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Sampling strategy, quantification, characterization and hazard potential assessment of greywater from ships in the Baltic Sea

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ABSTRACT

Ship-generated greywater contains a variety of pollutants which, through various pathways, usually are discharged into the sea. To understand the seasonal variation in greywater volumes, the contaminant concentrations in, and the potential hazard of, ship-generated greywater streams, a four-phase strategy for sampling, characterization and hazard assessment of greywater was developed and implemented. Eight greywater streams, sampled from five ships, were characterized for selected pollutants. The metals Zn, Cu, Mn and the metalloid, As, collectively contributed 98 % to the Hazard Index. Laundry greywater had the highest average concentration of phosphorus (42 mg/l) while galley greywater had the highest average concentration of nitrogen (30 mg/l). The geometric means of COD-Cr, BOD_5, TSS and P exceeded the IMO resolution MEPC 227(64) guideline values for sewage effluent from Advanced Wastewater Treatment Plants. The results establish the basis for and contribute to discussions on, the optimization of ship-generated greywater management and the establishment of potential regulatory strategies in the Baltic Region.

1. Introduction

Ships cause a variety of marine pollution, where the third highest toxicity potential among the ship-generated emissions in the Baltic Sea is greywater (GW) (Ytreberg et al., 2022). GW is sanitary wastewater with several sources but originating primarily from accommodation (sinks and showers), laundry facilities, and galley (kitchen area) (Jalkanen et al., 2021, United States Environmental Protection Agency (US-EPA), 2011a). GW constitutes the largest volume of sanitary wastewater generated on board ships, with an estimated volume of 5.5 million m^3 annually in the Baltic Sea and potentially discharged into the sea. Of this, 90 % is collectively generated by RoPax and cruise vessels (Ytreberg et al., 2020). Per capita GW generation rates on board passenger ships range from 157 to 235 l/person/day (Mikkola, 2020). The percentage contribution from the accommodation, laundry and galley sub flows is estimated as 64 %, 19 % and 17 %, respectively, on cruise ships (Mikkola, 2020) and 61 %, 8 % and 9 %, respectively on ferries (Juneau, 2021). GW management strategies on board vessels range from direct discharge after generation, mixing and treating with sewage, to mixing with food waste before discharge (Kalnina et al., 2021; Vaneeckhaute and Fazli, 2020). These various forms are either delivered to port reception facilities (PRFs) when at berth or into the sea during a voyage.

The discharge of GW into the sea implies that pollutants enter the marine environment and could have adverse effects. GW contains metals, nutrients, organic compounds such as *per*- and polyfluoroalkyl substances (PFAS), phthalates, persistent and mobile organic chemicals (PMOCs), polycyclic aromatic hydrocarbons (PAHs) (Jalkanen et al., 2021), benzophenone, bisphenol A, tonalide, caffeine and tris(2 chloro 1 methylethyl) phosphate (Westhof et al., 2016), suspended solids, carbon-based organics (US-EPA, 2008; Ytreberg et al., 2020), pharmaceuticals such as diclofenac and ibuprofen (Westhof et al., 2016) and microplastics (Folbert et al., 2022; Kalnina et al., 2022; Mikkola, 2020; Peng et al., 2022; Jang et al., 2024). The metals zinc (Zn) and copper (Cu) have been analyzed in ship-generated GW with the highest contribution to its cumulative environmental risk (Ytreberg et al., 2020). Elevated concentrations of Zn and Cu in the marine environment may impact marine organisms and their subsequent bioaccumulation in many different organs in fishes, molluscs and along the food chain, causing immune malformation (Alaska Department of Environmental Conservation (ADEC), 2010; Razak et al., 2021). These metals can also react with each other in complex mixtures creating different effects in

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recipient environments. Binary mixtures of Zn and Cu have shown slightly antagonistic reaction, based on the Concentration Addition (CA) model, which has influenced their toxicity on zebrafish larvae (Gao et al., 2018). Moreover, total annual load of nutrients from GW into the Baltic Sea was estimated as 159 tons of nitrogen (N) and 26.5 tons of phosphorus (P), which contribute to eutrophication (Ytreberg et al., 2020).

