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Cumulative environmental risk assessment of metals and polycyclic aromatic hydrocarbons from ship activities in ports

Anna Lunde Hermansson [a,*], Ida-Maja Hassellöv [a], Jukka-Pekka Jalkanen [b], Erik Ytreberg [a]

[a] Chalmers University of Technology, Department of Mechanics and Maritime Sciences, Gothenburg, Sweden
[b] Finnish Meteorological Institute, Erik Palmenin aukio 1, 00101 Helsinki, Finland

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ABSTRACT

Marine environmental risk assessments rarely consider the cumulative risk from multiple contaminants and sources. Ships give rise to a range of contaminants, originating from different onboard sources, resulting in contaminant loads to the marine environment. Here, the Ship Traffic Emission Assessment Model (STEAM), in combination with the hydrodynamic and chemical fate model MAMPEC, was used to calculate loads and predicted environmental concentrations (PECs) of metals and polycyclic aromatic hydrocarbons, in four ports. PECs were compared to the predicted no effect concentrations (PNEC) to assess environmental risk from the different onboard sources, both separately and cumulatively. The results show that three out of four ports were subject to unacceptable risk. This study highlights the importance of accounting for multiple contaminant sources when assessing the marine environmental risks of shipping and challenges the suitability of the proposed new international guidelines on how to assess risk of scrubber water discharge.

1. Introduction

In areas of high shipping intensity, e.g. frequently trafficked ship lanes and semi-enclosed areas such as harbours and ports, contaminants from ships may exert substantial pressures on the marine environment. Ports are constantly occupied by ships of various types and sizes that all contribute to the continuous load of contaminants, such as metals and polycyclic aromatic hydrocarbons (PAHs) from different sub-systems, i.e. contaminant sources, on the ship. Examples of contaminant sources are near-ship atmospheric deposition from exhaust; release of biocides from antifouling paints; discharge of residuals from onboard operations such as bilge water from the engine room and discharge of scrubber water, i.e. wash water from exhaust gas cleaning systems (Jalkanen et al., 2021). As both metals and PAHs, with their derivatives, can negatively impact marine organisms (Achten and Andersson, 2015; Morales et al., 2016; Honda and Suzuki, 2020), the input of these contaminants to the marine environment may result in an increased risk, where risk is considered unacceptable if contaminant concentrations exceeds the predicted no effect concentrations (PNECs).

Several studies have shown that the discharge of scrubber water may result in adverse effects on marine organisms (Koski et al., 2017; Ytreberg et al., 2019; Teuchies et al., 2020; Thor et al., 2021). As previous work shows, scrubbers contribute significantly to the environmental load of metals and PAHs (Lunde Hermansson et al., 2021; Ytreberg et al., 2022), and there is a need to adopt a more complete approach, including the contribution from several contaminants and ultimately mixture toxicity, when assessing the environmental risks associated to emissions from scrubbers and shipping in general. Many of the contaminants found in scrubber discharge water (e.g. benzo[*a*]pyrene (BaP), nickel (Ni)) are included as (priority) hazardous substances under the EU Water Framework Directive (WFD, 2013) where EU Member States have agreed to take action to phase out these substances and to stop the emissions.

The Marine Environment Protection Committee (MEPC) of the International Maritime Organization (IMO) has refined the guidelines regarding risk and impact assessments related to scrubber water discharge (MEPC, 2022). Although ships equipped with scrubbers constitute a small fraction of the fleet (<5 % of total number of ships (DNV-GL, 2021); and approximately 16 % of gross tonnage (Teuchies et al., 2020)), they represent 25 % of the global commercial shipping fleet's fuel consumption (IEA, 2020). The relative load of metals (e.g., copper (Cu) and vanadium (V)) and PAHs (e.g. anthracene (Ant)) from these vessels can be significant (Ytreberg et al., 2022). According to the recently approved IMO guidelines (MEPC, 2022), the Marine

* Corresponding author.
E-mail address: anna.lunde.hermansson@chalmers.se (A. Lunde Hermansson).

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Antifoulant Model to Predict Environmental Concentrations (MAMPEC, Deltares) shall be used to assess the environmental risk of scrubber water discharge with the purpose to aid member states when formulating local and regional regulations (Japan, 2019; MEPC, 2022). MAMPEC has been developed to calculate the predicted environmental concentrations (PECs) within and outside a port environment, in the so-called surrounding environment. Similar to environmental risk assessments of antifouling paints for commercial shipping within the European Union (EU, 2012; ECHA, 2022), the MEPC guidelines propose the maximum PECs in the surrounding environment from the MAMPEC model to be used in the assessment, indicating that the port itself is not considered worthy of protection which might lead to acceptance of very high contaminant loads. In a legal context, different ship activities are regulated separately (Table 1) and the environmental risk is assessed for the separate onboard systems one by one. For example, in the EU, antifouling paints are risk assessed using MAMPEC and regulated based on the analysis results but the assessment do not consider any other discharges/emissions from shipping (ECHA, 2017). A similar approach is now proposed for the environmental risk assessment of scrubbers (MEPC, 2022). The MEPC (2022) guidelines propose that the assessment should account for other contaminant sources and background concentrations and that worst-case emission factors should be used to assess reasonable worst-case scenarios. How the worst-case emission factors are determined is however not defined, only that they should be based on measurements and result from scientific reasoning (MEPC, 2022). To the knowledge of the authors, no assessments have been done to calculate the quantity of contaminants that would be allowed to be discharged in a port if the MAMPEC results for the surrounding environment are used. Therefore, this work will test the applicability of the new guidelines and assess if the use of the surrounding environment is an adequate approach to ensure that the discharge of scrubber water is not causing unacceptable risks in the marine environment.

As the total loads, derived from all contaminant sources, are not assessed together, the current environmental risk assessments of ship-activities are often incomplete. A reason for this may be the challenges to identify the combination of risks and the complexity of assessing several contaminants, their sources and possible mixture toxicity effects simultaneously. Taking one step towards an improved risk assessment, the aim of this study was to compare the loads of metals and PAHs from near-ship atmospheric deposition, bilge water discharge, scrubber water discharge and the release of biocides from antifouling paints and to assess the sources' relative contribution to the cumulative environmental risk. The approach allows for a more complete environmental risk assessment, where multiple ship-derived contaminant sources can be assessed with respect to metals and PAHs simultaneously.

2. Material and method

The environmental risk was assessed with a bottom-up approach (Fig. 1) in four different ports where; 1) the loads of metals and PAHs from ships were derived from Ship Traffic Emission Assessment Model (STEAM) in combination with emission factors from previous studies (Ytreberg and Hassellöv, 2020; Jalkanen et al., 2021; Lunde Hermansson et al., 2021); 2) based on the loads of metals and PAHs, the water column PECs inside the port and in the surrounding environment were calculated using MAMPEC and; 3) the risk characterisation ratios (RCRs) for the individual contaminants, for the specific contaminant-sources and for the sum of all contaminant sources were computed for each port.

