computed near field concentration distribution, versus the duration under which ecotoxicological studies were conducted (*e.g.*, exposure duration of 72 or 96 hours). To evaluate the impact based on 24 or 96 hour toxicological data may be conservative for motile organisms (*e.g.*, pelagic fish) which may pass quickly through the washwater plume; conversely, such an assessment may be non-conservative for sessile organisms (*e.g.*, demersal fish or benthic infauna).

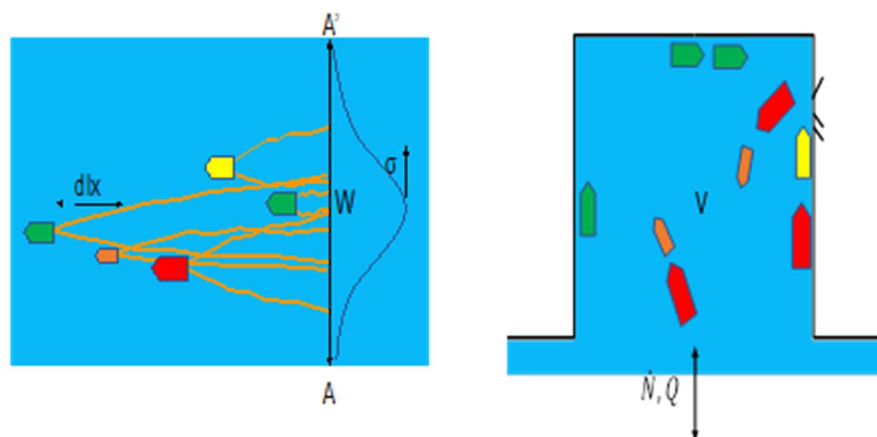


Figure 12. Schematics of (left) the near-field and (right) the far-field model in top-view. Vessels in different colors denote different ship types (thus engine load capacities). (Left) vessels cross a line AA' normal to a shipping lane of width W and follow a Gaussian distribution with a mean crossing location in the middle of the lane and a lateral spread with a standard deviation of  $\sigma$ . (Right) vessels move and berth inside an enclosed waterbody (of volume V) that receives on average number of vessels per time  $N$  and has a flushing rate of Q

#### 7.1.4 Far Field Models

Far field models could range in complexity, but the simplest would be a well-mixed box model in which steady-state and spatially averaged concentrations were computed as a function of the frequency of vessel arrivals, their type, the times they would spend in transit and at berth, the waterbody volume and hydraulic residence time, and the appropriate loss rate(s). Unlike the near field, the time scales in the far field (*e.g.*, given by residence times) could be days to weeks, during which time loss mechanisms such as biodegradation, sorption/deposition could be very important in reducing concentrations. The time that organisms were exposed to far field concentrations was also

important. Under the assumption that far field concentrations were both steady and spatially well-mixed, the exposure time of an organism that would reside permanently in the waterbody would exceed typical exposure times in laboratory ecotoxicological tests (e.g., 96 hours or 28 days) rendering the calculations non-conservative. However, a more realistic calculation should consider organism movement into and out of the waterbody.

#### 7.1.5 Available models

Ports and harbours differed in their geometry, and thus a model study conducted at one site may not be suitable for other sites. Thus, different and more generalized analyzes were required. Quite recently, two papers had been published where the GESAMP-BWWG Standard Harbour, as used in the ballast water management systems (BWMS) evaluation had been adapted to 2 commercial harbours of Hamburg in Germany and Koper in Slovenia (David, *et al.*, 2018 and Dock, *et al.*, 2019). The results showed that a model with success could be adapted to other situations. In these papers, the model MAMPEC had been used to perform the calculations. The first paper dealt with the environmental risk assessment and the second the risks for the general public. Below the Task Team discussed several models that had been, or could be, used to analyze EGCS effluent concentrations in ports and harbours.

##### 7.1.5.1. MAMPEC-BW

MAMPEC (Marine Antifoulant Model to Predict Environmental Concentrations) was a steady-state, 2D-integrated hydrodynamic and chemical fate model (Hattum, *et al.*, 2016). The original model was developed for the evaluation of anti-foulant paints in the framework of the biocides regulation in the EU. The MAMPEC-BW model was adapted for exposure assessment of chemicals discharged from ballast water management systems and had the same features as MAMPEC. Before 2011, MAMPEC was used on a voluntary basis by a number of applicants (summarized in Zipperle, *et al.*, 2011). On the request of the GESAMP-BWWG and IMO, a special standardized version of MAMPEC-BW for ballast water was created (MAMPEC-BW v3.0) in 2011, with a dedicated environment, a compound and an emission scenario. Currently the model was part of the evaluation methodology for Basic and Final Approval by the MEPC of IMO, based on the recommendations of the GESAMP-BWWG (MEPC, 2017).

Perhaps the most significant factor affecting predicted concentrations was the exchange rate between harbour and the surrounding environment (about 30% in the GESAMP-BWWG Standard Harbour scenario, MEPC, 2017). This was expressed as an exchange flow defined as the volume of water released over a tidal period. Six exchange mechanisms were considered--tidal exchange, horizontal circulation, vertical density exchange, other non-tidal exchange, flushing due to external flows, and wind-driven exchange—with the first three generally being the most important. Different approaches were used to estimate these six exchange flows ranging from simple analytical expressions (tidal exchange) to synthesis of model 3-D hydrodynamic runs (wind driven exchange). Each of the flows was then used to compute a longitudinal dispersion coefficient that produced the same exchange flow as given by the exchange flows themselves. The sum of the 6 dispersion coefficients was used to drive the 2-D transport model based on a given pollutant loading (magnitude and distribution) and the fate process contained in the model DELWAQ.

The MAMPEC model used an entry screen to define the compound. It had incorporated a database of about 40 chemicals that were defined as the most relevant disinfection by-products (DBP) in ballast water management systems (BWMS). The standard model harbour, GESAMP-BWWG Standard Harbour, was defined as a worst-case scenario with specified dimensions with a fixed exchange rate between the harbour and the surrounding area. Finally, there was the emission scenario, where the highest concentration in ballast water of the DBP under consideration was discharged in the harbour on a daily basis and with a pre-determined default value for the amount of water discharged. The output of the model was the yearly averaged concentration in the water and sediment phases. Sediment concentrations could also be computed for up to 10 years. In this way, also potential accumulation of the DBPs may be analyzed. The GESAMP-BWWG used in all cases a worst-case scenario, *e.g.* for the different DBPs no degradation was assumed. In relevant cases, if concentrations were determined in excess of the ecotoxicological reference values (Predicted No Effect Concentration, PNEC), degradation was taken into account as a second Tier. The Task Team on EGCS was of the opinion that the MAMPEC model may be used for the risk assessment of components of the effluent of EGCS, using various emission scenarios (*e.g.* ratio of the ships that used either open loop or closed loop mode). Also, the discharge of the BOTU may be analyzed with the model after thorough evaluation of the adjustments needed. One of the main differences with ballast water, was that EGCS emissions from ships would take place during the whole journey of the ship, much of the time being in open waters. For this, MAMPEC had the possibility to calculate the concentration also in a shipping lane. It had to be seen how this feature could be incorporated in a potential risk assessment approach for EGCS effluents. Schematics of the MAMPEC model applied to both a harbour and a shipping lane were shown in Figure 13.

MAMPEC was recently applied to four generic harbours visited by cruise ships (Faber, *et al.*, 2019). Concentrations of 11 trace metals and 16 PAHs in the water column and sediment were predicted. The predictions all fell well below applicable water quality and sediment quality standards. The authors claim that their results were conservative, but there are several aspects of their study which may not be conservative. The hydrodynamics varied significantly among the four sites with exchange rates (net volume of water exchanged at the mouth divided by the tidal period) ranging from  $\sim 1000 \text{ m}^3 \text{ s}^{-1}$  for the “Standard OECD-EU Commercial Harbour” down to  $\sim 30 \text{ m}^3 \text{ s}^{-1}$  for the “Baltic Commercial Port”, having the same horizontal area and depth. This was a huge difference, suggesting the need for additional validation. Dividing the exchange flow rate by the wastewater flow rate of  $\sim 0.1 \text{ m}^3 \text{ s}^{-1}$  ( $45 \text{ T (MWh)}^{-1}$  from Ulpre and Eames (2014) times a cumulative loading from all ships in the harbour of 8 MW) would give an average dilution varying from  $\sim 10,000$  to  $\sim 300$ . The higher value was comparable to the predicted no-effects dilution of 5000 based on the WET tests used by MEPC 74/INF24 (MEPC, 2019c), while the lower value was  $\sim 30$  times lower implying a potentially large impact. Yet, as mentioned above, the analysis of Faber, *et al.* (2019) based on individual pollutants showed all PECs were well below the relevant PNECs. This highlighted the substantial difference between analyses based on WET with a large assessment factor, and studies based on individual constituents.

MEPC 74/INF.24 (MEPC, 2019c) described another application of MAMPEC: to predict concentrations of nitrate, phosphate and pH in three bays in Japan rather than compare predicted concentrations against a national environmental standard depending on the category of sea area (*e.g.* some part of the bay was classified as industrial area, so that the severe standard for conserving fishery resources was not applicable). Also, the authors compared the predicted increase in concentrations to existing

background concentrations. The predicted increases after 10 years were all less than 1% of the existing background concentration suggesting that the impact would be acceptable. However, details of the study were scant. And as there was no national environmental standard for PAH in Japan, the model was applied for the phenanthrene equivalents of PAHs.

In summary, MAMPEC had an elegant structure that allowed application to a range of different waterbodies, with different pollution loadings. The Task Team was of the opinion it could be a promising model for future EGCS applications, but that it should be further validated, perhaps through a carefully designed field study focusing on EGCS loading.



**Figure 13.** Layout of the commercial harbour and the shipping lane in MAMPEC (Boon, *et al.*, 2008)

#### 7.1.5.2. STEAM3

In Finland, the model STEAM was developed. STEAM stands for Ship Traffic Emission Assessment Model. The first model, STEAM, used historic information of the automatic identification system (AIS), in which ships track their position to avoid collisions with other ships in the same area together with a technical database with emission factors from ships. The model STEAM2 was developed to analyze emission to the North Sea and the Baltic Sea and successively the model STEAM3 was developed to enable global emissions. (Endres, *et al.*, 2018 and references therein).

The authors were of the opinion that their simulation results for SO<sub>x</sub> and NO<sub>x</sub> were quite comparable with measured emission factors, but for PM the calculations were a factor of 2 to 3 higher than the measurements. They concluded that more real measurement data should be used as input (Johansson, *et al.*, 2017). Some of the assumptions in the model were the use of the cheapest fuel that met the requirements of the regulations, and an additional power consumption of 2 to 3% to run the EGCS (Johansson, *et al.*, 2017).

The STEAM3 model had primarily been developed for use in atmospheric pollution problems. With appropriate assumptions, alternative emission scenarios could be used for policy management in air pollution caused by international shipping. Whether or not the model would also be applicable for water pollution scenarios by EGCS may be further investigated (Johansson, *et al.*, 2017). The extension of the model with a module on EGCS occurred already in 2018. The STEAM model was constantly upgraded to include further areas of research, like, noise, OL/CL EGCS, antifouling paints residues, black/grey water discharges, food waste nutrients, ballast water discharges and stern tube oil leakages. (Jalkanen, personal communication, 2019)

### 7.1.5.3. DREAM

DREAM (Dose-related risk and effects assessment model; Reed and Hetland, 2002, Rye, *et al.*, 2008)) was developed by the Norwegian independent research organization SINTEF and was originally designed to describe the behavior of drilling discharges in the sea/ocean. Drilling discharges consisted of complex mixtures of chemical components and particles which might lead to toxic and nontoxic stress in the environment. In order to be able to evaluate the potential environmental consequences of such discharges in the water column and in sediments, a numerical model was developed. The model included water column stratification, ocean currents and turbulence, natural burial, bioturbation, and biodegradation of organic matter in the sediment.

Accounting for these processes, the fate of the discharge was modeled for the water column, including near-field mixing and plume motion, far-field mixing, and transport. The fate of the discharge was also modeled for the sediment, including sea floor deposition, and mixing due to bioturbation. Formulas were provided for the calculation of suspended matter and chemical concentrations in the water column, and burial, change in grain size, oxygen depletion, and chemical concentrations in the sediment. The model was fully 3-dimensional and time dependent. It relied on an external hydrodynamics model and a provided wind field to produce circulation. A Lagrangian approach was successively used to transport particles released to the water column, and an Eulerian approach with a fixed grid was used to calculate the fate of pollutants in the sediment. The model had been used to calculate the environmental risk, both in the water column and in sediments, from drilling discharges. It could serve as a tool to define risk mitigating measures, and as such it could provide guidance towards the “zero harm” goal. (Rye, *et al.*, 2008)

The authors were of the opinion that the DREAM model could be applied to evaluate the discharge of EGCS as the DREAM model was already used to model different chemical and particulate discharges and to simulate their transport and fate. Example applications of the model included the discharge of produced water and drilling wastes from oil and gas operations, mine tailings and dredge spoils, cooling and wastewater, and run-off from aquaculture (excess feed, feces, medical treatments). Current developments in DREAM included a toxicokinetic model that allowed tracking of body burdens in stationary 'numerical cages' which could be numerically deployed in the model area. (Brønner, 2019, personal communication).

### 7.1.6 Conclusions and Recommendations on Exposure Assessment

Concerning the exposure assessment of aquatic organisms and humans indirectly via the environment, the Task Team determined the following conclusions and corresponding recommendations:

1. In relatively open waterbodies with heavy ship traffic, dilution of EGCS washwater occurred mainly through turbulent mixing. Near the ship this mixing was generated mainly by the ship, its propeller, and the EGCS discharge, while farther away it was mainly from turbulent diffusion caused by wind, waves and currents. In order to better characterize the lateral and vertical mixing, the Task Team on EGCS recommended conducting a field tracer study in which a tracer was injected into the EGCS from one or more ships, and tracer concentrations were measured in cross sections at various distances aft of a ship employing EGCS.



2. One of the most significant factors affecting predicted concentrations in an enclosed waterbody was the exchange rate between the water body and the surrounding environment. MAMPEC had an elegant structure to account for such flows based on various physical processes. The models for each process had been developed by different researchers using different techniques and therefore, the Task Team on EGCS recommended having a more comprehensive model validation, perhaps involving a carefully conducted field study in one or more harbours, focusing on EGCS loading.
3. Whether or not EGCS washwater was harmful to aquatic health depends on the magnitude of predicted concentrations (or dilutions) compared with a reference standard. At least three approaches had been used in the literature: 1) comparison of predicted dilutions against predicted no effects dilution based on whole effluent toxicity testing, usually with a large assessment factor, 2) comparison of predicted concentrations for individual pollutants with corresponding predicted no effects concentrations, and 3) comparison of predicted concentrations with relevant “background” concentrations. The three approaches could lead to significant differences in risk assessment, and therefore, the Task Team on EGCS recommended having a rigorous comparison of the three approaches.
4. **In situations where exposure could be long term, physical/chemical/biological fate processes such as volatilization, photo-degradation, sedimentation, etc. could be important. Some studies had omitted these processes, while others had included them but with little description. As a recommendation the Task Team considered that a comprehensive analysis could be made to characterize the importance of the various processes, and the conditions under which they were expected to be important.**
5. MAMPEC (or MAMPEC-BW) had been used in several studies of EGCS in enclosed waterbodies, but there were several other models that could also be used, including STEAM3 and DREAM. The Task Team on EGCS recommended seeing a side by side comparison of these, and possibly other, models, not yet known to the Task Team.
6. The Task Team recommended the performance of a set of four to five well-established, good quality WET tests, including short term and long-term endpoints on species of three trophic levels. The Task Team recommended such testing to be performed using a range of representative EGCS washwater samples from open loop systems taken in representative areas.

## 7.2. Hazard Assessment, including CMR and PBT

### 7.2.1 Human Health Hazard Assessment for CMR Properties

The Task Team recognized that carcinogenicity, mutagenicity and reproductive toxicity (CMR) properties for the chemicals presented in chapter 6 may be categorized according to their classification in accordance with the Globally Harmonized System for Classification and Labelling of Chemicals (GHS) (United Nations, 2019). The harmonized classifications according to the European version of the GHS, the CLP Regulation where CLP stands for Classification, Labelling and Packaging of Chemicals (European Commission, 2008a) had been used for all selected trace metals except for arsenic and nickel. For arsenic the classification in the Integrated Risk Information System (IRIS) was used. USEPA classified arsenic as a human carcinogen (class A) (IRIS, 1991). For nickel the classification of the International Agency for Research on Cancer (IARC) had been taken into account. IARC classified nickel and nickel compounds as human carcinogens (IARC, 2012c). The CMR

properties of the selected trace metals were listed in Table 12 and of the PAHs in Table 13. Chemicals with harmonized classifications as Carcinogenic Category 1A and 1B, Mutagenic Category 1A and 1B and Reproductive toxicity Category 1A and 1B were regarded as CMR substances within the scope of the current report. In addition, IARC classification in Group 1 and 2B had been included in the categorization of a substance as carcinogenic (C). In several jurisdictions across the world, CMR substances were identified and risk management measures were directed towards minimization of use of such chemicals.

**Table 12. CMR properties for trace metals**

<b>Chemical</b>	<b>C</b>	<b>M</b>	<b>R</b>	<b>CMR</b>
<b>Arsenic</b>	1	0	0	1
<b>Cadmium</b>	1	0	1	1
<b>Chromium (vi)</b>	1	0	0	1
<b>Copper</b>	0	0	0	0
<b>Nickel</b>	1	0	0	1
<b>Lead</b>	0	0	1	1
<b>Selenium</b>	0	0	0	0
<b>Vanadium</b>	0	0	0	0
<b>Zinc</b>	0	0	0	0

The harmonized classification according to CLP had been used for benzo(a)pyrene (European Commission, 2008). IARC had classified a number of the PAHs, and a classification in Group 1 and Group 2B was taken into account within the scope of this report. Regarding the mutagenic properties of the PAHs, the work of several expert groups had been taken into account (refer to section on PAHs in 7.2.1.1).

#### 7.2.1.1 Extrapolation and Derivation of Reference Values

The next step after the hazard assessment was the hazard characterization which would include establishing guidance levels for the chemicals. These were levels below which no adverse health effects to humans were expected for chemicals with a threshold effect. However, in the case of substances that could cause cancer, the guidance levels were associated with a low, possibly hypothetical, acceptable risk since no lower safe limit existed. For chemicals with a threshold effect, on the other hand, it was sometimes possible to establish a value for tolerable daily intake (TDI) or for provisional tolerable weekly intake (PTWI) for the general public (WHO, 2010). For chemicals that may cause cancer to humans it was more appropriate to assign a slope factor. This was an estimate of the life-time cancer risk associated with a unit dose of a chemical through ingestion (or inhalation). The slope factor was defined as increased cancer risk from lifetime exposure to a substance by ingestion (or inhalation). It was expressed as an estimate of cancer risk associated with a unit concentration ( $\text{mg kg}^{-1} \text{bw d}^{-1}$ ) or risk per  $\text{mg kg}^{-1} \text{bw d}^{-1}$  (USEPA, 2005). The slope factor may be used to derive the dose ( $\text{mg kg}^{-1} \text{bw d}^{-1}$ ) associated with cancer at a specified risk level, for instance  $10^{-5}$ . It was also possible to use a Benchmark Dose (BMD) which was defined as the dose for a

predetermined level of response, such as 1% or 10% cancer incidence (WHO, 2010). For food contaminants JECFA established lower confidence limits (BMDL).

**Table 13. CMR properties for 16 EPA PAHs**

<b>Chemical</b>	<b>C</b>	<b>M</b>	<b>R</b>	<b>CMR</b>
<b>Naphthalene</b>	1	0	0	1
<b>Acenaphthylene</b>	NA	NA	NA	NA
<b>Acenaphthene</b>	0	0	0	0
<b>Fluorene</b>	0	0	0	0
<b>Anthracene</b>	0	0	0	0
<b>Phenanthrene</b>	0	0	0	0
<b>Fluoranthene</b>	0	0	0	0
<b>Pyrene</b>	0	0	0	0
<b>Benzo[a]anthracene</b>	1	1	0	1
<b>Chrysene</b>	1	1	0	1
<b>Benzo[b]fluoranthene</b>	1	1	0	1
<b>Benzo[k]fluoranthene</b>	1	1	0	1
<b>Benzo(a)pyrene (PAH)</b>	1	1	1	1
<b>Indeno[1,2,3-cd]pyrene</b>	1	1	0	1
<b>Dibenzo[a,h]anthracene</b>	1	1	0	1
<b>Benzo[ghi]perylene</b>	0	1	0	1

*NA = not available (no IARC classification)*

As EGCS washwater may contain many constituents the focus of this report and any future hazard and risk assessment should be on those chemicals that were of highest concern and for which information was available on their appearance and concentrations in EGCS washwater. The analyses focused, therefore, on the 16 EPA PAHs, and trace metals (As, Cd, Cr, Cu, Ni, Pb, Se, V, Zn).

### **Trace metals**

Metals found in EGCS washwater may be in dissolved or particulate form. Many trace metals may be found but for practical reasons the Task Team considers that the assessment should be limited to the metals that were in the analysis scheme of the 2015 EGCS Guidelines which were arsenic (As), cadmium (Cd), copper (Cu), chromium (Cr), nickel (Ni), lead (Pb), selenium (Se), vanadium (V) and zinc (Zn). As regards the hazards for humans, most metals were very data rich and hence the hazard evaluation had been focused on work done by internationally recognized bodies (WHO, IARC, US EPA, EFSA). Furthermore, it should be noted that some of the trace metals were essential to humans (copper, zinc and selenium) which would make the hazard evaluation more complicated, since the difference between the amount needed for proper physiological functioning (nutritional requirement) and the amount above which adverse effects may start to appear (maximum tolerable intake) was small (a factor of about 5 for copper).

### Arsenic (As)

WHO classified arsenic as carcinogenic (IARC, 2012b). Long-term exposure to arsenic in drinking-water was causally related to increased risks of cancer in the skin, lungs, bladder and kidneys. The excess lifetime risk of lung or bladder cancer at the current drinking water guideline value ( $10 \mu\text{g L}^{-1}$ ) was more than 3 cases per 1000 individuals (NRC, 2001). Note that this level was higher than the  $10^{-5}$  excess lifetime risk cancer for genotoxic carcinogens used in the Guidelines for drinking-water quality (WHO, 2017b).

Arsenic was found widely in the earth's crust in oxidation states of -3, 0, +3 and +5, often as sulfides or metal arsenides or arsenates. In water, it was mostly present as arsenate (+5), but in anaerobic conditions, it was likely to be present as arsenite (+3). It was usually present in natural waters at concentrations of less than  $1 - 2 \mu\text{g L}^{-1}$ . However, in waters, particularly in groundwater, where there were sulfide mineral deposits and sedimentary deposits deriving from volcanic rocks, the concentrations could be significantly elevated (WHO, 2017b).

Human and animal data indicated that over 90% of the ingested dose of dissolved inorganic trivalent (+3) or pentavalent (+5) arsenic was absorbed from the gastrointestinal tract. Organic arsenic compounds in seafood were also readily absorbed (75 – 85%). Absorption of less soluble forms, *e.g.* arsenic trioxide, was much lower (Ishinichi, *et al.*, 1986).

Arsenic had been found in the diet, particularly in fish and shellfish and cereal and cereal products, with particularly high concentrations in rice grains and rice-based products, and bran and germ. A range of benchmark dose lower confidence limit ( $\text{BMDL}_{01}$ ) values between  $0.3$  and  $8 \mu\text{g kg}^{-1} \text{bw d}^{-1}$  was identified for cancers of the lung, skin and bladder, as well as skin lesions.  $\text{BMDL}_{01}$  represented the benchmark dose of 1% extra risk. The estimated dietary exposures to inorganic arsenic for average and high level consumers in Europe were within the range of the  $\text{BMDL}_{01}$  values identified, and therefore there was little or no margin of exposure and the possibility of a risk to some consumers could not be excluded (EFSA, 2009a).

### Cadmium (Cd)

The WHO stated that there was evidence that cadmium was carcinogenic by the inhalation route (WHO, 2017b), and IARC had classified cadmium and cadmium compounds in Group 2A (probably carcinogenic to humans) (IARC, 2012a). However, there was no evidence of carcinogenicity by the oral route and no clear evidence for the genotoxicity of cadmium. The kidney was the main target organ for cadmium toxicity. The WHO drinking water guidance value of  $3 \mu\text{g L}^{-1}$  was based on the provisional tolerable monthly intake (PTMI) of  $25 \mu\text{g kg}^{-1} \text{bw}$  (WHO, 2011a), where 10% was allocated to drinking water due to the high intake of cadmium via food (WHO, 2017b). Data on human exposure to cadmium in the general population had been statistically associated with increased risk of cancer such as in the lung, endometrium, bladder, and breast (EFSA, 2009b).

Cadmium was released to the environment in wastewater, and diffuse pollution was caused by contamination from fertilizers and local air pollution. Contamination in drinking-water may also be caused by impurities in the zinc of galvanized pipes and solders and some metal fittings. Food was the main source of daily exposure to cadmium. Cadmium absorption after dietary exposure in humans was relatively low (3 – 5%) but cadmium was efficiently retained in the kidney and liver in the human body, with a very long biological half-life ranging from 10 to 30 years (EFSA, 2009). The

EFSA Panel on Contaminants in the Food Chain (CONTAM Panel) had established a tolerable weekly intake (TWI) for cadmium of 2.5  $\mu\text{g kg}^{-1}$  bw (EFSA, 2009). The mean exposure for adults across Europe was close to, or slightly exceeding, the TWI of 2.5  $\mu\text{g kg}^{-1}$  bw. Subgroups such as vegetarians, children, smokers and people living in highly contaminated areas may exceed the TWI by about 2-fold.

### Chromium III and VI (Cr)

IARC (IARC, 1996) had classified chromium(VI) in Group 1 (human carcinogen) and chromium(III) in Group 3 (not classifiable as to its carcinogenicity to humans). Chromium(VI) compounds were active in a wide range of *in vitro* and *in vivo* genotoxicity tests, whereas chromium(III) compounds were not. The WHO drinking water guidance value of 50  $\mu\text{g L}^{-1}$  for total chromium was first proposed in 1958 for hexavalent chromium, based on health concerns, but was later changed to a guideline for total chromium because of difficulties in analyzing for the hexavalent form only (WHO, 2017b). The EFSA Panel on Dietetic Products, Nutrition and Allergies (NDA) had evaluated whether Cr(III) could be considered to be an essential trace element for humans (EFSA, 2014a), but however noted that the attempts to create chromium deficiency in animal models had not produced consistent results. The NDA Panel considered that there was a possibility that Cr(III) was an essential trace element for humans, but that there was, as yet, no convincing evidence. The CONTAM Panel had derived a Tolerable Daily Intake (TDI) of 300  $\mu\text{g Cr(III) kg}^{-1}$  bw from the lowest No Observed Adverse Effect Level (NOAEL) identified in a chronic oral toxicity study in rats (EFSA, 2014d). The bioavailability of Cr(VI), that had been ingested, was minimized by the reduction to Cr(III). This reduction by saliva, gastric juices and intestinal bacteria, took place outside the cells (extracellular), and had been described as significant (De Flora, *et al.*, 1997 in Draft USEPA, 2010). A unit lifetime excess small intestine cancer risk of  $8 \times 10^{-4}$  at an average oral daily dose of 1  $\mu\text{g Cr(VI) kg}^{-1}$  bw  $\text{d}^{-1}$  was estimated by USEPA (Draft USEPA, 2010). This value had been adopted by ECHA (2013).

### Copper (Cu)

Copper was an essential trace element for humans, and it was required for electron transfer processes (EFSA, 2015a). Copper was a central component of many enzymes, such as those involved in neurotransmitter synthesis, in energy metabolism and in collagen and elastin cross-linking. Copper was not classified as carcinogenic to humans and animals (JECFA, 1982), and furthermore, copper salts were not embryotoxic in rodents. Except for individuals with Wilson's disease, copper did appear to be a cumulative toxic hazard for man. Adequate Intakes (AIs) had been established by EFSA based on mean observed intakes in several European Union (EU) countries, while taking into account that there was no evidence of overt copper deficiency in the European population. Data from balance studies were used as supportive evidence. AIs of 1.6  $\text{mg d}^{-1}$  for men and 1.3  $\text{mg d}^{-1}$  for women were proposed (EFSA, 2015a).

### Lead (Pb)

The WHO guidance value for drinking water of 10  $\mu\text{g L}^{-1}$  (WHO, 2017b) was no longer a health-based guideline value but was designated as provisional on the basis of treatment performance and

analytical achievability. Concentrations in drinking water were generally below  $5 \mu\text{g L}^{-1}$ , although much higher concentrations (above  $100 \mu\text{g L}^{-1}$ ) had been measured where lead service connections or fittings were present. The primary source of lead was from service connections and plumbing in buildings. Lead occurred primarily in the inorganic form in the environment. Human exposure was mainly via food and water, with some via air, dust and soil. Cereal products contributed most to dietary lead exposure, while dust and soil could be important non-dietary sources in children. The Panel on Contaminants in the Food Chain (CONTAM Panel) identified developmental neurotoxicity in young children and cardiovascular effects and nephrotoxicity in adults as the critical effects for the risk assessment (EFSA, 2010). A  $\text{BMDL}_{10}$  value had been derived by EFSA from a blood level of  $15 \mu\text{g Pb L}^{-1}$  (corresponding to a dietary intake value of  $0.63 \mu\text{g kg}^{-1} \text{bw d}^{-1}$ ) and based on effects on prevalence of chronic kidney disease JECFA had established the following values as a point of departure  $0.6 \mu\text{g kg}^{-1} \text{bw d}^{-1}$  loss of 1 IQ point in children;  $1.2 \mu\text{g kg}^{-1} \text{bw d}^{-1}$  for 1 mmHg increase in blood pressure (adults) (WHO, 2011b).

### Nickel (Ni)

Ni and Ni compounds had been classified by IARC (2012a) as human carcinogens causing cancers of the lung, nasal cavity and paranasal sinuses after inhalation (IARC Group 1 for nickel compounds, and IARC Group 2B for metallic nickel). The WHO drinking water guidance value of  $70 \mu\text{g L}^{-1}$  was based on the tolerable daily intake (TDI) of  $12 \mu\text{g kg}^{-1} \text{bw}$  (JECFA, 2011), where 20% was allocated to drinking water (WHO, 2017b). Food was the dominant source of nickel exposure in the non-smoking, non-occupationally exposed population; water was generally a minor contributor to the total daily oral intake. There were no maximum levels (MLs) for Ni in food. For drinking water, a parametric (maximum acceptable concentration) (WHO, 2017a) value of  $20 \mu\text{g Ni L}^{-1}$  in water intended for human consumption, and a ML of  $20 \mu\text{g Ni L}^{-1}$  in natural mineral waters were laid down in Council Directive 98/83/EC ('Drinking Water Directive', European Commission, 1998) and in Commission Directive 2003/40/EC ('Mineral Water Directive', EU, 2003), respectively. These maximum limits were well within the guideline value of  $70 \mu\text{g L}^{-1}$  set by the WHO (EFSA, 2015b). A tolerable daily intake of  $2.8 \mu\text{g Ni kg}^{-1} \text{bw d}^{-1}$  had been derived by EFSA while taking into account the lower 95 % confidence limit for a benchmark dose of  $0.28 \text{mg kg}^{-1} \text{bw}$  at 10 % extra risk ( $\text{BMDL}_{10}$ ) for post-implantation fetal loss in rats (EFSA, 2015b).

### Selenium (Se)

Selenium was an essential trace element for humans (EFSA, 2014b), and an Adequate Intake (AI) of  $70 \mu\text{g d}^{-1}$  for adults had been set. Selenium was part of a number of selenoproteins that displayed a variety of functions that had been identified in humans, including antioxidant effects, T-cell immunity, thyroid hormone metabolism, selenium homeostasis and transport, and skeletal and cardiac muscle metabolism.

### Vanadium

Vanadium had not been shown to be essential for humans (EFSA, 2006). Vanadium compounds administered orally produced adverse effects on kidneys, spleen, lungs and blood pressure in rats.

Reproductive and developmental toxicity had been shown in rats and mice. Gastrointestinal disturbances had been reported in humans. EFSA found the available data to be inadequate to derive a tolerable upper intake level. The daily intake from food of vanadium was estimated to be of the order of 10-20  $\mu\text{g d}^{-1}$ . This intake was below the lowest doses reported to cause adverse effects by at least three orders of magnitude (EFSA, 2004). The Food and Nutrition Board (FNB) recommended a maximum intake level of 1.8 mg V  $\text{d}^{-1}$  for adults (FNB, 2001). Vanadium pentoxide had been evaluated by IARC as “possibly carcinogenic to humans” (Group 2B based on inhalation studies in animals (IARC, 2003). The conclusion was based on “sufficient evidence” in experimental animals, as there was no human data.

### Zinc (Zn)

Zinc was an essential trace element for humans (EFSA, 2014c), and the Average Requirement (AR) range was 6.2-10.2 mg  $\text{d}^{-1}$  for women (reference weight: 58.5 kg), and 7.5-12.7 mg  $\text{d}^{-1}$  for men (reference weight: 68.1 kg). Zinc had a catalytic role in many enzymes. The human transcriptome had 2500 zinc finger proteins, with a broad intracellular distribution, and activities that included binding of RNA molecules, but also involvement in protein–protein interactions. The biological role of these proteins included transcriptional and translational control/modulation and signal transduction (EFSA, 2014c). A daily dietary requirement/maximum tolerable daily intake of 0.3-1 mg  $\text{kg}^{-1}$  bw was established by JECFA (1982).