The reception of ship-generated GW by ports is a concern for municipal authorities who are responsible for its efficient treatment and environmentally safe final disposal (Press et al., 2020). Municipal wastewater treatment plants (MWTP) treat all sources of wastewater from the municipalities, including wastewater from ships received by the ports. However, in addition to huge volumes of ship-generated GW delivered to PRFs, uncertainty about its quality may prevent its release into the city's sewerage system (Swedish Transport Agency, 2014; IMTC, 2015). This opposition may be unfounded if the quality of ship-generated GW is unknown. The quality of influent conveyed to MWTPs may present a risk to the cleaning process or to aquatic organisms in the recipient environment. For instance, MWTPs are not designed to remove metals, hence, discharging ship-generated GW with high metal concentrations into MWTPs could result to their discharge into the recipient environment through MWTP effluent (Swedish Water, 2019). The International Convention for the Prevention of Pollution from ships (MARPOL) Annex IV (IMO, 2003) and the European Union (EU) (EU Directive 2019/883 of 2019) oblige States to equip their ports with adequate reception facilities for ship-generated wastes. Additionally, ports in Sweden are also responsible for ensuring that wastewater received from ships meet the limit values for wastewater influent set by both the municipality's general regulations for water and sewage (i.e. Allmänna bestämmelser vatten och avlopp (ABVA)), and Svenskt Vatten (i.e., "Swedish Water", the representative of municipal wastewater service companies in Sweden) (Swedish Transport Agency, 2014). In case of a doubt on which rules to follow the stricter requirements take precedence (Swedish Water, 2019). According to the ABVA, the owner of the municipal wastewater treatment facility is not obliged to receive wastewater whose content differs significantly from domestic wastewater (Press et al., 2020). Therefore, to control the quality of wastewater channeled to the municipality, some Swedish ports (for instance, the ports of Trelleborg and Ystad in the Skåne county) have installed port-based wastewater treatment plants to pre-treat ship-generated wastewater before releasing into the municipality's sewerage system.

Despite emerging insights about the ship's GW system as one that generates a complex chemical mixture with probable dangerous consequences for the marine environment, there is no legal instrument regulating ship-generated GW management. Regional and national regulations exist e.g. in the Alaska Region, Australia and Canada (Juneau, 2021; Nowlan and Kwan, 2001; The Alaska State Legislature, 2022; Transport Canada, 2013; US-EPA, 2008) but they are limited to the removal of nutrients and in some cases, biochemical oxygen demand (BOD), chemical oxygen demand (COD) coliform bacteria, total suspended solids (TSS) and some organic compounds. For other contaminants like metals, pharmaceuticals, organic compounds and microplastics, no known standards exist. The advocacy for an international GW regulation is on the rise and environmental advocates suggest that regulating GW together with sewage (black water (BW)) would make existing standards more attainable and practicable (Chen et al., 2021). MARPOL Annex IV contains a series of Regulations only for BW management, while IMO Resolution MEPC 227(64) (guidelines on implementation of effluent standards and performance tests for sewage treatment plants) (IMO, 2012) stipulates limits for nutrients, BOD_5, COD, TSS, thermotolerant coliform and pH, in sewage effluent discharged from advanced wastewater treatment plants on passenger ships operating in the Baltic Sea special area. Resolution MEPC 227(64) guidelines may not be ideal because of the limited scope of contaminants it covers. However, in the absence of a GW regulation, these standards could reveal the probability of GW polluting, to the extent of its scope. Measured concentrations (MC) of contaminants in GW above the sewage effluent standards may be indicative of potential adverse effects similar to inadequately treated sewage.

The chemical composition of GW seems fuzzy and its probable negative impacts is a concern, especially due to the variability of its sources and its restricted shore-based final disposal strategies. Uncertainties also exist on how different ship-generated GW from its land-based counterparts is, and whether it is necessary for all ports to not only receive ship-generated wastewater, but to pre-treat before releasing it into the municipal sewerage system. While these questions remain unanswered within the Baltic Region and stimulate the need for chemical analysis of GW streams, variation in the configuration of collection, storage and discharge systems, and the absence of a harmonized GW sampling procedure on board passenger ships constitute major impediments to sampling and subsequent chemical analyses. These illuminate the need for thorough planning prior to sampling, and meticulous analysis and documentation of reliable data on ship-generated GW pollutant characteristics, which is currently lacking in the Baltic Region, and constitutes a knowledge gap that this study intends to fill.