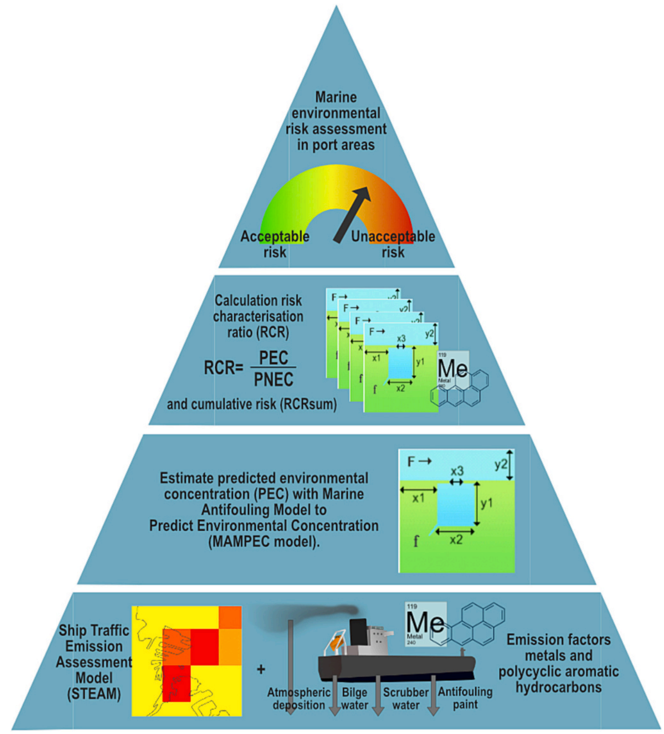

Fig. 1. Illustration of the bottom-up approach applied in this study where ship activity and emission factors were used as input to calculate the contaminant loads that enabled MAMPEC modelling to estimate predicted environmental concentrations (PECs). The PECs were compared to the predicted no effect concentration (PNEC) of the respective contaminant to calculate the risk characterisation ratio (RCR) and RCR_{sum}, corresponding to the cumulative risk within the port environment.

Conventional environmental risk assessment is focused on single-substances, meaning that the predicted (or measured) environmental concentration (P(M)EC) of a single substance (i) is compared to the predicted no effect concentration (PNEC) of that same substance (Backhaus and Faust, 2012). If PEC is larger than PNEC, then the PEC/PNEC ratio, i.e. the risk characterisation ratio (RCR), will be larger than 1, implying an unacceptable risk to the environment. There are different approaches available to account for mixture toxicities and to calculate a total RCR for a complex solution (Backhaus and Faust, 2012; Nys et al., 2017). As a 1st Tier conservative approach, the sum of all the individual PEC/PNEC values can be used as an estimate of the RCR_{sum} for the mixture (Eq. 1) (Backhaus and Faust, 2012).

Table 1
Overview of regulations associated to different onboard systems that contribute to the environmental load of metals and PAHs. Also listed are the approval mechanisms and monitoring systems. IMO = International Maritime Organization; MARPOL = International Convention for the Prevention of Pollution from Ships; MEPC = Marine Environment Protection Committee; AFS = Antifouling systems; ODME = Oil Detector Monitoring Equipment.

	Open & closed loop scrubber	Antifouling paint	Bilge water	Atmospheric emission[1]
Shipping regulations and/or guidelines	IMO: MARPOL Annex VI on SO_X emission to air; only guidelines on water discharge (MEPC.259(68) and MEPC.340(77))	IMO:AFS convention EU: Biocidal Products Regulation	IMO: MARPOL Annex I discharge of oil and MEPC.108 (49)	IMO: MARPOL Annex VI on SO_X, PM and NO_X emissions. EU: Sulphur directive
Approval mechanism and monitoring	Approval by flag-state Administration; monitoring by onboard scrubber unit system	EU: Product approval based on risk assessment. No monitoring.	ODME is used to ensure compliance and keep log on discharge.	On-board fuel sampling; NOx Technical Code 2008.

[1] Atmospheric deposition on sea surface is not regulated. Emissions from ship are only regulated with respect to SO_X, PM and NO_X.

$$RCR_{sum} = \sum \frac{PEC_i}{PNEC_i} \tag{1}$$

If $RCR_{sum} > 1$, this means that there is an unacceptable risk to the environment. The summation of RCRs (Eq. 1) is supported by the recently approved IMO guidelines for environmental risk and impact assessments of scrubber discharge water (MEPC, 2022), stating that:

> The cumulative effects of mixtures should be taken into account and a PEC/PNEC summation approach is recommended where PEC/PNEC ratios of all mixture components (PAHs and metals) are summed up to a final Risk Quotient.

The four port/harbour environments, included in the current study (see Table 2 and detailed description in Supplementary material A), consisted of two existing ports based on 2018 ship activity data (Port of Copenhagen and Port of Gdynia) and two modelled OECD default ports (OECD EU and Baltic) for comparison. Port of Copenhagen and the Port of Gdynia were selected as they were identified as hotspots with respect to discharge of open loop scrubber water in 2018 (STEAM dataset) and they were also identified as ports of interest within the EU project SHEBA (all input data in Supplementary material A). The OECD EU harbour is a modelled harbour environment, based on the properties of Port of Rotterdam (derived by Van der Plassche and Van der Aa, 2004), and the OECD Baltic harbour is an adapted version of the OECD EU harbour to more accurately represent the conditions within the Baltic Sea area (Supplementary material A) (Faber et al., 2021). The OECD Baltic has the same dimensions as the OECD EU harbour, but with a lower temperature, lower salinity and no tidal difference (i.e. lower water exchange rate) compared to the OECD EU harbour (Table 2).

To estimate the PECs in Port of Copenhagen and Port of Gdynia, activity data from STEAM 3.5.3 (updated 10.8.2021) and emission factors from previous studies (Ytreberg and Hassellöv, 2020; Jalkanen et al., 2021; Lunde Hermansson et al., 2021) were combined with the hydrodynamic and chemical fate model MAMPEC. For the default OECD ports, the ship activity data, i.e., number of ships and total power output, was estimated from previous studies and was assumed to be the same (Section 2.2 and Table 3).

2.1. Modelling approach

The MAMPEC model v 3.1 BW (Van Hattum et al., 2002), a 2D hydrodynamic and chemical fate model that assumes steady state, was used to calculate PECs of 9 metals and 16 PAHs (Table 4 and Supplementary Material A). MAMPEC is an established model used in regulatory assessments of antifouling paints and ballast water treatment systems (Van Hattum et al., 2002; Zipperle et al., 2011).

The MAMPEC model consist of three modules, *Environment*, *Compound* and *Emission*. In the *Environment* module, the Port of Copenhagen and Port of Gdynia were defined (Supplementary Material A) and the default OECD ports (OECD, 2005) were included for comparison. In the *Compound* module, the 9 metals and 16 PAHs were added individually which required compound specific characterisation (e.g. partitioning coefficients, solubility and degradation rate). The contaminants were selected for this study (Table 4 and Supplementary Material A) as they are known constituents of scrubber discharge water (Lunde Hermansson et al., 2021). Compound characteristics needed for the modelling in MAMPEC were collected from literature and the US-EPA software EPI-Suite (Supplementary material A).

The average daily load (g/day) of the metals and PAHs emitted to the different ports (described in Section 2.2) were used in the *Emission* module of MAMPEC. In this study, a total of five sources were included (Table 3), all contributing to the metal and PAH load to the port. The final output of MAMPEC is modelled environmental concentrations, i.e. average PECs, in water (total and dissolved species) and sediment. The water compartment was the focus of this study. As MAMPEC is limited to one contaminant and a constant daily load per model run, the results were transferred and compiled into an Excel sheet (Supplementary material B) where the principle of MAMPEC was applied but now with added flexibility of adding several contaminants and sources to calculate the RCR_{sum} and to assess the contribution of specific contaminants and sources to the risk.

The calculations in MAMPEC are based on linear correlation between the loads of the specific compounds, i.e. the mass added to the environment, and the resulting PEC (Fig. 2). Thus, from one run in MAMPEC, the slope (k_i) could be determined for each compound (i) for the predefined port environments. Then, the PEC could be calculated for all mass additions, allowing for change in both volume and concentration of the scrubber discharge water and the inclusion of more than one contaminant sources (Fig. 2 and Supplementary Info B).