### PAHs

EGCS washwater contained a range of PAHs as has already been described in chapter 6. Some PAHs were known carcinogens such as B(a)P, see Table 13 above. PAHs had been evaluated by the International Programme on Chemical Safety (IPCS) (WHO/IPCS, 1998), the Scientific Committee on Food (SCF) (SCF, 2002) and by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (JECFA, 2005). SCF concluded that the other 15 PAHs (benzo[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene, chrysene, cyclopenta[cd]pyrene, dibenzo[a,h]anthracene, dibenzo[a,e]pyrene, dibenzo[a,h]pyrene, dibenzo[a,i]pyrene, dibenzo[a,l]pyrene, indeno[1,2,3-cd]pyrene and 5-methylchrysene) showed clear evidence of mutagenicity / genotoxicity in somatic cells in experimental animals *in vivo*. Furthermore, all PAHs, with the exception of benzo[ghi]perylene, showed clear carcinogenic effects in various types of bioassays in experimental animals. SCF reasoned that these compounds may be regarded as potentially genotoxic and carcinogenic to humans and represented a priority group in the assessment of the risk of long-term adverse health effects following dietary intake of PAHs. SCF also suggested using benzo[a]pyrene as a marker of the carcinogenic PAHs in food. This decision was based on examinations of PAH profiles in food, and on evaluation of two coal tar mixtures in a carcinogenicity study in mice. JECFA re-evaluated PAHs in 2005 (JECFA, 2005) while using the assessments of IPCS and SCF for 13 PAHs, but also took into account more recent studies. Overall, the JECFA concluded that 13 PAHs were clearly genotoxic and carcinogenic. The conclusion made by the CONTAM Panel regarding a suitable indicator for PAH in food was that benzo[a]pyrene was not a suitable indicator (JECFA, 2005). The CONTAM Panel concluded, based on currently available data, relating to occurrence and toxicity, that PAH4 (the sum of benzo[a]pyrene, chrysene, benzo[a]anthracene and benzo[b]fluoranthene) and PAH8 (Sum of benzo[a]anthracene, benzo[a]pyrene,

benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, chrysene, dibenzo[a,h]anthracene and indeno[1,2,3-cd]pyrene) were the most suitable indicators of PAHs in food.

### Benzo(a)pyrene

IARC classified benzo(a)pyrene as carcinogenic to humans (Group 1) (IARC, 2010). The World Health Organization drinking water limit value of  $0.7 \mu\text{g L}^{-1}$  was a maximum concentration allowed in drinking water relating to an excess lifetime cancer risk of  $10^{-5}$  for the general population. A BMDL<sub>10</sub> value for an extra 10% risk compared to the background was estimated by EFSA (2008). The established BMDL<sub>10</sub> of  $0.07 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  (Culp, *et al.*, 1998) was chosen for benzo[a]pyrene as a marker for the carcinogenic PAHs in food.

**Table 14. Guideline values for selected CMR chemicals**

Chemical	CAS No	Guideline value DW WHO, 2017	BMDL <sub>10</sub> / BMDL <sub>01</sub> *	IRIS Slope factor	Cancer risk at $10^{-5}$ ( $\text{mg kg}^{-1} \text{ d}^{-1}$ )
Benzo(a)pyrene (PAH)	50-32-8	$0.7 \mu\text{g L}^{-1}$	$0.07 \text{ mg kg}^{-1} \text{ bw d}^{-1}$	1 per $\text{mg kg}^{-1} \text{ bw d}^{-1}$	1E-5
Arsenic	7440-38-2	$10 \mu\text{g L}^{-1}$	$0.3 - 8 \mu\text{g kg}^{-1} \text{ bw d}^{-1}$	1.5 per $\text{mg kg}^{-1} \text{ bw d}^{-1}$	NA <sup>1</sup>
Cadmium	7440-43-9	$3 \mu\text{g L}^{-1}$	$2.5 \mu\text{g kg}^{-1} \text{ bw w}^{-1}$ (TWI)	$1.8 \times 10^{-3}$ per $\mu\text{g m}^{-3}$	1.6E+0
Chromium (vi)	18540-29-9	NA <sup>2</sup>	carcinogen	$8 \times 10^{-4}$ per $\mu\text{g kg}^{-1} \text{ bw d}^{-1}$	1.2E+1

\* This value represent a BMDL of 01 for arsenic

<sup>1</sup> NA = not applicable, the cancer risk for arsenic was 3 cases per 1000 individuals and exceeds the cancer risk level of 1 case in 100 000 individuals

<sup>2</sup> NA = not applicable

**Table 15. Guideline values for selected chemicals with threshold effects**

Chemical	CAS No	Guideline value DW WHO, 2017	Guideline value food, PMTDI	BMDL <sub>10</sub>
Chromium total	7440-47-3	$50 \mu\text{g L}^{-1}$	NA	
Chromium (iii)	7440-47-3		$0.3 \text{ mg kg}^{-1} \text{ bw d}^{-1}$	
Copper	7440-50-8	$2000 \mu\text{g L}^{-1}$ ( $2 \text{ mg L}^{-1}$ )	$1.6 \text{ mg d}^{-1}$ for men and $1.3 \text{ mg d}^{-1}$ for women	
Lead	7439-92-1	$10 \mu\text{g L}^{-1}$	$0.6 \mu\text{g kg}^{-1} \text{ bw d}^{-1}$ loss of 1 IQ point in children; $1.2 \mu\text{g kg}^{-1} \text{ bw d}^{-1}$ for 1 mmHg increase in blood pressure (adults)	$0.63 \mu\text{g kg}^{-1} \text{ bw d}^{-1}$
Nickel	7440-02-0	$70 \mu\text{g L}^{-1}$ $20 \mu\text{g L}^{-1}$ (EFSA, 2018)	$2.8 \mu\text{g kg}^{-1} \text{ bw d}^{-1}$	$0.28 \text{ mg kg}^{-1} \text{ bw d}^{-1}$
Selenium	7782-49-2	$40 \mu\text{g L}^{-1}$	$70 \mu\text{g d}^{-1}$ (AI)	
Vanadium	7440-62-2	NA	$1.8 \text{ mg d}^{-1}$	
Zinc	7440-66-6	$>3000 \mu\text{g L}^{-1}$ ( $3 \text{ mg L}^{-1}$ )	$0.3 - 1 \text{ mg kg}^{-1} \text{ bw d}^{-1}$	

NA = not available

USEPA had established a slope factor for benzo(a)pyrene (2017) of 1 per  $\text{mg kg}^{-1} \text{ d}^{-1}$  based on the tumor response in the alimentary tract (forestomach, esophagus, tongue, and larynx) of female



B6C3F1 mice (Beland and Culp, 1998). This slope factor was selected as the factor with the highest value (most sensitive) among a range of slope factors derived (USEPA, 2017), see Table 14 and 15.

In addition to the sources for guidance values stated above, it should be mentioned that the European Commission had established maximum limits (MLs) for a number of food contaminants including heavy metals (lead, cadmium, mercury, tin (inorganic) and arsenic (inorganic)) and PAHs (EFSA, 2019).

## 7.2.2 Environmental Hazard Assessment for PBT Properties

Substances may be Persistent (*i.e.* poorly degradable in the environment), Bioaccumulating (accumulate in organisms and foodchains) and Toxic for humans and the environment (PBT). If certain criteria were met, substances may be classified as PBT. In Europe, criteria for PBT identification were laid down in Annex XIII of the REACH Regulation (EC No. 1907/2006) (European Commission, 2006). In several jurisdictions across the world, PBT substances were identified and risk management measures were directed towards minimization of environmental releases of such chemicals. The minimization need was due to the persistence of PBT substances in the environment, the unpredictability of long-term environmental risks and impacts and ability to be distributed over long ranges and across food chains. Assessment of persistence, bioaccumulation and toxic effects of chemicals was performed in several jurisdictions such as under the Stockholm Convention of the United Nations Environment Programme (UNEP) to protect the environment from persistent organic pollutants (POP's), the UNECE POP protocol and the Oslo-Paris Convention (OSPAR). Criteria applied were largely comparable but there were some differences. In the EU under REACH in addition to PBT also very Persistent and very Bioaccumulating (vPvB) substances were identified. Within the context of assessment of active substances in ballast water management systems, an assessment of PBT properties was performed. The criteria were also laid down in BMW.2/Circ.13/Rev.4 (MEPC, 2017).

### 7.2.2.1 Availability of Data

EGCS washwater may contain chemical constituents that were of concern regarding possible PBT properties. Metals, metal compounds and other inorganic compounds, NO<sub>x</sub>, SO<sub>x</sub> were not of concern as these constituents did not meet the PBT criteria. Hydrocarbon fuel oil constituents and combustion products may be of concern depending on their chemical characteristics such as carbon chain length and molecular structure. Linear, branched and cyclic hydrocarbons would normally not meet the PBT criteria as the rate of (bio-)degradation would be sufficient for breakdown in environmental compartments after release. Main constituents in EGCS washwater that were of concern regarding possible PBT properties were Polycyclic Aromatic Hydrocarbons (PAHs). PAHs were a class of hundreds of organic compounds that consisted of two or more aromatic rings fused in linear, angular, or clustered arrangements. The USEPA had listed 16 PAHs as priority pollutants in aquatic and terrestrial ecosystems. According to ECHA (2016) based on the available information from degradation experiments, B[a]P degraded very slowly in soil with half-lives of > 180 d and, therefore, met the P criterion. The bioaccumulation of B[a]P in aquatic species was measured and BCFs > 5000 obtained, which qualified for the B criterion. In addition, B[a]P was a very toxic substance. Based on the available information, the most sensitive organism to B[a]P was *Crassostrea gigas*. With a calculated EC<sub>10</sub> of 0.22 µg L<sup>-1</sup>. Also, its human health hazard classification as a CMR

compound contributed to its qualification for the T criterion. Similar assessments were available for other listed PAHs. PBT information available on the 16 USEPA PAHs was listed in Appendix, Table A1.

Eight of the 16 US EPA PAHs including benzo[a]pyrene were classified as PBTs in the EU in accordance with the REACH Regulation (ECHA (2008, 2016, 2017a, 2017b, 2018a, 2018b, 2018c, 2018d). In the USA benzo[a]pyrene and 8 other PAHs were included in EPA’s Persistent Bioaccumulative and Toxic Chemical Program covering 16 PBT chemicals and 5 PBT chemical compound categories (US EPA (2018)). The information in Table 16 provided an indication on the PBT status of 16 EPA PAHs currently available in the USA and the EU. The 16 EPA PAHs were a selection of a much larger group of PAHs that would be contained in EGCS washwater as the PAHs were fuel and lube oil constituents and may be formed during combustion of the fuel. Depending on molecular size and structure, PAHs would have a range of environmental fate properties. EGCS washwaters would exhibit a certain PAHs profile. For the purpose of a risk assessment focusing on PBT properties of PAHs it was, therefore, appropriate to consider the 16 EPA PAHs as indicators for the overall presence of the larger group of molecules. The finding that some PAHs should be considered PBT chemicals did not necessarily mean that other PAHs had the same property. However, since the larger group of PAHs had the same source in a risk assessment the group should be considered as a whole. No specific information relevant for PBT assessment was available in any of the MEPC and PPR documents provided to the Task Team.

**Table 16. Legally established PBT properties of 16 EPA PAHs in the US and EU**

PAH	CAS No.	PBT	Reference
<b>Naphthalene</b>	91-20-3	NA	
<b>Acenaphthylene</b>	208-96-8	NA	
<b>Acenaphthene</b>	83-82-9	NA	
<b>Fluorene</b>	86-73-7	NA	
<b>Anthracene</b>	120-12-7	Yes	ECHA (2008)
<b>Phenanthrene</b>	85-01-8	NA	
<b>Fluoranthene</b>	206-44-0	Yes	ECHA (2018a) US EPA (2018)
<b>Pyrene</b>	129-00-0	Yes	ECHA (2018b)
<b>Benzo[a]anthracene</b>	56-55-3	Yes	ECHA (2017a) US EPA (2018)
<b>Chrysene</b>	218-01-9	Yes	ECHA (2017b) US EPA (2018)
<b>Benzo[b]fluoranthene</b>	205-99-2	Yes	US EPA (2018)
<b>Benzo[k]fluoranthene</b>	207-08-9	Yes	ECHA (2018c) US EPA (2018)
<b>Benzo[a]pyrene</b>	50-32-8	Yes	ECHA (2016) US EPA (2018)
<b>Indeno[1,2,3-cd]pyrene</b>	193-39-5	Yes	US EPA (2018)
<b>Dibenzo[a,h]anthracene</b>	53-70-3	Yes	US EPA (2018)
<b>Benzo[ghi]perylene</b>	191-24-2	Yes	ECHA (2018d) US EPA (2018)

NA: Classification not assigned or unknown

### 7.2.3 Environmental hazard assessment for aquatic toxicity

Assessing the aquatic environmental hazards of EGCS washwater constituents may be performed either by examining individual chemical constituents and their respective toxicological properties or by whole effluent approaches. Information available from whole effluent toxicity testing on EGCS washwater was described in section 7.2.4. Both short-term and long-term effects should be considered and effects on organisms living in the water column as well as on sediment dwelling organisms should be considered. The latter was of specific relevance as in the intermediate or long-term some of the chemical constituents of EGCS washwater may be distributed from the water column to marine sediment. The primary focus of the assessment was to the marine aquatic compartment including sediment. However, the Task Team noted that because in some harbour or estuarine areas the alkalinity may be much lower and even be closer to brackish or freshwater conditions, a hazard assessment should also account for low-alkalinity conditions.

As EGCS washwater may contain many constituents, the focus of this report and any future hazard and risk assessment should be on those chemicals that were of highest concern and for which information was available on their appearance and concentrations in EGCS washwater. The analyses focused, therefore, on the 16 EPA PAHs, and trace metals (As, Cd, Cr, Cu, Ni, Pb, Se, V, Zn).

#### 7.2.3.1 Availability of data

##### PAHs

Polycyclic aromatic hydrocarbons were substances that had both natural and anthropogenic origins. They could be formed as a result of combustion, and were constituents of many petroleum products as well. PAHs had different physicochemical and environmental properties (*e.g.* log  $K_{ow}$ , log  $K_{oc}$ , solubility and Bio Concentration Factors (BCF)). Different PAHs exhibited different levels of toxicity and induced different kinds of effects in aquatic organisms (Incardona, *et al.*, 2004; Lee and Anderson, 2005) depending on their physico-chemical properties and the speed of microbial transformation (Heintz, *et al.*, 1999). PAH toxicity was related to both low molecular weight and high molecular weight PAHs. Toxicity studies were mostly related to oil spills. Toxicity tests on crude and heavy fuel oils had shown that alkyl-PAH were substantially more toxic than unsubstituted PAH (specifically alkyl-phenanthrene versus phenanthrene) (Turcotte, *et al.*, 2011; Barron, *et al.*, 2004). Similarly, Barron, *et al.* (2004) suggested that alkyl-phenanthrenes were a primary cause of toxicity of crude oil in the early life stages of fish. The high aromatic content of HFO caused a high concentration of naphthalene derivatives. For almost all PAHs, the highest concentrations of alkylated PAHs were present, *e.g.* the C3-alkylated naphthalene derivatives. In the BSH study (PPR, 2018) also methylated naphthalenes were analyzed and found in notable concentrations (2 – 19  $\mu\text{g L}^{-1}$ ) for 1-methyl and 2-methyl naphthalene) in OL EGCS discharge water. These physical properties of PAHs affected their bioavailability and toxicity to freshwater and marine organisms (Neff, *et al.*, 2004).

The ecotoxicity of PAHs to freshwater and marine organisms was widely investigated and a large database of ecotoxicological studies was available in literature. For the purpose of the EGCS washwater environmental risk assessment reference was made to a comprehensive literature review including many studies across the world performed by the Dutch Institute for Public Health and the

Environment (RIVM) (Verbruggen, 2012). The resulting conclusions on Predicted No Effect Concentrations could be found in section 7.2.3.2.

### Trace metals

Metals found in EGCS washwater may be in dissolved or particulate form. Many trace metals may be found but for practical reasons the Task Team considered that the assessment should be limited to the metals that are in the analysis scheme of the 2015 EGCS Guidelines which are arsenic (As), cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb), nickel (Ni), selenium (Se), vanadium (V) and zinc (Zn). As regarding the hazards for aquatic and sediment organisms, most metals were very data rich and to arrive at a proper hazard evaluation review of many studies or review of usability of hazard assessments provided by regulatory bodies across the globe would be needed which was not feasible within the timeframe provided for this report. Most information was relevant for freshwater ecosystems but also data relevant marine risk assessment was available. For metals hazard assessment of the marine aquatic compartment, a specific assessment would be needed. Due to high alkalinity and buffering capacity of seawater, the hazards assessment of metals was very different from freshwater assessment.

### Arsenic (As)

For arsenic an environmental hazard assessment was made in the framework of the International Commission for the Protection of the river Rhine (IKSR-CIPR-ICBR, 2009). The authors concluded that based on the available data algae, crustaceans and other invertebrates were among the most sensitive species for effects of arsenic. The lowest long-term No Observed Effect Levels were concluded to be at levels  $<10 \mu\text{g L}^{-1}$ . For marine organisms a lowest effect level (EC50) of  $11 \mu\text{g L}^{-1}$  was found for mortality of the copepod *Tigriopus brevicornis*. The authors concluded no valid sediment data were available for derivation of toxicity reference values. The data were used in a species sensitivity distribution to arrive at relevant environmental toxicity reference values.

### Cadmium (Cd)

An environmental hazard assessment of cadmium metal and cadmium oxide was available in a European Risk Assessment report (European Commission, 2007). For environmental hazard assessment of cadmium a comprehensive dataset was available. The authors concluded on a  $\text{PNEC}_{\text{water}}$  of Cd based on a median  $\text{HC}_5$  value from 44 chronic NOEC values, some of which were geometric species means. The  $\text{HC}_5$  was defined as a minimum effect value representative for the most sensitive species (5% of the population was more sensitive) derived by statistical species sensitivity distribution analyses. These data were derived from 19 tests with fish / amphibians, 22 tests with aquatic invertebrates and 8 tests with primary producers and represent 28 species in total. All these tests belonged to reliable data. The NOEC values were obtained from laboratory based, single species studies and refer to the dissolved fraction. An assessment factor of two was applied on the  $\text{HC}_5$  and a  $\text{PNEC}_{\text{water}}$  was set at  $0.19 \mu\text{g Cd L}^{-1}$ . The assessment included an evaluation of the effect of environmental parameters such as the water hardness on the toxicity. In the derivation of the EQS

reference values (European Commission, 2013) water hardness was included in the criteria covering 5 different water hardness classes. For sediment similar assessment was performed.

### Chromium (Cr)

For chromium and chromium compounds an environmental hazard assessment was made in the framework of the International Commission for the Protection of the river Rhine (IKSR-CIPR-ICBR, 2009). Long-term toxicity data for chromium III were concluded to be available for bacteria, algae, crustaceans and fish. The lowest No Observed Effect level reported for the crustacean *Daphnia magna* was at 47  $\mu\text{g L}^{-1}$ . The lowest NOEC for chromium VI was determined for reproduction effects in the crustacean *Ceriodaphnia dubia* at 4.7  $\mu\text{g L}^{-1}$ . The data were used in a species sensitivity distribution to arrive at relevant environmental toxicity reference values.

### Copper (Cu)

Copper was widely known as a compound with a relatively high aquatic toxicity and was generally accepted to be classified according to GHS as Acute Aquatic Category 1 and Chronic Aquatic Category 1. The European Commission's Scientific Committee on Health and Environmental Risks (SCHER, European Commission, 2009a) provided an opinion on a voluntary risk assessment performed by industry on copper and copper compounds. In the report, it was summarized that for the marine environment 57 chronic toxicity data (NOECs) on 24 species (4 algae, 18 invertebrates, and 2 fishes) were selected as highly reliable. The lowest copper effect levels were found in the low  $\mu\text{g L}^{-1}$  range. The lowest effect concentration was 5.2  $\mu\text{g Cu L}^{-1}$  normalized at Dissolved Organic Carbon (DOC) content of 0.2  $\text{mg L}^{-1}$ . On the basis of the available dataset species sensitivity distribution curves were generated to arrive at PNECs. For sediments, sufficient reliable toxicity data were concluded not to be available. Therefore, the so-called equilibrium partitioning method was applied to arrive at reference values.

As copper was an extremely data rich metal, normally species sensitivity distribution would be applied to arrive at a PNEC. The European Commission's Scientific Committee on Health and Environmental Risks (European Commission, 2009a) provided an opinion on a voluntary risk assessment performed by industry on copper and copper compounds. For the marine environment, 57 chronic toxicity data (NOECs) on 24 species (4 algae, 18 invertebrates, and 2 fishes) were selected as highly reliable using different statistical approaches,  $\text{HC}_{5-50}$  values (50 meant the median value) ranging from 4.8 and 5.2  $\mu\text{g Cu L}^{-1}$  were calculated from data normalized on 2  $\text{mg L}^{-1}$  DOC (corresponding to 1.3 to 1.4  $\mu\text{g Cu L}^{-1}$  normalized on 0.2  $\text{mg L}^{-1}$  DOC). Based on a large dataset on copper and copper compounds the SCHER arrived at  $\text{PNEC}_{\text{marine}}$  of 2.6  $\mu\text{g Cu L}^{-1}$ , applying an assessment factor of 2 to the lowest effect concentration of 5.2  $\mu\text{g Cu L}^{-1}$  normalized at Dissolved Organic Carbon (DOC) content of 0.2  $\text{mg L}^{-1}$ . For the freshwater compartment  $\text{HC}_{5-50\text{sediment}}$  values specific for a range of different scenarios were calculated ranging from 7.8 and 27.2  $\mu\text{g Cu L}^{-1}$ .

For marine sediments, the SCHER considered sufficient reliable toxicity data were not available. Therefore, a partitioning method was applied. Considering the characteristics of marine and estuarine sediments, PNECs of 338 and 144  $\text{mg Cu kg}^{-1}$  dry weight respectively were calculated. (European Commission, 2009a). For freshwater sediments the  $\text{HC}_{5-50\text{sediment}}$  was calculated at an indicative level of a PNEC of 1741  $\text{mg Cu kg}^{-1}$  organic carbon.

### Lead (Pb)

An environmental hazard assessment on lead and lead compounds was available in a voluntary risk assessment report performed by the metals industry. In an opinion by the Scientific Committee on Health and Environmental Risks (SCHER) the environmental effect data were summarized (European Commission, 2009b). The SCHER concluded that the risk assessment contained too few marine NOEC data to derive a reliable PNEC. It was acknowledged that sensitivity comparisons between freshwater and marine species might be influenced by biological (*e.g.* different physiology) and chemical factors (different bioavailability) and that using a combined freshwater and marine database was not the most scientific way forward. SCHER concluded to support the generation of more reliable NOECs for more marine species. A preliminary HC<sub>5</sub> of 6.1 µg L<sup>-1</sup> was derived from NOECs for 6 species (2 algae, 2 annelids and 2 crustaceans). This HC<sub>5</sub> reference value was higher than the annual average environmental quality standard of 1.3 µg L<sup>-1</sup> that was established in 2013 in the framework of the EU Water Framework Directive (See Table 18). The task team recognized there was a need to further investigate the justification of appropriate guidance values for lead in marine water also in relation to the background concentration of lead and recommended a more international approach instead of a regional approach.

### Nickel (Ni)

Soluble nickel compounds such as nickel oxide in the EU had a legally harmonized classification according to GHS of Aquatic Chronic 4, which meant that acute and long-term aquatic toxicity were not high but there were some grounds for concern related to the possibilities of nickel to cause long-term effects on aquatic organisms. In an European risk assessment report submitted by Denmark (European Commission, 2003b) the environmental hazards of nickel metal and 4 soluble nickel salts had been assessed. Chronic nickel toxicity data were available for fifteen marine species. The database used included a broad representation of temperate marine organisms, including unicellular algae, macroalgae, invertebrates, and fish. The marine toxicity database used for calculation of an HC<sub>5</sub> included 15 different organisms representing 6 different taxonomic groups (*i.e.*, algae, crustaceans, echinoderms, mollusks, annelids, and fish) and covering a range of different life forms, feeding strategies and trophic levels.

### Selenium (Se)

Van Vlaardingen (2005) identified acute toxicity data for cyanobacteria, protozoa, algae, macrophytes, rotifers, molluscs, crustaceans, insects, annelids, fish, and amphibians. Chronic toxicity data were found for bacteria, cyanobacteria, protozoa, algae, macrophytes, crustaceans, insects and fish. The base set for acute toxicity data was considered complete. Since chronic toxicity data were divided over 8 taxonomic groups, for 36 species, it was decided to use the robust dataset for statistical extrapolation for derivation of toxicity reference values.

### Vanadium (V)

Van Vlaardingen (2005) found acute toxicity data for protozoa, coelenterata, molluscs, annelids, crustaceans, insects, echinoderms and fish. Chronic toxicity data were found for algae, crustaceans and fish. The base set for acute toxicity data was considered not complete. However, the omission of algae in the acute data was compensated for by chronic data for algae. The lowest NOEC used for deriving toxicity reference values was  $41 \mu\text{g L}^{-1}$  in a study with *Jordanella floridae* in a test that started with eggs.

### Zinc (Zn)

For zinc an environmental hazard assessment was made in the framework of the International Commission for the Protection of the river Rhine (IKSR-CIPR-ICBR, 2009). A large database on zinc aquatic and sediment toxicity data was available but the primary focus was on freshwater species. Marine data were concluded to be available for crustaceans and algae only with 'species mean' NOEC values between 10 and  $2700 \mu\text{g L}^{-1}$ . The lowest available NOEC for a benthic species *Hyaella azteca* was  $488 \text{ mg kg}^{-1}$  dry weight (added zinc). No sediment data on marine sediment dwelling organisms was available.

### 7.2.3.2 Extrapolation and Derivation of Reference Values

#### PAHs

The environmental toxicity of PAHs was widely investigated and a large database of ecotoxicological studies was available in literature and in hazard and risk assessment reports from research institutions and within various legal frameworks across the globe. In a report by Verbruggen (2012), among other reference values so-called maximum permissible concentrations for ecosystems ( $\text{MPC}_{\text{eco}}$ ) were derived for the 16 EPA PAHs. These environmental risk limits were derived using data on ecotoxicology and environmental chemistry and represented the potential risk of substances to the ecosystem. They were presented as the scientific basis for setting environmental quality standards (EQSs) by policy makers.

Table 17 provided an overview of the relevant MPCs derived from Verbruggen (2012) that were accepted as a reasonable representation on the state of play of scientific knowledge on the environmental hazards of PAHs. The Task Team on EGCS considered that after 2012 more ecotoxicological data may have been generated. However, as the database used for the analysis in 2012 was already extensive and included many tests on various species covering all relevant trophic levels, new information would probably have little impact on the magnitude of the Maximum Permissible Concentrations. The Task Team on EGCS considered the following MPCs as relevant for an environmental risk assessment of EGCS washwater discharges:  $\text{MPC}_{\text{eco,water}}$  (for risk assessment of pelagic organisms in freshwater conditions),  $\text{MPC}_{\text{eco,marine}}$  (for risk assessment of pelagic organisms in marine water conditions),  $\text{MPC}_{\text{eco,sediment}}$  (for risk assessment of benthic organisms in freshwater conditions) and  $\text{MPC}_{\text{eco,marine sediment}}$  (for risk assessment of benthic organisms in marine conditions). As the MPC values presented for PAHs were all based on large datasets with an accepted assessment factor, as required, these entities should be considered to represent the PNEC, which was a more globally accepted term. For the sake of this report, the Task Team on EGCS qualified these regional terminologies as the PNECs to be used in potential risk assessments. The Task Team on EGCS noted that the methodology applied for derivation of MPCs was similar to the derivation of PNECs and

hence the MPCs presented in Table 17 would be used as PNECs in the risk assessment performed for EGCS washwater.

### Trace metals

Because metal toxicity was largely influenced by pH and alkalinity Predicted No Effect Concentrations (PNECs) for the marine aquatic compartment should preferably be derived based on test data with marine species. In case no test data on marine organisms was available the uncertainty should be accounted for in the derivation of toxicity reference values for the marine aquatic compartment.

In a risk assessment, factors such as water hardness, pH, dissolved organic carbon or other water quality parameters should be accounted for as these affect the bioavailability of metals.

**Table 17. Overview of the derived risk limits for 16 EPA PAHs individually. Concentrations in water were in  $\mu\text{g L}^{-1}$ , concentrations in sediment in  $\text{mg kg}^{-1}$  dw (standard sediment containing 10% organic matter). From Verbruggen (2012)**

PAH	CAS No.	MPC <sub>eco,water</sub>	MPC <sub>eco,marine</sub>	MPC <sub>eco,sediment</sub>	MPC <sub>eco,marine sediment</sub>
Naphthalene	91-20-3	2.0	2.0	0.16	0.16
Acenaphthylene	208-96-8	1.3	0.13	0.17	0.017
Acenaphthene	83-82-9	3.8	0.38	0.97	0.10
Fluorene	86-73-7	1.5	0.30	0.83	0.17
Phenanthrene	85-01-8	1.1	1.1	0.78	0.78
Anthracene	120-12-7	0.10	0.10	0.047	0.0047
Pyrene	129-00-0	0.023	0.023	1.67	0.84
Fluoranthene	206-44-0	0.12	0.12	4.11	4.11
Chrysene	218-01-9	0.07	0.007	1.64	0.16
Benz[a]anthracene	56-55-3	0.012	0.0012	0.35	0.04
Benzo[k]fluoranthene	207-08-9	0.017	0.017	0.79	0.79
Benzo[b]fluoranthene	205-99-2	0.017	0.017	0.79	0.79
Benzo[a]pyrene	50-32-8	0.01	0.01	0.49	0.49
Benzo[ghi]perylene	191-24-2	0.0082	0.00082	0.49	0.049
Dibenzo[a,h]anthracene	53-70-3	0.0014	0.00014	0.18	0.018
Indeno[1,2,3-cd]pyrene	193-39-5	0.0027	0.00027	0.38	0.038

Table 18 provided an overview of the PNECs for trace metals that would be used in the environmental risk assessment of EGCS washwater discharges. For most trace metals Maximum Permissible Concentrations (MPCs) were available derived in the framework of the EU Water Framework Directive (Directive 2000/60/EC) or in reports by scientific institutions. In general MPC could be used as equivalent to the PNEC as the methodology applied for deriving these limit values was the same as used for PNEC derivation. Environmental Quality Standards (EQS) for annual average concentrations (AA) and peak allowable concentrations (MAC) were laid down in the EU Water Framework Directive and also had a similar basis as MPC and could, therefore, also be used as PNEC for environmental risk assessment of EGCS washwater discharges.



The values presented in Table 18 applied to dissolved metal concentrations. For chromium the total of Cr(III) and Cr(VI) was included in the reference value. In metals risk assessment the natural background concentration played an important role. For cadmium and its compounds the EQS values varied depending on the hardness of the water as specified in five class categories (Class 1: < 40 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 2: 40 to < 50 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 3: 50 to < 100 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 4: 100 to < 200 mg CaCO<sub>3</sub> L<sup>-1</sup> and Class 5: ≥ 200 mg CaCO<sub>3</sub> L<sup>-1</sup>). Reference values presented in Table 18 for copper, cadmium, nickel and lead were inclusive of background concentrations while for the other six metals the background concentration was excluded and hence the reference values presented a PNEC for added risk due to the EGCS washwater discharge.

Because metal toxicity was largely influenced by pH and alkalinity Predicted No Effect Concentrations (PNECs) for the marine aquatic compartment should preferably be derived based on test data with marine species. In case no test data on marine organisms was available the uncertainty should be accounted for in the derivation of toxicity reference values for the marine aquatic compartment.

**Table 18. Overview of the derived PNECs or similar reference values for individual dissolved trace metals contained in EGCS washwater. Concentrations in water in µg L<sup>-1</sup>, concentrations in sediment in mg kg<sup>-1</sup> dw or otherwise indicated, various sources**

Trace metal	PNEC <sub>water</sub>	PNEC <sub>marine water</sub>	PNEC <sub>sediment</sub>	PNEC <sub>marine sediment</sub>	Reference
<b>Arsenic (As)</b>	Background + 0.5 <sup>3</sup> Background + 8.0 <sup>4</sup>	Background + 0.6 <sup>3</sup> Background + 1.1 <sup>4</sup>			IKSR-CIPR-ICBR (2009)
<b>Cadmium (Cd)</b>	≤0.08 – 0.25 <sup>11</sup> ≤0.45 – 1.5 <sup>21</sup>	0.2 <sup>13</sup> ≤0.45 – 1.5 <sup>14</sup>			European Commission (2013)
<b>Chromium (Cr)</b>	Background + 3.4 <sup>5</sup>	Background + 0.6 <sup>5</sup>			IKSR-CIPR-ICBR (2009)
<b>Copper (Cu)</b>	7.8 - 27.2 <sup>3</sup>	2.6 <sup>1</sup>	1741 <sup>41</sup>	144-338 <sup>1</sup>	European Commission (2009a)
<b>Lead (Pb)</b>	1.2 <sup>13</sup> 14 <sup>14</sup>	1.3 <sup>13</sup> 14 <sup>14</sup>			European Commission (2013)
<b>Nickel (Ni)</b>	4 <sup>13</sup> 34 <sup>14</sup>	8.6 <sup>13</sup> 34 <sup>14</sup>			European Commission (2013)
<b>Selenium (Se)</b>	Background + 2.1 <sup>6</sup>	Background + 2.1 <sup>6</sup>	Background + 1.3 <sup>6</sup>		Van Vlaardingen (2005)
<b>Vanadium (V)</b>	Background + 0.40 <sup>3</sup> Background + 2.2 <sup>4</sup>	Background + 0.4 <sup>5</sup>			Smit (2012) Van Vlaardingen (2005)
<b>Zinc (Zn)</b>	Background + 7.8 <sup>3</sup> Background + 15.6 <sup>4</sup>	Background + 3 <sup>6</sup>			IKSR-CIPR-ICBR (2009)

In a risk assessment factors such as water hardness, pH, dissolved organic carbon or other water quality parameters should be accounted for as these affect the bioavailability of metals.

<sup>1</sup> AA EQS for annual average dissolved concentration

<sup>2</sup> MAC EQS for maximum allowed dissolved concentration during peak emissions

<sup>3</sup> Including background concentration

<sup>4</sup> Indicative value in mg Cu kg<sup>-1</sup> organic carbon.

<sup>5</sup> Maximum Permissible Concentration

<sup>6</sup> MAC EQS for maximum allowed dissolved concentration applicable for peak emissions and as annual average concentration

Table 18 provided an overview of the PNECs for trace metals that would be used in the environmental risk assessment of EGCS washwater discharges. For most trace metals Maximum Permissible Concentrations (MPCs) were available derived in the framework of the EU Water Framework Directive (Directive 2000/60/EC) or in reports by scientific institutions. In general MPC could be used as equivalent to the PNEC as the methodology applied for deriving these limit values was the same as used for PNEC derivation. Environmental Quality Standards (EQS) for annual average concentrations (AA) and peak allowable concentrations (MAC) were laid down in the EU Water Framework Directive and also had a similar basis as MPC and could, therefore, also be used as PNEC for environmental risk assessment of EGCS washwater discharges. The values presented in Table 18 applied to dissolved metal concentrations. For chromium the total of Cr(III) and Cr(VI) was included in the reference value. In metals risk assessment the natural background concentration played an important role. For cadmium and its compounds the EQS values varied depending on the hardness of the water as specified in five class categories (Class 1: < 40 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 2: 40 to < 50 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 3: 50 to < 100 mg CaCO<sub>3</sub> L<sup>-1</sup>, Class 4: 100 to < 200 mg CaCO<sub>3</sub> L<sup>-1</sup> and Class 5: ≥ 200 mg CaCO<sub>3</sub> L<sup>-1</sup>). Reference values presented in Table 18 for copper, cadmium, nickel and lead were inclusive of background concentrations while for the other six metals the background concentration was excluded and hence the reference values presented a PNEC for added risk due to the EGCS washwater discharge.

