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Metal and PAH loads from ships and boats, relative other sources, in the Baltic Sea

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ABSTRACT

The Baltic Sea is a sensitive environment that is affected by chemical pollution derived from multiple natural and anthropogenic sources. The overall aim of this study was to estimate the load of metals and polycyclic aromatic hydrocarbons (PAHs) from shipping and leisure boating, relative other sources, to the Baltic Sea and to identify possible measures that could lead to major reductions in the loads of hazardous substances from maritime shipping and leisure boating. The use of copper-based antifouling paints, and operation of scrubbers in open loop mode, were the two most dominant identified sources of hazardous substances to the Baltic Sea. Open loop scrubbers accounted for 8.5 % of the total input of anthracene to the sea. More than a third of the total load of copper can be reduced if copper-free antifouling paints or other biocide-free antifouling strategies are used on ships and leisure boats.

1. Introduction

Baltic Sea is recognized as one of the most polluted sea areas in the world. Due to the northerly geographic location, natural hydrographic properties, large catchment area and the semi-enclosed character of this brackish inland sea, concentrations of hazardous substances are considerably higher in the Baltic Sea than in other seas (Pohl and Hennings, 2007). According to the latest integrated contamination status assessment in the Baltic Sea, conducted by the Baltic Marine Environment Protection Commission, also known as the Helsinki Commission (HELCOM), for the period 2011–2016, all Baltic Sea basins failed to reach good environmental status as defined by EU's Maritime Strategy Framework Directive (MSFD) (HELCOM, 2018). The assessment is based on thirteen core indicators comprising different hazardous substances and substance groups, including the polycyclic aromatic hydrocarbons (PAH) benzo(*a*)pyrene, anthracene and fluoranthene and the heavy metals mercury (Hg), cadmium (Cd) and lead (Pb). Other metals, e.g. copper (Cu) and zinc (Zn) are also routinely monitored in the Baltic Sea, and concentrations in sediment and water are frequently exceeding national threshold values (Ytreberg et al., 2021c). Therefore, Cu is proposed to be included as a core indicator in HELCOMs third holistic assessment (HOLAS III) (Lagerström et al., 2021a).

The Contracting Parties of HELCOM have agreed to monitor hazardous substances, assess the environmental status and to propose and undertake measures to prevent and eliminate pollution of the Baltic Sea environment. Compilations of pollution load data (PLC) have been an integral part of the HELCOM assessment system since 1987, focusing on annual and periodic assessments of inputs of nutrients and selected hazardous substances. Thus, load compilation (tonnes per year) of various heavy metals from riverine inputs to the Baltic Sea is available from 1995 to 2019 (HELCOM, 2019). In addition to the riverine input, HELCOM assessments also include input of metals from direct point sources (e.g. coastal waste water treatment plants and industries) (HELCOM, 2021b) and modelled Cd, Cu, Hg, Pb, and benzo[*a*]pyrene deposition on the Baltic Sea from the EMEP/MSC-E regional model (EMEP, 2021; HELCOM, 2021b). The EMEP/MSC-E modelling involves collection of emission inventory data, description of atmospheric transport and the deposition of air pollutants. Further, the results are validated against measurements of air quality and chemical analysis of precipitation. However, inclusion of ship emissions (and the corresponding deposition on the Baltic Sea) is limited to only a few substances, e.g. emissions of nitrogen oxides and N deposition (Gauss et al.,

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2020), or as for Cd, Pb, Hg, benzo[*a*]pyrene where the EMEP model only include domestic shipping and does not comprise international shipping (EMEP, 2020). In contrast, atmospheric deposition assessed through environmental monitoring typically include more substances and integrates atmospheric emissions from all sources, including shipping. However, it is not possible to resolve the emissions from the background.

Despite being two sectors known to emit both PAHs and metals (Turner et al., 2017a), the loads from maritime shipping and leisure boating have so far not been explicitly included in the HELCOM assessments, nor has any direct comparison to loads from other natural and anthropogenic sources been conducted in the Baltic Sea region. These knowledge gaps make it challenging to assess and quantify potential measures to reduce the input of hazardous substances. In a recent publication by Jalkanen et al. (2021), Baltic Sea ship emissions from different liquid waste streams (open and closed loop scrubber wash water, bilge water, grey water and sewage) and biocides from antifouling paints were calculated using the Ship Traffic Emission Assessment Model (STEAM). STEAM uses AIS (Automatic Identification System) data positions from all vessels equipped with AIS transceiver which allows vessel identification and consecutive determination of e.g. the vessels' passenger capacity, main engine power, gross tonnage, vessel size, hull surface area etc. based on vessel databases, like IHS Markit. STEAM allows for generation of activity, emission and discharge maps (volumes of different liquid waste streams) of individual vessels or group of vessels within the study domain, in this case the Baltic Sea area. The results from Jalkanen et al. (2021), which is based on ship activity in 2012, showed PAHs and/or metals to be present in open- and closed loop scrubber water, bilge water, grey water, sewage and antifouling paint.

Antifouling paints were identified to be the main source of both copper and zinc from shipping. Globally, cuprous oxide is the dominating biocide in antifouling paint, often in combination with zinc oxide to increase the overall toxicity and to control the leaching process (Amara et al., 2018). The use of antifouling paints has shown to cause elevated concentrations in marinas and natural harbours in the Baltic Sea region (Egardt et al., 2017; Kylin and Haglund, 2010; Lagerström et al., 2020a). According to a recent compilation of 145 commercially available antifouling paints for the shipping and leisure boat sector, the release rate of Cu can vary from 2 to 66 $\mu g/cm^2/day$ between antifouling products (Jalkanen et al., 2021). Typically, a higher release rate is needed to protect the hull surface from biological fouling in areas with high fouling pressure, while a lower release rate is needed in low-fouling areas such as the Baltic Sea (Lagerström et al., 2018). Recent studies suggest that a release rate of 2.2 $\mu g/cm^2/day$ and 5 $\mu g/cm^2/day$ to be sufficient to prevent macrofouling (e.g. barnacles and macroalgae) in the Baltic Proper and Kattegat, respectively (Lagerström et al., 2020b), indicating that most antifouling coatings for the shipping sector are excessively toxic when used on ships in the Baltic Sea region.

Other ship-related waste streams that contain hazardous substances are bilge water from engine spaces, grey water from showers, laundry, and galleys, sewage, and wash water from the use of exhaust gas cleaning systems (also known as scrubbers) (Magnusson et al., 2018; Teuchies et al., 2020; Ytreberg et al., 2020). Scrubbers are used to remove sulphur oxides (SO_X) by leading the engine exhaust through a fine spray of water (Turner et al., 2017b). Thereby, the emissions of SO_X to the air can meet the emission levels corresponding to combustion of fuel with 0.5 % sulphur content, which is the global limit, or 0.1 % sulphur inside sulphur emission control area (SECA), according to MARPOL Annex VI (MEPC.280(70)). Therefore, high sulphur fuel oil (HSFO) can still be used when a ship is operating with a scrubber as long as the atmospheric SO_X levels are met. However, the scrubbing process results in large volumes of acidic wash water, and while regulations are focused on SO_X removal from the exhausts, other pollutants e.g., PAHs and metals are also transferred to the wash water and discharged to the marine environment (Endres et al., 2018; Turner et al., 2017b). There are three types of wet scrubbers, open loop, closed loop, and hybrid scrubbers that can be operated in either open- or closed-loop mode (Turner et al., 2017b). Globally about 85 % of the scrubbers installed are open loop (IMO GISIS, 2021), but in the Baltic Sea, which is designated as a SECA, 83 % of the installations in 2018 where hybrid or closed loop systems (HELCOM, 2021a). Scrubbers operating in closed loop mode generate smaller volumes of wash water, but they can still be a substantial source of primarily metals, but also PAHs. Concerns have been raised that scrubbers, as a new direct source of PAHs and metals, is causing additional pressures to the already polluted Baltic Sea (Hassellöv et al., 2020; Thor et al., 2021; Ytreberg et al., 2019; Ytreberg et al., 2021b).