The PECs were then compared to their respective PNECs to calculate RCR and RCR_{sum} (Eq. 1). When available, the PNEC values were based on the annual average environmental quality standards (AA-EQS) published in the Water Framework Directive (Annex X in WFD (2013)). For the remaining substances, those included in the WFD as River Basin Specific Pollutants (RBSP) in Sweden was assigned the Swedish threshold value for Good Ecological Status in marine and coastal areas while the PNECs for the rest of the substances were collected from publicly available risk assessment reports (Supplementary material A). PNEC values are sometimes derived for the total concentration and sometimes for the dissolved concentration. If the PNEC of a compound was based on the dissolved fraction (e.g. Ni), this was also the fraction collected from the MAMPEC results. Most metals (except mercury (Hg) and V) are assessed based on the dissolved fraction while all the PAHs are assessed based on total concentration (see Supplementary materials A and B).

The Excel compilation could also be used to calculate the theoretical maximum volume of scrubber water that could be discharged per day ($Volume_{max}$) in a specific port environment without exceeding $RCR > 1$ (Eq. 2):

$$Volume_{max} = \frac{\left(\frac{RCR_i}{\sum RCR}\right) \times PNEC_i}{k_i} \tag{2}$$

where the $Volume_{max}$ can be interpreted as volume equivalents to derive a maximum allowable total addition to the environment, i.e., when input from other sources were added, the $Volume_{max}$ would decrease.

Although the MAMPEC model has been validated for some antifouling and ballast water products (Van Hattum et al., 2002; Zipperle et al., 2011; van Hattum et al., 2014), it has not been used to assess scrubber discharge water, which is a mixture of several different substances at various concentrations. Applying the RCR_{sum} approach, based

Table 2
Port environments with defined length and width used as input to MAMPEC and the calculated water exchange volume (and percentage) per tidal period.

Port name	Length (m)	Width (m)	Water exchange volume (m^3/tidal period)	% water exchange (% per tidal period)	Tidal difference (m)	Non-tidal difference (m)
OECD EU	5000	1000	5.1×10^7	68	1.5	0
OECD Baltic	5000	1000	1.3×10^6	1.7	0	0.08
Port of Copenhagen	1600	1000	2.9×10^5	2.4	0	0.3
Port of Gdynia	1300	2600	4.3×10^5	1.1	0	0.03

Table 3
Comparison of the different magnitude of contaminant sources within the pre-defined environments. HFO stands for Heavy Fuel Oil consumption and MGO/MDO stands for Marine Gas Oil/Marine Diesel Oil consumption.

	Total power output (approximate fraction of fuel and abatement method)	Bilge water volumes	Antifouling release rate		Closed loop volumes	Open loop volumes
	(MWh/day)	(m^3/day)	Cu (g/day)	Zn (g/day)	(m^3/day)	(m^3/day)
OECD-EU	295 (8.5 % HFO + scrubber and 91.5 % MGO/MDO)	13	22,570	4054	0.54	2160
OECD-Baltic	295 (8.5 % HFO + scrubber and 91.5 % MGO/MDO)	13	6909	2118	0.54	2160
Port of Copenhagen	100 (20 % HFO + scrubber and 80 % MGO/MDO)	11.3	5965	1065	0.06	917
Port of Gdynia	126 (5 % HFO + scrubber and 95 % MGO/MDO)	9.3	22,787	3750	0.009	287

Table 4
The annual load of metals and PAHs from open loop scrubber water discharge within the port of Copenhagen and port of Gdynia in 2018, based on data from STEAM. As a complement, the open loop contribution to the total load is showcased in the columns to the right.

	Annual load from open loop scrubber discharge (kg/year) in 2018		Percentage of the total annual load from ships within the port that is due to open loop scrubbers	
	Port of Copenhagen	Port of Gdynia	Port of Copenhagen	Port of Gdynia
Arsenic (As)	2.3	0.7	94 %	85 %
Cadmium (Cd)	0.3	0.1	99 %	99 %
Chromium (Cr)	5.0	1.6	98 %	96 %
Copper (Cu)	12	3.8	1 %	0.05 %
Lead (Pb)	2.9	0.9	100 %	99 %
Mercury (Hg)	3.0×10^{-2}	9.0×10^{-3}	100 %	100 %
Nickel (Ni)	16	5.0	98 %	95 %
Vanadium (V)	57	18	99 %	98 %
Zinc (Zn)	37	12	9 %	1 %
Acenaphthene (Ace)	6.4×10^{-2}	2.0×10^{-2}	91 %	79 %
Acenaphthylene (Acy)	4.0×10^{-2}	1.3×10^{-2}	96 %	89 %
Anthracene (Ant)	2.7×10^{-2}	8.0×10^{-3}	96 %	91 %
Benzo[*a*]anthracene (BaA)	4.0×10^{-2}	1.3×10^{-2}	99 %	97 %
Benzo[*a*]pyrene (BaP)	1.7×10^{-2}	5.0×10^{-3}	97 %	93 %
Benzo[*b*]fluoranthene (BbF)	1.3×10^{-2}	4.0×10^{-3}	97 %	92 %
Benzo[*ghi*]perylene (BghiP)	7.0×10^{-3}	2.0×10^{-3}	91 %	80 %
Benzo[*k*]fluoranthene (BkF)	0.003	1.0×10^{-3}	95 %	87 %
Chrysene (Chr)	6.4×10^{-2}	2.0×10^{-2}	98 %	96 %
Dibenzo[*a,h*]anthracene (DahA)	1.0×10^{-2}	3.0×10^{-3}	99 %	97 %
Fluoranthene (Fla)	5.4×10^{-2}	1.7×10^{-2}	94 %	85 %
Fluorene (Flo)	0.2	4.8×10^{-2}	92 %	80 %
Indeno[1,2,3-*cd*]pyrene (InP)	2.3×10^{-2}	7.0×10^{-3}	99 %	97 %
Naphthalene (Nap)	0.9	0.3	79 %	60 %
Phenanthrene (Phe)	0.5	0.2	97 %	91 %
Pyrene (Pyr)	0.1	3.3×10^{-2}	95 %	87 %

on the results from MAMPEC, is one way to include more than one contaminant from several different sources. The RCR_{sum} approach can however result in overestimation of toxicity according to Backhaus and Faust (2012). Since only a selection of contaminants have been included (9 metals and 16 PAHs), for which the potential synergistic effects are not accounted for, and not all operational discharges are represented (e.g. stern tube oil, cooling water and ballast water), the RCR_{sum} approach is considered appropriate to use as a conservative approach. If more data on additional contaminants and their ecotoxicological effects are collected, and more sophisticated environmental fate and transport models in water become available, this may have to be revised, but since an environmental risk assessment should be based on worst case scenarios (EC, 2018; MEPC, 2022), RCR_{sum} was considered the best available approach.

2.2. Definition of specific loads of different contaminant sources

For the Port of Copenhagen and Port of Gdynia, the annual volumes of bilge water effluents and scrubber water discharge were derived from STEAM. The annual mass-release of Cu and Zn from antifouling paints were also derived from the same dataset. In STEAM, vessel activity data was obtained from the HELCOM AIS data archive and consisted of over 660 million automatic position reports from ships received during Jan 1st and December 31st, 2018. The vessel technical database used to describe the features of the global fleet was obtained from IHS Markit. Air emissions and water discharges from ships modelled with STEAM were output to annual 0.02-degree grids. Contributions from ambient effects (wind waves, sea ice, currents) in STEAM were excluded from this study.