#### 7.2.4. Whole Effluent Toxicity (WET) Testing

As stated in section 6.4, it was impossible that the chemical analysis performed could identify all potential organic compounds that may have adverse effects on the marine ecosystem. The advantage of performing Whole Effluent Toxicity (WET) testing on the discharge washwater was that it integrated and addressed the potential aquatic toxicity of SO<sub>x</sub>, together with other exhaust gas components, including any unidentified PAHs, trace metals and other organic chemicals. Also, the aggregated ecotoxicity from contaminants both dissolved and adsorbed on particulate matter could be taken into account.

In the references provided by PPR together with scientific literatures collected by Task Team, there were four papers performing the WET testing using the actual discharge water from EGCS; Japan (2019), Ytreberg, *et al.* (2019), Koski, *et al.* (2017) and Magnussen, *et al.* (2018). The summary of those tests were shown in the Table 19.

Japan (2019) performed three acute toxicity WET tests using discharge sea water with dilution by clean sea water. They used three test organisms, micro algae (*Skeletonema costatum*), crustacean (*Hyale barbicornis*) and fish (*Oryzias javanicus*), that represented three taxa. The raw discharge water without dilution indicated a certain adverse effect in all three tests. Japan (2019) concluded that the lowest dilution needed to achieve NO mortality (NOEC) was 12.5% (8-fold dilution), also that the lowest EC50 delivered was 20% (5-fold dilution). The Task Team evaluated both the methodologies they applied and validity of their assessment. The Task Team concluded that both NOEC and EC50 reported had sufficient reliability from a methodological viewpoint. In addition, the Task Team concurred with the assessment in the report that the residual toxicity observed was predominantly caused by low pH of the discharge water, because a certain dilution by clean seawater the pH for the exposure recovered to be more than 6.5 in one hour.

However, the Task Team recognized that the discharge water was collected from the test plants set up on land; combination with a middle speed 4 cycle engine and EGCS originally designed for closed loop. The Task Team also recognized that the lower concentration of PAHs rather than those reported in the other documents. Therefore, the Task Team could not conclude that the residual ecotoxicity measured in Japan (2019) had sufficient representativity for discharge water from actual ECGSs onboard. Secondly, the Task Team considered that other adverse effects which led by the other than SO<sub>x</sub> may be masked by low pH, and neither be quantified in these tests. The Task Team considered that further tests and assessment will be needed from these viewpoints using pH adjusted discharge water in combination with performing long-term toxicity testing.

Ytreberg, *et al.*, (2019) stated that EGCS discharge water caused increased (positive) growth of two micro algae (*Nodularia spumigena* and *Melosira cf. arctica*). The test organisms were exposed for 13 days, using 5% (20-fold dilution) and 10% dilution (10-fold dilution) of the discharge water. Ytreberg, *et al.*, (2019) concluded that the two species respond differently: *N. spumigena* showed negative responses in photosynthetic activity (EC<sub>10</sub> = 8.6% for *N. spumigena*) and increased primary productivity (EC<sub>10</sub> = 5.5% for *M. cf. arctica*). However, Ytreberg *et al.* (2019) made only two diluted sections (5% and 10%), so that the Task Team considered the number or dilution series could be insufficient. Also, the tests used different endpoints for growth inhibition than those in internationally accepted standards. Therefore, the Task Team could not perform the further quantitative evaluation to calculate adverse effects.

Koski, *et al.*, (2017) performed three acute WET tests, using the discharge water of actual EGCS diluted by clean water. The test organisms used were micro algae (*Rhodomonas* sp.) for growth inhibition, and crustacean (*Acartia tonsa*) mortality of adults (copepodite) and eggs, respectively. The raw discharge water indicated strong residual ecotoxicity on all three test organisms, by which all the test organisms cannot survive or no growth during the exposure duration. On the other hand, with a certain dilution rate (e.g. 10% or 10-fold dilution), no adverse effects were observed. However, all three tests by Koski, *et al.* (2019) were not performed in accordance with internationally accepted standards, for example, log phase of the micro algae growth were NOT kept for the exposure duration of 12 days. Therefore, the Task Team concluded that there are uncertainties on the reported data to calculate EC<sub>50</sub> or EC<sub>10</sub>.

Magnussen, *et al.*, (2018) carried out chronic toxicity tests with juvenile copepods (*Calanus helgolandicus*) using discharge water in a range of dilution series. The Task Team noted that the LOECs for molality were 0.04% and 0.1% for CL (two different tests), and 1% for OL. However, because of the uneven geometric factor for dilution series, the Task Team considered that these toxicities may be overestimated. Also, the Task Team considered that the method to detect the statistical difference was not appropriate for molarity. Therefore, the Task Team concluded the LOEC using Metabolic rate as its endpoint was more appropriate, and in this case, LOEC were >2% and 0.5% for CL (two different tests), and >10% for OL. Magnussen, *et al.* (2018) concluded that not the increased acidity or the reduced alkalinity caused the chronic adverse effects, rather than chemical contaminants. the CL EGCS has a function that the washwater was post-treated and diluted prior to discharge, so that 99% of the organic compounds and large fraction of the metals are removed, even in this case, a higher residual toxicity was observed. Therefore, Magnussen *et al.* assume that the lower molecular weight organics, that are better soluble in water, and the high copper and mercury concentrations might cause the chronic toxicity. However, because of less exposure time, 1 week, the Task Team considered that the test should be classified as semi-chronic. Also, the Task Team

recognized that the methodology to determine the NOEC was not in accordance with internationally accepted standards, therefore, the Task Team could not fully evaluate their EC10 instead of LOEC.

**Table 19. Summary results of WET testing using the actual discharge water from EGCS**

Reference	Test	Test organisms	pH	End point	References/guidelines
Japan	Growth inhibition testing using micro Algae	<i>Skeletonema Costatum</i>	3.5	growth rate after 72 hours EC50 49%	ISO 10253:2016
Japan	acute toxicity testing using crustacean	<i>Hyale barbicornis</i>	3.5	mortality after 96 hours EC50 20%	USEPA OCSP 850.1020 (2016)
Japan	acute toxicity testing using fish	<i>Oryzias Javanicus</i>	3.5	mortality after 96 hours EC50 35%	OECD TG203(1992)
Ytreberg, et al. (2019)	growth inhibition testing using micro Algae	<i>Nodularia spumigena</i>	2.8	primary productivity after 72 hours, EC10=9%	
Ytreberg, et al. (2019)	growth inhibition testing using micro Algae	<i>Melosira cf. arctica</i>	NA	primary productivity after 72 hours, Positive EC10=6%	
Koski, et al. (2019)	growth inhibition testing using micro Algae	Rhodomonas sp.	NA	growth rate after 12 days because of insufficient data EC50 could not be delivered ( LOEC could be 10%)	NO standards were applied
Koski, et al. (2019)	Acute	<i>Acartia tonsa</i> (adult copepods)	3.2	mortality after 24 hours (NOEC could be 30%)	NO standards were applied
Koski, et al. (2019)	Acute	<i>Acartia tonsa</i> (egg)	3.2	Egg hatching after 24 hours (NOEC could be 50%)	NO standards were applied
Magnussen, et al. (2018)	Chronic	<i>Calanus helgolandicus</i> (juvenile copepods)	7.6	CL Metabolic rate after 7 days >2% and 0.5%	NO standards were applied
Magnussen, et al. (2018)	Chronic	<i>Calanus helgolandicus</i> (juvenile copepods)	7.6	OL Metabolic rate after 14 days >10%	NO standards were applied

From all the results, the Task Team considered that the acute adverse effects were dominantly caused by the low pH of the discharge water, associated with low dissolved oxygen concentrations. Among these four results of WET tests, because of the uncertainties identified above, the Task Team could not fully perform a comparison of the ecotoxicity reported. Even though, the Task Team draw a preliminary conclusion that acute toxicity caused by the discharge water of OL could be mitigated with around 10% dilution (10-fold dilution). The Task Team recognized that all EGCS comply with the discharge standard for the pH; more than 6.5 at 4.0 m distance from the discharge point. The Task Team also recognized that at least a 10-fold dilution was needed to reach the standard for the pH of discharge water reported in the papers. It should be noted that a 10-fold dilution caused only approximately one unit increase of pH, the rest of differential pH will be neutralized by the buffer effect of alkalinity. The compliance of the standards implies that the acute adverse effects of the discharge water could be sufficiently mitigated by the initial dilution at 4 m from discharges, together with neutralization by the buffering effect of alkalinity.

It should be noted that this mitigation process would be applied when the alkalinity of surrounding seawater was sufficiently high, to about 2,200  $\mu\text{g L}^{-1}$ , and the pH was higher than 8.2, as assumed in the 2015 Guideline. Therefore, in this extraordinary case of a surrounding environment, such as fresh water with less alkalinity, the adverse effects could not be sufficiently mitigated, and the area with lower pH could be expanded over a distance of more than 4 m.

For the chronic or long-term ecotoxicity, the Task Team could not reach a conclusion from the data mentioned above. The Task Team noted the preliminary assessment approach by Japan that applied an assessment factor (*i.e.* 1000 in accordance with the methodology of GESAMP BWWG (MEPC, 2017). Using this approach, the Task Team considered that a 1/10,000 ( $10^{-4}$ ) dilution would be sufficient to mitigate chronic effects from the WET testing considered above. This dilution ratio was lower than the NOEC of the semi chronic test for OL in Magnussen, *et al.* (2018). Therefore, the Task Team considered that further data and assessment will be needed to conclude on chronic or long-term effects by the discharge water.

#### 7.2.4.1 Conclusions and Recommendations for WET

The Task Team determined the following conclusions and corresponding recommendations with respect to WET testing:

1. The Task Team concluded that the concept of Whole Effluent Toxicity (WET) testing could be a worthwhile effort to explore further assessment to evaluate the aggregated ecotoxicity of the washwater of EGCS, which may be caused by SOx together with all contaminants.
2. The Task Team recognized that almost all WET tests were not performed in accordance with internationally accepted methodology as these did not exist. It was, therefore, difficult to make a quantitative comparison and full evaluation among the data.
3. Even with the uncertainties mentioned above, the Task Team considered that the acute adverse effects were dominantly caused by the low pH of the discharge water, associated with low dissolved oxygen concentrations.
4. If washwater was discharged to full strength seawater with high alkalinity (*i.e.* 2,200  $\mu\text{g L}^{-1}$ ), the acute adverse effects could be sufficiently mitigated.
5. In the extraordinary case of a surrounding environment, such as freshwater with less alkalinity, the adverse acute effects could not be sufficiently mitigated.

6. Also, there was a possibility that the ecotoxicity of the other contaminants was masked by the dominant adverse effects by the low pH, therefore the Task Team recommended that further tests will be needed from these viewpoints using pH adjusted discharge water.
7. For the chronic or long-term ecotoxicity, the Task Team could not reach a conclusion from the data mentioned above. The chronic WET testing, however, was quite expensive and should be performed according to good laboratory practice but could give reliable answers on how the general toxicity, including all the unknown substances, could be tackled. The Task Team recommended that a set of four to five well-established, good quality WET tests should be performed in different relevant areas to study the toxicity for aquatic organisms and subsequently ecosystems.

#### 7.2.5 Conclusions and Recommendations on Hazard Assessment

1. For human health hazard assessment, the Task Team recommended to establish guidance values to be used in the risk characterization. For use in the risk assessment for general public existing guidance values such as those established by for example JECFA or EFSA for food contaminants, such as PTWI or TDI, should be taken into account (see section 7.2.1.1). For chemicals with a non-threshold effect such as genotoxic carcinogens, it was considered more appropriate to use a slope factor or BMDI. The Task Team further recommended establishing exposure scenarios while taking into account exposure pathways for humans via the environmental and coastal amenities. For instance, exposure via food (secondary poisoning in conjunction with bioconcentration and biomagnification in aquatic organisms) as already mentioned earlier this report (section 3, section 5.3), may be a suitable way forward in a human health risk assessment approach.
2. For environmental hazard assessment the Task Team recommended to establish PNECs to be used in risk characterization. The Task Team considered PNECs should be developed for PAHs and all trace metals relevant in the scope of an environmental risk assessment for EGCS washwater discharges. Preferably PNECs should be derived for the marine aquatic and sediment compartment. As some enclosed areas such as harbours and estuaries could have freshwater rather than marine conditions, the Task Team considered derivation of PNECs for freshwater including sediment would be appropriate. The Task Team further considered that given the comprehensiveness of ecotoxicological datasets available for the chemicals under consideration and evaluations of the data being available in various legal frameworks by regulatory bodies, it would be advisable to refer to conclusions of such analyses in a EGCS risk assessment.
3. The Task Team noted that for PBT classified PAHs present in EGCS washwater, a risk assessment focusing on comparison of PECs with PNECs had limited or no meaning. It was generally understood by the scientific community that quantitative risk assessment for PBT chemicals could not yet be performed with sufficient certainty. In several legal frameworks across the globe regulating chemical supply and use identified PBTs were considered in a scheme targeted at minimization of use and emissions. The Task Team took note of these findings and concluded a risk assessment targeted at PBT chemicals in EGCS washwater should be targeted to PBT identification. The Task Team considered that in this respect a comparative analyses of PAHs emissions to the marine aquatic compartment in certain



regions or on a global scale compared with other anthropogenic sources could facilitate the discussions on appropriate risk management measures in the context of maritime policy.

### 7.3. Potential for Risk Assessment

The general risk assessment process was depicted in Figure 14. There were 2 important routes distinguished: the upper route would give the hazard identification based on (eco)toxicological information, while in the lower route the exposure assessment was indicated. The exposure was based on the emission of the substance under consideration and its physico-chemical characteristics. The ratio of exposure and hazard defined the risk assessment quotient: PEC/PNEC for the environment and the ratio exposure / guidance value for threshold substances (DNEL) or the ratio exposure / guidance value for genotoxic substances (DMEL) for the human health assessment. Each step may be refined to represent more realistic situations: the tiered approach.

In section, 7.1 the exposure assessment possibilities were outlined and in section 7.2 the hazard assessment possibilities.

#### 7.3.1 Human Health Risk Assessment Approaches

The Task Team noted that the SOLAS study (Endres, *et al.*, 2018) stated that IARC had identified 17 PAHs as a threat to human health, and that some of these substances were carcinogenic (Abdel-Shafy and Mansour, 2016). Furthermore, according to the report, PAHs were transported through the food web and end up in marine sediments where sediment dwelling and filter-feeding organisms, shellfish and fish may accumulate PAH. Fish and seafood containing PAH may then be consumed by humans. The Task Team agreed with the conclusion and suggested that in any future risk assessment proper exposure scenarios based on this conclusion may be established, i.e. exposure to trace metals and other chemicals via seafood consumption.

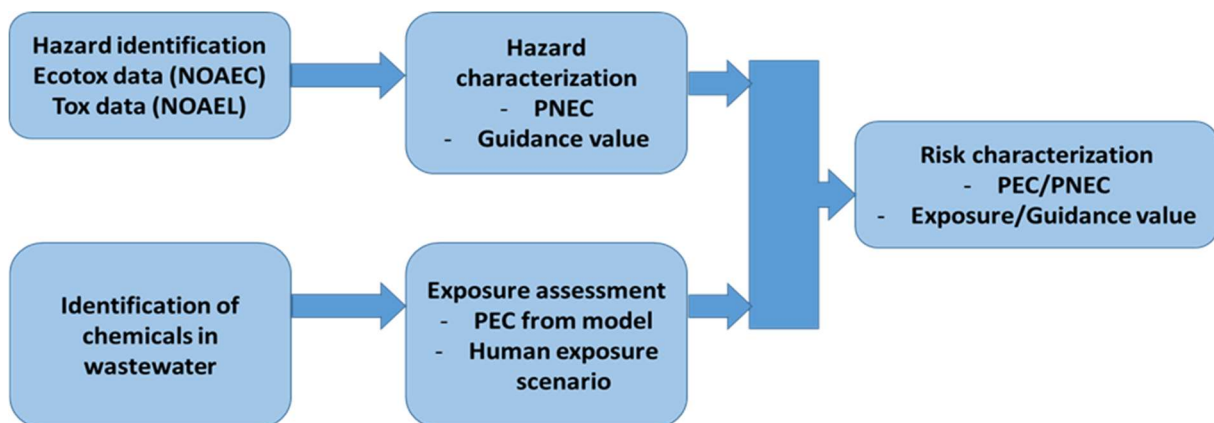


Figure 14. Steps in the risk assessment approach (modified from GESAMP (2019))

The Task Team noted that in the CESA study (MEPC, 2018) the concentrations of PAH, BTEX and metals were compared with the WHO guidelines for drinking water (WHO, 2017b). The Task Team recognized that this approach may be suitable for screening purposes but did not serve as a

complete risk assessment strategy. The reason was that a part of the quantitative exposure assessment part was missing, *i.e.* how many of the various chemicals were taken up in aquatic organisms and humans. Furthermore, the Task Team was of the opinion that the WHO guidelines for drinking water would not be suitable risk characterization guidance values as such, since they were expressed as concentrations ( $\text{mg L}^{-1}$ ). The underlying data that had been used to establish the drinking water guideline values may, however, be used to derive an appropriate guidance value, *i.e.*, an estimate of a substance that could safely be taken in daily per unit body weight (expressed as  $\text{mg kg}^{-1} \text{bw d}^{-1}$ ). This approach would be applicable for chemicals with a threshold effect.

The Task Team noted that the USEPA (2011) study analyzed data from EGCS water chemical analyses from three different vessels and compared them against US NRWQC (US National recommended water quality criteria). The study concluded that for heavy metals and PAH, IMO guidelines wastewater limits may not be sufficiently protective, since measured values exceeded US NRWQC. The Task Team acknowledges the use of the US NRWQC, but however reiterates its preference for the use of more suitable guidance values, such as guidance values derived for contaminants in food, to be used in a human health risk assessment.

### 7.3.2 Exposure via Different Sources and Background Concentrations

As already stated in section 7.2.1, there were risk management measures directed towards minimization of use of chemicals with CMR properties. Humans were exposed to contaminants in multiple ways other than through point sources of contamination, for example via food, drinking water and air. Since the total exposure from all sources should not exceed the tolerable daily intake (TDI), it was suggested (Naturvårdsverket, 2009) that exposure via an additional source of contaminants, such as from contaminated soil, should not account for more than 50% of the total TDI. Furthermore, it was suggested that for substances where the known background concentrations present were larger than 50% of the TDI, which was the case for lead, cadmium and mercury in that study, maximum 20% of the total TDI may originate from the contaminated area. For persistent organic contaminants such as dioxins and PCB, the corresponding figure was proposed to be 10%. It should be noted that for CMR substances no safe guidance levels could be assigned. For inorganic arsenic, there was little or no margin of exposure since the estimated dietary exposures for average and high level consumers in Europe were already within the range of the  $\text{BMD}_{L01}$  values identified, and therefore the possibility of a risk to some consumers could not be excluded (EFSA, 2009a).

### 7.3.3 Possibilities for Environmental Risk Assessment

Chemicals that were classified as PBT by several authorities across the world were given high priority for far reaching risk management measures such as use prohibitions and emission minimization requirements. PBT were generally understood as chemicals having a high potential risk to the environment because they are persistent and may bioaccumulate in food chains. Their risks were unpredictable on the long term and their presence in environmental compartments and biota was irreversible. The possibilities for quantitative risk assessment of PBT chemicals were limited but may progress in future. The Task Team considered currently the scientific methods for risk assessment of PBTs too limited to advance on this within the context of the ToR. Therefore, in an assessment of environmental risks of EGCS wastewater, a PBT assessment should focus on PBT identification only.



The Task Team considered that some of the PAHs contained in EGCS washwater in several jurisdictions were classified as PBT chemicals. The Task Team considered also that formation of PAHs was a phenomenon associated with combustion processes in general. PAH releases occurred as a result of various anthropogenic activities involving combustion of oil, coal or biomass and waste and also resulting from natural fires etc. Maritime shipping was a source of global PAH release. An increase in the use of EGCS would mean a shift from primarily atmospheric emissions towards seawater emissions via washwater. It was for policy makers to decide whether appropriate abatement would have to be applied to minimize emissions of PAHs by anthropogenic sources. The Task Team considered that in this respect a comparative analyses of PAHs emissions to the marine aquatic compartment in certain regions or on a global scale compared with other anthropogenic sources could facilitate the discussions on appropriate risk management measures in the context of maritime policy.

As regards the marine aquatic compartment and sediment compartment the Task Team considered that a quantitative risk assessment may be carried out on the short term and long-term toxicological effects on pelagic and benthic ecosystems. In doing this, the Task Team proposed that relevant exposure scenarios were defined to arrive at model-derived Predicted Environmental Concentrations (PECs) in seawater and sediment. This assessment should result in a range of PECs that were predicted based on EGCS washwater concentrations of chemical constituents. For each chemical, a PNEC would have to be derived or used from assessments available from other authorities (such as environmental quality standards or other appropriate guidance or reference values). PNECs were predicted concentrations at which marine aquatic ecosystems were considered not to be adversely affected if exposure to the chemical of concern was below that level. PNECs were established by applying a safety factor (assessment factor) to an ecotoxicological effect value found in tests on the most sensitive species. An assessment factor in general would be lower in case the size of the ecotoxicological dataset would increase. Finally, the risk assessment should include a comparison of PEC and PNEC values arriving at a so-called risk characterization ratio (PEC/PNEC) as indicated in Figure 14. In this deterministic approach, a risk characterization ratio below 1 depicted a numeric representation of a lack of risk. A ratio above 1 was considered a risk at ecosystem level.

#### *7.4. Recommendation on Risk Assessment*

1. Whether or not EGCS washwater was harmful to aquatic health depends on the magnitude of predicted concentrations (or dilutions) compared with a reference standard. At least three approaches had been used in the literature: 1) comparison of predicted dilutions against predicted no effects dilution based on whole effluent toxicity testing, usually with a large assessment factor, 2) comparison of predicted concentrations for individual pollutants with corresponding predicted no effects concentrations, and 3) comparison of predicted concentrations with relevant “background” concentrations. The three approaches could lead to significant differences in risk assessment, and therefore, the Task Team on EGCS recommended having a rigorous comparison of the three approaches.

## 8. UNCERTAINTY

In the area of EGCS there exist still many uncertainties: the amount of substances in the exhaust gas was rather small but the number of substances huge, their toxicological (human health) and ecotoxicological (aquatic organisms) effects are in many cases insufficiently known and the behavior in the environment ((bio-)degradation and sorption) was unknown for many substances, etc. In fact, every measurement and assumption have specific uncertainties. A measurement may have systematic errors and confounding errors in the best case according to a Gaussian distribution, while an assumption may be an educated guess or just a default value but mostly a range of potential values may exist in reality. In the case of a model application, many assumptions are needed for every parameter used. Sometimes, techniques like parameter estimation or sensitivity analysis are used to determine the most appropriate value of a parameter in a certain case, while in another case another value of a parameter maybe more appropriate. The number of possibilities in reality was numerous. Therefore, assumptions and simplifications are required to start to understand the systems under consideration. In this case, the possibilities of carrying out a risk assessment for EGCS, it will not be different. The data required to perform a risk assessment are divided in 3 types of data, emission estimation to determine the load the different chemicals into the aquatic ecosystem, the behavior estimation of the chemicals in the aquatic environment, how they will be distributed based on their physico-chemical characteristics and (bio-)degradation and sorption possibilities and finally the toxicological and ecotoxicological data the determine potential effects on the organisms in the aquatic environment and the subsequent effects on humans that are exposed through the environment. In addition, almost all WET tests were not performed in accordance with internationally accepted methodology, therefore, the comparison and full evaluation through the project was difficult.

EFSA (2018) and OECD (2019) have developed methods to evaluate the uncertainties in a scientifically sound way but it takes many efforts to work along the proposed lines. Therefore, the Task Team on EGCS did not pursue this item further. The Task Team recommended, if considered necessary, to include the uncertainty assessment of the risk assessment of EGCS emissions to aquatic systems in a potential subsequent ToR.

## 9. FURTHER RESEARCH

To the Task Team on EGCS it was quite clear that additional research on the risk assessment of EGCS effluent washwater was required to fully develop a scientifically sound evaluation. The question is, of course, how much input of manpower and financial resources are required to achieve an acceptable result for policy decisions. The Task Team concluded that a Tier 1 risk assessment would be considered possible, provided sufficient data related on :

- the information in this report on the availability of data of EGCS efficiency,
- information on chemical substances relevant for EGCS with regard to physico-chemical characteristics and (eco-)toxicological endpoints, and
- calculation methods to derive a scientifically sufficiently sound estimation of a PEC or a PNEC for the environmental evaluation and human exposure scenarios related to a PEC including establishing guidance values (*i.e.* TDI or slope factor/BMDL) for human health evaluation.

Sufficient data could be collected for some sample substances and with reasonably sound expert judgement of several parameters to perform this risk assessment. The Task Team considered it feasible to do this in a time period of one year. Several choices on the tools to be used should be made upfront. If a more sophisticated assessment would be considered necessary including *e.g.* the best possible choice for a mathematical model, the time required would increase quickly and may amount to several years of manpower and programming costs. Research results using mathematical modelling are often criticized because of the fact that these tools are simplifications of the real world and therefore always exceptions may be encountered where such a model would not be applicable or the model would need further adaptation to the local situations.

### 9.1 Conclusions and Recommendations on Future Research

The Task Team on EGCS came to the following more general recommendations related to future work, finally leading to a scientifically sound risk assessment methodology taking into account the current status.

1. The Task Team recommends converting for some example substances the efficiency data into a load to the environment. In principle, this could only be a rough estimate based on a limited number of ships and a limited number of measurements, but for a preliminary risk assessment, this may suffice.
2. The Task Team recommended establishing a data base of these chemicals and their properties *e.g.* in accordance with the GISIS data base on disinfection by-products for ballast water management systems evaluation.
3. The Task Team recommended to focus on certain key parameters, *e.g.* some PAHs (maximally the 16 EPA PAHs and the alkylated PAHs), some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter.
4. EGCS remove several substances in sufficient quantities from the exhaust gas flows from the engines onboard ships. The application of special features like EGR and SCR may even increase the efficiency, especially for NO<sub>x</sub>. Achieving a high efficiency for PM should be strived for as the chemicals adhered to PM, like heavy metals and PAHs could be better controlled in the water phase compared to the gaseous phase. That these substances have to be controlled further was not the question; the question was more what efficiency has to be achieved in controlling these chemicals as the costs may increase disproportionately above 95% or more depending on the chemical. Should the best available technology be applied or was the best available technology not entailing excessive costs (BATNEEC) sufficient, was a question that the Task Team would like to pose to the decision making bodies.
5. The Task Team also presented the following recommendations for further consideration in relation to the use and performance of EGCS. The Task Team decided not to take any position with respect to a complete ban of EGCS and to adhere to the ToR. Nevertheless, although the recommendations below were not directly related to risk assessments, they surely related to a potential reduction of the emission of chemicals to the aquatic environment and potential risks to aquatic organisms and subsequently humans:
  - .1 for HFO-fueled ships the sulfur emission should be controlled to a relevant minimum and other contaminants are removed to environmentally acceptable levels;
  - .2 application of EGR and SCR was preferred to minimize the emission of NO<sub>x</sub>;

- .3 closed loop EGCS are preferred because a greater part of contaminants are prevented from pollution of air and water, however, it should be noted that highly concentrated washwater will be emitted in small mass amounts;
  - .4 sufficient reception and processing facilities for residual sludge from closed loop EGCS should be available around the globe in important harbours. Smaller harbours may collect but not process the waste and have to transport the waste to the nearest facility.
6. If a follow-up to the work of the Task Team should be considered, the Task Team recommended the following items to be included in the ToR:
- .1 estimate reasonable emission data based on EGCS efficiency for some example substances, *e.g.* 16 EPA PAHs, some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter for discharge into a standardized location;
  - .2 collect for some example substances, *e.g.* 16 EPA PAHs, some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter sufficient data on physico-chemical properties, human and environmental effects to perform a risk assessment and develop a GISIS-like data base for them;
  - .3 develop a standardized emission scenario for the discharge situation of the above mentioned example chemicals;
  - .4 explore the possibilities to adjust the MAMPEC model in the current assessment situation;
  - .5 carry out a risk assessment with the developed substance data, the discharge situation and the environment situation, including a relevant scenario;
  - .6 report the findings in an information document to PPR 8 or PPR 9 depending on the urgency.

## 10. ROLE OF GESAMP

GESAMP, or in full the IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UN Environment/UNDP/ISA Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection, was part of the organizational structure of the United Nations, especially in the area of marine environmental protection. It provides advice to the UN member organizations on a wide variety of issues and concerns with the marine environment as a common denominator. GESAMP was not a political body but gives purely scientific advice that may have political implications. Therefore, the current report and the subject fit perfectly in this position in between policy driven organizations and the science driven necessity to provide sound defendable advice. GESAMP was able to put together in a relatively short timeframe a Task Team on EGCS and to provide a peer reviewed report that met the requirements of the ToR. However, as stated before the quality of the advice laid down in this report would have been higher if more time would have been allotted.

GESAMP was available for any follow-up activity in the area of EGCS as decided by PPR and MEPC. A new ToR has to be provided including a reporting time. Whether or not the Task Team could be the same as the current team might be dependent on the wording of the ToR and on which items emphasis should be put to meet the successive ToR. If the MEPC and PPR are of the opinion that more work should be done in this area of EGCS emission to the aquatic environment, GESAMP will be

able to support additional work with its expertise. Potential items for a potential next ToR are indicated in the recommendations.

## 11. CONCLUSIONS AND RECOMMENDATIONS

Considering the available time and complexity of the issue under consideration by the Task Team on EGCS the present information report was not fully conclusive and did not completely cover all aspects of potential environmental impacts of EGCS washwater effluents discharge.

Nevertheless, the Task Team on EGCS reached the following conclusions with respect to its ToR.

### 11.1 General

Related to the current work performed by the Task Team on EGCS, the following more general conclusions and recommendations were identified compared to the more specific conclusions and recommendations in 11.2 and 11.3.

1. Based on information available the Task Team was not able to conclude on the risks of EGCS discharges to the marine environment as it identified several uncertainties and data gaps.
2. In case a risk assessment would be carried out the Task Team advised that it should include an assessment of ecotoxicological risks to marine pelagic and sediment dwelling organisms, and of human health risks as a result of direct exposure through dermal contact and indirectly via intake of food.
3. In the documentation assessed by the Task Team in submissions to IMO or in other available scientific literature, the Task Team encountered useful information about the potential load to the environment of several chemicals based on measurements of the discharge concentration or an analysis of the efficiency of the EGCS used. Therefore, the Task Team recommended converting, for some example substances, the efficiency data into a load to the environment. In principle, this could only be a rough estimate based on a limited number of ships and a limited number of measurements, but for a preliminary risk assessment, this may suffice.
4. For many chemicals that were commonly identified in EGCS washwater, a full dataset on the required data to perform a (preliminary) risk assessment was not available. The Task Team found gaps in data for physico-chemical properties, and for (bio-)degradation and sorption data. With respect to human health hazards and environmental hazards (i.e. (eco)toxicological effect data) the Task Team considered for most relevant chemicals sufficient information was expected to be available within existing legal frameworks across the globe and in scientific literature. It was recommended to establish a data base of these chemicals and their properties *e.g.* in accordance with the GISIS data base on disinfection by-products for ballast water management systems evaluation. These data were considered essential for a potential risk assessment of these chemicals. Some datasets may, however, already be sufficiently complete to perform a preliminary risk assessment.
5. The potential presence of a large amount of individual chemicals in EGCS washwater could make it impossible to collect all data for these chemicals as identified in point 1. Therefore, it was recommended to focus on certain key parameters, *e.g.* some PAHs (maximally the 16

EPA PAHs and possibly the alkylated PAHs), some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter, whether or not as such or as a vector for other chemicals. For some of these, sufficient quality data were available. For the metals and PAHs, quite a lot of information on (eco-)toxicological data was available as several organizations evaluated them, *e.g.* USEPA, EU, OECD, WHO, although the speciation of metals are quite variable which would have to be considered in risk assessment.

6. Another building block of the risk assessment was the exposure assessment to estimate the predicted environmental concentration that should be compared to the effect data. The Task Team determined several calculation methods in the literature available to the Task Team. One was *e.g.* a direct comparison of the measured concentration in the discharge of the EGCS with some available results of ecotoxicological tests carried out with some test organisms. Although these tests may not be reliable in all cases, they provide a means of comparison. Other methods calculated the concentration based on a conceptual emission situation like the discharge in a predetermined body of water, *e.g.* a harbour. Several mathematical models were available that in principle may be made suitable for application in the current situation. The Task Team recommended that especially the MAMPEC model that was already used in the evaluation of ballast water management systems, may rather easily be made fit for purpose and this could then serve as a Tier 1 tool to estimate the PEC of the chemical under consideration. In the opinion of the Task Team, some other available models would require too much work and resources to make them operational.
7. Whether or not EGCS washwater was harmful to marine aquatic ecosystems would depend on the magnitude of predicted concentrations (or dilutions) compared with a reference standard. At least three approaches had been used in the literature: 1) comparison of predicted dilutions against predicted no effects dilution based on whole effluent toxicity testing, usually with a large assessment factor, and 2) comparison of predicted concentrations for individual pollutants with corresponding predicted no effects concentrations, and 3) comparison of predicted concentrations with relevant “background” concentrations. The three approaches could lead to significant differences in risk assessments, and therefore, the Task Team on EGCS recommended having a rigorous comparison of the three approaches.
8. The Task Team also concluded it could be worthwhile to explore further the concept of Whole Effluent Toxicity testing (WET). WET testing, if performed according to good laboratory practice, could give useful information on the general toxicity of EGCS washwater to aquatic organisms. WET testing would include all identified and unknown substances and would provide some insight into how complex interactions between chemicals contained in EGCS washwater would affect overall toxicity. The Task Team, therefore, recommended the performance of a set of well-established, good quality WET tests, including short term and long-term endpoints on species of three trophic levels. The Task Team recommended such testing to be performed using a range of representative EGCS washwater samples from open loop systems taken in representative areas.
9. With respect to potential effects to humans the Task Team concluded that a full human health hazard dataset was available for all chemicals under consideration. This information was generated in the framework of many legal frameworks and in existing scientific literature. The Task Team concluded a main challenge was to derive appropriate toxicological

reference values for the EGCS risk assessment scenarios focusing on humans exposed indirectly via the environment.