Since the activity of maritime shipping in the Baltic Sea has increased since 2012 (which is the year the results from Jalkanen et al. (2021) are based on), and the number of ships equipped with scrubbers has increased from 16 in 2012 to 178 in 2018 (HELCOM, 2021a; Jalkanen et al., 2021), an updated study of ship emissions to the Baltic Sea is called for. The overall aim of this study was to estimate the load of metals and PAHs from maritime shipping and leisure boating, relative other sources, to the Baltic Sea and specifically to: 1) identify the metals and PAHs with the highest load (tonnes/year) from ships and leisure boats to the Baltic Sea as waterborne pollution and 2) compare these loads to other sources (rivers, atmospheric deposition, and coastal point sources) in order to 3) suggest how maritime shipping and leisure boating can reduce their loads of metals and PAHs to the Baltic Sea.

2. Material and methods

The load of PAHs (16 US EPA priority PAHs) and metals (As, Cd, Cr, Cu, Pb, Hg, Ni, V and Zn) from atmospheric deposition, riverine inputs, point sources (coastal industries and wastewater treatment plants), maritime shipping and leisure boating to the Baltic Sea were calculated based on data from various sources (Table 1 and Fig. 1). In addition, calculations were made at subbasins level, i.e., Bothnian Bay, Bothnian Sea, Gulf of Finland, Gulf of Riga, Baltic Proper, Danish Straits and Kattegat. The most recent available data sources were used for the load compilation. However, that also implied a variability in time periods from when the data were collected, which also affect the quality of the load compilation. For maritime shipping, activity data and load estimations for 2018 were used, while the estimated annual average load from leisure boating were based on the Baltic Sea leisure boat fleet characterization and activity for the period 2010–2018. For maritime shipping, the analysis was delimited to direct discharges of bilge water, scrubber wash water, greywater, and sewage. Emissions/leakage from antifouling paint were also included, while discharges from ballast, cooling water, hull cleaning and tank cleaning were excluded due to data gaps or minimal contribution to the studied contaminants (Maljutenko et al., 2021). For leisure boats, only antifouling paint was included, while sewage, hull cleaning and wet exhaust were excluded due to lack of data.

For atmospheric deposition and point sources, annual loads were calculated based on data for the period 2016–2018. For riverine input, the average annual loads were determined based on the data for the period 2015–2017, since the reported loads for 2018 contained large data gaps. For arsenic (As) and vanadium (V), riverine input was only available from Sweden and therefore, a case study was designed to determine the loads of As and V from atmospheric deposition, riverine inputs, point sources (coastal industries and wastewater treatment plants), shipping and leisure boating to the Swedish Exclusive Economic Zone (EEZ). A detailed description about the data sources and methodology is given in Sections 2.1–2.3 and in Table 1.

2.1. Maritime shipping and leisure boating

The STEAM model, as described in Jalkanen et al. (2021), was used to calculate volumes of open and closed loop scrubber wash water, bilge water, grey water and sewage and mass of Cu and Zn from antifouling paints to the Baltic Sea, its subbasins and to the Swedish EEZ (Fig. 2)

Table 1
Data availability of metals and the 16 US EPA priority PAHs from different emissions sources. The PAHs were subdivided to low molecular weight (LMW) PAHs which represent compounds with 2–4 aromatic rings and high molecular weight (HMW) species which represent compounds with 5–6 rings.

Emission source		Data period	Metals	PAHs (LMW)	PAHs (HMW)	References
Riverine input		2015–17 (Baltic Sea) 2016–18 (Swe EEZ)	As[a], Cd, Cr, Cu, Hg, Ni, Pb, V[a], Zn	n/a	n/a	HELCOM (2021c) [m] SLU (2021) [m]
Atmospheric deposition[b]		2016–2018	As, Cd, Cr, Cu, Hg, Ni, Pb, V, Zn	Phenanthrene Anthracene Fluoranthene Pyrene	Benzo[*a*]anthracene Chrysene Benzo[*b*]fluoranthene Benzo[*k*]fluoranthene Benzo[*a*]pyrene Indeno[*1,2,3-cd*]pyrene Benzo[*ghi*]perylene Dibenz[*a,h*]anthracene	EBAS (2021) [c]
Ships	AF paint	2018	Cu, Zn			STEAM [m], this study
	Grey water	2018	As, Cd, Cr, Cu, Ni, Hg, Pb, V			STEAM [v], this study Jalkanen et al. (2021) [c]
	Sewage	2018	As, Cd, Cr, Cu, Ni, Hg, Pb, V			STEAM [v], this study Jalkanen et al. (2021) [c]
	Bilge water	2018	As, Cd, Cr, Cu, Ni, Hg, Pb, V	Naphthalene Acenaphthylene Acenaphthene Fluorene Phenanthrene Anthracene Fluoranthene Pyrene	Benzo[*a*]anthracene Chrysene Benzo[*b*]fluoranthene Benzo[*k*]fluoranthene Benzo[*a*]pyrene Indeno[*1,2,3-cd*]pyrene Benzo[*ghi*]perylene Dibenz[*a,h*]anthracene	STEAM [v], this study Jalkanen et al. (2021) [c]
	Scrubber water, open and closed loop	2018	As, Cd, Cr, Cu, Ni, Hg, Pb, V	Naphthalene Acenaphthylene Acenaphthene Fluorene Phenanthrene Anthracene Fluoranthene Pyrene	Benzo[*a*]anthracene Chrysene Benzo[*b*]fluoranthene Benzo[*k*]fluoranthene Benzo[*a*]pyrene Indeno[*1,2,3-cd*]pyrene Benzo[*ghi*]perylene Dibenz[*a,h*]anthracene	STEAM [v], this study Lunde Hermansson et al. (2021) [c]
Leisure boats AF paint		2010–2018	Cu, Zn	n/a	n/a	Johansson et al. (2020) [m]
Point sources		2016–2018	Cd, Cr, Cu, Hg, Ni, Pb, Zn	n/a	n/a	HELCOM (2021b) [m]

[a] Data only available from Sweden.
[b] Based on data from Swedish background sites [c], [v] and [m] indicates concentrations, volume and mass.

from the shipping sector. Emission and discharge factors, and model settings described in Jalkanen et al. (2021) were used, but with activity data for 2018 and with the exception of scrubber discharge rates (see Supporting Material). The loads of metals and PAHs were calculated by multiplying the total volumes of open- and closed loop scrubber wash water, bilge water, grey water, and sewage with the corresponding average concentrations of the contaminants present in the respective waste stream, i.e. using the same approach as Jalkanen et al. (2021). The average concentrations of PAHs and metals in the different waste streams were obtained from Jalkanen et al. (2021), except for open and closed loop scrubber water, which were obtained from Lunde Hermansson et al. (2021). For the scrubber waters, all reported samples (n = 89 open loop, and n = 25 closed loop) in Supporting Information of Lunde Hermansson et al. (2021) were used to calculate the average concentrations.

For leisure boats, only emissions of Cu and Zn from antifouling paints were included in the assessment. Leisure boats can also be a source of PAHs (Egardt et al., 2018) but was not included due to lack of emission factors. Inputs of Cu and Zn from antifouling paint on leisure boats were obtained from Johansson et al. (2020), where the total yearly emissions of Cu and Zn to the Baltic Sea are estimated, per boat category and per country (Sweden, Finland, Denmark, Germany and "other countries"). Thus, the data could only be used to determine total loads to the entire Baltic Sea area and not to its subbasins.