The volume-based emission factors (bilge water and scrubber water) and mass-release of biocides from antifouling paints are described in Jalkanen et al. (2021). In the updated version of STEAM, used in this study, the volume-based emission factor of scrubber water has been redefined to 90 m^3/MWh (compared to previous 45 m^3/MWh) for open loop scrubber and 0.45 m^3/MWh (compared to previous 0.3 m^3/MWh) for closed loop water. The selection of grids to be included within the Port of Copenhagen and Port of Gdynia and the extraction of selected data from STEAM were performed with MATLAB R2020a (Supplementary material A).

For both the OECD EU and Baltic harbours, the daily power output (MWh/day) was used to calculate the discharge volumes of bilge and scrubber water. Faber et al. (2019) estimated the daily auxiliary loading at berth (P_{aux}) in a port to correspond to 192 MWh/day based on a scenario of continuous (i.e. 24 h) 8 MW port load. According to fuel consumption for ships at berth versus the total fuel consumption, 60 % of the fuel consumption was represented by ships at berth in Port of Copenhagen while in Port of Gdynia, 70 % of the fuel consumption originated from ships at berth (Supplementary material A). Based on this, 65 % of the power output within a harbour was assumed to correspond to ships at berth and the total daily power output (P_{tot}) in the OECD harbours was calculated to 295 MWh/day (Eq. 3).

$$P_{tot}\left(MWh\left(day^{-1}\right)\right) = P_{aux}\left(192\ MWh\left(day^{-1}\right)\right)*\frac{100}{65} \quad (3)$$

Faber et al. (2019) argued that the berthing load used in their report (192 MWh/day) was relatively high, which indicates that a total power

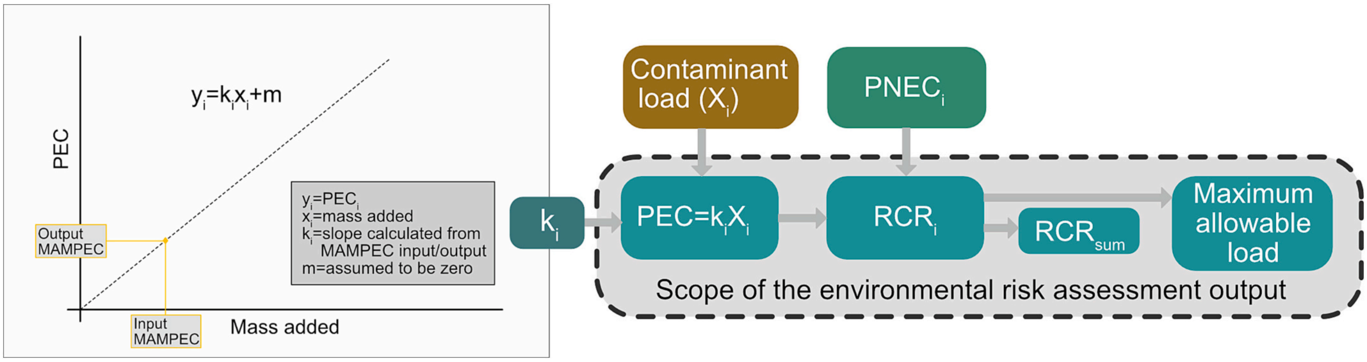

Fig. 2. The graph illustrates the linear relationship between contaminant load (x-axis) and predicted environmental concentration (y-axis) according to MAMPEC v.3.1. This information was used to calculate the slope (k) that was applied as input in environmental risk assessment (within the dashed square) to calculate the risk associated to several contaminants and sources.

output of 295 MWh/day in the generic OECD harbours would also be high. However, another study shows that the power output within the Scheldt estuary was 499 MWh/day and in Antwerp harbour docks the power output was 1293 MWh/day (Teuchies et al., 2020). This would instead suggest that the total power output of the OECD harbours of this study is rather low. Especially considering that the OECD ports are based on the port of Rotterdam which is the largest port in Europe, handling more than twice as much freight as Antwerp in 2019 (Eurostat, 2022).

The power output of the fleet operating with scrubbers was calculated by assuming that 8.5 % of the power output of the entire fleet were equipped with scrubbers (=25.2 MWh/day), and that closed loop scrubbers constituted 5 % of the scrubber fleet (=1.2 MWh/day). This was decided based on the share of the fleet equipped with scrubbers that arrived at port of Antwerp in 2019 (8.7 %) (Teuchies et al., 2020). Also, STEAM fuel consumption data from Port of Gdynia and Port of Copenhagen showed that ships equipped with scrubbers, i.e. ships running on heavy fuel oil (HFO), constituted 5 % and 20 % of the total fuel consumption respectively. As this study is based on 2018 data, and the installations of scrubbers have increased (Jalkanen et al., 2021), 8.5 % of total power output might be an underestimation. The power outputs of the open and closed loop scrubber fleet were multiplied by the discharge flow rates of the respective system; 90 m^3/MWh for open loop and 0.45 m^3/MWh for closed loop (Ytreberg and Hassellöv, 2020; Ytreberg et al., 2022), yielding the daily discharge volumes of 2160 m^3/day for open loop and 0.54 m^3/day for closed loop (Table 3).

The mass load for each compound from the scrubbers could then be calculated by multiplying the average concentration of compound *i* (μg/L) in the scrubber discharge water (Lunde Hermansson et al., 2021) with the calculated (for OECD harbours) or modelled (for the Ports of Copenhagen and Gdynia) daily discharge flow (m^3/day) (Table 3). More information regarding the concentrations of metals and PAHs in scrubber water is described Lunde Hermansson et al. (2021). Briefly, the data is based on several sampling campaigns on open and closed loop scrubbers between 1993 and 2018. Values reported as below limit of detection (<LOD) were treated as ½LOD.

The discharge volumes (L) of bilge water within the OECD harbours were calculated based on the same equations used in STEAM (Eq. 4) (Jalkanen et al. (2021), details in Supplementary material A):

$$\text{Bilge}_{\text{OECD}} = \left(\left(\left(0.1313 \times P_{\text{pass}} \right) + 373.4 \right) + \left(\left(0.0247 \times P_{\text{other}} \right) + 154.4 \right) \right) \times 0.75 \tag{4}$$

where P_{pass} is the power output (in kWh/day) of passenger ferries (assumed to correspond to 30 % of the total power output) and P_{other} is the power output of the remaining 70 % of the fleet. It was assumed that 25 % of the bilge water is delivered in port (Jalkanen et al., 2021 and refrences within) and a factor of 0.75 was added to the equation (Eq. 4).

The final loads from bilge water discharge were calculated by multiplying the average concentrations (Ytreberg and Hassellöv, 2020) with the estimated daily volume discharges, calculated for the OECD harbours (Eqs. 3 and 4) or collected from the STEAM 2018 data within the Port of Copenhagen and Port of Gdynia (Table 3).

To determine the loads of Cu and Zn from antifouling paints in the OECD harbours, the *OECD-EU Estuarine Commercial Harbour scenario* for emissions in the MAMPEC model, developed for antifouling paints, was used to derive the number of ships (24 per day), their respective surface area (between 1200 and 16,000 m^2 per ship) and the application factor (0.9) (OECD (2005) and Supplementary material A). The number of ships and their respective wetted surface area was used for both the OECD EU and Baltic harbours. The release rates of Cu and Zn from antifouling paints were adjusted to match the rates published in Jalkanen et al. (2021), where the *International* region (leaching rate Cu = 24.5 μg/cm^2/day and Zn = 4.4 μg/cm^2/day) was used for the OECD EU harbour and the *Baltic Proper* region (leaching rate Cu = 7.5 μg/cm^2/day and Zn = 1.4 μg/cm^2/day) was used for the OECD Baltic harbour (Table 3).