The aim of this study was twofold: first to develop a strategy for mapping sampling points, quantify, obtain and analyze GW specimens, and secondly, to assess the potential adverse effects of GW, including the different GW streams from ships operating in the Baltic Region. Two different approaches were employed to estimate the potential negative impacts of greywater, i) the hazard quotient (HQ) and hazard index (HI) approach to estimate the potential hazard of metals identified in GW and to compare the hazard potential of GW with other wastewater types (BW and mixed grey- and black water from ships, and domestic wastewater; and ii) comparison of measured concentrations of N, P, BOD_5, COD-Cr and TSS to the IMO Resolution MEPC 227(64) sewage effluent standards.

2. Materials and methods

A four-phase strategy was applied in this study, comprising the preparatory phase, onboard sampling, chemical analysis, and hazard assessment. The studied ships operate in different routes within the Baltic Sea.

2.1. Preparatory phase

Pre-sampling visits were carried out on six ships (S1-2 and S4-7) to map sampling points, examine the piping systems, and collect information on wastewater management strategies employed on board the ships. One ship (S3) was unavailable for a pre-sampling visit; instead, a request for pictures of the piping system and potential sampling points was made by email. Through onboard observations and non-structured interviews with the crew, basic statistics on systems generating GW and discharge volumes, as well as information regarding laundry practices and GW management strategies (e.g. laundry machines, dishwashers) were obtained. When the discharge volumes could not be obtained from the ships, the information was solicited from the ports. Based on this information, onboard sampling points were identified (Supplementary material 1 Fig. 1) and a sampling protocol was drafted (Supplementary material 1 Table 1). This phase also involved the acquisition, cleaning and labelling of sampling bottles (Supplementary material 1 Fig. 2) as detailed in the sampling protocol. To ensure homogeneity of the samples, a 10 L glass bottle served as the main sampling bottle from which subsamples were to be drawn.

Ship S1 is a non-commercial ship designated for special purposes with a limited staff capacity, therefore, it has no passengers. It operates regularly within the Swedish territorial sea for 7 days before berthing at the port for maintenance. S1 could be considered a proxy for cargo and container ships. S2 is a passenger/roro (RoPax) ship operating between Sweden and Germany along the Gothenburg – Kiel route. The ships S3 and S7 are RoPax ships operating between Sweden and Germany along the Trelleborg – Rostock trajectory. Ships S4, S5 and S6, which are "sister ships", are the largest RoPax ships with regards to passenger capacity among the studied ships, operating between Sweden and Finland along the Stockholm – Helsinki route. Supplementary material 1 Table 2 shows the profiles of the ships, and the GW streams sampled. In this study, 80 % of the ships were RoPax because, in addition to their availability and willingness to participate in the study, RoPax vessels generate the highest volume of GW in the Baltic Sea as they operate all year round (Ytreberg et al., 2020). Discharged volumes of GW were collected from ships S1 to S6 for various periods. In addition to quantification of the discharged volumes, the data was used to estimate the GW discharge rates.

2.2. Onboard sampling

Among the 7 ships shortlisted, 5 ships were sampled between February 2023 to March 2023. Two major types of sampling methods were used based on the configuration of the ships and the location of the sampling points: sampling from a holding tank, and sampling from the discharge pipes.

Sampling from the holding tank was the second method applied on one vessel (S1). Sampling was done at full tank capacity as well as full crew capacity while operating offshore. Due to the location of the sampling point, which was close to the bottom of the tank and restricted by pipes (Supplementary material 1 Fig. 1A), the large 10 L could not be filled, hence the smaller sampling bottles were filled directly from the discharge point. In addition, the tank was relatively small, so its content was considered homogenous.

Sampling points at the discharge pipes required the exercise to be executed during the release of GW to PRFs (the case of S2–S5). Where the ship had separate tanks for the accommodation, laundry, and galley GW streams (the case of S2), discharge was performed sequentially using the same pipeline, and sampling was done in the middle of each process. To minimize the risk of cross-contamination during sampling, the first stream was discharged completely, after which the second stream was allowed to flow for some minutes before sampling, making sure the pipe was completely cleared of the first stream. Supplementary material 1 Fig. 3 shows sampling on board the ships. On some ships, the GW discharge outlets were surrounded by intercrossing pipes that restrained accessibility. In such instances, it was not feasible to use the 10 L glass bottles, therefore, the manometer line next to the flowmeter was detached and extra extension pipes were connected to the sampling points to facilitate both the positioning and filling of the bottles. On S4 a metal pipe extension was connected, and on S5 a plastic tube extension was connected from the sampling point outward (Supplementary material 1 Fig. 3C and D, respectively).