### 11.2 With respect to Chapter 6

Task Team recognized also that current available evidence on chemicals in EGCS washwater effluents and its importance for the environment, should call for an increased and broad focus on this topic from both the science community and from policymakers. This would provide a framework for future work according to the following lines:

1. Well-documented and established ranges of contaminant ambient concentrations in the open sea, coastal waters and specific areas of bays, estuaries, harbours and ports were recommended for their use in the risk assessment efforts in areas where effluents of OL EGCS was discharged into the ambient environment;
2. Extensive chemical characterization of substances in EGCS washwater effluents, including toxic and genotoxic species (such as heterocycle and alkyl-PAH) was recommended and needed for better appraisal of contaminants emissions through EGCS and their environmental threats for marine life. In this context, EGCS washwater measurements should also include monitoring of particulate and dissolved contaminant concentrations, especially for PAHs, trace metals and all other relevant chemicals. The Task Team recognized also the necessary improvements and need for harmonized procedures in terms of washwater sampling and analysis to ensure better comparability in different data sets. With respect to these points the Task Team recognized also that the current available data and practice was insufficient;
3. Besides improving chemical emissions characterization, the development of spatial and temporal in situ measurements of contaminants along shipping lanes and in all areas of high volume of ship activities was considered important to gain a more comprehensive understanding of EGCS related pollution distribution and the environmental consequences of large-scale uses of EGCS. In respect to this, the ship emissions monitoring network may be further developed. Such developments were recommended by the Task Team, considering also an enhancement of cooperation with independent academic institutions;
4. **In terms of total amounts of contaminant discharges through EGCS, it appeared that large scale uses of these systems may lead to deterioration of environmental status, especially in the ecologically vulnerable and sensitive areas such as coastal waters, semi-enclosed seas and also in ports and harbours. In respects to this, the Task Team recommended further assessment and appraisal of contaminant loads discharged with EGCS washwater effluents.** The development of alternative effective methods and protocols based on combining ship traffic data with contaminants emission factors were fundamental for the improvement of EGCS contaminants loads monitoring and assessments. The spatial representation of related discharges of pollutants would require further modeling efforts combining ship traffic data with ship emission factors and in-situ measurements of contaminants. The amounts of EGCS contaminants should also be further taken into account in the appreciation of their environmental impacts and risk assessment approaches and methods;
5. Considering the complexity of the topic and its importance for the marine environment, the Task Team recognized a great need for more interdisciplinary research development to

better predict ship emissions and the use of EGCS and their chemicals impacts on acidification, eutrophication and related climate feed backs.

### 11.3 With respect to chapter 7

#### 11.3.1 Exposure Assessment

1. In relatively open waterbodies with heavy ship traffic, dilution of EGCS washwater occurred mainly through turbulent mixing. Near the ship this mixing was generated mainly by the ship, its propeller, and the EGCS discharge, while further away it was mainly from turbulent diffusion caused by wind, waves and currents. In order to better characterize the lateral and vertical mixing, the Task Team on EGCS recommended conducting a field tracer study in which a tracer was injected into the EGCS from one or more ships, and tracer concentrations were measured in cross sections at various distances aft of a ship employing EGCS.
2. One of the most significant factors affecting predicted concentrations in an enclosed waterbody was the exchange rate between the water body and the surrounding environment. MAMPEC had an elegant structure to account for such flows based on various physical processes. The models for each process had been developed by different researchers using different techniques and therefore, the Task Team on EGCS recommended having a more comprehensive model validation, perhaps involving a carefully conducted field study in one or more harbours, focusing on EGCS loading.
3. **In situations where exposure could be long term, physical/chemical/biological fate processes such as volatilization, photo-degradation, sedimentation, etc. could be important. Some studies had omitted these processes, while others had included them but with little description.** As a recommendation the Task Team considered that a comprehensive analysis could be made to characterize the importance of the various processes, and the conditions under which they were expected to be important.
4. MAMPEC (or MAMPEC-BW) had been used in several studies of EGCS in enclosed waterbodies, but there were several other models that could also be used, including STEAM3 and DREAM. The Task Team on EGCS recommended seeing a side by side comparison of these, and possibly other, models, not yet known to the Task Team.
5. The Task Team recommended that especially the MAMPEC model, that was already used in the evaluation of ballast water management systems may rather easily be made fit for purpose and this could then serve as a Tier 1 tool to estimate the PEC of the chemical under consideration.

#### 11.3.2 Hazard Assessment

1. For human health hazard assessment, the Task Team recommended to establish guidance values to be used in the risk characterization. For use in the risk assessment for general public existing guidance values such as those established by for example JECFA or EFSA for food contaminants, such as PTWI or TDI, should be taken into account (see section 7.2.1.1). For chemicals with a non-threshold effect such as genotoxic carcinogens, it was considered more appropriate to use a slope factor or BMDI. The Task Team further recommended establishing exposure scenarios while taking into account exposure pathways for humans via



the environmental and coastal amenities. For instance, exposure via food (secondary poisoning in conjunction with bioconcentration and biomagnification in aquatic organisms) as already mentioned earlier in this report (section 3, section 5.3), may be a suitable way forward in a human health risk assessment approach.

2. For environmental hazard assessment the Task Team recommended to establish PNECs to be used in risk characterization. The Task Team considered PNECs should be developed for PAHs and all trace metals relevant in the scope of an environmental risk assessment for EGCS washwater discharges. Preferably PNECs should be derived for the marine aquatic and sediment compartment. As some enclosed areas such as harbours and estuaries could have freshwater rather than marine conditions, the Task Team considered derivation of PNECs for freshwater including sediment would be appropriate. The Task Team further considered that given the comprehensiveness of ecotoxicological datasets available for the chemicals under consideration and evaluations of the data being available in various legal frameworks by regulatory bodies, it would be advisable to refer to conclusions of such analyses in a EGCS risk assessment.
3. The Task Team noted that for PBT classified PAHs present in EGCS washwater a risk assessment focusing on comparison of PECs with PNECs had limited or no meaning. It was generally understood by the scientific community that quantitative risk assessment for PBT chemicals could not yet be performed with sufficient certainty. In several legal frameworks across the globe regulating chemical supply and use, identified PBTs were considered in a scheme targeted at minimization of use and emissions. The Task Team took note of these findings and concluded a risk assessment targeted at PBT chemicals in EGCS washwater should be targeted to PBT identification. The Task Team considered that in this respect a comparative analyses of PAHs emissions to the marine aquatic compartment in certain regions or on a global scale compared with other anthropogenic sources could facilitate the discussions on appropriate risk management measures in the context of maritime policy.
4. The Task Team recommended establishing PNECs for PAHs and all trace metals in EGCS washwater to be used for environmental risk assessment. Preferably PNECs should be derived for the marine aquatic and sediment compartment and for freshwater conditions if deemed appropriate. The Task Team further considered that, given the comprehensiveness of ecotoxicological datasets available for the chemicals under consideration and evaluations of the data being available in various legal frameworks by regulatory bodies, it would be advisable to refer to conclusions of such analyses in a EGCS risk assessment.

### 11.3.3 Whole Effluent Toxicity

1. The Task Team concluded that the concept of Whole Effluent Toxicity (WET) testing could be a worthwhile effort to explore further assessment to evaluate the aggregated ecotoxicity of the washwater of EGCS, which may be caused by SOx together with all contaminants.
2. Task Team recognized that almost all WET tests were not performed in accordance with internationally accepted methodology as these did not exist. It was, therefore, difficult to make a quantitative comparison and full evaluation among the data.
3. Even with the uncertainties mentioned above, the Task Team considered that the acute adverse effects were dominantly caused by the low pH of the discharge water, associated with low dissolved oxygen concentrations.

4. If washwater was discharged to full strength seawater with high alkalinity (i.e. 2,200 µg L-), the acute adverse effects could be sufficiently mitigated.
5. In the extraordinary case of a surrounding environment, such as freshwater with less alkalinity, the adverse acute effects could not be sufficiently mitigated.
6. Also, there was a possibility that the ecotoxicity of the other contaminants was masked by the dominant adverse effects by the low pH, therefore the Task Team recommended that further tests will be needed from these viewpoints using pH adjusted discharge water.
7. For the chronic or long-term ecotoxicity, the Task Team could not reach a conclusion from the data mentioned above. The chronic WET testing, however, was quite expensive and should be performed according to good laboratory practice but could give reliable answers on how the general toxicity, including all the unknown substances, could be tackled. The Task Team recommended that a set of four to five well-established, good quality WET tests should be performed in different relevant areas to study the toxicity for aquatic organisms and subsequently ecosystems.
8. The Task Team, therefore, recommended that a set of four to five well-established, good quality WET tests should be performed in different relevant areas to study the toxicity for aquatic organisms and subsequently ecosystems. Almost all WET tests were not performed in accordance with internationally accepted methodology, therefore, the comparison and full evaluation through the project was difficult.

#### 11.3.4 Risk Assessment

1. Whether or not EGCS washwater was harmful to aquatic health depends on the magnitude of predicted concentrations (or dilutions) compared with a reference standard. At least three approaches had been used in the literature: 1) comparison of predicted dilutions against predicted no effects dilution based on whole effluent toxicity testing, usually with a large assessment factor, 2) comparison of predicted concentrations for individual pollutants with corresponding predicted no effects concentrations, and 3) comparison of predicted concentrations with relevant “background” concentrations. The three approaches could lead to significant differences in risk assessment, and therefore, the Task Team on EGCS recommended having a rigorous comparison of the three approaches.

#### 11.4 Recommendations for future work

The Task Team on EGCS came to the following more general recommendations related to future work, finally leading to a scientifically sound risk assessment methodology taking into account the current status.

1. The Task Team recommends converting for some example substances the efficiency data into a load to the environment. In principle, this could only be a rough estimate based on a limited number of ships and a limited number of measurements, but for a preliminary risk assessment, this may suffice.
2. The Task Team recommended establishing a data base of these chemicals and their properties *e.g.* in accordance with the GISIS data base on disinfection by-products for ballast water management systems evaluation.

3. The Task Team recommended to focus on certain key parameters, *e.g.* some PAHs (maximally the 16 EPA PAHs and the alkylated PAHs), some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter.
4. EGCS remove several substances in sufficient quantities from the exhaust gas flows from the engines onboard ships. The application of special features like EGR and SCR may even increase the efficiency, especially for NO<sub>x</sub>. Achieving a high efficiency for PM should be strived for as the chemicals adhered to PM, like heavy metals and PAHs could be better controlled in the water phase compared to the gaseous phase. That these substances have to be controlled further was not the question; the question was more what efficiency has to be achieved in controlling these chemicals as the costs may increase disproportionately above 95% or more depending on the chemical. Should the best available technology be applied or was the best available technology not entailing excessive costs (BATNEEC) sufficient, was a question that the Task Team would like to pose to the decision making bodies.
5. **The Task Team also presented the following recommendations for further consideration in relation to the use and performance of EGCS. The Task Team decided not to take any position with respect to a complete ban of EGCS and to adhere to the ToR. Nevertheless, although the recommendations below were not directly related to risk assessments, they surely related to a potential reduction of the emission of chemicals to the aquatic environment and potential risks to aquatic organisms and subsequently humans:**
  - .1 for HFO-fueled ships the sulfur emission should be controlled to a relevant minimum and other contaminants are removed to environmentally acceptable levels;
  - .2 application of EGR and SCR was preferred to minimize the emission of NO<sub>x</sub>;
  - .3 closed loop EGCS are preferred because a greater part of contaminants are prevented from pollution of air and water, however, it should be noted that highly concentrated washwater will be emitted in small mass amounts;
  - .4 sufficient reception and processing facilities for residual sludge from closed loop EGCS should be available around the globe in important harbours. Smaller harbours may collect but not process the waste and have to transport the waste to the nearest facility.
6. If a follow-up to the work of the Task Team should be considered, the Task Team recommended the following items to be included in the ToR:
  - .1 estimate reasonable emission data based on EGCS efficiency for some example substances, *e.g.* 16 EPA PAHs, some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter for discharge into a standardized location;
  - .2 collect for some example substances, *e.g.* 16 EPA PAHs, some trace metals, *e.g.* As, Cr, Cu, Ni, and V and particulate matter sufficient data on physico-chemical properties, human and environmental effects to perform a risk assessment and develop a GISIS-like data base for them;
  - .3 develop a standardized emission scenario for the discharge situation of the above mentioned example chemicals;
  - .4 explore the possibilities to adjust the MAMPEC model in the current assessment situation;

- .5 carry out a risk assessment with the developed substance data, the discharge situation and the environment situation, including a relevant scenario;
- .6 report the findings in an information document to PPR 8 or PPR 9 depending on the urgency.

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**13. APPENDIX**

Table A1. Log  $K_{ow}$ , freshwater solubility, and estimated acute and chronic toxicity of PAH frequently found in crude and refined petroleum. The 16 US EPA PAH are marked with an asterix. Solubility and toxicity values are micrograms per liter ( $\mu\text{g L}^{-1}$ ). Log  $K_{ow}$  values and solubilities are from Mackay, *et al.*, 1992, Neff and Burns, 1996, and Ran, *et al.*, 2002; (table from Neff, *et al.*, 2004)

PAH	Log $K_{ow}$	Freshwater solubility [ $\mu\text{g L}^{-1}$ ]	Acute Toxicity [ $\mu\text{g L}^{-1}$ ]	Chronic toxicity [ $\mu\text{g L}^{-1}$ ]
Naphthalene*	3.37	33.720	4.870	970
C1-Naphthalenes	3.87	27.160	1.420	284
C2-Naphthalenes	4.37	4.725	410	81
C3-Naphthalenes	4.9	2.100	130	17
C4-Naphthalenes	5.55	NA <sup>a</sup>	42	4.1
Biphenyl	3.95	7.728	1.420	250
Acenaphthylene*	4.07	16.688	1.181	180
Acenaphthene*	3.92	16.908	1.360	270
Dibenzofuran	4.12	4.225	860	135
Fluorene*	4.18	2.045	730	150
C1-Fluorenes	4.97	1.090	96	19
C2-Fluorenes	5.2	NA	56	11
C3-Fluorenes	5.5	NA	16	5.3
Anthracene*	4.54	79.6	300	60
Phenanthrene*	4.46	1.100	367	55
C1-Phenanthrenes	5.14	272	64	13
C2-Phenanthrenes	5.51	NA	26	5.1
C3-Phenanthrenes	6	NA	7.4	1.5
C4-Phenanthrenes	6.51	NA	2	0.4
Dibenzothiophene	4.49	1.136	350	70
C1-Dibenzothiophenes	4.86	NA	140	28
C2-Dibenzothiophenes	5.5	NA	27	5.4
C3-Dibenzothiophenes	5.73	NA	16	3.1
Fluoranthene*	5.22	261	55	11
Pyrene*	5.18	134	61	12
C1-Fluoranthenes/pyrenes	5.72	NA	15	3.1
Benzo[a]anthracene*	5.91	14.7	9.8	2
Chrysene*	5.86	6	11	2.2
C1-Chrysenes	6.42	62.2	2.7	0.53
C2-Chrysenes	6.88	25	0.8	0.16
C3-chrysenes	7.44	NA	0.2	0.04
C4-Chrysenes	8	NA	0.06	0.01
Benzo[b]fluoranthene*	5.8	4.1	14	2.9
Benzo[k]fluoranthene*	6	0.8	8.6	1.7
Benzo[e]pyrene	6.04	4	7.6	1.5
Benzo[a]pyrene*	6.04	1.4	7.6	1.5
Perylene	6.25	0.4	4.3	0.86
Indeno[1.2.3-cd]pyrene*	7	6	0.64	0.13
Dibenz[a.h]anthracene*	6.75	0.5	1.3	0.25
Benzo[ghi]perylene*	6.5	0.3	2.4	0.49

*a) Not available: No solubility found*

In the case of an oil spill, the distribution of dissolved PAHs was generally dominated by low molecular weight petrogenic compounds, mainly naphthalene (Naph), phenanthrene (Phe), fluorene (Flu) and their alkylated derivatives (González, *et al.*, 2006, Zhou, *et al.*, 2013).

The Task Team was of the opinion that this Table A1 was insufficiently evaluated.

Table A2. Trace metal and PAH concentrations ( $\mu\text{g L}^{-1}$ ) in open loop EGCS washwaters measured in the provided measurement campaigns

	CESA (1)	CESA	CESA	GER (2)	GER	GER	JP (3)	Koski (4)	Kjølholt (5)	Buhug (6)	Hansen (7)	Ushakov (8)
Metals ppb	Min	Max	mean	min	max	mean	mean	mean	mean	mean	mean	mean
Arsenic (As)	0	0	0.00	1	6.9	3.3	1.02	1.4	0.2	< 0.1	1.4	1.71
Cadmium (Cd)	0	0	0.00	0.01	0.07	0.03	0.035	BD	0	0.05	0.1	< 0.010
Chromium (Cr)	2	60	27.29				22.8	1.9	4.8	<1	5.6	1.87
Copper (Cu)	6	140	45.88	1.6	15.7	6.4	8.12	21	187.8	41.6	190	2.31
Iron (Fe)							997					
Lead (Pb)	20	120	72.25	0.04	2.12	0.08	1.755	0.61	17.05	5	26.4	0.64
Mercury (Hg)	8	8	8.00						0.086	< 0.1	<0.05	
Molybdenum, Mo												11.10
Nickel (Ni)	20	240	63.03	4.1	67.4	15.7	17.9	41	42.03	32.8	43.3	29.67
Vanadium (V)	20	860	213.3	10.6	290	78.4	58	162	164.3	35	164	111.09
Zinc (Zn)	20	2000	236.4	2.1	133	4.7	48.3	6.7	325	6	324	10.90
PAH ppb												
Acenaphthene	0.01	1.6	0.34									1.92
Acenaphthylene	0.02	0.58	0.16									0.0265
Anthracene	0.02	1.2	0.12									0.1205
Benzo(a)anthracene	0.02	1.2	0.23	<LOD	0.04	0.02	0.006					0.343
Benzo(a)pyrene	0.01	0.55	0.11	<LOD	0.1	0.04	0.014					1.094
Benzo(b)fluoranthene	0.01	0.37	0.10				0.012					<0.01
Benzo(g,h,i)perylene	0.01	0.36	0.08				0.014					0.095
Benzo(k)fluoranthene	0.01	0.09	0.04									0.0735
Chrysene	0.02	1.6	0.26									0.016
Dibenzo(a,h)anthracene	0.01	0.08	0.03				0.006					0.012
Fluoranthene	0.01	0.76	0.17									0.021
Fluorene	0.04	1.8	0.63									<0.01
Indeno(1,2,3-c,d)pyrene	0.01	0.14	0.04									<0.01
Naphthalene	0.02	14	3.65	0.57	9.47	3.02	0.006		0.48			<0.01
Phenanthrene	0.08	6.1	1.88	0.67	2.89	1.61	0.006					0.012
Pyrene	0.01	2.6	0.42				0.007					<0.01
PAH total	0.31	33.03	8.25	1.24	12.5	4.69	0.071		0.48			3.70

(1) MEPC 73/INF.5; (2) PPR 6/INF.20; (3) MEPC 74/INF.24; (4) Koski, *et al.*, 2017; (5) Kjølholt, *et al.*, 2012; (6) MEPC 56/INF.5; (7) Hansen, 2012; (8) Ushakov, *et al.*, 2019.



# Air emissions and water pollution discharges from ships with scrubbers

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Bryan Comer, PhD, Elise Georgeff, and Liudmila Osipova, PhD

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# TABLE OF CONTENTS

<b>Executive Summary</b> .....	<b>1</b>
<b>Introduction</b> .....	<b>2</b>
<b>Background</b> .....	<b>3</b>
<b>A history of IMO’s scrubber guidelines</b> .....	<b>7</b>
MEPC.130(53): 2005 guidelines—the first scrubber guidelines.....	8
MEPC.170(57): 2008 guidelines—where the first and only discharge criteria were established .....	9
pH .....	12
PAH .....	13
Turbidity .....	13
Nitrates .....	13
<b>Results</b> .....	<b>15</b>
Air emissions .....	15
Water pollutants.....	18
pH .....	21
PAHs .....	22
Turbidity .....	24
Nitrates .....	25
Heavy metals .....	26
<b>Conclusions</b> .....	<b>29</b>
<b>References</b> .....	<b>32</b>

## EXECUTIVE SUMMARY

Ships use scrubbers to comply with fuel sulfur standards by removing sulfur dioxide from the exhaust instead of using lower sulfur but more expensive fuels. Instead, ships with scrubbers can continue to use cheaper high-sulfur heavy fuel oil (HFO). The International Maritime Organization (IMO) allows the use of scrubbers as an equivalent compliance option because they are expected to reduce sulfur dioxide emissions by the same, or more, as using compliant fuels. However, when considering the total air pollution consequences of scrubbers, they may not be equivalent to using lower-sulfur fuels, such as marine gas oil (MGO). Additionally, while scrubbers are effective at reducing sulfur dioxide, the sulfur and other contaminants removed from the exhaust gas—including carcinogens such as polycyclic aromatic hydrocarbons (PAHs) and heavy metals—are dumped overboard in the form of washwater, also called discharge water. This happens even with so-called “closed-loop” scrubbers.

In this study, we estimated air and water emission factors for ships using HFO with scrubbers compared to other fuels based on the available literature and the methods of the *Fourth IMO Greenhouse Gas Study*. Regarding air emissions, we found that using scrubbers can substantially reduce sulfur dioxide emissions but carbon dioxide, particulate matter, and black carbon emissions were higher when using HFO with a scrubber than using MGO. For water pollutants, we found that scrubber discharges usually comply with IMO guidelines; however, compliance does not guarantee that scrubber discharges are safe. We found that all scrubbers (open-loop, closed-loop, and hybrid) discharge water that is more acidic and turbid than the surrounding water. Additionally, scrubbers emit nitrates, PAHs, and heavy metals, all of which can negatively affect water quality and marine life. Within Canada, this includes scrubber discharges in the Great Lakes, as well as British Columbia and the St. Lawrence Estuary, where endangered species like the Southern Resident killer whales and belugas already suffer from high levels of contamination, including from PAHs and heavy metals.

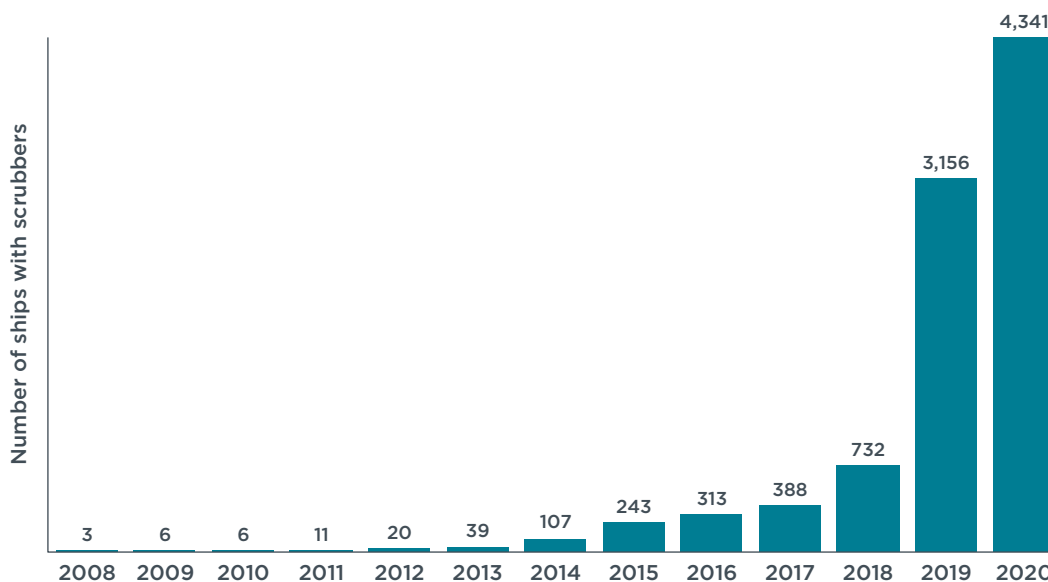
Based on this analysis, we make the following recommendations. We recommend individual governments continue to take unilateral action to restrict or prohibit scrubber discharges from both open-loop and closed-loop systems. We also recommend that the IMO focus on harmonizing rules for scrubber discharges including where, when, and even if those discharges should be allowed, and to do so with urgency. The IMO should consider prohibiting the use of scrubbers as a compliance option for new build ships and work to phase out scrubbers installed on existing ships. This is because we found that using HFO with scrubbers is not equivalently effective at reducing air pollution compared to using lower sulfur fuels, such as MGO. Additionally, scrubbers of all kinds (open, closed, and hybrid) directly contribute to ocean acidification and water pollution, whereas lower sulfur fuels do not. Until then, we recommend that individual countries, including Canada, take immediate actions to protect their air and waters from scrubber emissions and discharges. These actions could include one or both of the following: (1) an immediate prohibition on using scrubbers to comply with the Canadian portion of the North American ECA because they are not equivalently effective at reducing air pollution as ECA-compliant fuels; (2) an immediate prohibition on all scrubber discharges in Canadian ports, internal waters, and territorial seas because they contribute to acidification and water pollution that can negatively affect marine life.

## INTRODUCTION

In this report, the International Council on Clean Transportation (ICCT) provides expert advice to Environment and Climate Change Canada to enable them to update their Marine Emission Inventory Tool such that air and water pollution discharges from ships equipped with exhaust gas cleaning systems (EGCSs), also known as “scrubbers,” can be estimated for ships operating in Canadian waters.

## BACKGROUND

Ships use scrubbers as a way to comply with regional and global fuel sulfur standards by removing sulfur dioxide (SO<sub>2</sub>) from the exhaust rather than using lower sulfur fuels. In the North American Emission Control Area (ECA), the maximum allowable fuel sulfur content is 0.10% by mass. The ECA extends 200 nautical miles from the U.S. and Canadian coasts and includes all Canadian waters south of 60°N latitude. The American and Canadian Arctic regions are not covered by the ECA. Outside ECAs, the maximum allowable sulfur content for marine fuels is 0.50% as of January 1, 2020. Before 2020, the maximum allowable sulfur content was 3.50%. This tightening of the global fuel sulfur cap drove dramatic increases in scrubber installations, and the rapid uptake of scrubber installations and orders in the lead-up to 2020 is illustrated in Figure 1.



**Figure 1.** Number of ships with scrubbers by year. Source: DNV GL (2020)

While scrubbers are effective at reducing SO<sub>2</sub>, the sulfur and other contaminants removed from the exhaust gas—including carcinogens such as polycyclic aromatic hydrocarbons (PAHs) and heavy metals—are dumped overboard in the form of washwater, also called discharge water. Many of these contaminants in the washwater, including heavy metals and many PAHs, do not biodegrade and therefore amass in the environment and the food web. This makes these pollutants of particular concern for marine mammals. When marine mammals are exposed to these contaminants, usually through their food, the contaminants accumulate in their organs or are stored in their fat reserves. In lean times when food is scarce, or during pregnancy, the fat reserves are used, re-exposing the animal to the contaminants. Heavy metals, which are known to bioaccumulate in the liver, bone marrow, and kidneys in marine mammals, have been linked to carcinogenic effects and immune suppression in marine mammals (Dosi, 2000; Kakuschke & Prange, 2007). On the east coast of North America in the St. Lawrence estuary system, high PAH concentrations in beluga whales corresponded with higher rates of digestive tract cancers and tumor production (Guise, Lagacé, & Béland, 1994; Martineau et al., 2002). On the west coast, the endangered Southern Resident killer whales, found in the inlets and sounds of British Columbia, have a population critically at

risk with only 72 individuals remaining in 2020, according to the U.S. National Oceanic and Atmospheric Administration (NOAA) Fisheries Department (NOAA Fisheries, 2020). PAHs and trace metals are listed as direct impacts to the species in the Recovery Strategy in their Species at Risk Act designation, which notes that they are likely to be the most contaminated mammals in the world (Government of Canada, 2011; Ross, Ellis, Ikonomou, Barrett-Lennard, & Addison, 2000).

Georgeff, Mao, and Comer (2019) found that, in 2017, 30 scrubber-equipped ships emitted nearly 35 million tonnes of scrubber discharge water off the coast of British Columbia, including in and near critical habitats for threatened and endangered Northern and Southern Resident killer whales. Cruise ships were responsible for 90% of these discharges. The paper predicted that the International Maritime Organization (IMO) 2020 global fuel sulfur cap would result in 47 million tonnes of scrubber discharges in that area in 2020 as more ships, particularly container ships, bulk carriers, and roll-on/roll-off ferries, begin to use scrubbers. This figure includes ships that use open-loop scrubbers, which continuously discharge contaminated washwater, and from hybrid scrubbers than are operated in open-loop mode. No ships used closed-loop scrubbers in that area. Forthcoming research from the ICCT will also show that in addition to discharges off Canada's west coast, ships are also using scrubbers on the east coast, including in the St. Lawrence estuary, home to endangered beluga whales (Osipova, Georgeff, & Comer, forthcoming).

Some ships are using closed-loop scrubbers or hybrid scrubbers in closed-loop mode, mainly when operating near shore or in port. Closed-loop scrubbers recirculate the washwater, but a small volume of bleed-off water is still emitted. Unlike open-loop systems, closed-loop systems store scrubber sludge (also called residuals) on board for on-land disposal. Although closed-loop scrubbers can operate in zero-discharge mode for short periods (Kjøholt, Aakre, Jürgensen, & Lauridsen, 2012), they most often emit highly concentrated and highly contaminated bleed-off, making "closed loop" a bit of a misnomer. While closed-loop scrubbers do remove some solids, the sludge ultimately ends up in a landfill, usually as hazardous waste (Kjøholt et al., 2012). Open-loop scrubbers typically do not have water treatment systems to remove solids before discharge, contrary to many schematics of scrubbers in the literature. The water flow rate of open-loop systems is often too high to allow for onboard treatment (European Sustainable Shipping Forum, 2017). Instead, whatever sludge could be captured from open-loop systems remains suspended in the washwater and is discharged overboard.

In response to the rapid uptake and use of scrubbers to comply with the IMO's 2020 global fuel sulfur limit, and concerns about the cumulative effects that more ships using scrubbers discharging acids, PAHs, heavy metals, and other pollutants could have on the marine environment, many countries are limiting or prohibiting scrubber discharges in their exclusive economic zones (EEZs), territorial seas, internal waters, canals, and/or ports, as shown in Table 1. We note that Canada has no such restrictions, despite significant and growing scrubber discharges, including 5.1 million tonnes in critical habitat for threatened and endangered Northern and Southern Resident killer whales off the coast of British Columbia as of 2017 (Georgeff, 2020).

**Table 1.** Locations where scrubber discharges are restricted or prohibited as of September 2020

Country	Details
<b>Argentina</b>	Prohibits open-loop (OL) discharge water in internal waters, territorial seas, and EEZs
<b>Australia</b>	Ships using scrubbers must notify Australian Maritime Safety Authority before port arrival
<b>Bahrain</b>	Prohibits OL discharges in territorial seas and EEZs unless they can be proven to comply with the 2015 IMO guidelines
<b>Belgium</b>	Discharges prohibited in ports, internal waters, and within 3 nautical miles (nm) of shore
<b>Bermuda</b>	Prohibits OL discharges in territorial seas; closed-loop (CL) discharges allowed with prior approval
<b>Brazil</b>	Discharges prohibited at Vale bulk terminals/ports; discharges discouraged within 24 nm of shore
<b>China</b>	Prohibits OL discharges in internal rivers and Domestic Emission Control Areas
<b>Egypt</b>	Discharges prohibited in territorial seas, ports, and the Suez Canal
<b>Estonia</b>	Discharges prohibited in ports and estuaries unless the ship owner can demonstrate that the discharge does not cause significant adverse effects
<b>Finland</b>	Discharges prohibited in the port of Porvoo
<b>France</b>	Prohibits OL discharges in some ports and rivers, including Bordeaux, Port Jérôme-sur-Seine, River Seine, and Le Havre
<b>Germany</b>	Discharges prohibited in internal waterways
<b>Gibraltar</b>	Prohibits OL discharges in waters of Gibraltar
<b>Hong Kong</b>	Use of scrubbers requires an exemption
<b>Ireland</b>	Discharges prohibited in ports of Dublin, Waterford, and Cork
<b>Latvia</b>	Discharges prohibited in territorial seas and ports
<b>Lithuania</b>	Discharges prohibited in ports
<b>Malaysia</b>	Prohibits OL discharges in territorial seas except for ships transiting the Malacca Strait that are not bound for a Malaysian port
<b>Norway</b>	Prohibits OL discharges in World Heritage Fjords sea areas of Geirangerfjord and Nærøfjord
<b>Oman</b>	Discharges prohibited in territorial seas
<b>Pakistan</b>	Prohibits OL discharges in the ports of Karachi and Bin Qasim
<b>Panama</b>	Prohibits OL discharges in the Panama Canal
<b>Portugal</b>	Prohibits OL discharges in port
<b>Qatar</b>	Discharges prohibited in territorial seas
<b>Saudi Arabia</b>	Prohibits OL discharges in port
<b>Singapore</b>	Prohibits OL discharges in port
<b>Spain</b>	Prohibits OL discharges in the ports of Algeciras, Cartagena, and Huelva
<b>Sweden</b>	Discharges prohibited in the ports of Brofjorden, Gävle, Norrköping, Umeå, Sundsvall, Skellefteå, and Stockholm
<b>United Arab Emirates</b>	Prohibits OL discharges in the port of Fujairah
<b>USA</b>	California: Prohibits the use of scrubbers to comply with fuel sulfur limits within 24 nm Connecticut: Discharges prohibited in ports and waters of the state Hawaii: Discharges allowed, but special reporting required

Sources: Damgaard (2020) and Standard Club (2020)



Given this trend toward unilateral action by individual countries, the EU-28 and European Commission (EC) in 2019 proposed that IMO's Marine Environment Protection Committee (MEPC) undertake a new output to "evaluate and harmonize the development of rules and guidance on the discharge of liquid effluents from EGCS, including conditions and areas under which liquid effluents from EGCS can be discharged, and to regulate as appropriate access for ships equipped with such systems on that basis" (MEPC 74/14/1, para. 2). In their submission proposing a new output on harmonizing rules and guidance for EGCS discharges, the EU and EC explain that the only guidelines for EGCSs that currently apply are the 2015 guidelines, but that they do not have additional protections for sensitive areas. They also state that "it is questionable if the current criteria are fit for purpose in the current scenario, where a significant uptake of scrubbers or other technologies that discharge effluent into the marine ecosystem is occurring" (MEPC 74/14/1, para. 27).