The daily atmospheric deposition was derived from; 1) the power output within the port environment (Table 3), 2) the emission factors (mg/MWh) of metals and PAHs to air from Lunde Hermansson et al., 2021 and 3) an estimated fraction of near-ship deposition, based on a study by Badeke et al. (2021), where near-ship downward dispersion of inert tracers was modelled. The atmospheric deposition was assumed to be 0.1 % of the total air emissions from the ships within the port environment. All combustion of HFO was assumed to be connected to a scrubber where most metals and some PAHs were scavenged by the scrubber water, i.e. being discharged directly to the water and not deposited after exhaust-emissions (Lunde Hermansson et al. (2021) Table A.5). As STEAM does not provide power output from the main and auxiliary engines separately, the power output within the ports of Copenhagen and Gdynia had to be calculated from fuel consumption to enable air emission calculations of metals and PAHs. In this study, a default of 250 kg fuel/MWh was used for all fuel types (Supplementary material A). Although this may seem high, with 180 kg/MWh being a more commonly used default value for fuel consumption (Moldanová et al., 2009; Zetterdahl et al., 2016; Teinilä et al., 2018), Faber et al. (2021) and Tronczynski et al. (2022) mention that the fuel consumption increases at lower loads which supports the assumption of higher fuel consumption in the port scenarios. This is also supported by the 2nd IMO Green House Gas study report from 2009 that shows the development of specific fuel consumption as a function of engine generation/size, where the fuel consumption ranges from 165 g/kWh (2-stroke low-speed engine) to 250 g/kWh (4-stroke medium-/highspeed engine (<1000 kW)).

The atmospheric emission factors obtained from Lunde Hermansson et al. (2021) were collected from studies carried out when ships were running in cruising mode, i.e. with an engine load >50 %. This is not the default mode within ports, where it is more common that ships are at berth or manoeuvring. More data is needed to verify atmospheric emission factors of metals and PAHs under different engine loads, using different fuel grades. To the knowledge of the authors, Lunde Hermansson et al. (2021) is the only available study where emission factors of metals and PAHs to both water and air are presented simultaneously and, to avoid double counting of the loads, this motivates the use of those emission factors.

3. Result and discussion

If the new MEPC guidelines on environmental risk assessment of scrubber water discharged are followed (MEPC, 2022), e.g. assessing the surrounding PECs, in the port of Copenhagen, the volumes of scrubber water discharge that would be considered to have an acceptable risk to the environment would surpass 200,000 m^3 day^{-1} (almost 90 million m^3 $year^{-1}$). This corresponds to half of the total discharge volume of scrubber water in the entire Baltic Sea area during 2018, amounting to almost 200 million m^3 (based on STEAM model output). The 200,000 m^3 day^{-1} also corresponds to a daily load of over 40 kg V, 9 kg Cu, 12 kg Ni and almost 1.5 kg of the sum of the 16 PAHs, only for the port of Copenhagen. For comparison, this hypothetical allowable input of Cu and Ni to the port of Copenhagen corresponds to 20 % (for Cu) and 40 % (for Ni) of the total input from all coastal point sources within the Baltic Sea Area in 2018 (HELCOM, 2021). This example shows that assessing the environmental risk using the PEC in the surrounding environment in MAMPEC, in accordance with the new MEPC guidelines, will not provide adequate protection of the marine environment.

The discharge of open loop scrubber water accounts for a substantial fraction of the load of both metals and PAHs to the ports of Copenhagen and Gdynia. Despite the relatively low share of the ships' estimated total power consumption (5 % in port of Gdynia and 20 % in the port of Copenhagen), the discharge of open loop scrubber water accounts for a significant fraction (often >90 %) of the annual load for most of the contaminants (Table 4). The remaining fractions come from the other contaminant sources where the load of Cu and Zn is predominantly derived from the use of antifouling paints while the PAHs are also found in bilge water and from atmospheric deposition.

Ten of the most toxic contaminants found in scrubber water, based on the 9 metals and 16 PAHs covered in this study, contribute to >95 % of the cumulative risk associated with open loop scrubber water discharge (Fig. 3). Several of the ten most toxic compounds (Fig. 3) are directly linked to the combustion of HFO. V can be traced back to the parent fuel, where the V content can be correlated to the sulphur content of the fuel (Moldanová et al., 2011; Celo et al., 2015). Also, the emission factors of heavier PAHs (InP, DahA, BaP, BghiP and chrysene (Chr)) are 2–20 times higher from HFO combustion as compared to marine gas oil (MGO) combustion (Lunde Hermansson et al. (2021) and references therein). This indicates that, prohibiting the use of scrubbers would reduce the environmental load to the ports, and thus RCR_{sum}, with respect to at least six out of the ten most toxic compounds from the operational discharge of scrubber water. However, the emerging hybrid fuels, i.e. residual fuel blends labelled very low sulphur fuel oils (VLSFO, max 0.5 % S) and ultra-low sulphur fuel oils (ULSFO, max 0.1 % S), will still be allowed on the market. The emission factors, and subsequent deposition on the sea surface, of metals and PAHs from hybrid fuels must be further studied to be included in future analysis.

Cu and Zn in the scrubber water can also originate from the antifouling systems of the vessels, where the cooling water system, which is often connected to the scrubber water intake, is enriched with these metals due to the use of sacrificial anodes (Kjølholt et al., 2012; Koski et al., 2017) and marine growth protection systems (Growcott et al., 2016). As both Cu and Zn are identified as contributing substantially to the RCR_{sum} from open loop scrubber water discharge (Fig. 3), more attention to the cooling systems as a source of contaminants, e.g. discharge volumes and contaminant concentration, should be prioritized by policymakers and efforts should be made to also include cooling water discharge in environmental risk assessments of shipping.

It is important to note is that the four heavier PAHs and arsenic (indicated with * in Fig. 3 and listed in Table 5) are often found at concentrations lower than the limit of detection (<LOD) in scrubber discharge water, with the concentrations then treated as ½LOD. Despite this, the low PNEC values (Supplementary material A), i.e. high toxicity, of these compounds result in significant contribution to the RCR_{sum} of open loop scrubber water. For BaP, the number of samples <LOD is around 54 % while for dibenzo[*a,h*]anthracene (DahA), almost 80 % is <LOD (Table 5 and Supplementary material A). If values <LOD are omitted (reducing the number of samples, n), the average concentration of arsenic (As), BaP and benzo[*ghi*]perylene (BghiP) increases by a factor of 2 while Indeno[1,2,3-*cd*]pyrene (InP) and DahA decreases, due to some high LODs (Table 5). In Table 5, the average concentrations are also shown for the assumptions where <LOD are treated as zero (<LOD = 0) and where <LOD are treated as being equal to LOD (<LOD = LOD) to present a potential range of concentrations in the scrubber water. If <LOD is treated as 0, the RCR_{sum} from discharge of open loop scrubber water would decrease with approximately 50 % in all ports. If <LOD is treated as LOD, the RCR_{sum} from discharge of open loop scrubber water would instead increase with around 50 % (40–75 %) in all ports. More sensitive analytical methods are required to achieve lower limits of