Even though pre-cleaned bottles were used, all the bottles were rinsed three times with GW before sampling, as a quality control measure. A green glass 1 L bottle was filled with milli-Q water and used as a blank during each sampling phase. Mixed GW streams made up of the 3 main sub-flows (accommodation/laundry/galley (A/L/G)) were obtained from ships S1, S4 and S5, a mixed GW stream containing 2 sub-flows (accommodation/laundry (A/L)) was obtained from ship S3, while separate sub-flows were obtained from ships S2 (laundry, accommodation, and galley) and S3 (galley). On average, the GW streams were classified into 5 categories, namely: laundry ($n = 1$), accommodation ($n = 1$), galley ($n = 2$), mixed A/L ($n = 1$) and mixed A/L/G ($n = 3$). At the end of the sampling process, 8 samples were obtained from five ships, and each sample was further distributed into 14 sampling bottles of various sizes (Supplementary material 1 Fig. 4) according to the designated chemical analysis to be performed. The samples were packed in boxes containing ice blocks and shipped to accredited laboratories for analysis.

2.3. Chemical analysis

A commercial laboratory, ALS Global, analyzed metals, nutrients, Total Suspended Solids (TSS), organic matter and oxygen-consuming substances (OMOCS) in the matrix. Nutrient analysis allows for the determination of various forms of nitrogen and phosphorus in wastewater. Total nitrogen is the sum of Total Kjeldahl Nitrogen (TKN) and Nitrite-Nitrate nitrogen (US EPA, 1993) while total phosphorus is the sum of orthophosphate, polyphosphate, and organic phosphate in wastewater (Marshall, 2008). Analytical tests aimed at determining the concentrations of OMOCs have been used to establish the relative pollutant content of wastewater samples, and four laboratory tests have been customarily used. They include chemical oxygen demand – chrome (COD-Cr), biochemical oxygen demand (BOD), total organic carbon (TOC) and fat. COD-Cr measures oxygen consumption during the decomposition of organic and oxidation of inorganic matter, using dichromate as an oxidizing agent, within a short time such as in hours. BOD determines the concentration of oxygen required by microorganisms to decompose organic matter in wastewater during a period of 5 days (BOD_5) or 7 days (BOD_7). TOC measures the amount of organic carbon present in all the dissolved compounds in wastewater. TOC is considered the most complete of these analytical techniques as it detects all kinds of organic matter in wastewater (Aguilar-Torrejón et al., 2023). Fat analysis measures Fats, Oils and Greases (FOG) in wastewater which are problematic for sewerage systems by causing clogging (Mohana et al., 2023). TSS measurements capture the total concentration of non-soluble solids suspended in the greywater (Verma et al., 2013). ALS Global is accredited for all analyses except fat. As far as possible, the analyses were done per standardized methods as shown in Supplementary material 1 Table 3.

When a specific metal was reported as under the limit of detection (LOD) in at least one of the GW streams, half the LOD was used as a default value, according to Johnson (2018), Cohen and Ryan (1989), Ytreberg et al. (2020) and the Press et al. (2020) citing the Swedish EPA's guidance on environmental report of 20-02-2008 which states that "*when calculating annual values based on concentrations below the detection limit, the value corresponding to half the detection limit should be used in the calculation, on the condition that at least one concentration value is above the detection limit*". However, if the concentration of a specific metal was reported as <LOD in all eight GW streams, the data set was excluded from the hazard potential assessment. The uncertainty underlying this approach was monitored by ensuring that the percentage of data <LOD, and the relative contribution of such metals to the overall Hazard Index were minimal. The results of COD-Cr, TSS and nutrients were compared with the IMO MEPC 227(64) sewage effluent standards. For BOD_7 the results were converted to BOD_5 using the conversion factor, $BOD_5 = BOD_7/1.15$, as recommended by HELCOM (2011), before comparing with the IMO MEPC 227 (64) sewage effluent standards.