2.2. Riverine inputs

National data of riverine input of metals reported by the HELCOM contracting parties to HELCOM PLC Annual was collected for the period 2015–2017 (HELCOM, 2021c). The data portal contains metal loads per country and water basin. However, there were some data gaps. No data on metal loads from Danish rivers were available. Chromium (Cr) was not reported from Russia. Hg was not reported from the Neva River, which is the largest Russian river. For Cu and nickel (Ni), no data were available from Russian rivers in 2017. Since the Russian input of Cu and Ni corresponded to 11–38 % of the total riverine input to the Baltic Sea

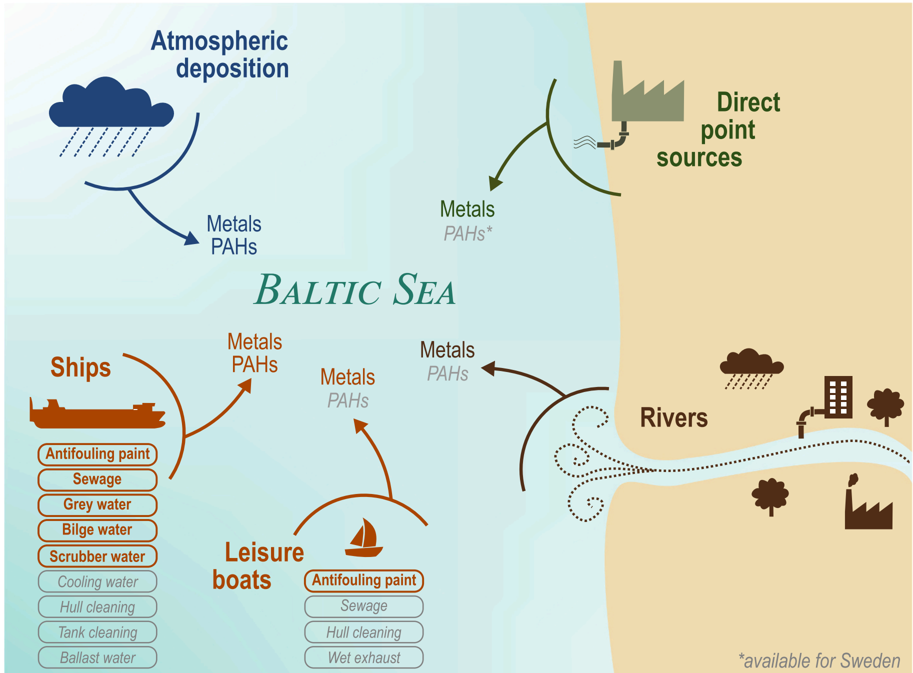


Fig. 1. Direct discharges and atmospheric deposition of metals and PAHs to the Baltic Sea. Grey text in italic indicates substances and sources where data is lacking and thus not included.

in previous years (2010–2016), the lack of data in 2017 made a large impact on the total average annual load estimation. Thus, it was decided to use the average yearly Russian load of Cu and Ni from 2010 to 2016 as an estimation of the 2017 load. For the Swedish case study, riverine loads of metals, reported to HELCOM PLC Annual (Baltic Sea) for 2016–2018 were downloaded from the SLU (2021) database. The PAHs are not included in the international reporting obligations to HELCOM PLC and there are no available Swedish data for the riverine load of PAHs.

2.3. *Atmospheric deposition*

Currently, only Cd, Cu, Hg, Pb and benzo[*a*]pyrene are included in EMEP-models to estimate deposition loads on the Baltic Sea region (EMEP, 2021; HELCOM, 2021b). Therefore, a simplified extrapolation approach was used, where annual atmospheric loadings of metals and PAHs to the surface water of the Baltic Sea were calculated by extrapolating the deposition fluxes of 9 metals and 12 PAHs at background stations to the surface area of the Baltic Sea subbasins (Fig. 2). This is a similar approach as e.g. Leister and Baker (1994) used to calculate annual loadings of PAHs to Chesapeake Bay, and the results should be treated as indicative. The data is accessible through the Swedish database for Air quality (SMHI, 2021) and the EMEP database, EBAS (2021).

The monthly atmospheric deposition of PAHs (dry and wet deposition on 1 m^2 large deposition surface) was calculated from the daily average value (μg/m^2/day) for each sampling period (month) and multiplied with the number of days for each period. The monthly deposition fluxes of PAHs were then summed up to a yearly deposition.

The monthly atmospheric deposition of metals was calculated from the concentration in precipitation (ng/ml), the precipitation rate (mm) and the funnel area of the bulk sampler (m^2) during each sampling period (month), as:

$$\text{Precipitation rate}\,(\text{mm}) = \text{Precipitation volume}\left(\text{m}^3\right)/\text{sampler area}\left(\text{m}^2\right)/1000 \quad (1)$$

$$\text{Deposition}\left(\mu\text{g}/\text{m}^2\right) = \text{concentration}\,(\text{ng}/\text{ml})^*\text{precipitation rate}\,(\text{mm}) \quad (2)$$

Monthly deposition fluxes of metals were summed up to a yearly deposition flux.

To estimate the atmospheric deposition of PAHs, mercury, and other metals to the specific subbasins of the Baltic Sea and to the Swedish EEZ, annual deposition fluxes (2016–2018) from the monitoring sites were multiplied with the area of each subbasin. For each subbasin the deposition flux from one or two monitoring site(s), located closest to the basin was assigned, see Fig. 2. An alternative option would be to use chemical transport modelling of ship emissions (Karl et al., 2019), but as described previously these do not provide emissions or deposition fluxes for PAHs or metals from the international shipping sector. In addition, only Cd, Cu, Hg, Pb and benzo[*a*]pyrene are included in EMEP-models (EMEP, 2021; HELCOM, 2021b).

2.4. *Point sources*

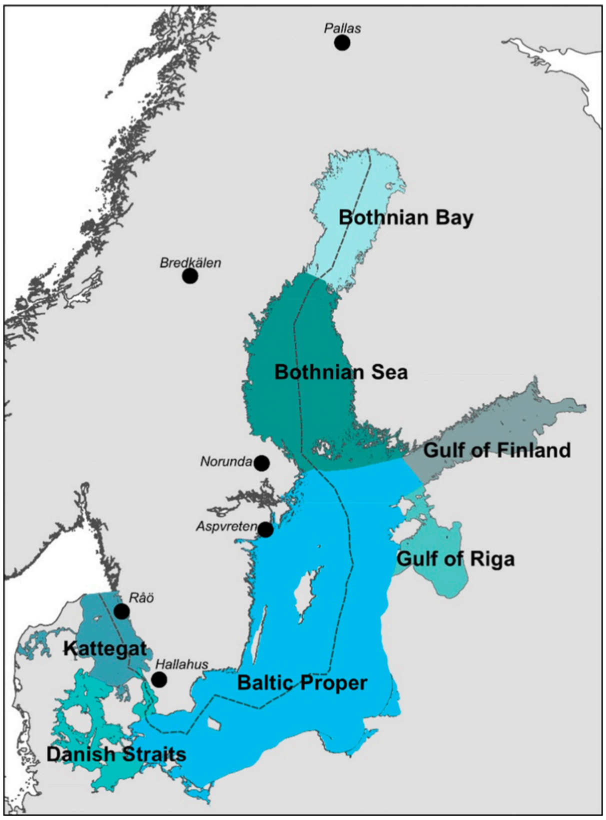
The loads of metals from point sources with outlets directly to the coast were compiled from HELCOM (2021b). The data includes loads from larger coastal municipal wastewater treatment plants (WWTPs) and industrial point sources.

Data on PAH loads to the Baltic Sea from point sources were only available from Sweden and comprised only a few facilities (industries and wastewater treatment plants) in the Kattegat and Oresund region (Swedish EPA, 2021). Only fluoranthene and the sum of so called PAH4 (benzo(*a*)pyrene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, indeno (*1,2,3-cd*)pyrene) are reported, which makes the comparison with the other emissions sources difficult. In addition, the loads are insignificant (g/year) as compared to atmospheric deposition and direct loads from maritime shipping. Therefore, PAH loads from point sources were excluded in this study.