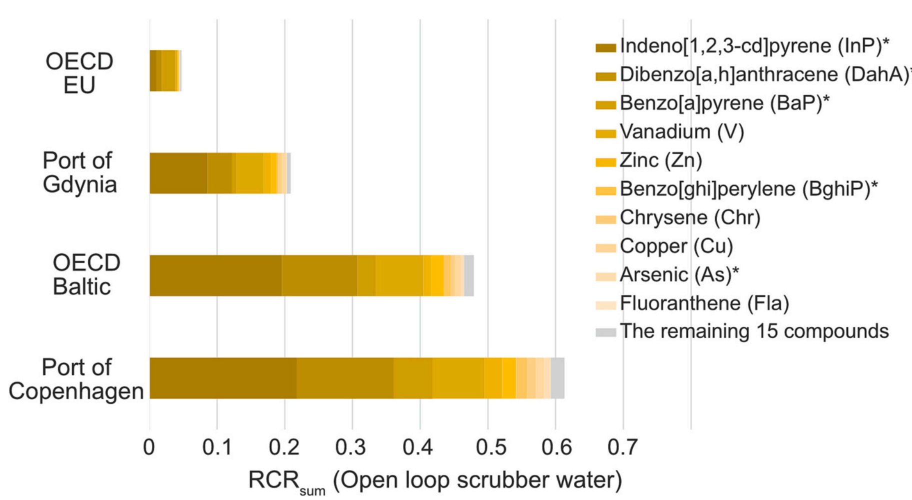

Fig. 3. The top-10 highest contributors to the cumulative risk characterisation ratio of metals and PAHs from open loop scrubber water. The compounds indicated by *, have been measured at concentration < LOD in >50 % of the occasions.

detection and quantification. Further research is needed to validate and, if more ecotoxicological data have become available, potentially update the PNEC values presented in Table 5 (and Table 4 in Supplementary material A). It is however beyond the scope of this work to assess the derivation of the PNEC values for the contaminants.

The discharge of scrubber water is only one of several sources of contaminants from ship-related operations within and outside a port. With the cumulative environmental risk assessment approach presented in this study (Supplementary material B), it is possible to assess several sources and contaminants, including their respective contribution to RCR_{sum}, simultaneously. The maximum allowable volume of scrubber water that can be discharged in a port, without exceeding $RCR_{sum} > 1$, can also be estimated (Fig. 4). For example, when no other sources or background concentrations are included in the assessment, the starting RCR_{sum} is zero and the calculated maximum allowable volume is the volume-limit of open loop scrubber water that can be discharged without exceeding RCR > 1. When other contaminant sources are included, the receiving capacity of the port environment, i.e. the maximum allowable discharge volume of scrubber water, will decrease when the RCR move closer to 1, i.e. the limit for unacceptable risk.

This is illustrated for the OECD Baltic harbour in Fig. 4 where the maximum allowable discharge volume of open loop scrubber water is 4500 m^3/day if no other contaminant sources or background concentrations are included in the risk assessment. When the load of metals and PAHs from near-ship atmospheric deposition, bilge water discharge, antifouling paint and closed loop scrubber water discharge is included, the maximum allowable volume of open loop scrubber water that can be discharged, before exceeding RCR > 1, decrease by almost 70 %, from 4500 m^3/day to approximately 1500 m^3/day (Fig. 4). This means that the calculated daily discharge volume of open loop scrubber water to the OECD Baltic harbour of 2160 m^3/day (Table 3) will result in an RCR > 1, implying that the resulting concentrations of metals and PAHs will pose an unacceptable risk to the port environment. If the other contaminant sources had not been included, the assessment would falsely result in an $RCR_{sum} < 1$ and the conclusion would have been that the risk is acceptable, indicating that no mitigations are neceesssary.

The maximum allowable discharge volume of scrubber water varies greatly between the different ports (Fig. 5). These results highlight the importance of using region-specific harbour environments, representing local conditions e.g. tidal difference and water exchange rates, when estimating environmental risks from ship emissions. For example, if the assessment was based on the OECD EU harbour, a 10-fold increase in maximum acceptable discharge volume as compared to the OECD Baltic harbour would be allowed (Fig. 5 to the left). Also, if the risk associated to open loop scrubbers is assessed based on the generic OECD EU harbour, the annual maximum allowable volume of open loop scrubber water that could be discharged within a single port environment would be almost 17 million m^3, corresponding to almost 3 t of V, 800 kg Ni, 600 kg Cu, 50 kg of naphthalene (Nap) and 25 kg of Phe during a year. The allowable input of Cu and Ni to the OECD EU harbour would then be in the same order of magnitude as the amounts emitted from all coastal point sources in Denmark during 2018, i.e. 760 kg (Cu) and 1200 kg (Ni) (HELCOM, 2021). Thus, it should be questioned how appropriate the use of the OECD EU harbour is for assessing environmental risks from ship contaminations.

Although the OECD Baltic harbour better correspond to the two existing ports characterised within this study, the maximum allowable volume is still approximately three times higher in the generic OECD Baltic harbour compared to the ports of Copenhagen and Gdynia (Fig. 5 to the right). These results indicate that the use of generic OECD harbours underestimate the risk associated to ship operations in ports and is thus not aligned with presenting reasonable worst-case scenarios when performing environmental risk assessments (EC, 2018; MEPC, 2022).

The complex composition of the scrubber water is also important as several contaminants are all contributing to the toxicity and effects of the discharged effluent. Previous studies (Lunde Hermansson et al. (2021) and references therein) have shown great variation in concentration of metals and PAHs in scrubber discharge water. The choice of scrubber water concentrations will have a direct impact on the RCR calculations and thus the maximum allowable volumes of scrubber water discharge. As an example, if the upper level of the 95 % confidence interval of the concentrations in open loop scrubber water is used instead of the average concentration (as can be supported by MEPC (2022) if the upper level are deemed more appropriate worst-case emission factors), the maximum allowable volume of open loop scrubber water that can be discharged is reduced by 30 % (OECD EU) to 40 % (OECD Baltic) (Supplementary material B).

Other contaminants, e.g. alkylated PAHs, are not included in the current environmental risk assessment. Studies have shown that alkylated PAHs are present in scrubber water (Thor et al., 2021; Du et al., 2022) and that some of them have a higher toxicity than their parent compounds (Achten and Andersson, 2015; Cong et al., 2021; Thor et al., 2021). If more contaminants are added to the assessment, that would also reduce the maximum allowable volume. To add new contaminants to the approach presented in this work, new PNEC values based on ecotoxicological data must be derived first. This was however beyond the scope of this study.

According to the results from MAMPEC, a large fraction of the total load of contaminants is predicted to be flushed out of the port environment. It is important to note that the contaminants transported out of the port will imply a risk in another environment, i.e. the surrounding environment. Using the Port of Copenhagen as an example again, with the contaminant loads calculated from STEAM 2018 data together with the emission factors and concentrations, the annual load of the sum of the 16 PAHs to the surrounding environment is above 300 g. For metals, the highest loads to the surrounding environment are represented by Cu (1200 kg/year), Zn (170 kg/year), V (52 kg/year) and Ni (12 kg/year). According to the European Pollutant Release and Transfer Register Annex II, listing the threshold values that requires industries to report their emissions of pollutants in environmental reports, the annual load of Cu must be reported if it exceeds 50 kg. The ship-related loads of Cu from Port of Copenhagen to the surrounding environment, based on

Table 5
The average concentrations used in the analysis compared to the average concentration if all <LODs were omitted associated to five of the compounds contributing most to the risk where >50 % of measurements were <LOD. The range and median values of the LODs. For comparison is the PNEC values listed for each compound.