MEPC 74 approved this new output on harmonizing rules and guidance for EGCS discharges, and tasked the Pollution Prevention and Response (PPR) subcommittee to work on the issue, with a target completion year of 2021. PPR 7 refined the title and scope of the output, which is expected to be approved at MEPC 75 (November 16–20, 2020) and will likely be sent back to PPR 8 to continue working on the topic. This provides an opportunity to develop guidance on when, where, or even if discharges should be allowed. It is likely that this work will focus on guidance for discharges in ports, harbors, estuaries, and busy shipping lanes, but Friends of the Earth International et al. (PPR 7/12/4) suggested that near shore areas, polar regions, and areas of cultural and ecological sensitivity and significance should also be considered.

## A HISTORY OF IMO'S SCRUBBER GUIDELINES

The IMO first decided to regulate sulfur oxides (SO<sub>x</sub>) from ships in the 1997 Protocol to the International Convention for the Prevention of Pollution from Ships (MARPOL), which included MARPOL Annex VI. Annex VI entered into force in May 2005 and contains regulations that limit SO<sub>x</sub> and nitrogen oxides (NO<sub>x</sub>) from ship exhaust. Sulfur oxides are primarily controlled by limiting the sulfur content of fuels, with one limit globally and another inside Sulfur Emission Control Areas (SECAs). Originally, scrubbers were to be allowed only within SECAs. However, a few months after Annex VI entered into force, IMO began revising it. In the revision, the IMO agreed that the maximum fuel sulfur content of marine fuels and the maximum NO<sub>x</sub> emissions from marine engines would become more stringent over time. Additionally, ships would be allowed to use scrubbers globally, not just in SECAs under an “equivalence” provision added as Regulation 4. The revisions also introduced ECAs, which set stronger limits for not only SO<sub>x</sub>, but also NO<sub>x</sub>. Currently, there are four ECAs (Table 2). These revisions to MARPOL Annex VI were adopted in 2008 and entered into force in July 2010.

Despite scrubbers being allowed as an alternative SO<sub>x</sub> compliance option under Regulation 4 of MARPOL Annex VI, port and coastal states are free to unilaterally limit or prohibit the use of scrubbers in their jurisdictions. Today, scrubber discharges are limited or prohibited in the territorial seas, internal waters, ports, or canals of at least 29 countries (Table 1). Canada currently has no restrictions on scrubbers.

**Table 2.** Current Emission Control Areas

Region	Applied for	Adopted	Enforced
<b>Baltic Sea</b>	1995 (SECA) 2016 (ECA)	1997 (SECA) 2017 (ECA)	2006: 1.5% max S 2010: 1% max S 2015: 0.1% max S 2021: Tier III NO <sub>x</sub>
<b>North Sea</b>	2000 (SECA) 2016 (ECA)	2005 (SECA) 2017 (ECA)	2007: 1.5% max S 2010: 1% max S 2015: 0.1% max S 2021: Tier III NO <sub>x</sub>
<b>North America (United States &amp; Canada, except the Arctic)</b>	2009 (ECA)	2010 (ECA)	2012: 1% S max 2015: 0.1% S max 2016: Tier III NO <sub>x</sub>
<b>United States Caribbean Sea (Puerto Rico &amp; U.S. Virgin Islands)</b>	2010 (ECA)	2011 (ECA)	2014: 1% S max 2015: 0.1% S max 2016: Tier III NO <sub>x</sub>

The IMO has established EGCS guidelines<sup>1</sup> for certain pollutants and other parameters (e.g., pH and temperature) for scrubber discharge water, but these guidelines are voluntarily applied by flag states, do not cover all pollutants (heavy metals are not explicitly included; turbidity is used as a proxy), and lack rigorous scientific justification. Endres et al. (2018) concluded that despite the existing IMO guidelines, “there is still the

<sup>1</sup> IMO proposed revised 2020 guidelines for scrubbers at the 7th session of its Pollution Prevention and Response Sub-Committee (PPR 7); while they have not yet been adopted by the Marine Environment Protection Committee, we expect them to be approved at MEPC 75. Nevertheless, the discharge criteria established in the 2015 guidelines, as found in IMO Resolution MEPC.259(68), remains unchanged. The text of the 2015 guidelines are available here: <http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Documents/MEPC.259%2868%29.pdf>

risk for acidification, eutrophication, and accumulation of PAHs, PM [particulate matter], and heavy metals in the marine environment” (p. 139).

The first IMO scrubber guidelines can be found in Resolution MEPC.130(53), adopted in 2005. IMO subsequently published 2008, 2009, and 2015 guidelines in Resolutions MEPC.170(57), MEPC.184(59), and MEPC.259(68). Draft 2020 guidelines have been proposed in Annex 9 of document PPR 7/22/Add.1 and are expected to be approved by MEPC 75 in November 2020.

Regarding air emissions, all guidelines require that scrubbers result in SO<sub>2</sub>/carbon dioxide (CO<sub>2</sub>) ratios that are less than or equal to those that would result from burning compliant fuels. These limits are based on sulfur content and are summarized in Table 3. Note that only the 0.50% and 0.10% values are relevant after January 1, 2020. As such, the rest have been grayed. While all scrubbers tend to easily meet these SO<sub>2</sub> limits, researchers have found that when accounting for total sulfur emissions (gaseous + particle phase), scrubbers may emit more total sulfur than compliant fuel (Johnson et al., 2017). The guidelines set no limits on any air pollutant other than SO<sub>2</sub>.

**Table 3.** Air emissions limits for ships with scrubbers

Fuel sulfur content (% m/m)	SO <sub>2</sub> (ppm)/CO <sub>2</sub> (% v/v)
4.50	195.0
3.50	151.7
1.50	65.0
1.00	43.3
0.50	21.7
0.10	4.3

### MEPC.130(53): 2005 GUIDELINES—THE FIRST SCRUBBER GUIDELINES

In the original scrubber guidelines, found in Resolution MEPC.130(53) and adopted by MEPC 53 on July 22, 2005, scrubbers were expected to be used solely inside of SECAs, as allowed under the Protocol of 1997, which entered into force on May 19, 2005. Under the original guidelines, scrubber washwater was to be monitored for pH and oil content, but no numeric discharge criteria were proposed for either parameter. Instead, section 17 states the following:

#### 17. Wash Water

*EGCS-SO<sub>x</sub> unit's wash water systems should:*

- (a) eliminate, or reduce to a level at which they are not harmful, hydrocarbons, carbon residue, ash, vanadium, other heavy metals, and other substances contained within EGCS-SO<sub>x</sub> unit's wash water that may have an adverse impact on ecosystems if discharged overboard,*
- (b) ensure that the approach adopted, to control wash water quality and residual waste is not achieved in a way that causes pollution in other areas or environmental media,*
- (c) also taking into account guidelines to be developed by the Organization.*

Regarding scrubber residues (sludge), section 18.1 makes it clear that they should be disposed of on land and not discharged overboard or incinerated on board:

*18.1 Residues generated by the EGCS-SO<sub>x</sub> unit should be land disposed. Such residues should not be discharged to the sea or incinerated on board.*

### **MEPC.170(57): 2008 GUIDELINES—WHERE THE FIRST AND ONLY DISCHARGE CRITERIA WERE ESTABLISHED**

In 2008, there were only three ships with scrubbers, according to DNV GL (2020). In the 2008 guidelines, found in Resolution MEPC.170(57), which were adopted by MEPC 57 on April 4, 2008, the first discharge criteria were set, but only when the “EGC System is operated in a [*sic*] ports, harbours, or estuaries” (section 10.1.1). It includes criteria for pH, PAH, turbidity/suspended particulate matter, and nitrates. Although subsequent guidelines have expanded the discharge limits to apply beyond ports, harbors, and estuaries, the discharge limits first established in these 2008 guidelines have never been revised to be more stringent.

The 2008 guidelines were adopted at MEPC 57, but the work on setting discharge criteria had begun in 2006. MEPC 55, which was held October 9–13, 2006, established a correspondence group on Washwater Criteria for Exhaust Gas-SO<sub>x</sub> Cleaning Systems. In establishing these discharge criteria, the correspondence group considered proposals from the United Kingdom (MEPC 55/4/5) as well as Finland and Norway (MEPC 55/4/7).

The UK document proposed that discharge criteria be established for pH and oil concentration (measured as PAH). They proposed that the pH of the discharge plume should not exceed 0.2 pH units below the background water conditions at a distance of 1 meter from the ship. They also proposed a 30 ppb (approximately equal to 30 µg/L) limit for PAHs, associated with a 50 tonnes per megawatt hour (t/MWh) flow rate. The same UK document shows that the 2000 EU Water Framework Directive sets drinking water standards of 0.01 ppb for total PAH. The 1992 Australian Water Quality Guidelines set a 3 ppb limit. In the 1992 Convention on the Protection of the Marine Environment of the Baltic Area, the Baltic Marine Environment Protection Commission, better known as HELCOM, set a 15 ppb limit for PAHs. The UK document provides the results of a 2004 study of discharges from an open-loop scrubber fitted to a European ferry, the *Pride of Kent*. In that study, the authors found that the maximum PAH concentration was 24 ppb, and that was in the residue settling tank. Typical PAH concentrations were 3–4 ppb compared with <0.6 ppb at the inlet, they said. It is perplexing why the UK would propose a limit of 30 ppb PAH at 50 t/MWh flow rate for ships with scrubbers, a level unlikely to be exceeded, given that typical concentrations were between 3 and 4 ppb. Indeed, as we will show in the results, we found that ships rarely exceed the PAH limits, which under the current guidelines allow discharges of approximately 50 µg/L (~50 ppb) at a 45 t/MWh flow rate.

The Norway and Finland document (MEPC 55/4/7) also proposed discharge criteria based on testing data from two ships, one ferry and one oil tanker, each outfitted with prototype open-loop scrubbers. The tests were conducted in 1991 and 1993. The minimum pH after the scrubber was recorded as 2.7. They assert that, due to dilution, even a pH of 0 would not result in a pH of less than 6.8, which is the most conservative

Predicted No Effect Concentration (PNEC<sup>2</sup>) they found in the literature, at a distance of at least 20 meters from the ship. The maximum PAH concentration in the scrubber washwater was 0.25 µg/L (~0.25 ppb), compared with the most conservative PNEC they could find in the literature, which was 3.3 µg/L. The Norway and Finland document suggests that, due to dilution effects, PAHs could be discharged at concentrations of approximately 6,200 µg/L while maneuvering or in transit, or more than 460 µg/L during quayside maneuvering and still not exceed the PNEC. Based on this, they recommend three tiers of criteria that port states could choose, with each level being 10 times more protective than the other. For pH, they suggested no limit. For PAH, they suggested a limit of 450, 45, or 5 µg/L (presumably rounded up from 4.5 µg/L), depending on the level of protection the port state would like to impose. They also proposed possible discharge criteria for heavy metals including nickel (Ni), vanadium (V), copper (Cu), lead (Pb), mercury (Hg), and cadmium (Cd) in units of µg/L, following the same tiered approach. However, individual heavy metal discharge criteria never made it into any scrubber guidelines because onboard monitoring is thought to be challenging. It should be understood that the modeling exercise presented in the Finland and Norway document, which showed no predicted adverse effects even at high pollution concentrations, is based on pollution discharges from one ship, whereas ports, harbors, estuaries, nearshore areas, and shipping lanes now experience scrubber discharge loads from multiple ships. Moreover, the number of ships with scrubbers is growing, as shown in Figure 1.

Ultimately, the correspondence group established by MEPC 55 did not propose specific discharge criteria limits. However, the group reported that most group members agreed that pH and oil concentration were two key performance parameters for scrubbers. The correspondence group suggested that a working group be established at MEPC 56 to finalize the discharge criteria.

At MEPC 56, which was held July 9–13, 2007, the Working Group on Air Pollution considered the report of the IMO Correspondence Group that MEPC 55 had established on Washwater Criteria for Exhaust Gas-SO<sub>x</sub> Cleaning Systems (their report is found in document MEPC 56/4/1) and developed a draft set of washwater discharge criteria for pH, oil (using PAHs as a proxy), heavy metals (using turbidity as a proxy), and nitrates. The report of the Working Group on Air Pollution (MEPC 56/WP.6) does not explain how it arrived at the discharge criteria for these parameters.

The criteria agreed to in the MEPC 56 Working Group on Air Pollution in the report are summarized in the annex to document BLG-WGAP 2/4. As stated in that document, MEPC 56 recommended a minimum outlet pH of 6.5 and a maximum difference between inlet and outlet of 2 pH units while the ship was at berth or at anchor in a port, harbor, or estuary. (In the eventual 2008 guidelines, this 2 pH difference would apply only to ships while maneuvering or in transit.) We note that because pH is a logarithmic scale, a difference of 2 pH units is equal to a 100-fold difference in acidity. They also suggested that, while underway in all areas, the pH should be maintained at a level that avoids acute effects on aquatic ecosystems, damage to antifouling systems, and accelerated corrosion of critical metal components. These considerations were lost in the eventual 2008 guidelines.

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2 PNEC is the limit below which no adverse effects from exposure are measured.

For PAHs, MEPC 56 suggested a limit of 15 ppb at a discharge rate of 45 t/MWh. This would be weakened to 50 µg/L under the 2008 guidelines. For turbidity, they recommended a maximum of 25 formazin nephelometric units (FNU), which remained, although an alternative limit of 25 nephelometric turbidity units (NTU) was added under the 2008 guidelines. For nitrates, they suggested no nitrate limit for EGCS units designed to reduce oxides of nitrogen by less than “[10] per cent” (BLG-WGAP 2/4, annex 2, p. 2). Otherwise, they suggested that the discharge limit should be less than that associated with a “[10] per cent” removal of NO<sub>x</sub> from the exhaust. No scrubbers are designed to remove NO<sub>x</sub>, so no nitrate discharge limits for scrubbers would be needed had the first clause remained. This first clause would later be removed, and the second clause was weakened to allow 12% removal of NO<sub>x</sub> or 60 mg/L of nitrates, whichever is greater, under the 2008 guidelines. The MEPC 56 Air Pollution Working Group advised MEPC not to adopt the draft 2008 guidelines yet and to instead send them to the second intersessional meeting of the Bulk Liquids and Gases Working Group on Air Pollution (BLG-WGAP 2) for further review and refinement.

BLG-WGAP 2 met from October 29, 2007, to November 2, 2007, in Berlin to work on the 2008 scrubber guidelines based on the draft washwater criteria developed by MEPC 56. BLG-WGAP 2 was instructed by MEPC 56 to finalize the draft revision to the 2005 guidelines found in MEPC.130(53), to finalize discharge criteria for EGCS from MEPC 56, and to include them in the draft amended 2008 guidelines. BLG-WGAP 2 did not finalize the draft washwater discharge criteria, so they were sent to BLG 12, which was held in February 2008, and they were also sent directly to MEPC 57, which was held in April 2008.

BLG 12 had for their consideration the draft discharge criteria from BLG-WGAP 2 in annex 6 to document BLG 12/6/Add.1. However, the discharge criteria BLG 12 ultimately recommended to MEPC 57 in document BLG 12/WP.6/Add.4 were weaker than those proposed by BLG-WGAP 2. The report of the BLG 12 Air Pollution Working Group (BLG 12/WP.6) contains no explanation or justification for this decision. The discharge criteria agreed to by BLG 12 were ultimately adopted, without revision, by MEPC 57 as the 2008 guidelines in Resolution MEPC.170(57) on April 4, 2008. Since then, the guidelines have been reviewed three times (2009, 2015, and 2020), and the discharge criteria have never been revised.

Below, for each parameter—pH, PAH, turbidity, and nitrates—we compare the recommendations of BLG-WGAP 2, as found in document MEPC 57/4/1, to the 2008 guidelines that MEPC 57 agreed to in Resolution MEPC.170(57). Table 4 details changes to the discharge criteria over time for these pollutants as well as heavy metals, compared with the number of ships with scrubbers installed during the year in which the revised guidelines were adopted. As the table shows, despite a review of the guidelines in 2009, 2015, and 2020, the discharge criteria that were initially established in the 2008 guidelines have never been revised and no numeric discharge criteria have ever been established for any heavy metal. Meanwhile, the number of ships with scrubbers has grown from three ships in 2008 to more than 4,300 ships in 2020.

**Table 4.** How IMO scrubber discharge criteria have changed over time, compared with the number of ships with scrubbers installed

Pollutant	MEPC.130(53): 2005 guidelines	MEPC 57/4/1: proposed discharge criteria from BLG-WGAP 2 for 2008 guidelines	MEPC.170(57): 2008 guidelines, as adopted by MEPC 57	MEPC.184(59): 2009 guidelines	MEPC.259(68): 2015 guidelines	PPR 7/22/ Add.1, Annex 9: Draft 2020 guidelines
<b>pH</b>	Eliminated or reduced “to a level at which they are not harmful.”	pH $\geq$ 6.5 stationary; max $\Delta$ 2 pH units when moving	pH $\geq$ 6.5 stationary; max $\Delta$ 2 pH units when moving.  OR  pH $\geq$ 6.5 in the plume at 4 m while stationary	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>
<b>PAH</b>	Eliminated or reduced “to a level at which they are not harmful.”	Max $\Delta$ 15 ppb PAH <sub>16</sub> at 45 t/MWh	Max $\Delta$ 50 $\mu$ g/L (-50 ppb) of PAH <sub>phe</sub> at 45 t/MWh	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>
<b>Turbidity</b>	Eliminated or reduced “to a level at which they are not harmful.”	Max $\Delta$ < 25 FNU or NTU; minimize suspended PM, including heavy metals and ash	Same as MEPC 57/4/1	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>
<b>Nitrates</b>	Eliminated or reduced “to a level at which they are not harmful.”	Not > that associated with a [10%] removal of NO <sub>x</sub> from the exhaust, or beyond [1] mg/L at 45 t/MWh, whichever is greater.	Not > that associated with a 12% removal of NO <sub>x</sub> from the exhaust, or beyond 60 mg/L at 45 t/MWh, whichever is greater.	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>	<i>Unchanged from 2008 guidelines</i>
<b>Heavy metals</b>	Eliminated or reduced “to a level at which they are not harmful.”	No limits	No limits	No limits	No limits	No limits
<b>Ships with scrubbers</b>	2	3	3	6	243	4,341 installed or on order through 2020

### **pH**

BLG-WGAP 2 recommended that scrubber washwater have a pH of not less than 6.5 while at berth, but when maneuvering and in transit the limit would be a maximum difference of 2 pH units between inlet and outlet. MEPC 57 agreed but kept an alternative compliance option introduced by BLG 12 that would allow setting the scrubber’s overboard pH discharge limit based on whatever pH achieved a minimum pH of 6.5 in the plume at a distance of 4 meters from the overboard discharge point. This introduces myriad confounding factors. The overboard discharge limit, in this case, would depend on the alkalinity of the inlet water, wind, waves, depth, sampling location, and other parameters. Moreover, setting the overboard pH discharge limit based on achieving a minimum pH of 6.5 at 4 meters from the overboard discharge point *ensures* that the pH will be less than 6.5 at the overboard discharge point and is therefore less protective. Given that the pH of seawater is typically around 8.0, and that the pH scale is logarithmic, even achieving a pH of 6.5 means that the overboard discharge is 32 times more acidic than seawater. Additionally, ships typically mix the scrubber outlet water with “reaction water,” which is usually ambient seawater, before discharging it



overboard, artificially raising the pH before it is monitored, while emitting the same total amount of acids overboard.

Note that the U.S. Environmental Protection Agency (EPA) under its 2013 Vessel General Permit (VGP) requires a pH of no less than 6.0 at the overboard discharge point, or a maximum difference of 2 pH units during maneuvering and transit. However, the EPA does not allow the second provision (i.e., a pH of no less than 6.5 at 4 meters) because the minimum pH of 6.0 at the point of discharge is weaker than the IMO's minimum pH of 6.5 at overboard discharge and likely results in a pH greater than 6.5 at 4 meters. The EPA (2013) explains in its VGP fact sheet that allowing a minimum pH of 6.0 while disallowing the 4-meter provision is simpler, while essentially consistent with the IMO guidelines. However, in October 2020, the EPA issued a proposed rule that would harmonize its pH requirement with the IMO's 2015 guidelines (U.S. EPA, 2020). The EPA is accepting comments through November 2020.

### **PAH**

BLG-WGAP 2 agreed that PAH was an appropriate indicator of oil content for scrubber washwater. They suggested that the U.S. EPA's 16 criteria PAHs ( $PAH_{16}$ ) should be measured and that washwater criteria for PAH be further reviewed at BLG 12. At BLG 12,  $PAH_{16}$  was replaced with phenanthrene equivalence ( $PAH_{phe}$ ) and the discharge limit was weakened. The original discharge limit was 15 ppb (approximately equal to 15  $\mu\text{g/L}$ ) of  $PAH_{16}$ ; in other words, the sum total of EPA's 16 criteria PAHs. This was replaced with 50  $\mu\text{g/L}$  of  $PAH_{phe}$ . Both limits were associated with a normalized washwater discharge rate of 45 t/MWh. Both the BLG-WGAP 2 recommendations and the 2008 guidelines explain that the PAH concentration should be measured downstream of any water treatment equipment, but upstream of any dilution or reactant dosing prior to discharge.

### **Turbidity**

Both MEPC 57/4/1 and MEPC.170(57) set the limit at 25 NTU or FNU, although we found no justification for this limit. Additionally, "the discharge water treatment system should be designed to minimize suspended particulate matter, including heavy metals and ash," although there are no specific numeric limits associated with this. Also, open-loop systems do not typically have discharge water treatment systems.

### **Nitrates**

For nitrates, BLG-WGAP 2 had draft limits in bracketed text associated with no more than a 10% removal of  $NO_x$  or 1 mg/L, whichever is greater. The bracketed text means the group could not agree on an exact limit and the "whichever is greater" language already sets a weaker standard than had it been phrased as "whichever is lower." During BLG-WGAP 2, the European Association of Internal Combustion Engine Manufacturers (EUROMOT) wanted to weaken the provision further by increasing the limit to that associated with a 20% removal of  $NO_x$ . Ultimately, BLG 12 agreed to somewhat weaken the draft limit from 10% to 12%, but also to dramatically increase the allowable nitrate concentration from 1 mg/L to 60 mg/L. Scrubber discharges can comply with the guidelines for nitrate concentrations under either limit. In practice, the concentration limit is easier to demonstrate compliance with, rather than trying to estimate what nitrate concentration would be associated with a 12% removal of  $NO_x$ . Additionally, because scrubbers are not designed to remove  $NO_x$  and, as we will show in the results, are expected to have no impact on  $NO_x$  emission factors, the relevant nitrate limit is 60 mg/L, because it is the greater of the two. The 2008 guidelines did not explain



whether the nitrate limit was based on the discharge concentration or the difference between inlet and outlet concentrations. It was clarified in the draft 2020 guidelines that the limit is based on the latter. This clarification itself is a weakening of the nitrate limit, because seawater often contains nitrates. However, it is understandable that the guidelines would be interested in preventing additional nitrates from the scrubber system. We should note that washwater discharges contain both nitrates and nitrites; the IMO guidelines cover only nitrates. The United States, in its 2013 VGP, requires the sum of nitrates and nitrites to be less than 60 mg/L.

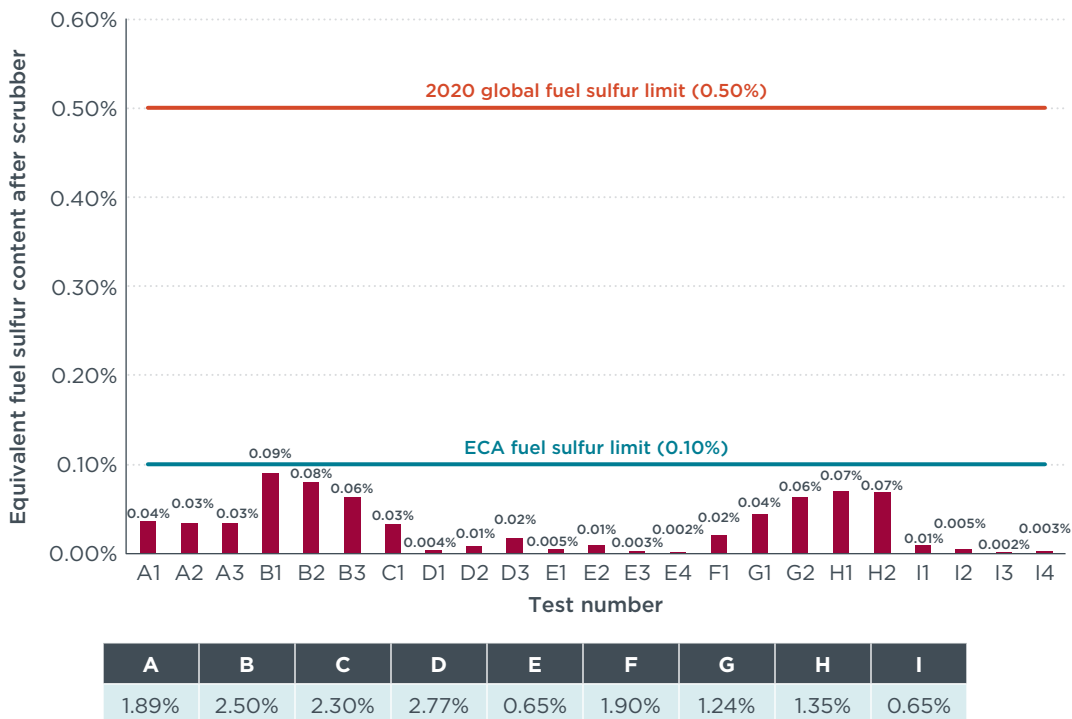
## RESULTS

This section summarizes the air and water emissions associated with scrubbers based on a review of the available literature and our own calculations.

### AIR EMISSIONS

We found eight studies representing 23 samples that contained information on air emissions from scrubbers. We compared the emissions from ships with scrubbers to expected values for other marine fuels, based on the emission factors in the *Fourth IMO Greenhouse Gas Study* (Faber et al., 2020). A detailed spreadsheet containing information about ship type, engine, scrubber type, and emission factors is provided in the supplemental material.

We calculated the equivalent fuel sulfur content of ships with scrubbers based on the SO<sub>2</sub> emissions after the scrubber and the engine’s specific fuel oil consumption (SFOC, measured in grams of fuel per kilowatt hour, g/kWh).<sup>3</sup> As shown in Figure 2, we found that all ships with scrubbers emitted SO<sub>2</sub> in amounts low enough to achieve equivalent fuel sulfur contents that were lower than both the 2020 global fuel sulfur limit of 0.50% and the ECA fuel sulfur limit of 0.10%. The original fuel sulfur content is presented in the table directly below the chart in the figure. While ships with scrubbers achieve lower SO<sub>2</sub> emissions than if they had used lower-sulfur fuels, other air pollutants are higher for ships with scrubbers than using ECA-compliant fuels, such as marine gas oil (MGO), as we explain next.



**Figure 2.** Equivalent fuel sulfur content after the scrubber, with original fuel sulfur contents in the table.

<sup>3</sup> Equivalent fuel sulfur content (% m/m) = gSO<sub>2</sub>/kWh ÷ (SFOC × 0.97753 × 2).

Table 5 shows the relative emissions in the exhaust for a ship using 2.60% sulfur heavy fuel oil (HFO) with a scrubber compared with other marine fuels, including 2.60% sulfur HFO without a scrubber, 0.50% sulfur very low sulfur fuel oil (VLSFO), 0.10% sulfur marine gas oil (MGO), and 0.07% sulfur MGO (global average fuel sulfur content as of 2019).

**Table 5.** Relative emissions change after the scrubber when using HFO (2.6% S) compared with other fuels

<b>Comparison: 2.6% S HFO + scrubber versus...</b>	<b>SO<sub>2</sub></b>	<b>CO<sub>2</sub></b>	<b>PM<sub>10</sub></b>	<b>PM<sub>2.5</sub></b>	<b>NO<sub>x</sub></b>	<b>CO</b>	<b>BC (SSD)</b>	<b>BC (MSD)</b>
<b>HFO (2.6% S)</b>	-98%	+2%	-79%	-79%	0%	-11%	-9%	-11%
<b>VLSFO (0.50% S)</b>	-90%	+4%	-59%	-59%	0%	-11%	unknown	unknown
<b>MGO (0.10% S)</b>	-52%	+4%	+61%	+61%	0%	-11%	+353%	+81%
<b>MGO (0.07% S)</b>	-31%	+4%	+69%	+69%	0%	-11%	+353%	+81%

We found that scrubbers can substantially reduce SO<sub>x</sub> emissions, with average SO<sub>2</sub> emissions 31% lower than 0.07% sulfur MGO. Based on SO<sub>2</sub> emissions and fuel consumption, we calculated the equivalent fuel sulfur content, as shown in Figure 2. One must remember that scrubber SO<sub>2</sub> performance depends on a number of factors. The performance will vary based on the sulfur content of the fuel, engine power, engine load, scrubber water flow rate, and the alkalinity of the inlet or recirculating water. While all of the scrubbers tested meet the 0.10% ECA sulfur limit, it is possible that scrubber parameters may be adjusted to only just meet the relevant sulfur limits. For example, if a ship is operating outside of an ECA, the scrubber flow rate may be adjusted down to allow SO<sub>2</sub> emissions that would correspond to 0.50% sulfur fuel. In that case, the SO<sub>2</sub> emissions reductions from scrubbers compared with VLSFO and MGO would be overestimated when the ship is operating outside of ECAs. If scrubber operations are modified to allow higher sulfur emissions outside of ECAs, direct PM emissions would also increase. Therefore, although we found that using 2.6% sulfur HFO with a scrubber can reduce PM emissions compared with using 0.50% sulfur VLSFO, this reduction would be overestimated if scrubber parameters are adjusted to allow higher emissions outside of ECAs. Likewise, our finding that PM emissions for ships using 2.6% sulfur HFO with a scrubber were nearly 70% higher than MGO, on average, would be an underestimate, meaning that PM emissions from ships using HFO with scrubbers could be even higher on the high seas.

For climate pollutants, including CO<sub>2</sub> and black carbon (BC), using HFO with scrubbers results in higher emissions than MGO. Average CO<sub>2</sub> emissions were 4% higher using HFO with a scrubber compared with MGO. BC emissions using HFO with a scrubber were expected to be 81% higher than using 0.07% sulfur MGO in a medium-speed diesel (MSD) engine and more than 4.5 times higher than using MGO in a slow-speed diesel (SSD) engine. This is because both MSD and SSD engines emit substantially more BC emissions when using residual fuels such as HFO compared with distillate fuels like MGO (Comer, Olmer, Mao, Roy, & Rutherford, 2017; Faber et al., 2020; Olmer, Comer, Roy, Mao, & Rutherford, 2017). Therefore, even though the scrubber removes some BC from the exhaust (roughly 10%), ships using HFO with scrubbers still emit more BC than those using MGO.

Emissions of NO<sub>x</sub> were sometimes lower and sometimes higher after the scrubber; however, based on the studies we reviewed, we found the average effect to be 0%. We do not expect scrubbers to have a significant direct impact on NO<sub>x</sub> emissions because

NO<sub>x</sub> formation is more sensitive to other parameters, including combustion temperature. We also found that scrubbers seem to somewhat reduce carbon monoxide (CO) emissions (-11% on average) across fuels. The mechanism by which scrubbers reduce CO emissions deserves further investigation. Based on these findings, Table 6 provides recommended emission factors for ships using HFO in combination with scrubbers.

**Table 6.** Recommended emission factors (g/kWh) for ships using HFO + scrubbers

Engine type	Engine age	SFOC (g/kWh)	Sulfur content	Carbon factor, Cf (gCO <sub>2</sub> /g fuel)	Engine RPM	SO <sub>2</sub>	CO <sub>2</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	NO <sub>x</sub>	CO	BC
SSD	<1984	205	2.60%	3.114	<130	0.19	650	0.30	0.28	18.2	0.48	0.04
SSD	1984-1999	185	2.60%	3.114	<130	0.17	586	0.30	0.27	18.2	0.48	0.03
SSD	2000-2010	175	2.60%	3.114	<130	0.16	554	0.30	0.27	17.1	0.48	0.03
SSD	2011-2015	175	2.60%	3.114	<130	0.16	554	0.30	0.27	14.5	0.48	0.03
SSD	2016+ outside ECA	175	2.60%	3.114	<130	0.16	554	0.30	0.27	14.5	0.48	0.03
SSD	2016+ in ECA	175	2.60%	3.114	<130	0.16	554	0.30	0.27	3.4	0.48	0.03
MSD	<1984	215	2.60%	3.114	720	0.20	681	0.30	0.28	14.1	0.48	0.09
MSD	1984-1999	195	2.60%	3.114	720	0.18	618	0.30	0.28	14.1	0.48	0.08
MSD	2000-2010	185	2.60%	3.114	720	0.17	586	0.30	0.27	12.1	0.48	0.08
MSD	2011-2015	185	2.60%	3.114	720	0.17	586	0.30	0.27	9.7	0.48	0.08
MSD	2016+ outside ECA	185	2.60%	3.114	720	0.17	586	0.30	0.27	9.7	0.48	0.08
MSD	2016+ in ECA	185	2.60%	3.114	720	0.17	586	0.30	0.27	2.4	0.48	0.08

In Table 7, we have estimated the expected life-cycle CO<sub>2</sub> emissions from ships using HFO with scrubbers compared with other fuels. We have taken into account the relative energy density and carbon factor (Cf) of each fuel based on the *Fourth IMO Greenhouse Gas Study* (Faber et al., 2020). We have also shown how SFOC changes based on fuel type and whether or not a scrubber is used. We assumed an SSD engine built in the year 2001 or newer. These SFOCs are consistent with the *Fourth IMO Greenhouse Gas Study*. We have added a 2% fuel consumption increase for HFO with scrubbers compared with HFO without scrubbers, consistent with our findings in Table 5, which show that using HFO with a scrubber emits 2% more CO<sub>2</sub> emissions than HFO without a scrubber. For VLSFO, we assume that it is an 80/20 blend of MGO and HFO to achieve a maximum 0.50% sulfur content.