3. Results and discussion

3.1. *Loads of metals to the Baltic Sea and its subbasins*

The single largest estimated annual metal load to the Baltic Sea, from all sources, was Zn (3932 t), which exceeds the sum of all other metals



HELCOM PLC Subbasins	PAH	Metals	Mercury
Bothnian Bay	Pallas	Pallas	Pallas
Bothnian Sea	Aspvreten/Norunda	Bredkälen	Bredkälen
Gulf of Finland	Aspvreten/Norunda	Aspvreten/Norunda	Average Bredkälen/Råö
Baltic Proper	Aspvreten/Norunda	Aspvreten/Norunda	Average Bredkälen/Råö
Gulf of Riga	Aspvreten/Norunda	Aspvreten/Norunda	Average Bredkälen/Råö
Kattegat	Råö	Råö	Råö
Danish Straits	Hallahus	Hallahus	Hallahus

Fig. 2. HELCOM 7 subbasins (Bothnian Bay, Bothnian Sea, Gulf of Finland, Gulf of Riga, Baltic Proper, Danish Straits and Kattegat) and the Swedish Exclusive Economic Zone (—). Monitoring background stations used for the estimates of deposition fluxes to the specific subbasins. The measurement at Aspvreten ended 2017 and from 2018 the sampling is performed at Norunda station.

load (≈2800 t, Table 2). The second largest load was Cu (1560 t) followed by Ni (675 t), Pb (273 t) and Cr (183 t). The load of vanadium (V) (66 t) was approximately twice as large as the loads of As and Cd, (26 t) respectively. The load of Hg was <5 t.

The total annual load of metals from maritime shipping and leisure boats to the Baltic Sea is dominated by Cu and Zn (Fig. 3). Cu showed the largest annual load (575 t) followed by Zn (166 t), V (35 t) and Ni (10 t). The other metals (Cr, Pb, As, Cd and Hg) contributed to considerably lower loads (in total 7 t). Due to the high shipping activity and the size of the water basin, inputs of metals from maritime shipping are concentrated to the Baltic Proper, e.g. 41 % of the total input of Cu from shipping occur in this subbasin (Table 2). Contrary, the largest metal load from riverine inputs is to the subbasin Gulf of Finland, except for Cr which was not reported from Russia. The loads of metals from points sources were, in comparison to the riverine input, low and accounted for between 1 and 5 % of the total riverine input to the Baltic Sea.

Regarding atmospheric deposition of metals to the Baltic Sea, the extrapolation of data from only a handful of monitoring stations to the large areas of the subbasins, and the location of the monitoring sites, all affect the accuracy of the results. The deposition of metals presented in Table 2 should therefore be considered as indicative. A comparison can however be made to the MSC-E modelled deposition. MSC-E applies a more sophisticated modelling approach, which includes atmospheric transportation/transformation and validates the modelled quantities against large number of measurement data (Gauss et al., 2020). Despite the different approaches, the MSC-E model showed deposition on the Baltic Sea of Cd and Hg to be on average 4.1 and 2.8 t/year, respectively, during the period 2016–2018 (HELCOM, 2021b). This is in the same order of magnitude as the deposition load estimated in this study (3.7 and 1.8 t/year respectively for Cd and Hg). For Cu, the MSC-E model also showed good agreement (average 87 t/year during the period 2016–2018) (EMEP, 2021) with what was estimated in this study (116 t/year). For Pb, a larger discrepancy was observed, where MSC-E modelled deposition was higher (143 t/year) (HELCOM, 2021b) than this study (58 t/year).

3.2. Load of PAHs to the Baltic Sea and its subbasins

The total estimated annual load of PAHs from maritime shipping and atmospheric deposition was close to 20 t per year (Table 3). The highest total loads were from fluoranthene and phenanthrene (3.7 and 3.3 t, respectively), followed by pyrene (2.6 t), benzo[*b*]fluoranthene (2 t) and chrysene (1.9 t). The loads from benzo[*a*]pyrene, benzo[*ghi*]perylene and indeno[*1*,*2*,*3*-*cd*]pyrene were in the range 1–1.4 t. The annual loads of naphthalene, benzo[*k*]fluoranthene and benzo[*a*]anthracene were in the range 0.45–0.9 t, while anthracene, dibenz[*a*,*h*]anthracene, fluorene, acenaphthene and acenaphthylene all were below 0.2 t each.

The direct annual load of PAHs from maritime shipping to the Baltic Sea is shown in Fig. 4. The dominating PAHs were all low molecular weight (LMW) compounds (consisting of 2–3 aromatic rings), with the highest load from naphthalene (560 kg) and phenanthrene (290 kg). Besides shipping, atmospheric deposition was the only source of PAHs that could be assessed in the study. As for the metals, the highest load of PAHs from both deposition and maritime shipping occurred in the Baltic Proper. In comparison with the other subbasins, Bothnian Bay and Bothnian Sea received low loads of all investigated PAHs from maritime shipping. This can be explained by the low usage of open and closed loop scrubbers in this area which are the dominating source of PAHs from maritime shipping (see section 0). It should also be emphasized that atmospheric deposition in this study was derived based on monitored deposition of PAHs at six land-based background stations. Thus, exhaust emissions of PAHs to the atmosphere from shipping and the corresponding deposition on the Baltic surface may not be fully accounted for.

For the PAHs, benzo[*a*]pyrene is the only compound included in the EMEP MSC-E model, and the deposition to the Baltic Sea has been rather steady since the mid-1990s with average annual deposition of 2.7 t/year (HELCOM, 2021b). This is about three-times more than the deposition estimated in this study (1 t/year) (Table 3).

3.3. Comparisons of loads

3.3.1. Metals

The metals with the highest load from maritime shipping and leisure boating are Cu, Zn and V (Fig. 3). Therefore, these metals were further selected to assess their relative contribution between the different sources. The total annual load of Cu to the Baltic Sea was 1560 t, of which riverine input contributed to 54 % of the total input (850 t) while maritime shipping and leisure boating together accounted for 37 % of the load (575 t) (Fig. 5). The origin (natural or anthropogenic) of the riverine input of Cu to the Baltic Sea is not known. However, diffuse emissions of Cu to Swedish catchment areas (inland waters) from various sources has been compiled by Ejhed et al. (2010). The result (based on data from 1985 to 2004) showed leaching from forest land (52 t/year), stormwater (38 t/year), leaching from agriculture (30 t/

Table 2
Inputs of metals, tonnes/year, from shipping and leisure boats, rivers, point sources and deposition to the Baltic Sea basins. Data presented as annual average input, in tonnes per year, ±1 SD for the period 2016–2018 (deposition and point sources) and 2015–2017 (rivers). For shipping, only direct discharges to the sea are considered an no uncertainties are presented since the loads are from 2018 only.