2.4. Hazard potential assessment

The European Union Marine Strategy Framework Directive (MSFD) (European Commission, 2013) stipulates eleven descriptors representing the concept of Good Environmental Status (GES) in the European sea areas and under descriptor 8, a set of criteria was developed for metals and organic compounds in the marine environment known as the Environmental Quality Standards (EQS) or Predicted No Effect Concentration (PNEC). In Sweden, the Swedish Agency for Marine and Water Management (SwAM) implements the EQS by setting PNEC values which, when compared with measured concentrations of metals, reveals the probable adverse effects of GW contaminants on marine life. To estimate the potential hazard of ship-generated GW, a hazard potential assessment (Ramírez-Morales et al., 2020) was carried out specifically for metals detected in GW. The Hazard Quotient (HQ) approach was used for individual metals as per European Commission (2003), Lucas et al. (2016), Orias and Perrodin (2013), and Ramírez-Morales et al. (2020) to rank the metals according to the magnitude of the hazard. HQ is defined as "the ratio of the potential exposure to a substance and the level at which no adverse effects are expected" (Goumenou and Tsatsakis, 2019) and obtained by comparing the average measured concentration (MC) of a specific substance with its predicted no-effect concentration (PNEC) value (Orias and Perrodin, 2013; Orias and Perrodin, 2014), as shown in Eq. (1). In the absence of the relevant parameters for the computation of the PNEC values, applicable regulatory documents, as well as prior literature, were explored for existing PNEC values (European Commission, 2013; HVMFS, 2013:19; HVMFS, 2012:18; European Chemical Agency (ECHA) Chemicals Database (ECHACHEM); Ytreberg et al., 2020; Tulcan et al., 2021), therefore, PNECs of 11 metals were obtained as shown in Table 7.

$$Hazard\ Quotient\ (HQ) = \frac{AMC}{PNEC} = \frac{Average\ Measured\ Concentration}{Predicted\ No\ Effect\ Concentration} \tag{1}$$

The hazard potential yielded by the metals identified in GW, which is the sum of the HQs for individual metals (ΣHQ) (Carazo-Rojas et al., 2018; Lucas et al., 2016; Ramírez-Morales et al., 2020), also known as the Hazard Index (HI) (Price, 2023; US-EPA, 2011b) was calculated based on the formula presented in Eq. (2). HIs are computed based on the Concentration Addition (CA) concept (Backhaus and Faust, 2012; Bopp et al., 2016; Norwood et al., 2003) assuming that the toxicity of the mixture is additive and has a similar mode of action. The HI approach establishes that if the sum of the HQs is maintained below 1, then adverse effects are no more likely to occur than if each chemical's exposure occurred separately (Price, 2023). Therefore, an HQ or HI ≥ 1 represents a high hazard potential, which implies that a particular substance or a mixture of multiple chemicals is likely to cause adverse effects on the aquatic environment if exposed to 100 % GW. Moreover, 1 $>$ HQ or HI ≥ 0.1 means a medium hazard potential and HQ or HI < 0.1 implies a low hazard potential (Carazo-Rojas et al., 2018; Lucas et al., 2016; Ramírez-Morales et al., 2020).

$$Hazard\ Index(HI) = \sum\nolimits_{i=1}^{n} (HQs)i \tag{2}$$

Therefore, the HQs of 11 metals, the cumulative HI of GW, the HIs of the 5 GW categories and the relative contribution of each metal to the HIs were computed.

2.4.1. Statistical analysis

One-way analysis of variance (ANOVA) was performed using IBM SPSS Statistics (version 28) to identify significant effects ($p < 0.05$) in HI between the different groups (i.e. greywater from ships ($n = 142$), blackwater from ships ($n = 36$), mixed grey- and blackwater from ships ($n = 297$) and domestic wastewater (mixed grey- and blackwater from houses) ($n = 12$) (Supplementary material 2). HI was calculated based on the concentrations of Cu, Zn and Mn, since these metals jointly contributed 95 % of the cumulative Hazard Index of GW. The homogeneity of variances was examined using Levene's test. The results of the Levene's Test were not significant, i.e. the two variances are approximately equal, and the assumption is met to perform an ANOVA.

3. Results and discussion

3.1. Greywater systems and onboard management strategies

The pre-sampling visit on the studied ships confirmed that the layout of the piping systems on board the ships was non-identical, and the ships' GW management strategies varied. S1 discharges all GW into the sea, S2 discharges some GW in the sea and some to PRFs, while S3–S6 discharge all GW to PRFs. Supplementary material 1 Table 4 summarizes the systems and GW management strategies on board the studied ships. A common practice observed on all the studied ships was that all heavy laundry, including bed linen and towels, is done ashore, leaving only the crews' and sometimes a few passengers' clothing being laundered on board. Therefore, it could be assumed that relatively lower quantities of laundry detergents are used on board compared to ships on which all the laundry is done on board. Moreover, according to the Chief Engineer (pers comm.), the GW piping system on ship S4 stretches several meters long and usually there is the issue of clogging in the pipes that reduces their diameter, therefore, products are dozed in the sinks and flushed weekly to clean the clogged pipes. It was reported that the use of these products poses difficulties because it is hard to find "environmentally friendly" products and that protein-based products have been used but they are barely as effective as expected even when dosed in high quantities. The exact dosage for the said products was unclear.