Combustion emissions in grams of CO<sub>2</sub> per kilowatt-hour out (gCO<sub>2</sub>/kWh out) are calculated by multiplying Cf (gCO<sub>2</sub>/g fuel) by SFOC (g fuel/kWh out).

Upstream emissions (gCO<sub>2</sub>/kWh out) are calculated as follows:

$$U_{out} = U_{in} \times \frac{EC}{1000} \times SFOC$$

U<sub>out</sub> = upstream emissions (gCO<sub>2</sub>/kWh out)

$U_{in}$  = upstream emissions ( $gCO_2/MJ$  in) from GREET (Argonne National Laboratory, 2019), which is 13.5 for MGO and 10.7 for HFO; VLSFO is assumed to be 12.9, reflecting an 80/20 mix of MGO and HFO.

EC = energy content ( $kJ$  in/ $g$  fuel) as found in Table 7; dividing by 1,000 converts to units of  $MJ$  in/ $g$  fuel

SFOC = specific fuel oil consumption ( $g$  fuel/ $kWh$  out), which is listed by fuel in Table 7

As shown in Table 7, the expected combustion emissions for ships with HFO and scrubbers are higher than using MGO, while the upstream emissions are lower. Adding the two together, we find that the total well-to-wake (WtWa) emissions for a ship using HFO with a scrubber are expected to be 1.1% higher than using MGO.

**Table 7.** Life-cycle  $CO_2$  emissions for ships using HFO + scrubbers relative to other fuels

Fuel	Energy content ( $kJ/g$ fuel)	Cf ( $gCO_2/g$ fuel)	SFOC ( $g$ fuel/ $kWh$ out)	(gCO <sub>2</sub> /kWh out)			WtWa relative to MGO
				Combustion	Upstream	Well-to-wake (WtWa)	
MGO	42.7	3.206	165	529	95	624	0.0%
VLSFO	42.2	3.188	167	532	91	624	-0.1%
HFO	40.2	3.114	175	545	75	620	-0.6%
HFO + scrubber	40.2	3.114	178	554	77	631	+1.1%

## WATER POLLUTANTS

We reviewed 17 studies and found that only 10 had enough information to assess whether scrubber discharges were complying with IMO guidelines. We evaluated each study based on whether it included relevant information on the ship, fuel sulfur content, scrubber type, engines, engine operating parameters, discharge water flow rate, and transparency of results, as shown in Table 8.

With the exception of the BP-funded study by Hufnagel, Liebezeit and Behrends (2005), which contained complete details, many industry-funded studies such as Faber et al. (2019) and Carnival (2019) lacked the necessary information to determine the total mass of pollution discharges and to assess whether they satisfied IMO guidelines. For example, in Faber et al. (2019), 253 samples were analyzed, but only generalized information on ship types and engine loads at berth were provided. No flow rate was reported, which makes it impossible to determine if the discharges comply with the IMO guidelines. Nevertheless, Faber et al. (2019) improperly compared unadjusted per-liter concentrations of PAHs and other pollutants to the discharge criteria in the 2015 IMO guidelines; this was improper because they did not normalize the pollutant concentrations to a specific washwater flow rate. The IMO guidelines limit PAH concentrations to  $50 \mu g/L$  at a normalized washwater flow rate of  $45 t/MWh$ . Faber et al. (2019) explained that the PAH concentrations in their study “were not normalized” (p. 38). They used this to argue that the samples that had PAH concentrations greater than  $50 \mu g/L$  may still comply with the guidelines, when exactly the opposite could be true. Without normalizing the pollutant concentrations to a specific washwater flow rate, no conclusions can be drawn regarding compliance with, or exceedance of, IMO guidelines.

We omitted three other studies that were at least partially funded by industry. One from Japan's Ministry of Land, Infrastructure, Transport and Tourism (MLIT, 2018), as well as Wärtsilä (2010) and Koski, Stedmon, and Trapp (2017). While the MLIT (2018) study included information for many of the evaluation criteria, we could not fully understand the experimental set-up and therefore excluded it. From what we can understand, MLIT (2018) evaluated the characteristics of scrubber discharge water generated in the lab using a 257 kW, medium-speed laboratory engine and a hybrid scrubber. While MLIT (2018) provided measured values for certain discharge criteria, it was not clear if they related to open-loop or closed-loop operations, or what engine power and flow rate were associated with those values. Wärtsilä (2010) did not report measured values for any discharge criteria. Koski et al. (2017) did not provide information on the associated flow rate, making it impossible to calculate the total mass of pollutants discharged.

Government-funded studies typically contained more details, although some government-funded studies did not include enough information, including U.S. EPA (2011), which did not contain information on fuel type, sulfur content, or flow rate. Additionally, Ytrebert et al. (2019), which was funded by the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning, focused on how microplankton respond to scrubber discharge water exposures, rather than evaluating scrubber performance against IMO's discharge criteria. With these exceptions, government-funded studies were the most useful for this analysis. In some cases, such as Teuchies, Cox, Van Itterbeeck, Meyseman and Blust (2020), which was funded by the independent municipal Antwerp Port Authority, the study included detailed supplemental material containing raw data that was made publicly available. Except for Teuchies et al. (2020), the downside is that the government-funded studies often were limited in scope. Only a handful were able to measure more than one ship, and almost all measured a ship in European waters.

**Table 8.** Evaluation of the quality of sources containing information on scrubber discharges

Source	Includes ship ID information (e.g., IMO number)?*	Includes fuel type and sulfur content?	Includes scrubber type?	Includes engine power?	Includes flow rate?	Includes raw data?	Grade (% based on a max score of 12)	Enough information to be used in this study?	Funding source
Hufnagl, Liebezeit, & Behrends (2005)	2	2	2	2	2	2	100%	Y	Industry
Kjølholt et al. (2012)	2	2	2	2	2	2	100%	Y	Government
Zhu et al. (2016)	2	2	2	2	2	2	100%	Y	Government
Hansen (2012)	2	2	2	2	2	1	92%	Y	Government
Ushakov, Senersen, Einang, & Ask (2020)	2	2	1	2	2	2	92%	Y	Government
Wärstila (2010)	2	2	2	2	2	0	83%	N	Industry
Teuchies et al. (2020)	0	2	1	2	2	2	75%	Y	Government
Koski, Stedmon, & Trapp (2017)	2	2	2	2	0	1	75%	N	Government/Industry
Germany (2018)	0	2	2	0	2	2	67%	Y	Government
Winnes et al. (2018)	2	2	1	2	0	1	67%	Y	Government
Magnusson, Thor, & Grandberg (2018)	2	0	1	1	2	2	67%	Y	Government/Industry
Buhaug, Fløgstad, & Bakke (2006)	2	1	1	2	0	2	67%	Y	Government/Industry
MLIT (2018)	0	2	2	1	1	1	58%	N	Government/Industry
U.S. EPA (2011)	2	0	2	2	0	1	58%	N	Government
Ytreberg et al. (2019)	0	2	1	2	1	1	58%	N	Government
Carnival (2019)	0	0	1	0	0	1	17%	N	Industry
Faber et al. (2019)	0	0	1	0	0	1	17%	N	Industry

\*Grading scale for all criteria: 2 = all relevant data provided; 1 = some relevant data provided; 0 = no relevant data provided.

We identified 10 studies containing a total of 112 discharge samples that were of high enough quality to compare scrubber discharges to the discharge criteria in the IMO guidelines. In this section, we compare reported values from the literature against the discharge criteria for pH, PAH, turbidity, and nitrates contained in the draft 2020 guidelines, which can be found in document PPR 7/22/Add.1, annex 9. These are the same as the limits first established in the 2008 guidelines, which are found in Resolution MEPC.170(57).

**pH**

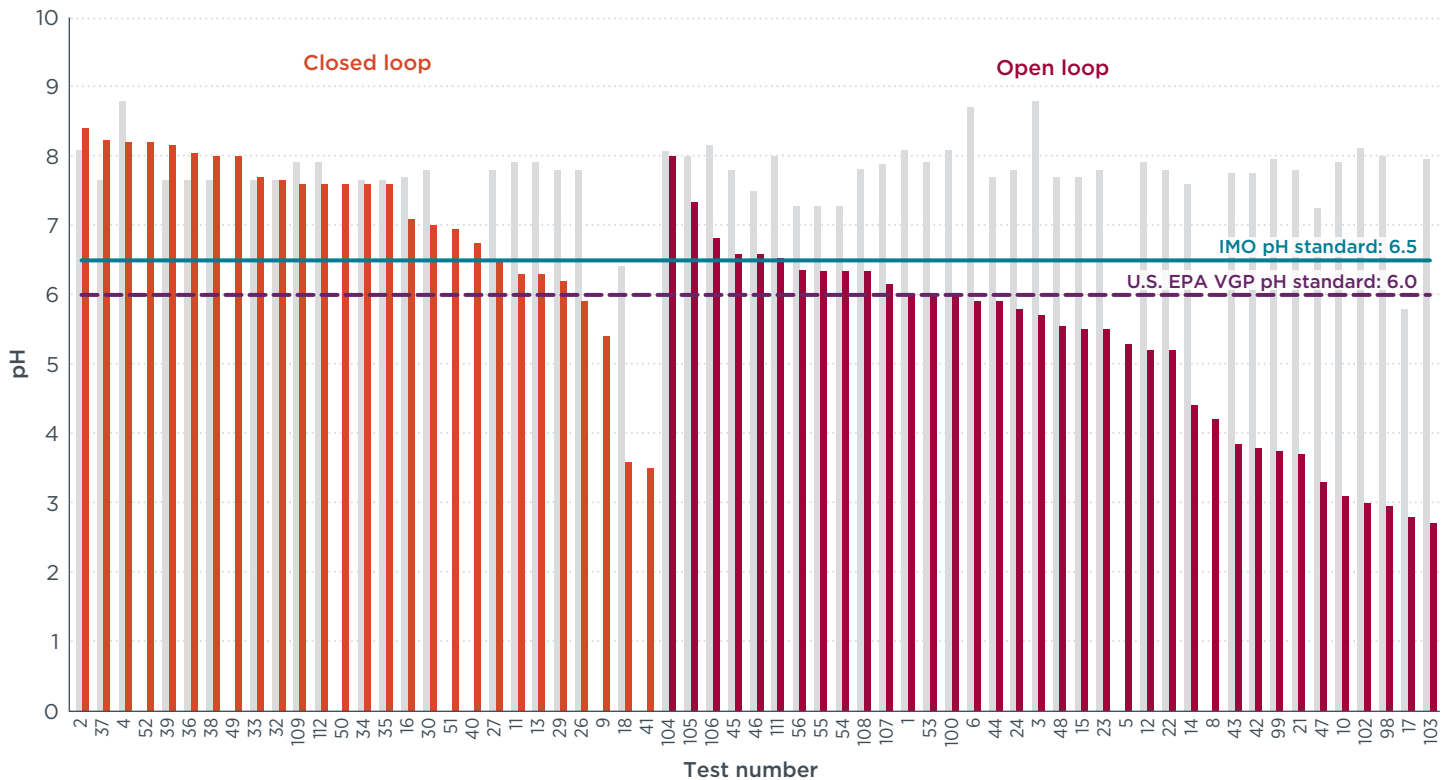
Ten studies representing 63 samples contained usable information on pH. The pH was measured at the overboard discharge point for all but one sample (test number 111). Twenty-seven samples were from closed-loop scrubbers, and 36 were from open-loop or hybrid scrubbers operating in open-loop mode. The pH was higher (less acidic) for closed-loop systems because the pH can be more directly controlled using alkaline materials, such as caustic soda, before discharging (Figure 3). The median pH for closed-loop systems was 7.59, while it was 5.63 for open-loop systems.

Of the 27 samples from scrubbers operating in closed-loop mode, all but seven had a pH  $\geq 6.5$ , which would comply with the IMO guidelines for when the ship is stationary. All but four samples had a pH  $\geq 6.0$ , which would comply with the EPA's 2013 VGP. It was not always clear in the literature if the ships were stationary, maneuvering, or in transit during the sampling. Nevertheless, all but one of the closed-loop samples also had a delta pH of less than 2, which would comply with both the IMO guidelines and the EPA 2013 VGP for ships that are maneuvering or in transit.

The pH was lower (more acidic) for open-loop systems, because the buffering solution is seawater, which has variable alkalinity. The pH also depends on the amount of reaction water, which is usually ambient seawater, mixed in before monitoring. As a result, only six out of 36 samples from open-loop scrubbers had a pH of  $\geq 6.5$ , while 14 had a pH  $\geq 6.0$ . Only 13 of 36 samples had a delta pH of less than 2, meaning that, had the ship been moving, 23 of 36 samples would have failed to comply with the IMO guidelines. Only one measurement in one study reported pH from a sample taken 4 meters away from the overboard discharge point (Ushakov et al., 2020); that was reported to have a pH of 6.52, which is high enough to comply with both the IMO guidelines and the EPA VGP.

Overall, closed-looped scrubbers performed the best in terms of pH, with 74% of samples having a pH  $\geq 6.5$  and 85%  $\geq 6.0$ . Additionally, 96% of closed-loop samples had a delta pH  $< 2$ . Open-loop scrubbers, on the other hand, performed poorly, with only 17% of samples having a pH  $\geq 6.5$  and 39% having a pH of  $\geq 6.0$ . Only 36% of open-loop samples had a delta pH less than 2. This is despite the practice of diluting the discharge with additional seawater before monitoring. Blending scrubber discharge water with ambient seawater prior to dumping it into the sea does not change how much acid is added to the surrounding waters; it merely raises the pH before it is monitored for comparison with the guidelines. Port State control officers may need to consider how to ensure that ships are complying with the delta 2 pH limit during maneuvering and transit of waters under their jurisdiction.





**Figure 3.** pH in scrubber discharge water. Gray bars show pH values before entering the scrubber system; orange and red bars show pH values after scrubbing process for closed- and open-loop scrubbers, respectively. Blue line indicates a pH of 6.5, consistent with IMO guidelines, and the purple dashed line is equal to a pH of 6.0, consistent with EPA 2013 VGP.

### PAHs

Four studies representing 60 samples contained usable information on PAHs. Ten samples were from closed-loop scrubbers, and 50 were from open-loop scrubbers or hybrid scrubbers operating in open-loop mode (Germany, 2018; Kjølholt et al., 2012; Teuchies et al., 2020; Ushakov et al., 2020). Nearly all samples were below the  $\text{PAH}_{\text{phe}}$  limit. At 50  $\mu\text{g}/\text{L}$  and 45 t/MWh, the maximum allowable discharge under the IMO guidelines is equivalent to 2,250,000  $\mu\text{g}/\text{MWh}$ . As shown in Figure 4, 93% of samples complied with the IMO guidelines (note the log scale). Open-loop scrubbers emitted greater amounts of  $\text{PAH}_{\text{phe}}$  compared with closed-loop systems, oftentimes an order of magnitude higher. The median  $\text{PAH}_{\text{phe}}$  value for closed-loop systems was 6,630  $\mu\text{g}/\text{MWh}$ , while it was 118,760  $\mu\text{g}/\text{MWh}$  for open-loop systems.

Only four samples exceeded the discharge criteria for  $\text{PAH}_{\text{phe}}$ , and they were from open-loop scrubber measurements taken on board ships by Germany's Federal Maritime and Hydrographic Agency (Germany, 2018). The report, which tested wastewater using onboard monitoring systems and additional in-situ measurements on board five ships, noted discrepancies between the two methods. It found that the onboard monitoring data showed lower  $\text{PAH}_{\text{phe}}$  values than the in-situ data. Worryingly, it also found that the onboard monitoring system seemed to be malfunctioning for two of the five ships, where PAH outlet concentrations were lower than inlet concentrations. This is highly unlikely, given that seawater has very low ambient concentrations of PAHs, so this suggested to the researchers that it was a calibration problem. While the onboard

monitoring never found exceedances of the PAH<sub>phe</sub> limits, the in-situ measurements showed that PAH<sub>phe</sub> concentrations were greater than 50 µg/L in seven out of nine tests (two tests for each of four ships, plus one test for the fifth), but this was without normalizing the results to 45 t/MWh, which is what the guidelines are based on. We normalized them and found that four test points were above the discharge criteria, as shown in Figure 4.

The remaining studies that recorded open-loop discharges (Kjølholt et al., 2012; Teuchies et al., 2020; Ushakov et al., 2020) found PAH<sub>phe</sub> emissions ranging from 7,000 to 1,600,000 µg/MWh, with an average of 900,000 µg/MWh. The large range indicates that open-loop PAH<sub>phe</sub> discharges are inconsistent.

The two studies that reported closed-loop scrubber PAH<sub>phe</sub> data (Germany, 2018; Teuchies et al., 2020) recorded PAH<sub>phe</sub> discharges from the bleed-off water to be below the IMO guideline limits, within the range of 1,800 to 24,000 µg/MWh. Germany (2018) tested one ship with a closed-loop scrubber and, like the open-loop scrubbers they evaluated, noted significant discrepancies between the ship's onboard monitoring and the in-situ measurements for the closed-loop PAH<sub>phe</sub> data. The in-situ PAH<sub>phe</sub> measurements were as much as 33 times higher than those reported by the onboard monitoring system. Teuchies et al. (2020) compared their closed-loop PAH<sub>phe</sub> measurements with the water quality standards of the European Water Framework Directive and noted that “the concentrations of most PAHs and all metals in closed loop bleed-off largely exceeded their WQS [water quality standards] and are expected to be acutely toxic for most aquatic organisms” (Teuchies et al., 2020, p. 7).

As previously mentioned, the current IMO guidelines are based on PAH<sub>phe</sub>. Phenanthrene, which is a molecule of three fused benzene rings and is classified as a low molecular weight PAH of 178 g/mol, is one of 16 PAHs that is customarily analyzed. Out of the 16 PAHs, the molecular weights range from 128 g/mol for 2-ring naphthalene, to 276 g/mol for 6-ring Benzo[g,h,i]perylene. The tendency to bioaccumulate and to resist biodegradation generally increases with increasing molecular weight (Adeniji, Okoh, & Okoh, 2018). Selecting phenanthrene as the surrogate for all PAHs in discharge water has unclear origins. According to the U.S. EPA, the IMO's basis for selecting PAH<sub>phe</sub> seems to be based on the fact that phenanthrene was found to be the most abundant PAH in the analysis of washwater during trials on the vessel *Pride of Kent*, which is reviewed in this report as Hufnagl et al. (2005). Recall that the United Kingdom used the *Pride of Kent* data in the submission to MEPC 55 that suggested a 30 ppb (~30 µg/L) limit for PAHs.

The U.S. EPA seems to find the IMO guidelines inadequate, given that monitoring 16 criteria PAHs is required in the 2013 EPA VGP. Bosch et al. (2009) critiqued the idea of “phenanthrene equivalents” as a proxy for measuring hydrocarbon emissions (i.e., oil), stating that the concept needs to be explained or replaced, due to the unknown amounts of other PAHs being emitted. Additionally, PAHs, phenanthrene and otherwise, are difficult to analyze on board. In some studies, discharge water samples were taken from the site and chemically analyzed in a lab. The onboard measurements depend on the measurement of the phenanthrene fluorescent intensity, and the results of that are dependent on the solubility of PAH<sub>phe</sub> and proper calibration of the instrument (Tomioka & Hashima, 2019). Germany (2018) suggested higher calibration and maintenance frequency of the systems for onboard measurements after seeing the large discrepancies in detail between onboard and laboratory analyses.

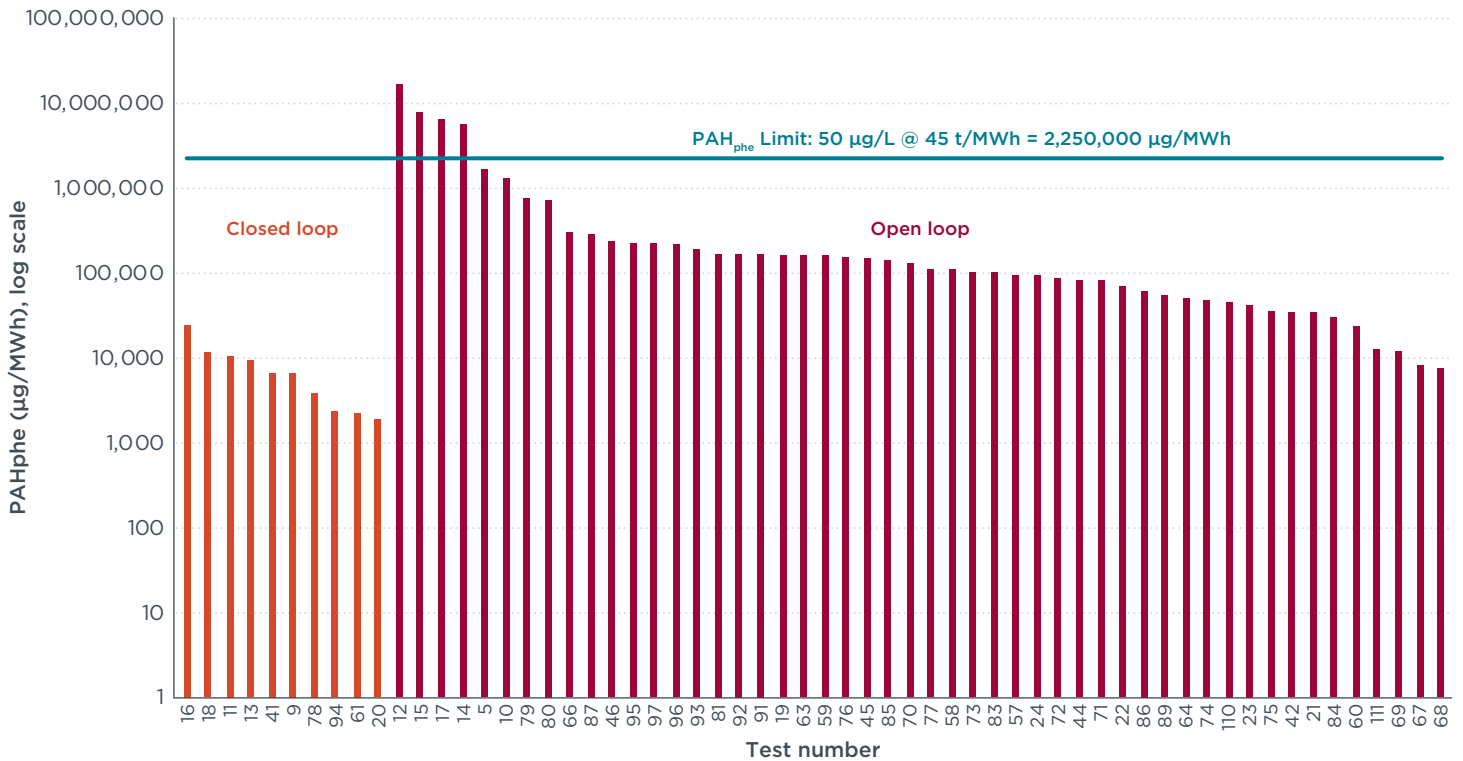


Figure 4. PAH<sub>phe</sub> in scrubber discharge water.

### Turbidity

Six studies representing 17 samples contained usable information on the turbidity of scrubber discharge water. Eight samples were from closed-loop scrubbers, and nine from open-loop or hybrid scrubbers operating in open-loop mode. The median turbidity for closed-loop systems was 9.9 NTU and it was 1.1 NTU for open-loop systems.

Closed-loop discharges had higher turbidity than open-loop discharges. It may be that there is higher turbidity in the closed-loop bleed-off water because it is more highly concentrated than open-loop discharges. It could also be that because water is recirculated, it becomes more turbid over time, despite water treatment designed to remove suspended solids as sludge.

The turbidity measurement units (FNU and NTU) both measure turbidity based on light scattering, although FNU uses infrared light and NTU uses white light. Two studies, Hansen (2012) and Ushakov et al. (2020), measured turbidity using FNU (see test numbers 100 for Hansen and 110 and 111 for Ushakov et al.). The one sample that measured above the IMO guideline's discharge criteria of 25 came from Germany (2018), which found an increase of 26.6 NTU from inlet water to outlet water for a closed-loop scrubber. Magnusson et al. (2018) found that the water treatment system used to collect residues from the closed-loop system they tested reduced turbidity in the discharge 96%, but even then the overboard discharge was at least 7.3 NTU higher than the surrounding seawater. Because no zeros were recorded, every discharge increased turbidity compared with the ambient seawater.

The IMO guidelines state that “the discharge water treatment system should be designed to minimize suspended particulate matter, including heavy metals and ash”

(PPR 7/22/Add.1, annex 9, p. 21). In practice, while closed-loop scrubbers intentionally separate out suspended particulate matter and store it onboard as sludge for on-land disposal, open-loop systems typically do not. A survey of scrubber manufacturers showed that open-loop systems typically do not collect sludge, implying that suspended particulate matter, including heavy metals and ash, are discharged overboard and not actually passed through a water treatment system (European Sustainable Shipping Forum, 2017). If solids were separated out, turbidity would be reduced, and heavy metals could be reduced as well because they can be attached to suspended solids. However, because the discharge water has a lower pH, metals can more easily dissolve into the water, rather than being held in the sediments. This was seen in a study by Wärtsilä (2010), which found high concentration of metals even though turbidity was well below the IMO discharge criteria. The U.S. EPA (2011) noted that there is no correlation between turbidity and particle concentration. Ushakov et al. (2020) questioned the scientific significance of measuring turbidity. They noted that the measured values depend on the scattering of light and the light source used, which can be influenced by seawater organics. Smaller particles in the discharge water would have low influence on the turbidity and could be missed, even though they may be contributing to pollution. Lastly, bubbles were a common source of interference in several studies, including Zhu et al. (2016), U.S. EPA (2011), and Wärtsilä (2010).

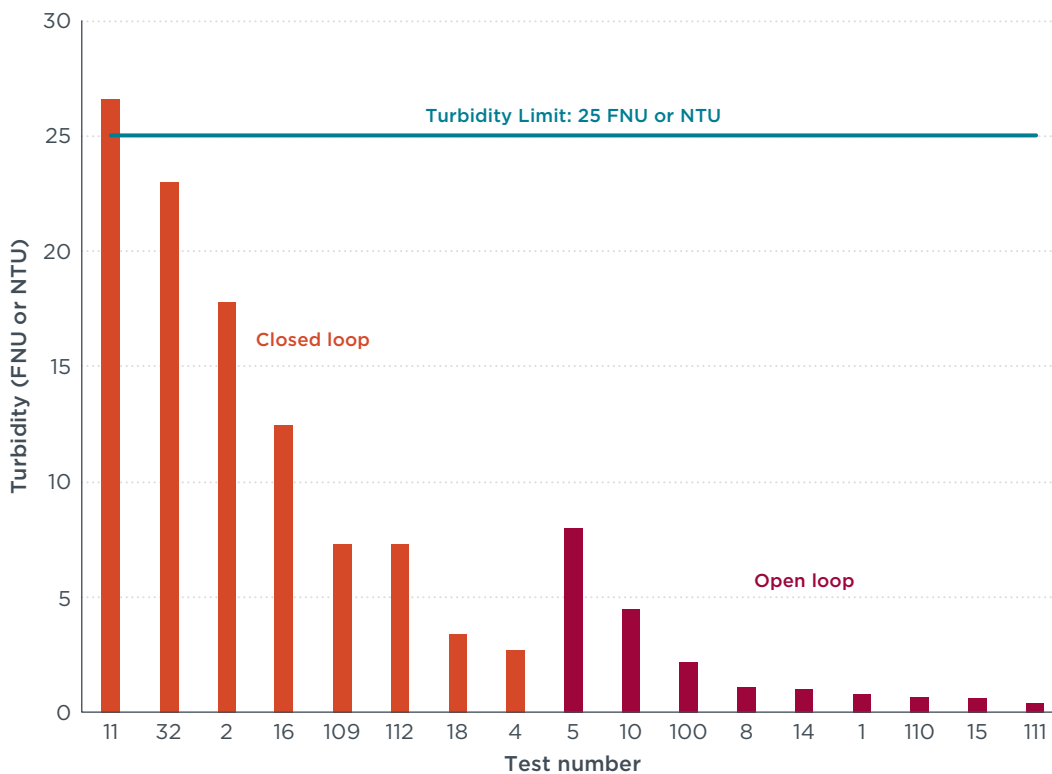


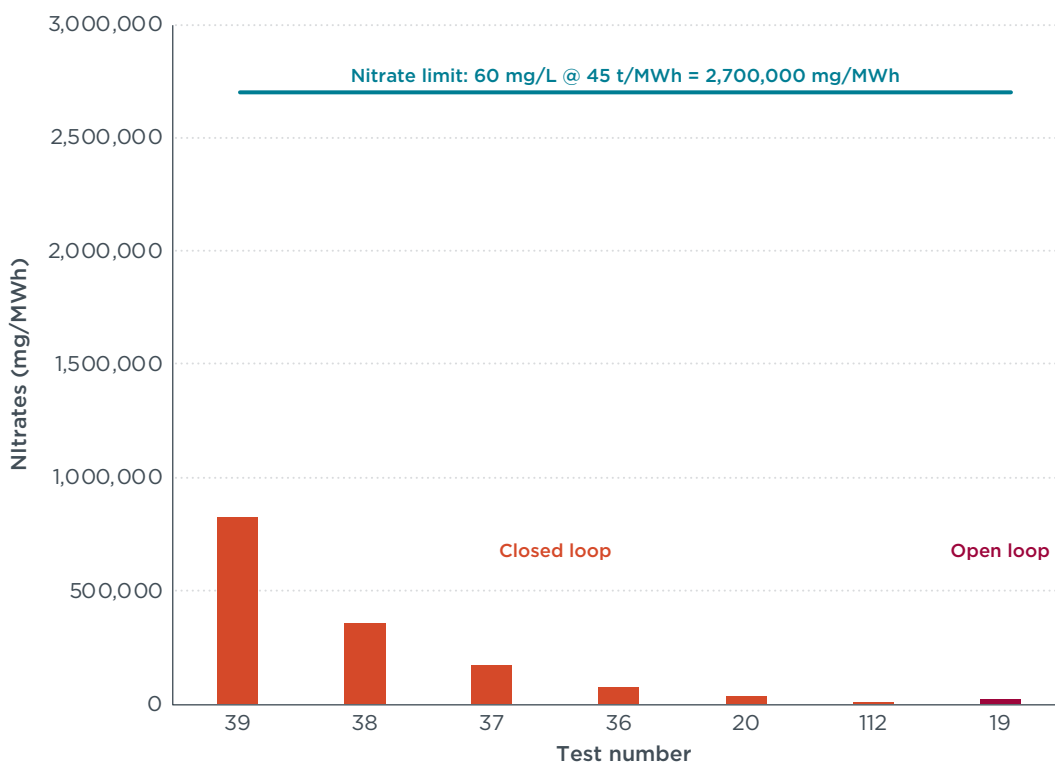
Figure 5. Turbidity in scrubber discharge water.

### Nitrates

Four studies representing seven samples reported nitrates (Germany, 2018; Kjølholt et al., 2012; Magnusson et al., 2018; Zhu et al., 2016), and all but one were from closed-loop systems. No samples exceeded the IMO guidelines discharge criteria for nitrates, which at 60 mg/L at 45 t/MWh is equivalent to 2,700,000 mg/MWh. Given that there was only

one value associated with open-loop discharges, it is not possible to compare discharge values between closed-loop and open-loop systems in detail. The median closed-loop discharge was approximately 125,000 mg/MWh. The sole open-loop discharge is 19,800 mg/MWh.

Nitrates and nitrites are essential for marine primary production, but an excess can accelerate eutrophication. Washwater discharges contain both nitrates and nitrites; however, the IMO guidelines cover only nitrates. The United States, in its 2013 VGP, requires ships to meet the same standard as the IMO guideline for nitrates, but it is the sum of nitrates and nitrites. Nevertheless, we have shown that scrubber discharges do not usually contain enough nitrates to exceed the limit in the IMO guidelines.

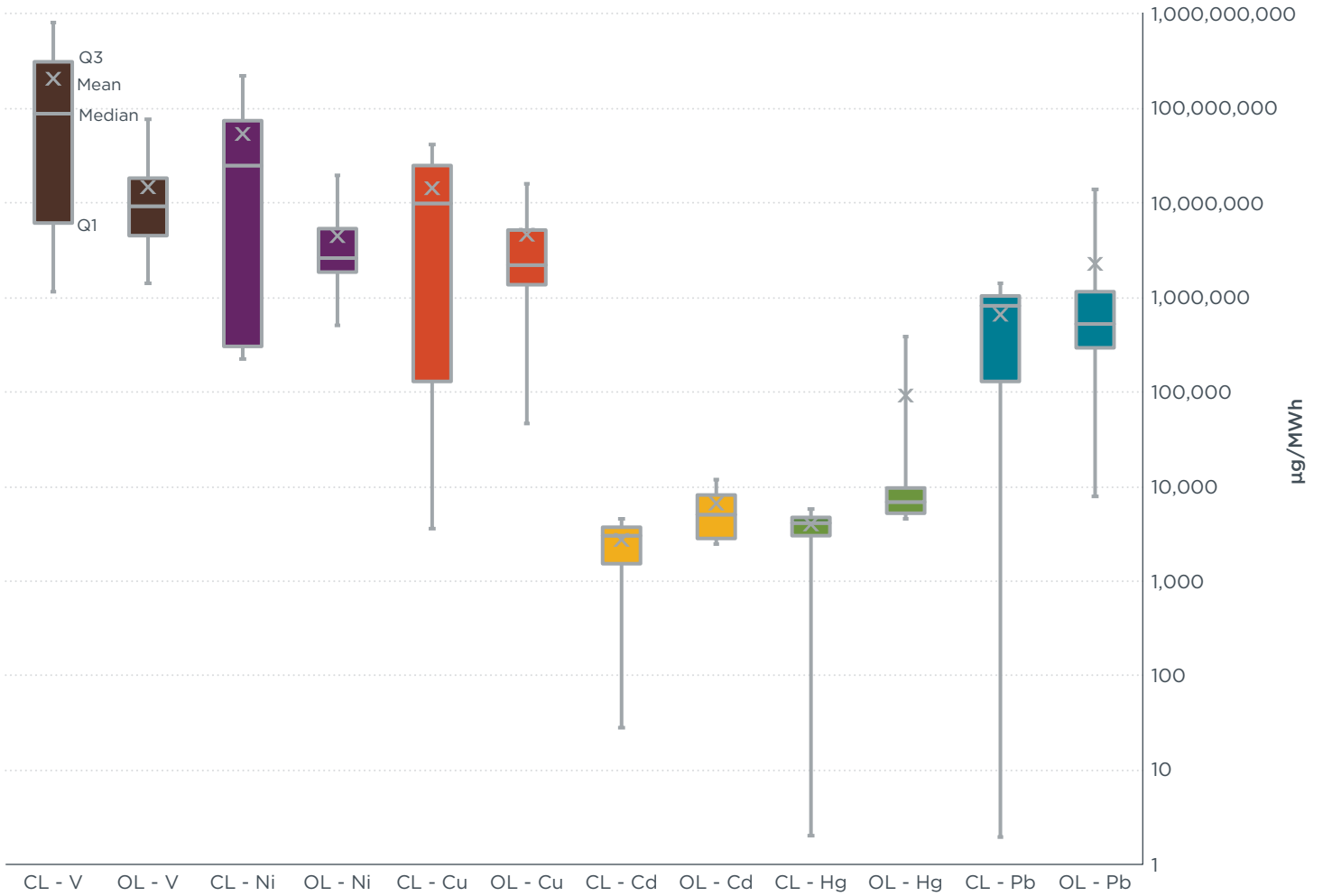


**Figure 6.** Nitrates in scrubber discharge water.

### **Heavy metals**

We evaluated discharges of six heavy metals: vanadium, nickel, copper, cadmium, mercury, and lead. We found seven studies, representing 58 samples, that had reported values for at least one of these metals. Vanadium, which is found in HFO, was the most studied metal with 58 samples, 46 being from open-loop mode. As shown in Figure 7, vanadium had the highest average discharges of the metals studied, with closed-loop systems emitting more than open-loop, but the open-loop discharge values showed less variability. Nickel and copper displayed similar patterns of higher, more varying average values in closed-loop mode, but vanadium was discharged at significantly higher amounts than nickel and copper (note the log scale). Other metals, such as cadmium, mercury, and lead, were observed in smaller amounts, but had higher average discharges from open-loop scrubbers than closed loop. Open-loop discharges are more acidic, which could lead to larger amounts of dissolved heavy metals in the discharge water.