Source	Subbasin	Annual input (tonnes/year)								
		As	Cd	Cr	Cu	Pb	Hg	Ni	V	Zn
Ships	Bothnian Bay	0.001	<0.001	0.01	12	0.001	<0.001	0.02	0.05	2.2
	Bothnian Sea	0.09	0.01	0.2	27	0.1	0.001	0.6	2.1	6.5
	Baltic Proper	0.9	0.1	1.9	213	1.1	0.01	6.0	21	52
	Gulf of Finland	0.2	0.02	0.4	110	0.2	0.002	1.1	3.9	23
	Gulf of Riga	0.005	<0.001	0.01	10	0.006	<0.001	0.03	0.1	1.9
	Danish Straits	0.2	0.02	0.4	80	0.2	0.002	1.3	4.4	17
	Kattegat	0.1	0.02	0.3	66	0.2	0.002	0.9	3.2	14
	Total	1.4	0.16	3.2	518	1.9	0.02	10	35	117
Leisure boats	Total	n/a	n/a	n/a	57	n/a	n/a	n/a	n/a	49
Riverine input	Bothnian Bay	n/a	1.5 ± 0.6	61 ± 11	150 ± 20	23 ± 3.9	0.3 ± 0.1	225 ± 50	n/a	550 ± 150
	Bothnian Sea	n/a	1.2 ± 0.3	41 ± 22	100 ± 30	21 ± 8.9	0.2 ± 0.04	100 ± 20	n/a	360 ± 140
	Baltic Proper	n/a	2.8 ± 1.6	33 ± 3.2	120 ± 20	41 ± 5.5	1.0 ± 1.1	100 ± 14	n/a	280 ± 150
	Gulf of Finland	n/a	15.3 ± 4.4	13 ± 6.3	390 ± 40	100 ± 40	0.7 ± 1.0	168 ± 75	n/a	1700 ± 1100
	Gulf of Riga	n/a	0.4 ± 0.2	7.9 ± 2.0	60 ± 8.7	20 ± 30	0.7 ± 1.2	11 ± 12	n/a	160 ± 70
	Danish Straits	n/a	0.1 ± 0.01	1.0 ± 0.5	5.4 ± 0.9	0.7 ± 0.2	0.01 ± 0.0	4.3 ± 1.1	n/a	15 ± 2.8
	Kattegat	n/a	0.3 ± 0.1	6.8 ± 1.3	30 ± 5.5	7.7 ± 1.1	0.1 ± 0.02	16 ± 3.3	n/a	80 ± 15
	Total	n/a	22 ± 3.7	160 ± 50	850 ± 80	210 ± 30	2.9 ± 0.1	625 ± 130	n/a	3100 ± 900
Point sources	Total	n/a	n/a	4.5	19	2.6	0.11	15	n/a	156
Atmospheric deposition	Bothnian Bay	0.8 ± 0.3	0.2 ± 0.03	0.9 ± 0.3	8.0 ± 0.9	4.1 ± 0.8	0.1 ± 0,04	6.1 ± 2.5	1.5 ± 0.3	22 ± 2.9
	Bothnian Sea	1.8 ± 0.2	0.5 ± 0.3	1.7 ± 0.7	12 ± 4.9	4.8 ± 2.6	0.3 ± 0.1	5.0 ± 3.1	1.6 ± 0.7	71 ± 11
	Baltic Proper	16 ± 10	1.7 ± 0.4	8.9 ± 2.9	57 ± 21	31 ± 7.7	0.9 ± 0.2	9.3 ± 2.8	19 ± 6.9	254 ± 81
	Gulf of Finland	2.3 ± 1.4	0.2 ± 0.05	1.3 ± 0.4	8.2 ± 3.0	4.5 ± 1.1	0.1 ± 0.03	1.3 ± 0.4	2.7 ± 1.0	36 ± 11
	Gulf of Riga	1.4 ± 0.9	0.2 ± 0.03	0.8 ± 0.3	5 ± 1.9	2.8 ± 0.7	0.1 ± 0.02	0.8 ± 0.3	1.7 ± 0.6	23 ± 7.2
	Danish Straits	1.3 ± 0.2	0.4 ± 0.2	0.8 ± 0.2	14 ± 9	5.7 ± 0.4	0.1 ± 0.01	1.5 ± 0.5	2.3 ± 0.5	51 ± 17
	Kattegat	1.4 ± 0.3	0.5 ± 0.2	0.9 ± 0.1	11 ± 0.8	5.0 ± 0.2	0.1 ± 0.01	1.4 ± 0.2	2.1 ± 0.4	52 ± 13
	Total	25 ± 12	3.7 ± 0.3	15 ± 3	116 ± 33	58 ± 9.6	1.8 ± 0.4	25 ± 2.2	31 ± 8.4	510 ± 100
Total, all sources	Total	26	26	183	1560	273	5	675	66	3932

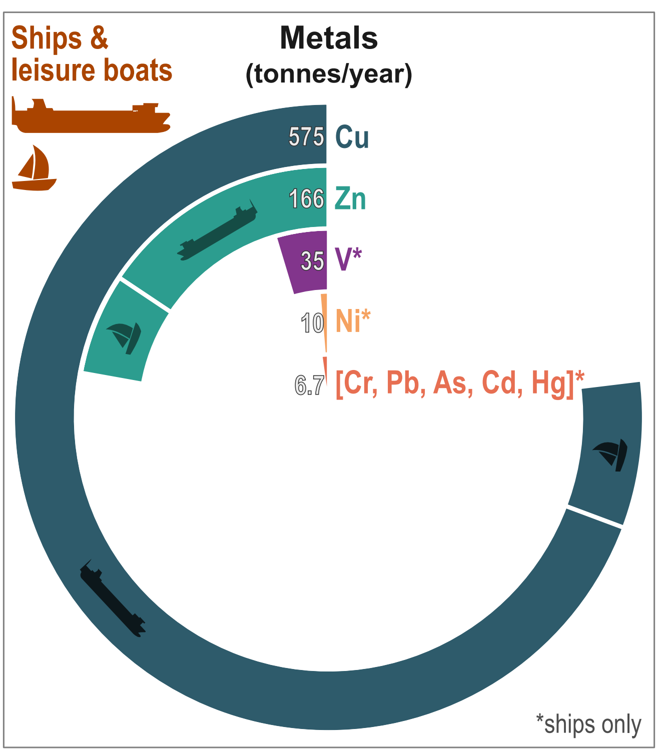


Fig. 3. Yearly Inputs of metals to the Baltic Sea from maritime shipping (in 2018) and from leisure boats (average yearly load based on fleet characteristics 2010–2018).

year) and remaining land areas (21 t/year) to be the largest diffuse sources of Cu to Swedish inland waters. The extent of which the diffusive Cu load (in total 143 t) to Swedish inland waters is being transported to the Baltic Sea is unknown. For comparison, the load of Cu to Swedish EEZ from ships coated with antifouling was determined to be 119 t annually (Supporting Material Table S1).

The load of Cu to the Baltic Sea from maritime shipping and leisure boating is almost five times as large as the estimated indicative load from atmospheric deposition (116 t). The bulk (98 %) of the Cu load from maritime shipping and leisure boating is from antifouling paints, where maritime shipping and leisure boating annually account for 509 and 57 t, respectively. Although the share from leisure boats is almost one order of magnitude lower as compared to maritime shipping, the emission from leisure boats is concentrated during a five-months summer season and occurs primarily in sensitive coastal areas.

Discharge water from scrubbers operating in open loop mode is the second largest source of Cu from shipping and constitutes about 7 t annually. This is about one third of the contribution from all direct point sources to the Baltic Sea (19 t annually). The load of Cu from grey water, sewage and bilge water is low and comprises to <3 t annually in total. The load of metals and PAHs from shipping is based on shipping activity during 2018. Based on this data set, only 178 vessels operated in the Baltic Sea with scrubbers and their discharge of open loop scrubber water to the total Baltic Sea was in this study estimated to 0.19 billion m^3/year (Supporting material Table S2). However, statistics from IMO GISIS (2021) have shown a rapid global increase in scrubber installations from about 260 vessels in 2018 to almost 3500 vessels in operation with a scrubber in 2020 (Fig. 6A). A similar increase has been seen in the Baltic Sea, where the number of vessels equipped with a scrubber has increased from 178 vessels in 2018 to 462 vessels in 2020 (Fig. 6B) (HELCOM, 2021a). Therefore, the discharge of scrubber wash water both on a global scale and to the Baltic Sea have increased the last years. For example, in a recent study by Osipova et al. (2021), the annual discharge volumes of open loop discharge water to the Baltic Sea was estimated to 0.3 billion m^3/year. The result from this study was based on 2019 activity data and with the assumptions that ships that had or were expected to have scrubbers installed by the end of 2020 was operating in open loop mode with a flow rate of 45 m^3/MWh. The applied flow rate of

Table 3
Input of naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[*1,2,3-cd*]pyrene (InP), benzo[*ghi*]perylene (BghiP), dibenz[*a,h*]anthracene (DahA) from shipping and atmospheric deposition to the Baltic Sea basins. Data presented as annual average input, in kg per year, ±1 SD for the period 2016–2018. For shipping, only direct discharges to the sea are considered an no uncertainties are presented since the loads are from 2018 only.