	Average concentration (ALL; <LOD = ½LOD) (μg/L)	Average concentration (excluding <LODs) (μg/L)	Min average concentration (ALL; <LOD = 0) (μg/L)	Max average concentration (ALL; LOD=) (μg/L)	PNEC (μg/L)
Arsenic (As)	6.8 (n = 66)	11 (n = 24)	3.9	9.7	0.55
Benzo[*a*]pyrene (BaP)	0.05 (n = 64)	0.11 (n = 28)	0.04	0.06	1.7×10^{-4}
Indeno[1,2,3-*cd*] pyrene (InP)	0.07 (n = 63)	0.03 (n = 14)	0.01	0.13	2.7×10^{-4}
Benzo[*ghi*]perylene (BghiP)	0.02 (n = 63)	0.05 (n = 22)	0.02	0.03	8.2×10^{-4}
Dibenzo[*a,h*] anthracene (DahA)	0.03 (n = 63)	0.02 (n = 10)	0.00	0.04	1.4×10^{-4}

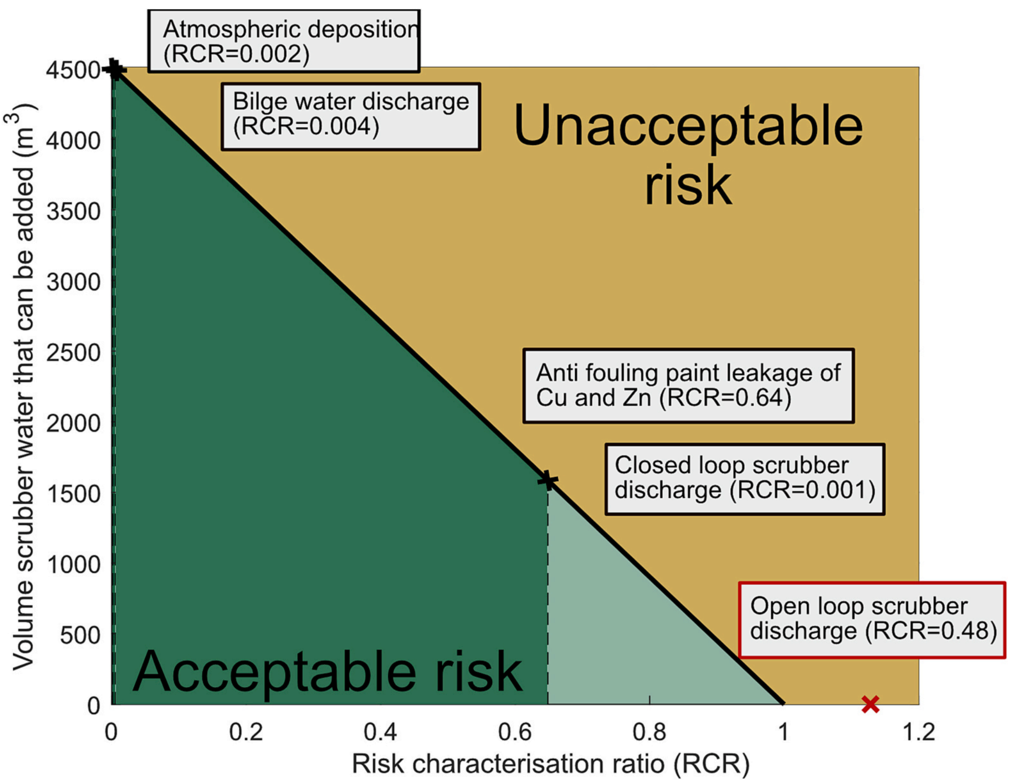

Fig. 4. The area left to the black line represent acceptable risk scenarios and the area to the right (yellow) represent scenarios of unacceptable risk in the OECD Baltic harbour environment. With every added contaminant source, the maximum allowable volume of scrubber water that can be discharged without exceeding $RCR_{sum} = 1$ decreases. When all five sources are included, $RCR_{sum} > 1$ (represented by red cross) and there is an unacceptable risk to the environment. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

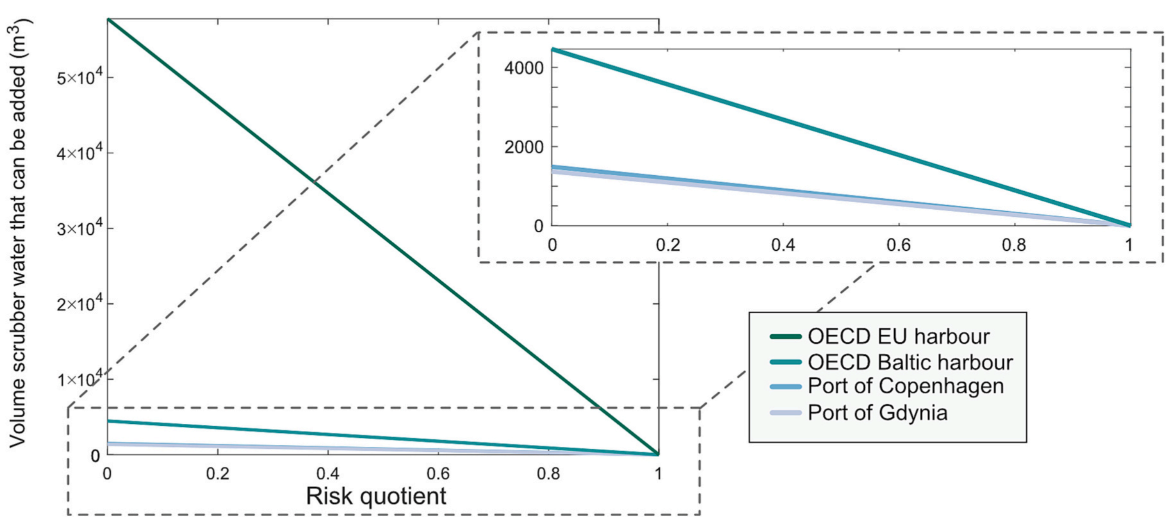

Fig. 5. Comparison of the maximum allowable open loop scrubber volumes, equivalent to contaminant load, that can be discharged to the defined environment. In the inset, the OECD EU harbour is excluded and focus on the three remaining ports for clearer comparison, ranging from 0 to 4500 m^3.

2018 STEAM data and MAMPEC model runs, are exceeding that by 24 times. Similarly, the reporting threshold values for Zn and Ni are 100 kg/year and 20 kg/year respectively.

The different port environments have different water exchange rates and are exposed to different levels of contaminant loads, resulting in RCR_{sum} from 0.2 (OECD EU harbour) up to 13 (Port of Gdynia) (Fig. 6). In all scenarios, the discharge of open loop scrubber water and the release of Cu and Zn from antifouling paints are the largest contributors to the RCR_{sum} (Fig. 6), suggesting that the greatest reduction of RCR_{sum} can be reached if these activities are regulated more strictly or even prohibited. This is also in accordance with Ytreberg et al. (2022) who came to the conclusion that scrubbers and antifouling paints are the main contributors to the metal and PAH load to the Baltic Sea and that a restriction of these inputs would significantly reduce the total loads to the Baltic Sea.