However, it appears that per MWh, closed-loop systems contribute greater mass of heavy metals than open-loop systems. With that said, additional work is needed to fully understand why closed-loop discharges exhibit greater variability. Currently there are no IMO guidelines for any heavy metal.



	CL - V	OL - V	CL - Ni	OL - Ni	CL - Cu	OL - Cu	CL - Cd	OL - Cd	CL - Hg	OL - Hg	CL - Pb	OL - Pb
<b>Q1</b>	6.2E+06	4.5E+06	3.1E+05	1.8E+06	1.3E+05	1.4E+06	1.5E+03	2.9E+03	3.0E+03	5.3E+03	1.3E+05	2.9E+05
<b>Median</b>	8.9E+07	9.3E+06	2.5E+07	2.6E+06	1.0E+07	2.2E+06	3.0E+03	5.0E+03	4.1E+03	6.8E+03	8.2E+05	5.2E+05
<b>Q3</b>	3.1E+08	1.8E+07	7.3E+07	5.3E+06	2.5E+07	5.2E+06	3.8E+03	8.3E+03	4.7E+03	9.8E+03	1.0E+06	1.1E+06
<b>Mean</b>	2.1E+08	1.5E+07	5.4E+07	4.4E+06	1.4E+07	4.5E+06	2.5E+03	6.1E+03	3.5E+03	8.4E+04	6.5E+05	2.2E+06

**Figure 7.** Heavy metal discharges (µg/MWh) for closed-loop (CL) and open-loop (OL) scrubbers, with values in the table. The box shows the interquartile range. The whiskers show the minimum and maximum values. The mean is marked by the X and the median is the horizontal line inside each box.

Table 9 includes recommended scrubber discharge water emission factors for each pollutant. They are based on rounded median values from the results presented in this

section. Some emission factors are more certain than others. We found more data on pH, PAHs, and heavy metals, but less on turbidity and nitrates. The open-loop nitrate emission factor is based on one measurement and should be considered the least certain. On the other hand, the PAH<sub>phe</sub> open-loop emission factor is based on 50 samples and should be considered the most certain. These emission factors can be used to get an understanding of the magnitude of water pollution from scrubbers, as well as trends over time. They will be particularly useful if paired with geospatial ship activity data so that the location and amount of discharges can be estimated. This could help determine the amount of pollution in ports, harbors, estuaries, rivers, critical habitats for marine life, Marine Protected Areas, Particularly Sensitive Sea Areas, and other areas of interest.

**Table 9.** Recommended scrubber discharge water emission factors

Scrubber mode	pH	PAH <sub>phe</sub> (µg/MWh)	Turbidity (NTU)	Nitrates (mg/MWh)	Heavy metals (µg/MWh)					
					Vanadium	Nickel	Copper	Cadmium	Mercury	Lead
<b>Closed loop</b>	7.6	6,600	10	125,000	88,850,000	24,540,000	9,990,000	3,000	4,000	818,000
<b>Open loop</b>	5.6	119,000	1	20,000	9,310,000	2,590,000	2,180,000	5,000	7,000	519,000

## CONCLUSIONS

This report assessed the impacts of scrubbers on air emissions and water pollution. Regarding air emissions, we found that scrubbers can substantially reduce  $\text{SO}_2$  emissions, with emissions from ships using 2.6% sulfur HFO with a scrubber averaging 31% lower than 0.07% sulfur MGO. We also found that scrubbers seem to somewhat reduce CO emissions (-11% on average), although the mechanism by which this occurs deserves further investigation. For other pollutants, including  $\text{CO}_2$ , PM, and BC, using HFO with scrubbers results in higher emissions than MGO. Average  $\text{CO}_2$  emissions were 4% higher using HFO with a scrubber compared with MGO. On a life-cycle basis, well-to-wake  $\text{CO}_2$  emissions are expected to be 1.1% higher than using MGO. PM emissions from using HFO with a scrubber were approximately 70% higher than MGO, on average. BC emissions using HFO with a scrubber were expected to be 81% higher than using MGO in an MSD engine and more than four times higher than using MGO in an SSD engine. Emissions of  $\text{NO}_x$  were sometimes lower and sometimes higher after the scrubber; however, based on the studies reviewed, we found the average effect to be 0%. We do not expect scrubbers to have a significant direct impact on  $\text{NO}_x$  emissions because  $\text{NO}_x$  formation is more sensitive to other parameters, including combustion temperature.

Regarding water pollutants, we found that all scrubbers—open loop, closed loop, and hybrid—discharge water that is more acidic and turbid than the surrounding water. Additionally, all scrubbers emit nitrates, PAHs, and heavy metals. The acids that scrubbers emit contribute to ocean acidification. Discharge from open-loop scrubbers was typically more acidic than bleed-off water discharges from closed-loop systems. Turbid water degrades water quality and the suspended PM in turbid water can contain PAHs and heavy metals. We found that closed-loop bleed-off water was more turbid than open-loop discharges. We did not have enough information to determine which system—open or closed—emits more nitrates. Discharging nitrates contributes to acidification and can lead to eutrophication.

The amount of pollution that is discharged, as well as its ecological impacts, will depend on the characteristics of the inlet and receiving waters. Ships use scrubbers not only on the open ocean, but also in places with brackish and fresh water; in Canada, these include the St. Lawrence and Fraser estuaries, as well as the Great Lakes. Brackish and fresh waters are less alkaline than sea water, and this can affect the performance of the scrubbers. These waters may also already be contaminated by PAHs and heavy metals, meaning scrubber discharges will add additional pollution burdens to marine life. PAHs are carcinogenic and heavy metals are toxic, and both can accumulate in the water, sediments, and marine life. They bioaccumulate up the food chain and have been linked to cancer and immune system suppression in marine mammals including in killer whales and belugas. Open-loop systems emit substantially more PAHs than closed-loop systems, often orders of magnitude higher, whereas closed-loop systems tended to emit more heavy metals; this is an unexpected finding, given that closed-loop systems are meant to collect PM, which could include heavy metals, in onboard sludge tanks. One possible explanation is that the recirculating water collects more heavy metals before it is discharged as bleed-off. However, we found that the variability in closed-loop heavy metal discharges was greater than open-loop systems. Therefore, more work is needed to fully understand if open-loop or closed-loop systems emit different amounts of heavy metals.



In general, scrubber discharges from both open-loop and closed-loop systems usually comply with IMO guidelines. However, we question whether complying with the IMO guidelines should be taken as evidence that scrubbers are doing no harm to the aquatic environment. We discovered that the discharge criteria set out in IMO's guidelines were weakened at the very first opportunity. The first IMO scrubber guidelines were set in 2005 and did not include numeric discharge criteria but did state that pollutants should be eliminated or reduced to a level at which they are not harmful. Since then, the guidelines have only been weakened. The first numeric discharge criteria for pH, PAHs, turbidity, and nitrates were included in the 2008 guidelines, which were adopted by MEPC 57. The pH, PAH, and nitrate discharge criteria that were ultimately agreed to by MEPC 57 based on the outcomes of BLG 12 were substantially weaker than those proposed by the second intersessional BLG Working Group on Air Pollution (BLG-WGAP 2). Neither BLG 12 nor MEPC 57 gave any explanation for why these criteria were weakened from those proposed by the intersessional working group.

One could consider these results and conclude that the IMO guidelines simply need to be reviewed again and strengthened. However, we would argue that history has shown that the IMO guidelines were established at a limit that ensures that scrubber technologies can meet them. Given opportunities to strengthen the discharge criteria in 2009, 2015, and 2020, IMO member states declined, citing too little scientific evidence to revise them. The result is that the discharge criteria have not been strengthened since they were established. Meanwhile, the number of ships with scrubbers has grown exponentially, from three ships in 2008 to more than 4,300 in 2020. The guidelines ignore the cumulative effects of many ships operating and discharging in heavily trafficked areas, something to be expected given this rapid increase in the number of ships with scrubbers. Given that the IMO completed its most recent review of the guidelines at PPR 7 in 2020 and that MEPC will likely adopt them without further revision, we do not expect another opportunity to review and revise the discharge criteria at the IMO level for at least several years. During that time, thousands of ships will continue to use scrubbers that are designed to discharge acids, nitrates, solid particles, PAHs, and heavy metals to the marine environment, including in ports, harbors, estuaries, near shore areas, and busy shipping lanes where the combined effects could rapidly accumulate. This includes places like the Great Lakes, as well as British Columbia and the St. Lawrence estuary, where endangered species like the Southern Resident killer whale and belugas already suffer from high levels of contamination, including from PAHs and heavy metals.

We recommend that individual governments continue to take unilateral action to restrict or prohibit scrubber discharges from both open-loop and closed loop systems. We also recommend that the IMO focus on harmonizing rules for scrubber discharges including where, when, and even if those discharges should be allowed, and to do so with urgency. The IMO should consider prohibiting the use of scrubbers as a compliance option for newbuild ships and work to phase out scrubbers installed on existing ships. This is because we have found that using HFO with scrubbers is not equivalently effective at reducing air pollution compared to using lower sulfur fuels, such as MGO. Additionally, scrubbers of all kinds (open, closed, and hybrid) directly contribute to ocean acidification and water pollution, whereas lower sulfur fuels do not. Until then, we recommend that individual countries, including Canada, take immediate actions to protect their air and waters from scrubber emissions and discharges. These actions could include one or both of the following: (1) an immediate prohibition on using

scrubbers to comply with the Canadian portion of the North American ECA because they are not equivalently effective at reducing air pollution as ECA-compliant fuels; (2) an immediate prohibition on all scrubber discharges in Canadian ports, internal waters, and territorial seas because they contribute to acidification and water pollution that can negatively affect marine life.

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# Three decades of trace element sediment contamination: The mining of governmental databases and the need to address hidden sources for clean and healthy seas

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## ABSTRACT

Trace elements (TEs) frequently contaminate coastal marine sediments with many included in priority chemical lists or control legislation. These, improved waste treatment and increased recycling have fostered the belief that TE pollution is declining. Nevertheless, there is a paucity of long-term robust datasets to support this confidence. By mining UK datasets (100s of sites, 31 years), we assess sediment concentrations of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) and use indices (PI [Pollution], TEPI [Trace Element Pollution] and  $I_{geo}$  [Geoaccumulation]) to assess TE pollution evolution. PI and TEPI show reductions of overall TE pollution in the 1980s then incremental improvements followed by a distinct increase (2010–13). Zn, As and Pb  $I_{geo}$  scores show low pollution, whilst Cd and Hg are moderate, but with all remaining temporally stable.  $I_{geo}$  scores are low for Ni, Fe and Cr, but increasing for Ni and Fe. A moderate pollution  $I_{geo}$  score for Cu has also steadily increased since the mid-1990s. Increasing site trends are not universal and, conversely, minimal temporal change masks some site-specific increases and decreases. To capture this variability we strongly advocate embedding sufficient sentinel sites within observation networks. Decreasing sediment pollution levels (e.g. Pb and Hg) have been achieved, but stabilizing  $I_{geo}$  and recently increasing TEPI and PI scores require continued global vigilance. Increasing Ni and Fe  $I_{geo}$  scores necessitate source identification, but this is a priority for Cu. Local, regional and world analyses indicate substantial 'hidden' inputs from anti-fouling paints (Cu, Zn), ship scrubbers (Cu, Zn, Ni) and sacrificial anodes (Zn) that are also predicted to increase markedly. Accurate TE input assessments and targeted legislation are, therefore, urgently required, especially in the context of rapid blue economic growth (e.g. shipping).

## 1. Introduction

Coastal regions contain some of the most ecologically productive habitats and are critical for their goods and services (Hassan et al., 2005; Watson et al., 2020). Yet, coastal sediments are often heavily contaminated by metals and metalloids from human activities, leading to sediment accumulation (Bryan and Langston, 1992) and human health risks from bioaccumulation (e.g. Liu et al., 2020). These trace elements (TEs) are also some of the most significant aquatic pollutants with Johnson et al. (2017)

placing 10 in the top 20 most toxic substances. To reduce concentrations and toxicological effects and to achieve healthy and clean oceans many TEs are included in priority chemical lists (e.g. US EPA, 2015) or other legislation (e.g. EU, 2000). In addition, governments have expected that technological advances in manufacturing and improved waste management and recycling will also reduce inputs. This has fostered a belief in some that TE contamination is generally resolved (i.e. a legacy); compounded by attention being focused elsewhere (e.g. microplastics).

Biomarker programmes such as 'Mussel Watch' can provide evidence

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of TE temporal decline, yet they have significant biological limitations (see Farrington et al., 2016). Selected bivalves are also ‘disconnected’ from benthic sediment habitats, for example, mussels attach to hard substrates in the water column (Gosling, 1992). Bivalves play a significant role in assessing contamination (see Beyer et al., 2017), but their utility lacks sediment-specific habitat relevance. Without a recognised mussel-equivalent sentinel infaunal species, research has focused on sediment matrix concentrations. One approach is to reconstruct historical contamination using sediment cores (e.g. Kang et al., 2018). However, frequent human disturbance disrupts the profile; limiting relevance to areas of minimal anthropogenic activity. Long term datasets that have been analysed are very locally focused (i.e. within one estuary) and/or have a limited sampling timescale (e.g. Traven et al., 2015; Ribeiro et al., 2018). Even those at the *trans*-national level are also spatially and temporally limited. For example, OSPAR (2019a) only uses the Marine Environment Monitoring and Assessment National (MERMAN) database (see below), preventing potential comparison and augmentation with examples from regional scale inshore and coastal areas. Thus globally, there is an urgent need for assessments over appropriate spatial (i.e. 1000s km of coastline and open ocean) and temporal (multiple decades) scales to confirm TE concentration reductions and, therefore, that the ecological health of these systems is improving.

UK coastal and marine monitoring data are held in two key public repositories: a) the Environment Agency (EA) and b) the MERMAN database managed by the British Oceanographic Data Centre (BODC) under the Clean Safe Seas Environmental Monitoring Programme (CSEMP) (Cefas, 2012). From these we generated a merged EA-MERMAN database focused on the Channel, southern North and Celtic Seas ensuring we reflect the global spectrum of contamination levels (e.g. Bryan and Langston, 1992; Haynes et al., 1995; Caplat et al., 2005; Naidu et al., 2012; Pan and Wang, 2012; Qian et al., 2015). This area also contains current and historical examples of nearly all direct anthropogenic TE inputs and key atmospheric sources (Rainbow, 2018; OSPAR, 2019a,b). By mining these data sources through our merged dataset, we first assess the evolution of sediment contamination for nine TEs (As, Cd, Cr, Cu, Fe, Pb, Hg, Ni and Zn) across multiple sites and decades. Our subsequent application of pollution indices at a broad spatial scale and over an extended period is pertinent to TE persistence, aligns with national and international legislative developments and incorporates decadal technological changes.

Whilst the inputs of contaminants from direct, riverine and atmospheric anthropogenic sources have been regularly estimated, e.g. see OSPAR (2019b,c) and Richmond et al. (2020) for the UK; recently Tornero and Hanke (2016) identified sea-based sources as also potentially significant contributors. From the eight sources stated we selected shipping due to its prominence in the Channel region. In addition, supporting data for shipping would likely be available to calculate input estimates. Thus, our final aim is to quantify TE inputs from three fundamental, but rarely studied components (termed ‘hidden’ here), of shipping activity: a) anti-fouling coatings, b) ship exhaust cleaning systems (termed scrubbers), and c) sacrificial hull anodes at local, national and global spatial scales and then to predict inputs for 2040. Combined, these data deliver critical information for evaluating the current policies for some of the most toxic chemicals delivered to marine benthic systems. Data provide a road map for identifying best practice for using existing databases for contamination studies. Outputs will also be key for updating legislative frameworks for achieving clean and healthy seas now, but most importantly, in the future as global blue economic investment (with shipping having a pivotal role) is predicted to expand rapidly (OECD, 2016).

## 2. Material and methods

### 2.1. Data mining

Upon request the EA provided both inland and marine data for

sediment and non-sediment samples for South and South West English regions. To obtain data from only transitional, estuarine and coastline waters, georeferenced sites were mapped (Google Earth, Google LLC). An iterative process was required for quality assessment including geographic locations and sample material codes etc. The BODC dataset (from 1999) required less pre-processing with georeferenced MERMAN sites mapped. The two datasets were merged and then standardized (e.g. concentration units, duplicated sites). Our resulting EA-MERMAN database consists of 45,962 data points (334 sites) for 29 chemicals over 31 years (1983–2013). We selected the nine most monitored TEs, together representing 87% of data (320 sites). Finally, to remove outliers (i.e. samples with extremely high values) all TE values higher than or equal to 99.85 quantile were removed (approximately seven data points per TE).

### 2.2. General sediment sample processing

Sediment samples collected for the MERMAN database were acquired and processed following guidelines (Cefas, 2012). Briefly, sediment samples were sieved (wet or dry) and the <63  $\mu\text{m}$  fraction retained for - usually - total digestion (td) using hydrofluoric acid (HF). The analytical technique chosen is not mandatory, but most laboratories now use Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for TE determination; Hg can be determined by cold vapour atomic absorption spectrometry or atomic fluorescence. The protocol for samples collected by the EA has evolved since the 1980s with the Cefas (2012) processes applicable, but to some data only (including coastal sediment data collected under the protocols, stored in both databases). EA sediment samples were formerly wet sieved and the <90  $\mu\text{m}$  fraction retained, but this was changed to retention of the <63  $\mu\text{m}$  fraction in the 1990s. To focus on TEs considered as biologically-available, extraction used strong acid digestion (sad) with hot  $\text{HNO}_3$  or aqua regia. When sediments were digested using HF, they were distinguishable in the EA database. HF, unlike hot  $\text{HNO}_3$ /aqua regia, dissolves TEs bound to silicate structures. However, we have considered hot  $\text{HNO}_3$ /aqua regia and HF digestions equivalent for Cu, Zn, As, Cd, Hg, Ni and Pb since those TEs are a small part of the sediment matrix; confirmed by comparing extraction efficiencies (Cook et al., 1997; Hall, 2017). For Fe and Cr, differences in sediment extraction efficiencies were > 10%, therefore, data from each extraction procedure were analysed separately. Our quality assurance processes ensured there was no step-change effect for changeover of analytical technique e.g. flame atomic absorption to ICP-MS (most laboratories); Hg would be analyzed with cold vapour technique (EA, pers. comm.). TE concentrations are given on a dry weight basis ( $\text{mg kg}^{-1}$ ). Median detection limits (DL, in  $\text{mg kg}_{\text{dw}}^{-1}$ ) were 0.1 for Cd, Cr (sad) and Hg, 1 for Cu, 2 for As, 3 for Fe (sad), 5 for Ni and Zn and 8 for Pb. For TE concentrations below the analytical procedure DL, we used half the DL values (US EPA, 1991).

### 2.3. Sediment quality guidelines (SQGs) and background levels

TE concentrations were compared to SQGs. Canadian SQGs consist of Interim Marine Sediment Quality Guidelines/Threshold Effect Levels (ISQG/TEL), thereafter called TEL and Probable Effect Levels (PEL). Below the TEL, adverse effects on the biota rarely occur; between the TEL and the PEL, adverse effects on the biota may occasionally occur; above the PEL, adverse effects on biota are frequent (CCME, 2001; Simpson et al., 2013). Although Hübner et al. (2009) stated that TEL/PEL-based SQGs are appropriate assessment criteria for contaminants in sediment with a proven scientific basis, Cefas (2012) considers as assessment criteria the Effects Range Low (ERL, developed by the US EPA [1991]) and the Background Assessment Concentrations (BAC) developed within the OSPAR Commission framework, where concentrations below the ERL rarely cause adverse effects in marine organisms and concentrations below the BAC are said to be near background.

Sediment contamination assessments require determination of TE



background concentrations (B) for uncontaminated sediments (Brady et al., 2015; Birch, 2017). Usually historical data are not available, hence a non-contaminated analogue is needed: unpolluted reference sites (e.g. Brady et al., 2015); crustal composition (e.g. Brady et al., 2015) or sedimentary rock composition (e.g. Dung et al., 2013). However, when the data represent a comprehensive cross section of a large region, B can also be defined as equal to a given dataset quantile. For example, Rainbow et al. (2011) calculated metal B values as equal to the 33rd percentile values of their concentration data and defined B as “typical” of the least disturbed areas in a region. Applying this here meant that most TEs were close to or above TEL values (S1 Table 1). Except in areas with naturally elevated background concentrations, baseline levels should not lead to any (or very little) adverse biological levels. Therefore, we have redefined B as the 20th percentile values of the concentration data, resulting in the B for As remaining slightly above the TEL, whilst all other TEs were below (S1 Table 1).

#### 2.4. Sediment quality indices

Among multiple sediment quality indices that calculate contamination, the use of the Contamination Factor (CF) and related 7-level Geoaccumulation Index ( $I_{geo}$ ) pollution scale are considered the preferred single-element indices (Brady et al., 2015). The  $I_{geo}$  pollution scale classifies sediments from unpolluted to very strongly polluted (Müller 1986).  $I_{geo}$  scale levels correspond to given CF values ( $I_{geo} = \log_2(CF/1.5)$ ). The CF of a TE  $n$  is the ratio between its concentration in the sediment  $C_n$  and its natural background concentration  $B_n$  ( $CF = C_n/B_n$ ) (Tomlinson et al., 1980) and the arbitrary 1.5 factor minimizes the impact of lithogenic enrichment and enrichment caused by sediment inputs from several sources. The relationship between CF values and  $I_{geo}$  scale levels is from Legorburu et al. (2013), but using a blue-red colour scale. The Nemerov Pollution Index (PI) (Nemerow, 1991) is the most comprehensive method for overall sediment quality and has been increasingly used in recent years (see Brady et al., 2015). PI uses the average of CF ( $CF_{average}$ ) of a suite of TEs focusing on the impact of contamination of one TE by using the maximum CF ( $CF_{max}$ ) ( $PI = ((CF_{average}^2 + CF_{max}^2)/2)^{1/2}$ ). The 5-level sediment quality scale classifies sediments from unpolluted ( $PI < 0.7$ ) to severely polluted ( $PI > 3$ ) (Brady et al., 2015; Qingjie et al., 2008). As the PI is weighted by the effect of a single TE and values are also strongly dependent on Bs, it may lead to an overestimation of the overall TE pollution status. A complementary index to the PI, is the Trace Element Pollution Index (TEPI) of Richir and Gobert (2014). TEPI does not rely on pre-determined background concentrations and does not give higher importance to a more polluting TE. TEPI is the weighted product of mean normalized (Moreda-Piñeiro et al., 2001) TE concentrations  $C_f$  of the  $n$  TE under study ( $TEPI = (C_{f1} * C_{f2} \dots C_{fn})^{1/n}$ ). This weighted index also enables reliable comparison of TE pollution levels even if the list of monitored TEs used differ (Richir and Gobert, 2014). The 5-level environmental quality scale of Richir et al. (2015) applied to TEPI values relies on the quantile method and classifies sediments from Very Low Contamination Level (VLCL) to Very High Contamination Level (VHCL). To utilize a more complete matrix for the PI and TEPI, site TE concentrations were averaged every five years (seven for 1983–1989, four for 2010–2013). The PI and TEPI were then calculated for all sites that, for those periods, had a minimum of six mean concentrations for the nine TEs monitored.

#### 2.5. Input estimates (antifouling paints, scrubbers and anodes)

The Solent separates the Isle of Wight from mainland England and is contained within the Channel; the water body between France and England and a component of the ecoregions of the Greater North Sea and Celtic Seas (ICES, 2020). Both the Channel and Solent are protected by many conservation designations, but also host several international commercial ports and high levels of recreational/commercial boating activity. To count the number of vessels in the Solent on any given day

satellite images (Google Earth, 2019) were analysed. A 2,304 km<sup>2</sup> grid (256 cells) overlaid on the map defined the Solent and within each cell all vessels below high water line (includes those located on intertidal habitats, but not dry dock) were counted (see S2 Table 1). To calculate the proportions of each vessel type, AIS (Automatic Identification Systems) tracking software (<https://www.vesselfinder.com/>) was used once a day for 10 days with vessel details extracted (S2 Table 2). These data were then used to calculate underwater surface area (UWSA) using specific vessel-type formulas (S2 Table 3). Daily AIS count data (signals that were not vessels or unspecified were removed) were also purchased from MarineTraffic for a 65,000 km<sup>2</sup> region defined as the Channel (S2 Table 1). Recreational vessel numbers (USA: Boating Industry Market, and ICOMIA report) were included as was the global fleet of merchant vessels (S2 Table 1). Mean, maximum and minimum predictions for changes in vessel numbers by 2040 (USA recreational and global merchant fleets) were calculated using the corresponding mean, maximum and minimum values calculated for the percentage increase/decrease between consecutive years in the number of vessels for the source data. These were then applied to each year up until 2040.

To estimate inputs from different sources and into different areas we have provided realistic (means), maximum and minimum scenarios. Anti-fouling (AF) data (e.g. Cu and Zn based anti-fouling paint use, release rates etc.) were combined with the Underwater Wetted Surface Area (UWSA) and vessel numbers (data source details given in S2 Tables 1–4) to calculate AF paint Cu and Zn inputs (example calculations provided in S2). For scrubber inputs we used Cu, Zn and Ni discharge rates and then used the mean, maximum and minimum calculated values for the proportion of vessels using scrubbers, combining scrubber use, type (open v. hybrid/closed), wastewater discharge rates (assuming a 10 mW engine) and vessel count data (S2 Tables 1, 5). The number of anodes required to protect a merchant vessel was calculated based on a hull's current requirement combined with the proportion of each vessel type and the number of vessels in an area (S2 Tables 1, 2, 6), whilst mean Zn release rates generated the equivalent for recreational vessels (S2 Tables 1, 2, 6).

#### 2.6. Statistical analysis

The trend analyses of TE  $I_{geo}$  values were performed using the five-year moving average method. TE data for sites monitored for a minimum six years were analysed using median regression with bootstrapped standard errors, with  $p$ -value threshold of 0.05 in agreement with OSPAR (2019a) excluding sites for Cd (21 sites), Hg (12 sites) and Cr (1 site) where  $\geq 25\%$  of data were below DL. Quantile (including median) regression is robust to outliers; avoids parametric distribution assumptions; estimates rates of change in all parts of the response variable distribution and is invariant to monotonic transformations (Cade and Noon, 2003; Koenker and Bassett, 1978). Data processing and analysis were performed with R (R Core Team, 2020) in RStudio (RStudio Team, 2019), using R's base function and additional functions mainly from packages: dplyr (Wickham et al., 2020), tidyr (Wickham and Henry, 2020), stringr (Wickham, 2019), pracma (Borchers, 2019), matrixStats (Bengtsson, 2020) and quantreg (Koenker, 2019).

### 3. Results and discussion

#### 3.1. EA-MERMAN Database: A globally relevant resource

Our combined EA-MERMAN database is a searchable source of sediment contamination data covering approximately 80,000 km<sup>2</sup> of ocean and nearly 2,000 km of coastline over 31 years. Of the 39,906 data points for the nine TEs, 96% correspond to coastal sites (i.e. sites in transitional, estuarine and coastline waters), 4% as open sea sites (distant/remote from coastline). A mean of  $4,434 \pm 607$  SD observations per TE (both digestion procedures considered together for Fe and Cr) indicates a similar TE monitoring effort, but effort over time was highly

variable for sites. For example, 49 sites were monitored for at least eight years; in contrast, 230 sites were surveyed only twice or less (median values, all TEs considered). Infrequent sampling dominates the open sea sites (97% were sampled once, usually in 2008, 2009 or 2012). Monitoring effort also varies more broadly across the decades; effort was relatively low from 1983 to 1993, then increased substantially remaining high until the 2010s. It is not surprising that sampling effort varies spatially and temporally and differs by TE. Evolving legislative and scientific drivers and improvements in analysis methods are all important. However, to harness the power of long term databases to address critical local, regional and global 21st century environmental problems, monitoring regimes must be coordinated and supported beyond the term of individual governments and funding (Hawkins et al., 2013). We, therefore, strongly advocate that sufficient sentinel sites representing key regional locations/habitats and levels of anthropogenic activity are (or continue to be) regularly monitored.

### 3.2. TE concentrations

Given the extensive temporal and spatial coverage of the dataset, variability in TE concentrations is, not surprisingly, considerable (see S1 Table 1). For example, concentrations of Cu ranged from 0.4 to 4,007 mg kg<sup>-1</sup>. TE median concentrations were maximal for Fe (25,366 or 30,900 mg kg<sup>-1</sup> depending on sad or td, respectively), minimal for Hg (0.18 mg kg<sup>-1</sup>) and decrease in the following order: Fe > Zn > Cr > Pb > Cu > Ni > As > Cd > Hg (S1 Table 1). The fraction of samples below the DL was <1% for all TEs except Hg (12.2%) and Cd (23.5%). This confirms that, despite the technological changes in detection methods over the sampling period, the full concentration range of the vast majority of TEs was measured. Concentrations of TEs from some sites and sampling times were extremely high (e.g. 54.2 mg kg<sup>-1</sup> for Cd in a site contaminated by abandoned metal mines [Yim, 1976], see S1 Table 1). TE concentrations are globally comparable, with mining or point source pollution usually responsible for extremely high values (Bryan and Langston, 1992; Haynes et al., 1995; Caplat et al., 2005; Naidu et al., 2012; Pan and Wang, 2012; Qian et al., 2015). However, the

contribution of any outlier that remained after applying the filtering process, is also down weighted due to database size, pollution indices (except the PI) and statistical methods used.

The percentage of samples from the full dataset within each I<sub>geo</sub> and CF category presented in Table 1 identify three clear TE groupings. Cr, Fe and Ni show no to moderate pollution levels for the vast majority of samples (percentage of samples below I<sub>geo</sub> 1–2 and CF 3–6 scores: Cr: 93–97%; Fe: 99%; Ni: 90%). Zn, Pb and As have a much higher proportion of samples that are moderately or strongly polluted (percentage of samples I<sub>geo</sub> 1–4 and CF 3–24 scores: As: 15%; Pb: 27.9%; Zn: 24.7%). Finally, 22, 36 and 45% of samples show moderate-strong to very strong pollution levels for Cu, Hg and Cd, respectively. These data reflect the diverse TE-specific enrichment and elevated concentrations in sites impacted by anthropogenic processes over a 31-year period (and older industrial inputs for undisturbed sediments) across the region (see Brady et al., 2015; Qian et al., 2015).

### 3.3. Temporal evolution of global pollution

The TEPI and the PI capture a global measure of TE contamination with the results (medians, quartiles, ranges and outliers) presented in Fig. 1. During the 1983–89 period TEPI median values were in the High Contamination Level class, but subsequently reduced to the Medium Contamination Level. Over the next 25 years it remained within this class limit, but with a continuous improvement as well as a reduction in variability as indicated by the reducing quartile ranges. A very similar pattern for the PI is also evident, but with median values always above, sometimes close (2005–09 period) to the severe pollution threshold. Although the 1980s reduction may have been due to lower sampling effort, subsequent incremental improvements are to be commended. However, the region is still severely polluted (PI) or moderately contaminated (TEPI) depending on the index used. (The difference in severity is the due the preponderance given to the TE with the highest CF and the definition of TE background concentrations for the PI). The overall slowly decreasing trends of PI and TEPI data confirm elevated TEs in sediments across a multi-decadal time scale despite substantial

**Table 1**

Percentage of sites (all sites together, coastal sites or open sea sites) monitored on a yearly basis, integrated over the full 31-year monitoring time series, distributed among the seven pollution levels of the Geoaccumulation Index scale, for each studied TE. Digestion techniques: strong acid digestion (sad) or total digestion (td) are specified and considered separately for Cr and Fe.