Source	Subbasin	Annual input (kg/year)															
		Nap	Acy	Ace	Flo	Phe	Ant	Fla	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	InP
Ships	Bothnian Bay	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
	Bothnian Sea	36	2	2	6	18	1	2	4	1	2	1	<1	1	<1	<1	1
	Baltic Proper	340	15	22	55	180	9	19	37	14	22	5	2	6	3	3	9
	Gulf of Finland	65	3	4	10	34	2	4	7	3	4	1	<1	1	1	1	2
	Gulf of Riga	2	<1	<1	<1	1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
	Danish Straits	62	3	4	10	31	2	3	6	2	4	1	<1	1	1	1	2
	Kattegat	55	2	4	9	28	1	3	6	2	3	1	<1	1	<1	<1	1
	Total	560	24	37	90	290	15	31	60	23	36	8	3	10	5	5	14
Atm. Dep.	Bothnian Bay	n/a	n/a	n/a	n/a	220 ± 30	12 ± 2	120 ± 10	80 ± 2	50 ± 50	70 ± 60	50 ± 3	20 ± 1	30 ± 10	4 ± 2	30 ± 5	30 ± 4
	Bothnian Sea	n/a	n/a	n/a	n/a	570 ± 50	30 ± 10	750 ± 120	510 ± 150	170 ± 100	370 ± 140	400 ± 200	160 ± 80	210 ± 90	30 ± 10	260 ± 140	290 ± 160
	Gulf of Finland	n/a	n/a	n/a	n/a	220 ± 20	12 ± 4	290 ± 50	200 ± 60	70 ± 40	140 ± 50	160 ± 80	60 ± 30	80 ± 3+	10 ± 4	100 ± 50	110 ± 60
	Baltic Proper	n/a	n/a	n/a	n/a	1500 ± 130	80 ± 30	2000 ± 300	1400 ± 400	460 ± 250	1000 ± 400	1100 ± 600	420 ± 220	540 ± 200	70 ± 30	700 ± 400	750 ± 430
	Gulf of Riga	n/a	n/a	n/a	n/a	140 ± 10	8 ± 2	180 ± 30	120 ± 30	40 ± 20	90 ± 30	100 ± 50	40 ± 6	50 ± 20	6 ± 2	60 ± 30	70 ± 40
	Kattegat	n/a	n/a	n/a	n/a	140 ± 40	5 ± 1	150 ± 40	100 ± 20	50 ± 20	90 ± 20	60 ± 20	30 ± 10	40 ± 10	7 ± 3	40 ± 20	40 ± 10
	Danish Straits	n/a	n/a	n/a	n/a	1200 ± 30	8 ± 2	250 ± 6+	160 ± 40	50 ± 20	130 ± 50	110 ± 40	40 ± 10	60 ± 20	10 ± 4	70 ± 20	70 ± 20
	Total	n/a	n/a	n/a	n/a	3000 ± 200	160 ± 40	3700 ± 500	2500 ± 600	900 ± 350	1900 ± 500	2000 ± 900	760 ± 360	1000 ± 400	130 ± 40	1300 ± 600	1400 ± 700
Total, all sources	Total	560	24	37	90	3290	175	3731	2560	923	1936	2008	763	1010	135	1305	1414

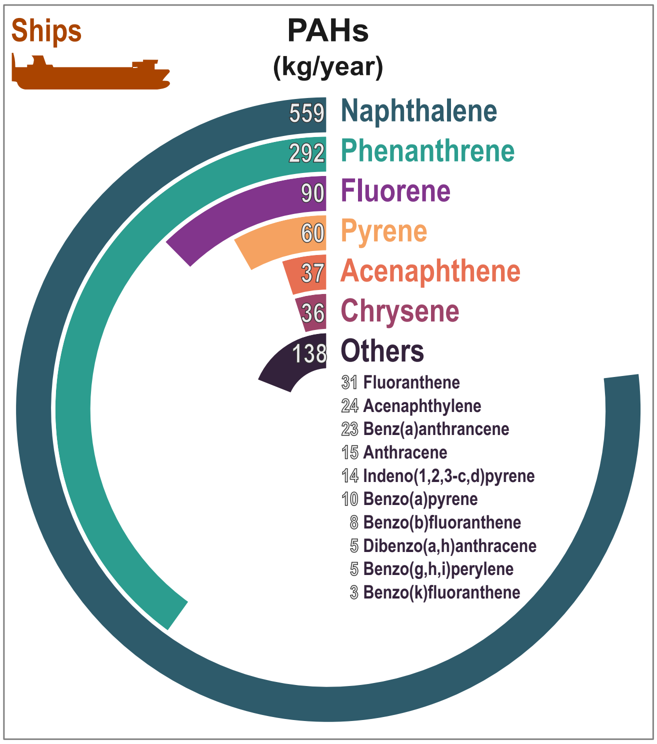


Fig. 4. Direct inputs of PAHs (kg/year) to the Baltic Sea from maritime shipping in 2018.

45 m^3/MWh, based on an old IMO document dated to 2008 (IMO, 2008), may however be out of date and when a flow rate of 90 m^3/MWh was applied instead, the total volume would be 0.6 billion m^3/year. This is about three times higher than the estimate in this study (0.19 billion m^3/year), indicating that the load of all pollutants from scrubber operated in open loop mode, to the Baltic Sea, could be significantly (threefold) higher.

For the load of Zn, riverine input is the largest route of entry to the Baltic Sea and constitutes about 80 % of the total loads to the sea (Fig. 5). The second largest source is atmospheric deposition (13 %), while the contribution from direct point sources and ships and leisure boats was similar, 4 % and 3.6 %, respectively. Similar to Cu, the largest source of Zn from maritime shipping and leisure boating was antifouling paints, followed by a lower share from scrubbers.

For vanadium, the assessment could only be performed for the Swedish EEZ due to lack of information of riverine loads from other HELCOM contracting parties. The Swedish EEZ is receiving more than one third of the total discharged open loop water to the Baltic Sea (Supporting material Table S2). The results showed maritime shipping, and the use of open loop scrubbers account for 18 % of the total input to Swedish EEZ (Fig. 5 and Supporting Material Table S3). This can partly be explained by the relatively high concentrations of vanadium in conventional HSFO as compared to marine gas oil (MGO) which has a direct impact on emission factors of V to air which is on average over 100 times higher from HSFO as compare to MGO (Lunde Hermansson et al., 2021). Today, HSFO can only be used by ships equipped with scrubbers and during operation in open loop mode they will thus continue to significantly contribute to the load of V in the Baltic Sea. In addition, if the assessment was performed based on the discharge volumes from Osipova et al. (2021) and the higher discharge rate (90 m^3/MWh), the load of V would be four times higher and thus constitute about 40 % of the total input to Swedish EEZ.

3.3.2. Polycyclic aromatic hydrocarbons (PAHs)

The share of PAHs from maritime shipping as compared to the total load from all sources, ranged from 0.4 to 8.9 %, and the input was almost exclusively (≥98 %) derived from scrubbers operating in open loop mode (Fig. 7). For the HELCOM core indicators anthracene, benzo[*a*]pyrene, and fluoranthene the share from maritime shipping was 8.5 %, 1.0 % and 0.8 %, respectively. However, as stated previously, the estimated atmospheric deposition of PAHs on the Baltic Sea holds large uncertainties. Emission factors of the 16 US EPA priority PAHs to air during combustion of HSFO (with and without scrubber) and low sulphur (<0.5 %) MGO have been compiled by Lunde Hermansson et al. (2021). The results showed the combustion of HSFO to have a significantly higher air emission factors of all PAHs as compared to MGO, e.g. the emission factor for anthracene was 8.2 mg/MWh and 2.2 mg/MWh when operating with HSFO (without scrubber) and MGO, respectively. If the ship is operating with a scrubber (and HSFO), the emission factors to water and air was estimated to 1.8 mg/MWh and 6.4 mg/MWh, respectively. However, since no atmospheric chemical transport model was used in this work to indicate deposition of PAHs other than benzo[*a*]pyrene in the Baltic region, it is beyond the scope of this study to account for ship emissions of PAHs other than the direct discharge via scrubber water and bilge water.

3.4. Possibilities to reduce the inputs of selected metals and PAHs

The results from this study show maritime shipping to be a significant source of certain metals and PAHs to the marine environment, where scrubbers operating in open loop mode, and Cu-based antifouling paints are the main waste streams. Alternatives to Cu-based antifouling paints exist, including biocide-free silicone foul-release coatings and inert coatings combined with in-water hull cleanings. A recent review of studies on the ecotoxicity of biocide-free silicone coatings has shown biocide-free silicone foul-release coatings to be substantially less toxic to marine organisms compared to conventional Cu-containing coatings (Lagerström et al., 2021b). Additionally, a study included one such coating found it to be more effective than Cu-based coatings in preventing biofouling when coated panels were exposed statically for 12 months in Gothenburg harbour (Kattegat) (Oliveira and Granhag, 2020). However, neither foul-release coatings nor Cu-based antifouling coatings are suitable for ice-going vessels since the ice will damage and/or remove the paint. Therefore, inert coatings are more appropriate to be used on ships sailing in the northern part of the Baltic Sea during wintertime. This is also accounted for in the STEAM model (and in this study) where it is assumed that only 20 % and 50 % of the ships that exclusively operate in the Gulf of Bothnia and Baltic Proper, respectively, are coated with Cu-based antifouling paints (Jalkanen et al., 2021). Cu is currently evaluated for inclusion as a HELCOM core indicator, with a proposed threshold value of 30 mg/kg dry weight normalized to 5 % total organic carbon (TOC). The proposed threshold value has been compared with surface sediment concentrations in the Baltic Sea in a recent report by Lagerström et al. (2021a). The result showed 76 % of the data points to exceed the proposed threshold value. Thus, to improve the status of Baltic Sea it is important to actively reduce the input of Cu where the antifouling paints, have been identified to be the largest anthropogenic source.