3.2. Greywater discharged volumes

S1 operates approximately 13 days a month and 160 days a year, discharging about 1.60 m^3 of GW daily. The annual discharge volume from S1 was estimated as 256 m^3/y. With an average daily crew of 14 persons, this volume corresponds to a per capita annual discharge rate of 114 l/p/y from S1. The total volumes of GW discharged from the RoPax ships S2–S6 from 2019 to 2022 was approx. 503,000 m^3. This corresponds to an average annual discharge volume of about 126,000 m^3/y from five RoPax ships. It also constitutes about 12 % and 14 % of the GW volumes generated by RoPax vessels operating in the Baltic Sea in 2012 (Ytreberg et al., 2020) and 2022 (Jalkanen et al., 2023), respectively. The average annual GW volumes discharged from the ships S2 – S6 during the period 2019–2022 were approximately 13,000 m^3/y, 13,700 m^3/y, 37,300 m^3/y, 29,900 m^3/y, and 31,500 m^3/y, respectively (Fig. 5A). These correspond to per capita discharge rates of 54 l/p/d, 138 l/p/d, 62 l/p/d, 49 l/p/d, 57 l/p/d (Supplementary material 1 Table 5). GW discharge volumes on all the ships were affected by the Covid 19 pandemic between 2020 and 2021 where the lowest discharge volumes were registered. Exceptionally, S3 recorded the highest GW

Table 7
A comparison between the average concentrations of some metals with the Swedish Water's guideline values for wastewater from operations (Swedish Water, 2019; Swedish Transport Agency, 2014).

Contaminants	Concentration in ships' GW ($n = 8$) (µg/l)		Swedish Water's guideline standards (µg/l)	ABVA Helsingborg (µg/l)	ABVA Trelleborg (µg/l)
	Approx. average	Highest values			
Ag	0.25	0.25	10	50	50
Cd	0.04	0.11	0.1[a]	0.5	Should not exist
Cr	2.30	6.58	10	10	50
Cu	32.1	96.3	200	200	200
Hg	0.01	0.01	0.1[a]	0.5	Should not exist
Ni	8.14	27.5	10	50	50
Pb	0.55	1.91	10	50	50
Zn	78.2	300	200	500	200
BOD_5/COD ratio	0.4–0.6	0.6	>0.5	>0.5	–
Fat content	43,000	140,000	–	<150,000	100,000

[a] Not allowed in industrial processes wastewater discharged into the wastewater network.