All sites together														
Pollution Level	I <sub>geo</sub>	Colour scale	CF	As (796)	Cd (1,210)	Cr (sad) (842)	Cr (td) (201)	Cu (1,070)	Fe (sad) (676)	Fe (td) (198)	Pb (1,064)	Hg (1,126)	Ni (1,066)	Zn (1,073)
Unpolluted	0		<1.5	53.9	14.6	43.9	76.1	28.7	58.9	82.3	36.2	25.7	38.5	33.6
Unpolluted - moderate pollution	0-1		1.5-3	27.9	18.0	49.2	20.4	25.8	39.6	17.7	34.4	15.0	51.3	39.1
Moderate pollution	1-2		3-6	6.8	22.6	6.9	3.5	22.6	1.3	0	20.8	23.0	9.3	19.2
Moderate - strong pollution	2-3		6-12	5.9	25.0	0	0	11.6	0.1	0	6.8	23.5	0.9	5.1
Strong pollution	3-4		12-24	2.8	14.4	0	0	6.4	0	0	1.7	9.9	0	1.3
Strong - very strong pollution	4-5		24-48	0.8	2.9	0	0	3.1	0	0	0.2	1.9	0	0.9
Very strong pollution	>5		>48	2.0	2.5	0	0	1.9	0	0	0	1.1	0	0.7

Coastal sites														
Pollution Level	I <sub>geo</sub>	Colour scale	CF	As (706)	Cd (1,121)	Cr (sad) (842)	Cr (td) (112)	Cu (980)	Fe (sad) (676)	Fe (td) (112)	Pb (974)	Hg (1,037)	Ni (976)	Zn (981)
Unpolluted	0		<1.5	52.3	12.6	43.9	65.2	28.5	58.9	74.1	36.0	21.2	41.1	33.8
Unpolluted - moderate pollution	0-1		1.5-3	27.8	17.0	49.2	28.6	23.0	39.6	25.9	32.2	14.7	48.2	36.8
Moderate pollution	1-2		3-6	7.1	22.3	6.9	6.2	24.0	1.3	0	22.5	24.9	9.7	20.6
Moderate - strong pollution	2-3		6-12	6.7	26.8	0	0	12.4	0.1	0	7.3	25.4	1.0	5.5
Strong pollution	3-4		12-24	3.1	15.5	0	0	6.7	0	0	1.7	10.7	0	1.4
Strong - very strong pollution	4-5		24-48	0.8	3.1	0	0	3.4	0	0	0.2	2.0	0	1.0
Very strong pollution	>5		>48	2.3	2.7	0	0	2.0	0	0	0	1.2	0	0.8

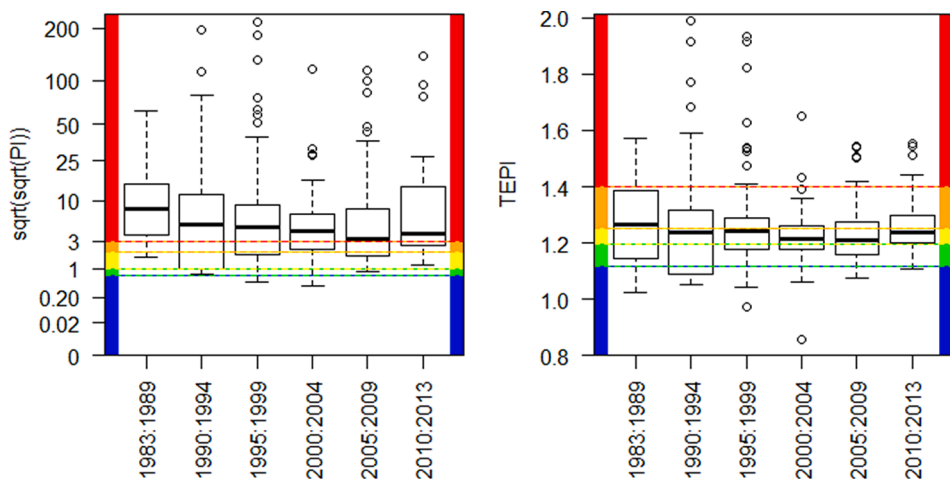
  

Open sea sites														
Pollution Level	I <sub>geo</sub>	Colour scale	CF	As (90)	Cd (89)	Cr (sad) (0)	Cr (td) (89)	Cu (90)	Fe (sad) (0)	Fe (td) (86)	Pb (90)	Hg (89)	Ni (90)	Zn (92)
Unpolluted	0		<1.5	66.7	40.4	-	89.9	31.1	-	93	37.8	77.5	10	30.4
Unpolluted - moderate pollution	0-1		1.5-3	28.9	30.3	-	10.1	56.7	-	7	57.8	19.1	85.6	64.1
Moderate pollution	1-2		3-6	4.4	25.8	-	0	7.8	-	0	2.2	1.1	4.4	4.3
Moderate - strong pollution	2-3		6-12	0	3.4	-	0	2.2	-	0	1.1	2.2	0	1.1
Strong pollution	3-4		12-24	0	0	-	0	2.2	-	0	1.1	0	0	0
Strong - very strong pollution	4-5		24-48	0	0	-	0	0	-	0	0	0	0	0
Very strong pollution	>5		>48	0	0	-	0	0	-	0	0	0	0	0

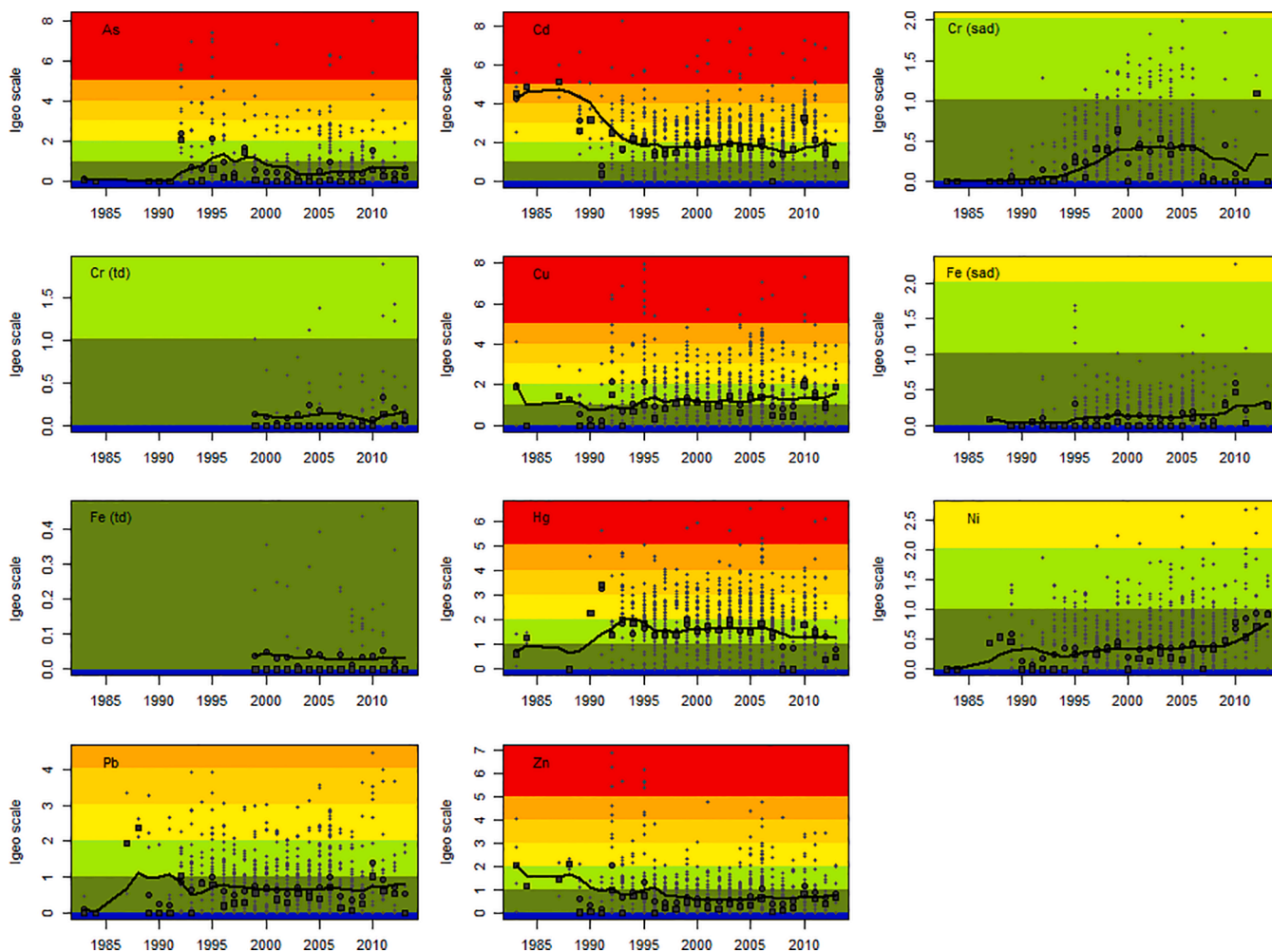
I<sub>geo</sub> = Geoaccumulation Index.

CF = Contamination Factor.

Number in brackets = number of sites monitored, on a yearly basis, over the 31-year time series.



**Fig. 1.** Boxplots (median [bold line]; Q1 and Q3 [boxes], ranges [whiskers] and outliers [open circles]) of Nemerov Pollution (PI) and Trace Element Pollution (TEPI) indices (PI boxplots are double square root transformed). Indices were generated with sites monitored for minimum six trace elements out of nine per four to seven-year periods. Pollution scales for TEPI classify sediments (see Richir et al., 2015 for details) according to 5 levels: blue: very low contamination level (VLCL, data below the 1st quartile mean); green: low contamination level (LCL, data between the 1st and 2nd quartile means); yellow: medium contamination level (MCL, data between the 2nd and 3rd quartile means); orange: high contamination level (HCL, data between the 3rd and 4th quartile means); red: very high contamination level (VHCL, data above the 4th quartile mean). For the PI (see Brady et al., 2015): blue: unpolluted ( $PI < 0.7$ ); green: slightly polluted ( $0.7 < PI < 1$ ); yellow: moderately polluted ( $1 < PI < 2$ ); orange: heavily polluted ( $2 < PI < 3$ ); red: severely polluted ( $PI > 3$ ). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** TE sediment contamination evolution using the 7-level Geoaccumulation Index (Igeo) pollution scale classifying sediment from unpolluted (blue) to very strongly polluted (red) (see Table 1 and Müller, 1986 for details). Dark squares: annual medians; dark circles: annual means; light grey crosses: site-specific annual means; full black lines: 5-year moving averages. Digestion techniques: strong acid digestion (sad) or total digestion (td) are specified and considered separately for Cr and Fe. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

investment in legislation controlling anthropogenic inputs, improvements in technology, changes in regional manufacturing effort and increased recycling. However, what is most concerning is that for 2010–13 there was a distinct increase in indices' median scores and associated quartile range for PI. Recalculating the TEPI excluding Ni and Cu (which from Fig. 2 show increasing  $I_{geo}$  values), puts the median TEPI and PI values for 2010–2013 closer to that of 2000–2004 (difference between periods decreases by 47% for TEPI and 19% for PI). It, therefore, seems probable that changes in Ni and Cu make an important contribution to the recent increased TEPI and PI scores for this period (S1 Figs. 1 and 2). Considering sites are located within a region of mature (often declining) industry and where some of the most comprehensive legislation for contaminants has been enacted, it is highly likely that the trends identified here are, at best, replicated across equivalent areas, or at worst much more dramatic. Our data highlight the urgent need to assess *current* regional inputs of *all* TEs into our seas, thus confirming if the national and international legislation is effective for those that are currently priority pollutants (e.g. within the EU), but also extend to those that are not (e.g. Cu and Fe).

### 3.4. Temporal evolution of TEs

Temporal trends per TE are also explored, and using the  $I_{geo}$  data summarized in Table 1 we calculated annual medians, means, site-specific annual means and 5-year moving averages (Fig. 2).  $I_{geo}$  score moving averages for two TEs (Cd and Hg) have remained stable since the 1990s, confirming the challenge for regulators to achieve improvements in the face of extreme persistence. However, some reductions e.g. Cd from the 1980s to early 1990s, Hg around the mid-1990s and in 2007–08 are visible. For Cd, this may be due to the lower level of sampling in the early 1990s. The 5-year moving averages for As, Pb and Zn also show a very stable level of pollution confirming persistence within benthic sediments. Nevertheless, as the mean and median  $I_{geo}$  scores are <1 (unpolluted to moderate polluted; dark green colour scale, see Table 1) the broadscale level of contamination is of low concern even though national/international agencies have highlighted them as priority chemicals (US EPA, 2015; EU, 2000). Although the mean and median  $I_{geo}$  scores are also <1 for Ni, Fe and Cr, Fig. 2 clearly shows a continuous increase over time for Ni is moving the  $I_{geo}$  score towards the moderate pollution threshold. Fe extracted with strong acids is also increasing especially from the mid-2000s, although it is some way from reaching the  $I_{geo}$  moderate pollution threshold. Most concerning is Cu as the mean  $I_{geo}$  score is within the moderately polluted scale, but it is also increasing from the mid-1990s.

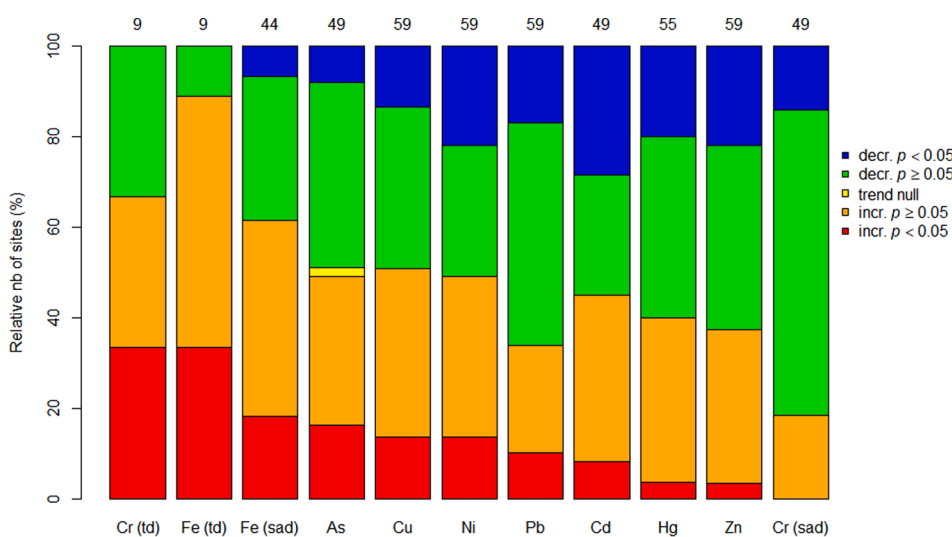


Fig. 3. Relative proportions of sites monitored for each TE over a minimum of six years, which show a net increase (incr.  $p < 0.05$ , red) or decrease (decr.  $p < 0.05$ , blue), an increase (incr.  $p \geq 0.05$ , orange) or decrease (decr.  $p \geq 0.05$ , green) or no evolution (trend null, yellow) of their sediment TE concentrations. Numbers above bars are number of sites included in the median regression analysis. Digestion techniques: strong acid digestion (sad) or total digestion (td) are specified and considered separately for Cr and Fe. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



coastal and biological observation networks that can focus on the local, but also provide the foundation for assessing broad-scale changes (She et al., 2019).

### 3.5. Pollution sources

Reducing marine TE contamination is essential for healthy coastal ecosystems; this drives the comprehensive policy and regulatory frameworks to achieve unpolluted seas. However, despite some reductions, data from Figs. 2 and 3 confirm that elevated TE pollution at sites has exerted effects on the benthos via the sediment matrix and bioaccumulation for the period 1981–2013. It may seem disheartening to think that TE persistence should constrain our ability to improve benthic systems, however, as 135 of 500 trends (both positive and negative) in the sediment concentrations had  $p < 0.05$  (Fig. 3), with proportionally more decreases (17%) than increases (10%) change does happen. Change is also reflected at the regional level as shown by the multi-element index and  $I_{geo}$  values (Figs. 1 and 2). Together they confirm that, despite TE persistence, future contamination levels can improve.

Critical to reducing TE concentrations in sediment is an accurate and up to date assessment of the input sources. Whilst significant data have been collected through a multitude of national and international programmes it is recognized some TE inputs are not monitored (e.g. Fe), whilst there is low confidence in some data (GESAMP, 2015; EEA, 2019). We have, therefore, highlighted the main sources that may elucidate the temporal evolution of each TE. Using the data from S1 Table 2, managers can also explore local input sources that may be key drivers for temporal change at the site, estuary, harbour and sub-region scales.

Regional and continental Pb inputs from the atmosphere (Pacyna et al., 2009; OSPAR, 2015; Richmond et al., 2020), rivers (Tappin and Millward, 2015; OSPAR, 2019b) and direct discharges (OSPAR, 2019b) have all declined considerably in the last 35+ years. Concentrations in biota (Raimundo et al., 2011) and sediments (Cossa et al., 2018) have also seen significant reductions. Pb phase out from gasoline has been the main driver, but other direct industrial inputs have also declined (Pacyna et al., 2009). This supports the relatively high number of sites (17% of these with  $p < 0.05$ ) with decreasing trends (Fig. 3). Nevertheless, Rusiecka et al. (2018) reported northeast Atlantic surface water concentrations to still be significantly elevated, implicating long distance transport. Long range circulation (1,000s of kms) and resuspension from sediments could explain the latter stabilization of the  $I_{geo}$  score (Fig. 2).

Generally, Hg marine inputs and concentrations in sediment and biota have all declined significantly (Pacyna et al., 2007, 2009; Tappin and Millward, 2015; Obrist et al., 2018; OSPAR, 2019a; Richmond et al., 2020). Nevertheless, riverine inputs and direct discharges have stabilized in some European countries (OSPAR, 2019b) and increasing atmospheric loads are reported in Asia (Obrist et al., 2018). Global vigilance is, therefore, required to maintain the declines from the mid-2000s onwards (Fig. 2) and increase the number of sites with significantly decreasing trends (Fig. 3).

Cd is also seen as a success story with significant declines in atmospheric and riverine inputs aligning to the decline in  $I_{geo}$  score in the 1990s (Fig. 2). Subsequent reductions have been limited (Pacyna et al., 2007, 2009; Tappin and Millward, 2015; OSPAR, 2015; Richmond et al., 2020) and concomitant reductions in biota and sediment have also stalled in some locations (Johnstone et al., 2016). This is reflected in our data where the  $I_{geo}$  score since mid-1990s has failed to reduce further. This highlights the need to increase efforts locally, nationally and globally on key current sources: e.g. combustion of fuel and fertilizer use (OSPAR, 2008; Richmond et al., 2020).

In Europe, direct and riverine Zn discharge levels vary between countries, however, a general decline can be detected (OSPAR, 2019a). Atmospheric emissions for the UK have also declined, but with no

further reductions from the mid-2000s (Defra and BEIS, 2019). The 5-year mean regional  $I_{geo}$  score (Fig. 2) has remained low after reductions in the early 1990s and combined with the low number of sites with significantly increasing trends (Fig. 3) would indicate that improvements have been maintained. Nevertheless, relatively large inputs from current sources including buildings, vehicle emissions, combustion (see Davis et al., 2001; Duan and Tan, 2013) as well as contributions from 'hidden' inputs (e.g. anti-fouling paints and anodes (see section below)) will make any further reductions very challenging to achieve.

Input data for As are relatively sparse as they are not collated for direct and riverine discharges (OSPAR, 2019a). However, Pacyna et al. (2009) supported by Harmens et al., (2007) show clear reductions in European emissions from the 1950s. Non-ferrous smelting and fossil fuel production are the main sources globally of anthropogenic arsenic, with the replacement of coal burning by natural gas a major factor in the 4-fold decline in UK emissions since 1970 (Faust et al., 2016). Although, the 5-year average regional  $I_{geo}$  score has remained low after a peak in the early 1990s (Fig. 2), relatively high numbers of sites with increasing trends ( $p < 0.05$ ) (Fig. 3) require further investigation and could mirror local scale changes as reported by Kang et al. (2018).

Cr input data are sparse for similar reasons to As, but domestic and industrial effluents and fuel combustion are identified as key sources (Pacyna et al., 2009; Cheng et al., 2014). The 5-year average regional  $I_{geo}$  score for Cr (sad) increased from the mid-1990s to mid-2000s (Fig. 2), but a subsequent decline would suggest that inputs are stable and this is supported by Kang et al. (2018). However, the high numbers of significantly increasing trends per site (Fig. 3) for the more recent sediment total digestion offset the more important proportion of sites showing significant decreases for strong acid digestion techniques. Cr requires further investigation at the local scale as increases have been associated with point sources including oil installations (Celis-Hernandez et al., 2018) and the metallurgical industry (Cheng et al., 2014).

Anthropogenic activities (combustion; mining and smelting) may have led to a tripling of atmospheric soluble Fe ocean deposition since the industrial revolution (Ito, 2015; Myriokefalitakis et al., 2015). Fe is not routinely measured in sediments, but the increasing  $I_{geo}$  score (Fig. 2) support these input increases. Increasing Fe concentrations may have contradictory effects on sediment toxicity. Firstly, Fe oxides reduce TE bioavailability and consequent toxicity (Xiangdong et al., 2001). However, increases in Fe sediment concentration could have direct impacts, as Johnson et al. (2017) ranked it as 8/60 for toxicity. Experimentally investigating the effects of Fe on benthic species is, therefore, essential. Finally, the increase in Fe may also impact all coastal habitats. Dissolved  $Fe^{II}$  is an essential and limiting nutrient that stimulates primary productivity (e.g. Smetacek et al., 2012). Like other TEs, sediment-bound Fe can be resuspended (Kalnejais et al., 2010). Under these conditions it could be converted to  $Fe^{III}$ , thus sediment-derived Fe could be a significant driver in the increasing global problem of harmful algal blooms (Smetacek and Zingone 2013).

Anthropogenic sources dominate Ni release (Clayton and Clayton, 1981; Pacyna et al., 2007, 2009; Strincone et al., 2013). Although official European emissions data suggest a 55% reduction between 1995 and 2000 (Ilyin et al., 2006), measurements in mosses did not show significant recent reductions (Harmens et al., 2007). National and transnational assessments are required to contextualize the increasing Ni  $I_{geo}$  score from Fig. 2, but also at the site level to identify local inputs. Cempel and Nikel (2006) stated waste water effluents to be major marine inputs, but increased emissions from shipping (Turner et al., 2017) could be key (see section below). A continuation of these inputs will take the  $I_{geo}$  score across the moderately polluted threshold, and although DeForest and Schlekot (2013) indicate Ni has relatively low toxicity, studies on benthic species via sediment exposure are very limited.

Like Zn, direct and riverine discharges of Cu in Europe are highly variable across countries (OSPAR, 2019b). Harmens et al. (2007) did report a significant decreasing trend for concentrations in European moss (1995–2000), but atmospheric emissions for the UK are only

**Table 2**

Assessment of TE inputs ( $t \text{ year}^{-1}$ ) into different regions/countries from AF (antifouling paints), scrubbers (vessel exhaust cleaning systems) and anodes (sacrificial hull anodes) with direct and river discharges for the Channel region and UK (2015), and atmospheric inputs for UK (2018) included for comparison. Values are realistic (mean), minimum and maximum scenarios in parentheses, except OSPAR data which are sometimes given as ranges. Data for 2040 based on predicted changes to vessel numbers and installation/use of scrubbers. See [S2 Tables 1–6](#) for details of each component calculation and supporting references, except [OSPAR \(2019b, c\)](#) and atmospheric inputs ([Richmond et al., 2020](#)). For details of vessel types and counting methods for each source category/input region, see [S2 Table 2](#).

Method	Source	Input region	2020 Cu	2040 Cu	2020 Zn	2040 Zn	2020 Ni	2040 Ni
AF	All vessels	Solent	94 (3–378)	135 (3–678)	27 (0.7–128)	39 (0.8–231)	–	–
Scrubbers	Merchant vessels (AIS only)	Solent	0.4 (0.03–2)	5 (0.3–27)	0.8 (0.02–3)	9 (0.3–30)	0.2 (0.02–0.6)	2 (0.2–7)
Anode	All vessels	Solent	–	–	349 (227–490)	503 (271–880)	–	–
AF	All vessels (AIS only)	Channel	22 (0.6–102)	32 (0.7–184)	6 (0.1–35)	9 (0.2–62)	–	–
Anode	All vessels (AIS only)	Channel	–	–	82 (46–133)	119 (55–238)	–	–
Scrubbers	Merchant vessels (AIS only)	Channel	5 (0.3–28)	58 (0–306)	10 (0.3–31)	107 (0–344)	2 (0.3–7)	24 (0–77)
OSPAR Direct	UK	Channel	0.25	–	0.24	–	–	–
OSPAR Direct	France + UK	Channel	0.25 (- + 0.25)	–	0.24 (- + 0.24)	–	–	–
OSPAR Riverine	UK	Channel	17	–	68	–	–	–
OSPAR Riverine	France + UK	Channel	57–59	–	258–259	–	–	–
OSPAR Riv. + Dir.	UK	UK	296–338	–	1,028–1,184	–	–	–
Atmospheric	UK	UK	269	–	460	–	98	–
AF	Recreational vess.	UK	340 (15–1,011)	310 (5–1,526)	98 (4–344)	89 (1–519)	–	–
Anode	Recreational vess.	UK	–	–	908 (476–1,362)	828 (144–2,053)	–	–
AF	Recreational vess.	Europe	3,624 (159–10,778)	3,307 (48–16,255)	1,041 (39–3,665)	950 (11–5,526)	–	–
Anode	Recreational vess.	Europe	–	–	9,671 (5,069–14,507)	8,825 (1,531–21,876)	–	–
AF	Recreational vess.	USA	9,828 (431–29,230)	8,968 (130–44,085)	2,823 (105–9,938)	2,576 (32–14,989)	–	–
Anode	Recreational vess.	USA	–	–	26,228 (13,746–39,342)	23,933 (4,151–56,365)	–	–
AF	Recreational vess.	World	19,627 (861–58,371)	17,909 (260–88,036)	5,637 (211–19,846)	5,144 (64–29,932)	–	–
Anode	Recreational vess.	World	–	–	52,376 (27,450–78,564)	47,793 (8,290–118,474)	–	–
AF	Merchant vess.	World	1,711 (51–6,408)	2,505 (63–10,563)	491 (12–2,178)	719 (15–3,591)	–	–
Anode	Merchant vess.	World	–	–	6430 (4,213–7,243)	9,414 (5,161–13,095)	–	–
Scrubbers	Merchant vess.	World	397 (28–1,797)	4,307 (31–19,917)	728 (25–2,027)	7,898 (278–22,459)	164 (21–451)	1,777 (236–5,004)

declining minimally (Defra and BEIS, 2019). Concentrations in biota (OSPAR, 2016) and sediments have seen increasing trends from multiple sites (OSPAR, 2016; Kang et al., 2018). Cu is used in many applications including: fertilizers (Jensen et al., 2016); vehicle brakes (Davis et al., 2001) and also combustion processes (Bhuiyan et al., 2018). Although brake wear is responsible for the vast majority of the UK's atmospheric emissions, it has remained relatively constant (e.g. around  $200 \text{ t y}^{-1}$  for 30 years in the UK) (Richmond et al. 2020). This and the decline/regulation of industrial uses indicate that other sources are driving the increasing regional  $I_{\text{geo}}$  score since the 1990s (Fig. 2), but also site-specific changes (Fig. 3).

### 3.6. 'Hidden' sources: Antifouling paints, scrubbers and anodes

For Cu, Ni and Zn a number of rarely-studied sources could be significant contributors to the sediment concentration evolution. Table 2 values are based on robust empirical data combined with selected literature (S2 Tables 1–6) generating realistic (i.e. mean) inputs with minimum and maximum scenarios reflecting source data variability. Since the Tri Butyl Tin-AF coatings ban, Cu-based AF (and combined with Zn-based products too) now dominate (Amara et al., 2018). Using our Google maps Solent vessel count combined with the other data (see S2 Tables 1–4) the realistic (mean) amount of Cu released is  $94 \text{ t y}^{-1}$ . This is double that reported by OSPAR (2019b) for the whole of the Channel region from both riverine and direct discharges and is a third of all UK-wide direct and riverine discharges. In contrast, Solent input levels of Zn are much lower ( $27 \text{ t y}^{-1}$ ), but still represent 10% of the Zn entering the Channel region from both France and UK (Table 2). The discrepancy in the amounts calculated for the Solent and Channel region is due to the vessel count method as the AIS data greatly underestimate the number of vessels per region. In coastal areas this is likely to be due to most vessels not having a legal requirement for AIS data transmission (Cope et al., 2020) or turning off the system to preserve battery life (C. Thisby, pers. comm.). At the larger spatial scales Cu and Zn amounts are substantial, but still likely to be significant underestimates due to limited recreational boat and global merchant fleet data. By 2040 the predicted increase in the merchant vessel fleet (a mean of  $1.8\% \text{ y}^{-1}$ , S2 Table 1) not surprisingly generates substantial increases for AF inputs globally, but applying a predicted  $-0.3\% \text{ y}^{-1}$  contraction of the USA recreational fleet to all other regions sees concomitant declines. Nevertheless, recreational boats contribute an order of magnitude more Cu and Zn than the merchant fleet.

Shipping contributes significant emissions to coastal environments (e.g. Aulinger et al., 2016; Xiao et al., 2018) and in response to legislation (IMO, 2008a), there has been a noteworthy increase in scrubber installations (Turner et al., 2017). A few studies have directly measured the metal concentrations in scrubber discharge water (see references in S2 Table 5), but no scale-up has been attempted. Although TE concentrations in waste water are high (see S2 Table 5), the small proportion of merchant vessels (current estimate: 3.4%) fitted with scrubbers and that merchant vessels make up a small fraction of those counted in the Solent means that, in comparison to AF, the amounts of Cu, Zn and Ni discharged are low. At the local (Solent) level, scrubber discharge contribution to the TE sediment concentrations is, therefore, small and orders of magnitude lower than AF. However, within the Channel and globally, scrubbers contribute nearly double the Zn from AF and 25% of the Cu. The future percentage of vessels fitted with scrubbers is a topic of intense discussion with divergent business responses for embracing scrubber technology. If an industry prediction of 25% by 2040 (S2 Table 5) in conjunction with the yearly increase in merchant fleet size does transpire, scrubbers will contribute nearly double the Cu from AF for the global merchant fleet and  $> 10\text{x}$  the amount of Zn. Globally, Ni inputs will also increase from 164 t in 2020 to 1,777 t in 2040.

Finally, the dissolution of sacrificial metal anodes are intrinsic to boat protection. Only a very limited number of studies have calculated Zn inputs (see references in S2 Table 6) and these were at the very local

level (e.g. Rees et al., 2017) or outdated (e.g. Boxall et al., 2000). At all spatial scales and for all vessel types, inputs of Zn from anodes are an order of magnitude greater than equivalent AF sources. For example, anodes from the UK recreational fleet release 908 t of Zn per year, compared to 98 t from AF. The level of input is equivalent to that reported by OSPAR (2015) for direct and riverine discharges for the whole of the UK and similar to a recent calculation by Rees et al. (2020). Despite the predicted contraction in fleet size, recreational vessels will continue to deliver considerable quantities of Zn in 2040 from anodes. However, the input of Zn from scrubbers for the global merchant fleet becomes equally important due to the increase in the predicted proportion of the fleet fitted with open scrubbers.

Table 2 clearly shows that inputs of Cu (AF) and Zn (AF, anodes) are already substantial at multiple spatial scales. The Cu  $I_{\text{geo}}$  score increase (Fig. 2) is, therefore, supported by the dominance of Cu-based AF coatings on vessels and is highly likely to contribute to the increases in median TEPI and PI values for 2010–2013 (Fig. 1). This is also reinforced by the fact that 88% of the sites with increasing Cu concentrations ( $p < 0.05$ ) were transitional/coastal water, close to urban areas with substantial vessel numbers. However, S1 Table 2 shows that spatial consistency of each TE's temporal evolution (i.e. multiple sites located within a harbour/estuary/sub-region showing consistent trends) is not always evident. For example, across 20 sites in the Solent both increasing and decreasing trends for Cu are present. A much more detailed local input analysis combined with an understanding of complex hydrodynamic and sedimentation processes will be required to fully contextualise the contribution of diffuse versus local (e.g. point discharges) sources. Regardless of the local level complexity, non-toxic AF coatings or other AF fouling control methods (e.g. cleaning and fouling waste capture) should be considered a priority for legislators to address these substantial and pervasive inputs (e.g. Bergman and Ziegler, 2019).

Although the overall inputs of Zn are much higher due to the contribution from anodes, it is not immediately clear why a lower proportion of trends show increases with  $p < 0.05$  (Fig. 3) and the  $I_{\text{geo}}$  score (Fig. 2) remains fairly constant. Declines in UK atmospheric emissions (Defra and BEIS, 2019) may have been counteracted by the AF and anode inputs, but legislation to expedite appropriate cleaning control along with education for recreational vessel owners on anode use would be important first steps (Rees et al., 2017). The increasing  $I_{\text{geo}}$  score for Ni is more challenging to explain based on the relatively low scrubber inputs for the Solent ( $0.2 \text{ t y}^{-1}$ ) and the Channel ( $2 \text{ t y}^{-1}$ ) from Table 2. Extensive burning of higher sulphur fuel oil by the majority of merchant vessels generates significant Ni atmospheric emissions (e.g. Peltier and Lippmann, 2010; Zhao et al., 2013). These would be deposited to the marine environment and concentrated by hydrodynamic currents to more localised areas (see S1 Table 2), although unknown point sources may also be responsible for spatial inconsistencies. Our data predict that scrubber discharges could be a significant future direct input source of Ni (as well as Zn and Cu and other TEs) to marine environments, which requires waste water discharges to be regulated for metals (rather than just guidelines at present IMO, 2008b). However, unless there is a much more rapid conversion to low sulphur (or alternative) fuels, Ni and other metals will still continue to input the marine environment, but indirectly from atmospheric deposition.

## 4. Conclusions

Mining sediment contamination databases from national public sources is a powerful tool to assess the contamination evolution at scales that are challenging to address. Integration within coastal observatory networks will significantly enhance their value and application, enabling managers to link ecosystem management approaches with multiple TE contamination levels that together have significant cumulative effects. However, this will require investment in data alignment and quality processes supported by long term monitoring of sufficient sentinel sites.

Our long-term spatiotemporal analysis identifies considerable improvements in contamination (declining TEPI and PI) at the regional level since the 1980s. Nevertheless, the increase in TEPI and PI for 2010–13, increasing  $I_{geo}$  scores for Cu, Fe and Ni; stabilization of other TE  $I_{geo}$  scores after declines (Cd, Zn, Hg, Pb) combined with the numbers of sites that have increasing trends all indicate continued/increasing inputs. Accurate input assessments including ‘hidden’ sources are, therefore, urgently required especially in the context of the global focus on the blue economy (e.g. UN Decade of the Oceans) and predicted increases in shipping, coastal aquaculture, marine electricity generation etc. to support growth (OECD, 2016). Ultimately, this must inform appropriate legislation and control measures to protect ecologically and economically valuable benthic systems across the globe.

### CRedit authorship contribution statement

**Richir Jonathan:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing. **Bray Simon:** Methodology, Validation, Writing - review & editing. **McAleese Tom:** Data curation, Formal analysis, Investigation. **Watson J. Gordon:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2020.106362>. The geo-spatial information contained in geographic KML files enables the visualization of sites from their geolocalisation on Google Earth.

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