Besides antifouling paint, scrubbers operating in open loop mode are the largest contributing waste stream from shipping with respect to metals and PAHs from maritime shipping. This is noteworthy because ships equipped with scrubbers only constitutes a small fraction (178 ships in 2018) of the fleet that operates in the Baltic Sea Area (8900 ships carrying an IMO number in 2018) (HELCOM, 2021a). Most of the ships equipped with a scrubber operated with a hybrid system ($n = 140$), 30 were open loop systems while 8 installations where of closed loop type. Thus, only 17 % of the ships have no possibility to operate in closed loop mode. Hence, if open-loop mode operation would be restricted in the Baltic Sea, the vast majority of the vessels could still be able to operate in closed loop mode but with less direct marine environmental impact. However, the scrubber effluent storage tank capacity might then become

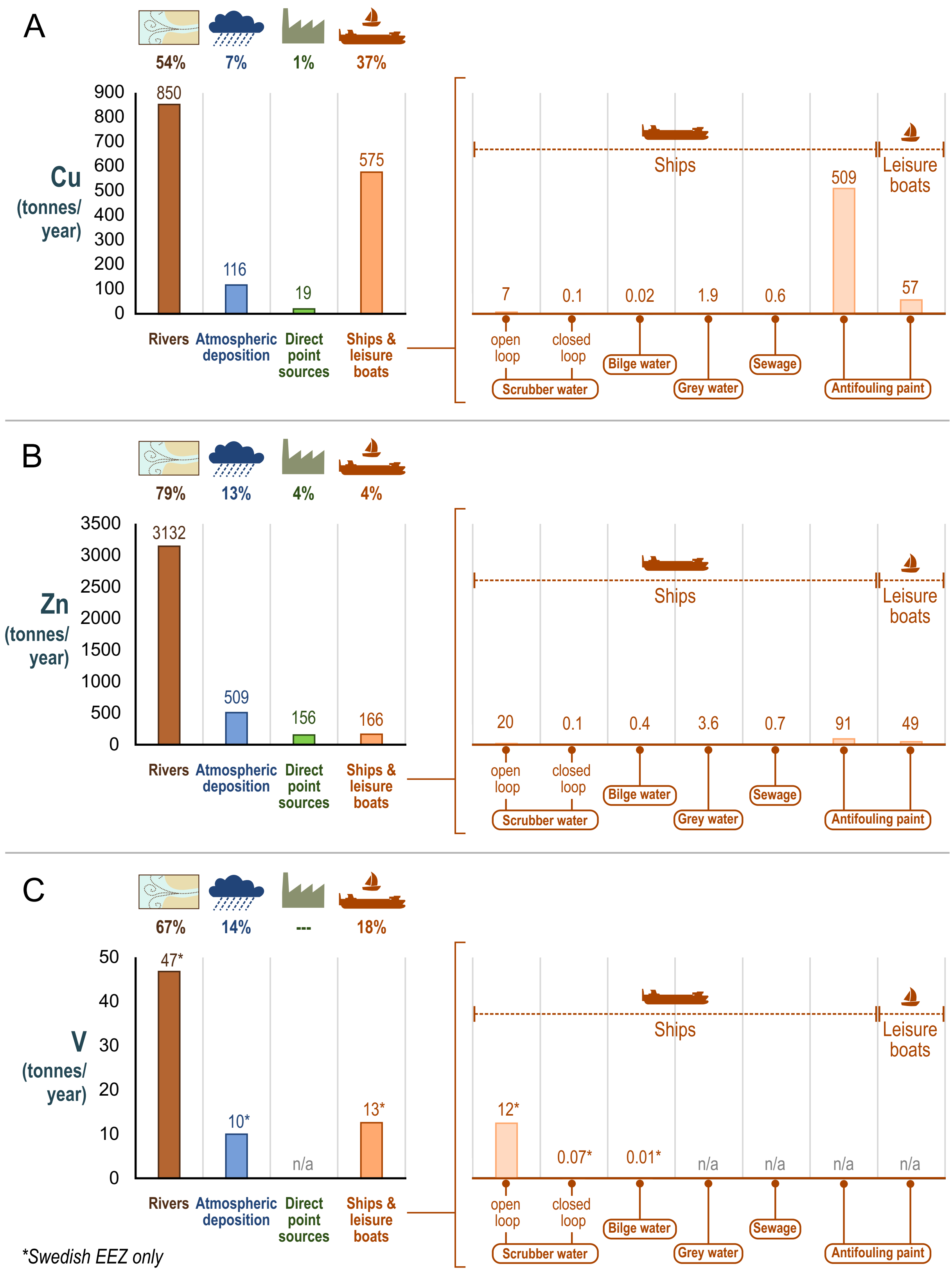


Fig. 5. Comparison of loads (tonnes/year) of Cu (A), Zn (B) and V (C) from rivers, atmospheric deposition, direct point sources, and direct discharges from ships and leisure boats to the Baltic Sea. For V, the assessment was performed in the Swedish Exclusive Economic Zone (EEZ) only, since data was missing for the other HELCOM contracting parties. The loads from atmospheric deposition should be treated as indicative due to large uncertainties.