In the four different ports, the atmospheric deposition had minor contributions (approximately 0.1 %) to RCR_{sum} (Fig. 6). However, during conditions of more efficient near-ship deposition, this contribution would increase. As an example, when PAH mass fluxes around Shanghai water systems was modelled with a fugacity model, >30 % of the 5–6 ring PAHs, emitted from ships, was estimated to deposit on the water surfaces (Su et al., 2021). In the same study, >1 % of the ship emissions were deposited for all of the PAH groups, indicating that the 0.1 % assumed in this study might be an underestimation. As shown by Badeke et al. (2021); stack height, ship shape, wind speed and wind direction will affect the dispersion of the emissions and in some

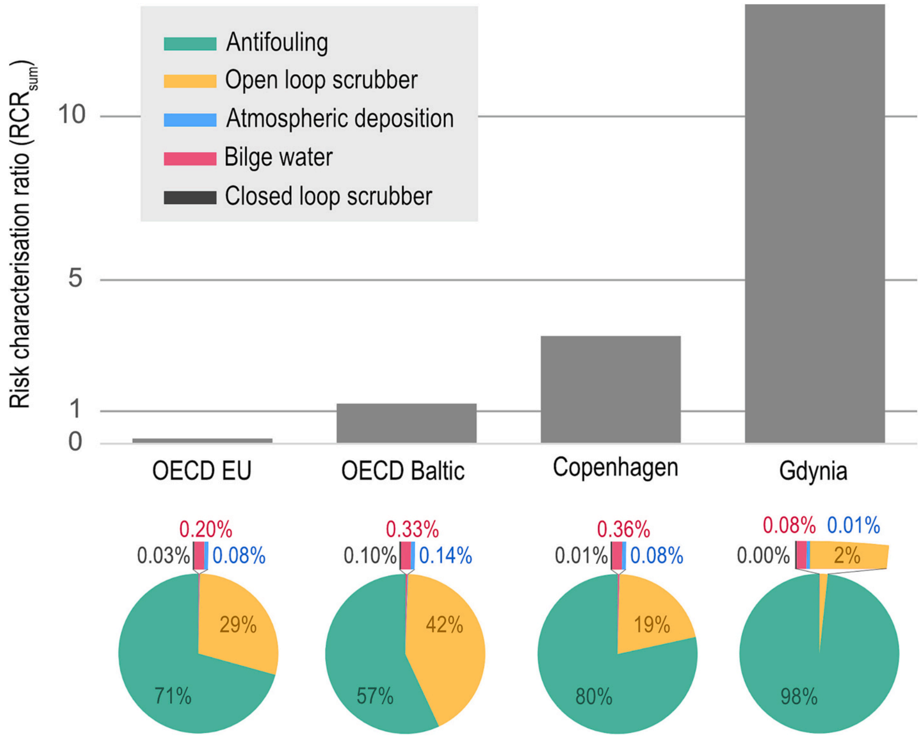

Fig. 6. The sum of risk characterisation ratio (RCR_{sum}) in the different ports and the contribution of the different contaminant sources. The pie charts showing the elative contribution of contaminant sources to the total risk (RCR_{sum}) in the different ports.

scenarios, the downward dispersion could constitute up to 55 % of the emissions. More work is needed to better account for the near-ship deposition but was included in this study as a conceptual contributor to highlight the importance of including different types of contaminant sources, all originating from ships. This study focuses on the local contaminant sources from ships, i.e. the ships within the port area, meaning that atmospheric long-range transportation, other industries or background concentrations are not included in the calculation of RCR and RCR_{sum}. The RCR_{sum} thus represent the cumulative risk contribution of the selected onboard systems (Table 3) and contaminants (Table 4).

This work is based on the calculations and output from MAMPEC, and all uncertainties from MAMPEC will propagate to the results and further calculations. One of the main issues with the MAMPEC model is the assumption of spatial and temporal homogeneity, where input, output and general conditions are assumed to be constant, resulting in a predicted concentration at steady state. Zipperle et al. (2011) carried out a sensitivity assessment, changing the decay rate and flushing within the standard OECD Commercial harbour, and concluded that the average concentrations could be well estimated by MAMPEC but that the maximum concentration was not. It was however advised that the lower flushing and decay rate should be applied to ensure that the results are based on realistic worst-case scenarios.

In the Port of Copenhagen, the daily ship-generated load of metals that will deposit on the sediment surface within the port environment varies between 10 % (V) to 70 % (Pb) of the total load from ships, equalling 15 g/day for V and almost 6 g/day for Pb. PAHs are also subject to degradation in the water and sediment, with half-lives ranging from a couple of days up to several years (Supplementary material A) and the fraction of the total ship-generated contaminant load that sedimented was generally lower for PAHs (0–35 %) with a trend of higher degree of sedimentation by the heavier molecular weight PAHs. The sediment can act as a sink, where most contaminants accumulate over time, and might thus be a better indicator than the water column when assessing the risk in an environment. However, due to the lack of appropriate PNEC values within the sediment compartment it was decided to limit the assessment of risk to the water column.

The MEPC (2022) guidelines propose that background concentrations of chemical substances should be added, which has not been done within this work. The results in Fig. 4 and Fig. 6 reflects the risk associated with the added contaminant load of metals and PAHs from ships. If backgound concentrations were to be added, the initial conditions in Fig. 4 would not be $RCR = 0$ and, depending on the background concentrations, the maximum allowable volume of scrubber water before exceeding $RCR > 1$ would be reduced. Determining background concentrations in port areas requires more knowledge of pre-industrial concentrations and contaminant loads from other activities. However, as RCR_{sum} is exceeded in both the Port of Copenhagen and the Port of Gdynia based solely on added input of contaminants from ships, it is reasonable to assume that the results would be reinforced if background concentrations were included.

For comparison, there are not many sampling campaigns of metals and PAHs conducted within the port areas. However, in the Port of Copenhagen, most of the predicted metal concentrations are lower than what have been measured by Koski et al. (2017) (sampled in 2016). According to the measurements from Koski et al. (2017), the RCR_{sum} for As, Cr, Cu, V and Zn is between 6 and 12, being two to four times higher than the RCR_{sum} estimated from this study for 9 metals and 16 PAHs. Similarly, the average concentrations from monitoring of cadmium (Cd), lead (Pb) and Zn in the Port of Gdynia (Popek et al., 2021) are up to 1000 times higher than the estimated PECs for these compounds calculated in this study. This indicate that 1) the approach used in the current study is underestimating environmental concentrations in the selected port and/or 2) the ports of Copenhagen and Gdynia are already severely polluted from shipping and other sources. More effort is needed to compile datasets that includes monitoring data, determination of background concentrations and other contaminant sources. Despite the uncertainties and lack of data, this study shows that there is an unacceptable risk in three out of four port environments when assessing the cumulative risk of metals and PAHs.

4. Conclusions

The approach presented in this work is based on the initial output from MAMPEC together with ship activity data (STEAM) and offers the opportunity to calculate PECs for several different substances emitted from different contaminant sources simultaneously. The calculations also provide information on the total added load of contaminants and the different contaminant sources' relative contribution to risk. All of which can then be used when evaluating risks as well as proposing mitigation strategies.

Environmental risk assessments, in accordance with the new the MEPC (2022) guidance document (i.e. assessing the surrounding rather than the port environment with MAMPEC), will not provide adequate protection of the marine environment. Based on environmental risk assessments in ports, the contribution of near-ship atmospheric deposition, bilge water discharge, release of Cu and Zn from antifouling paints and the discharge of scrubber water result in unacceptable risk in three out of four port areas. Antifouling paint and open loop scrubber water discharge are the two main contributors to RCR_{sum}. Moreover, the discharge of scrubber water represents an additional risk to the surrounding environment and should be prohibited in areas that fail to reach Good Environmental Status according to Paragraph 7.4 in MEPC (2022). This study shows that there is a significant contaminant load of metals and PAHs from ships in ports and that a large fraction of the load will be transported to the surrounding environment. Therefore, a more holistic assessment of ship-activities is needed to fully understand the impact on the marine environment.

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CRediT authorship contribution statement

Anna Lunde Hermansson: Methodology, Investigation, Writing – original draft, Writing – review & editing, Visualization, Formal analysis. **Ida-Maja Hassellöv:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition. **Jukka-Pekka Jalkanen:** Writing – review & editing, Resources. **Erik Ytreberg:** Conceptualization, Investigation, Writing – review & editing, Supervision, Funding acquisition.

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.

Data availability

I have shared the data as Supplementary Material A and B (attached).

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