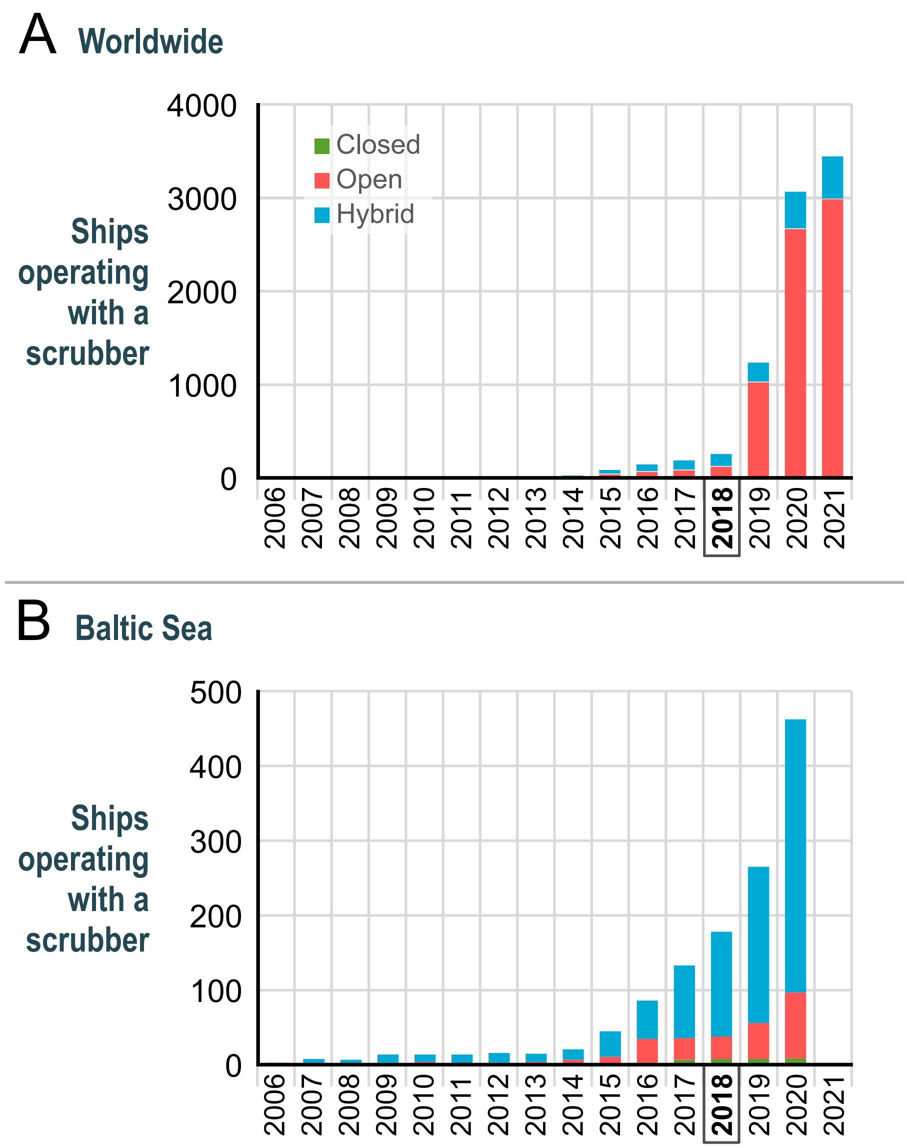


Fig. 6. Ships operating with a scrubber worldwide (A) and in the Baltic Sea (B), per scrubber type. Ships with scrubbers in operation in 2018 highlighted in bold in the figure.

an issue.

Nevertheless, the installation of scrubbers allows for a continuous use of HSFO which in turn is associated with higher emissions of both metals and PAHs as compared to distillate fuels (Lunde Hermansson et al., 2021). Although the atmospheric emissions of SO_X meet the IMO's limits with the use of scrubbers, other hazardous substances are being enriched and discharged to the marine environment, especially during the, more commonly used, open loop mode operation. For example, the total load of Cu, V and anthracene was calculated based on the emission factors in Lunde Hermansson et al. (2021) and the discharge rate of 90 m^3/MWh (open loop) and 0.45 (closed loop) m^3/MWh. The result showed the loads to be 13 (Cu), 4 (V) and 10 (anthracene) times higher if a ship is operating in open loop mode as compared to closed loop mode. This is similar to what Ytreberg et al. (2021a) concluded, i.e. that the marine ecotoxicological impact of open loop discharge water is six times more severe compared to closed loop systems when determined through Life Cycle Impact Assessment (LCIA). It must also be emphasized that emissions of PAHs and most metals to the atmosphere are significantly higher when a ship is operating with HSFO as compared to MGO, regardless if the vessel operating with HSFO is equipped with a closed loop scrubber system or not (Lunde Hermansson et al., 2021). Thus, to minimize the environmental load of PAHs and metals, the best practice would be not to use HSFO at all. Even though the loads of PAHs and metals from atmospheric deposition hold large uncertainties, the results from this study suggest open loop scrubber wash water to be a major source of PAHs to the marine environment, e.g. 8.5 % of the total load of anthracene in 2018 is caused by wash water from scrubbers operated in open loop mode in the Baltic Sea, despite the relatively low number of ships being equipped with scrubbers. As previously emphasized, the discharge of scrubber wash water has increased since 2018 and is predicted to be threefold as high when ships that had, or were expected to have, scrubbers installed by the end of 2020 are using open loop scrubbers with a flow rate of 90 m^3/MWh. Concerns have also been raised regarding the challenge to predict environmental impacts due to the chemical cocktail of acidifying, eutrophying and hazardous substances present in scrubber discharge water (Hassellöv et al., 2020). For example, in a study on copepods, none of the individual PAHs or heavy metals analysed in the scrubber wash water occurred in concentrations which could explain the toxic responses (Thor et al., 2021).

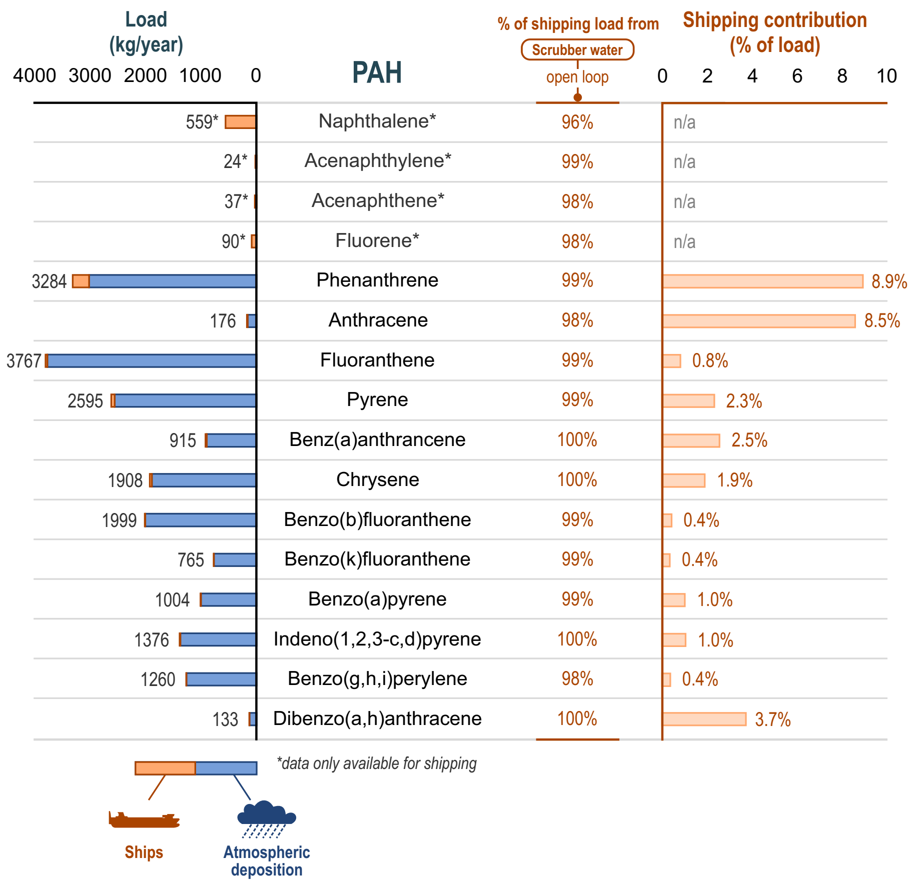


Fig. 7. Comparison of loads (kg/year) of PAHs from direct discharges from ships and atmospheric deposition to the Baltic Sea. The loads from atmospheric deposition should be treated as indicative due to large uncertainties.

4. Conclusions

This study shows maritime shipping and leisure boating to be two significant sources of PAHs and metals to the Baltic Sea. Together, the two sectors account for more than one third of the total Cu load to the Baltic Sea, mainly due to the use of Cu-based antifouling paints. Biocide-free alternatives do exist, including foul-release coatings, which have shown to be as effective as Cu-based coatings to prevent biofouling. However, as long as Cu-based coatings are allowed to be used in the Baltic Sea, biocide-free strategies have difficulty in gaining market shares. The use of open loop scrubbers on ships was identified as a major source of vanadium and anthracene to the Baltic Sea. Switching to closed loop mode, which 83 % of the Baltic Sea scrubber fleet has the possibly to do, could reduce the loads of most PAHs and metals with up to 90 %. In future HELCOM pollution load compilations, maritime shipping and leisure boating should be included as they here have been identified as the largest individual anthropogenic sources of e.g. Cu to the Baltic Sea.

CRediT authorship contribution statement

Erik Ytreberg: Conceptualization, Methodology, Writing – original draft, Formal analysis, Investigation, Funding acquisition. **Katarina Hansson:** Writing – review & editing, Methodology, Formal analysis, Investigation. **Anna Lunde Hermansson:** Writing – review & editing, Visualization, Formal analysis. **Rasmus Parsmo:** Writing – review & editing, Formal analysis. **Maria Lagerström:** Writing – review & editing, Visualization. **Jukka-Pekka Jalkanen:** Writing – review & editing, Methodology, Investigation. **Ida-Maja Hassellöv:** Writing – review & editing, Conceptualization, Methodology, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2022.113904.

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