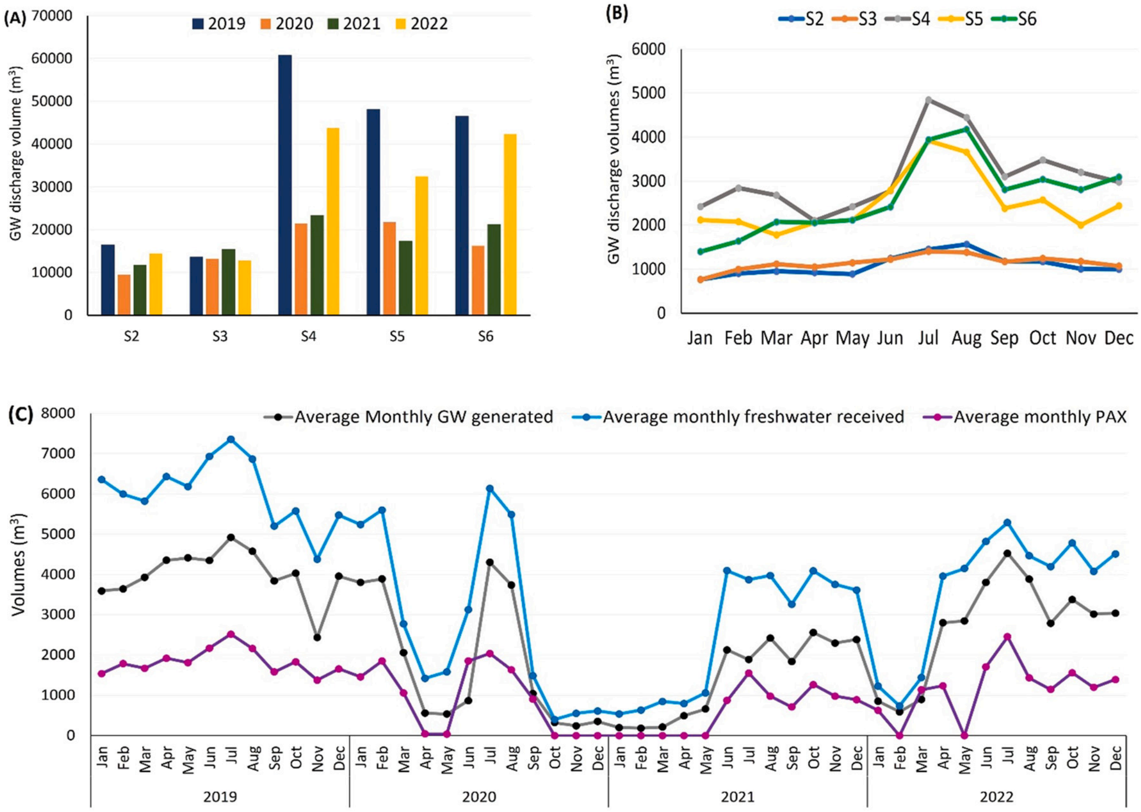

Fig. 5. (A) Greywater discharged volumes from five RoPax ships operating in the Baltic Sea during the period 2019–2022. (B) Variation in monthly GW discharged volumes from five RoPax ships (S2, S3, S4, S5 and S6) operating in the Baltic Sea from 2029 to 2022. (C) Monthly and annual variations in GW discharged volumes (grey line), freshwater reception volumes (blue line) and passengers (PAX) counts (purple line) on a RoPax ship (S5) operating in the Baltic Sea for a period of 4 years from 2019 to 2022. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

discharged volumes in 2021 within a 12-year period from 2012 to 2023 (Supplementary material 1 Fig. 6C). This ship may have implemented stringent Covid-19 protective measures to gain the confidence of passengers during this period when many ships were laid off. Considering 2019, the year before the Covid pandemic distorted passenger traffic trend, the per capita discharge rate from the five RoPax vessels were 68 1/p/d, 137 1/p/d, 100 1/p/d, 79 1/p/d and 84 1/p/d.

The data shows recurring and episodic variability in GW discharge volumes which depends on vessels type, size, number of persons, activities, water use practices, water-saving and GW management strategies implemented on board, as well as the season. There was great variation in the monthly discharged volumes which was mainly due to seasonal changes in weather conditions affecting movements and passenger counts (Jalkanen et al., 2023). The average monthly GW discharge volumes from ships S2–S6 during different periods between 2012 and 2023 ranged from approx. 743 m^3 in January to approx. 4800 m^3 in July (Supplementary material 1 Table 5). While maximum volumes were discharged in July and August, minimum discharged volumes varied among the ships between December to January and March to April. More passengers travel during the summer, therefore peak volumes observed in July and August from all the ships were evident (Fig. 5B).

GW generation and discharge are also a function of the volume of fresh water received from the port, and both also depend on the number of passengers on board. On S5, a total of 126,000 m^3 of GW was generated from 181,000 m^3 of freshwater received from 2019 to 2022, constituting 70 % of freshwater received. All three parameters were affected by the Covid 19 pandemic from October 2020 to May 2021 where the number of passengers (Pax) was zero (Fig. 5C). During this period 5450 m^3 of freshwater was obtained from the port and 2670 m^3 of GW was generated, constituting 49 % of freshwater received. This shows about 97 % and 98 % reduction in freshwater received and GW generated, respectively, due to limited number of crew on board during the Covid 19 pandemic.

Regarding GW and BW proportions, a total of 113,000 m^3 of wastewater was generated on board S2 from 2018 to 2023. Of this, approx. 82,900 m^3 (73 %) was GW and approx. 30,000 m^3 (27 %) was BW. On S3, a total of 167,000 m^3 of wastewater was generated from 2012 to 2023; 146,000m^3 (87 %) was GW and 21,400 (13 %) was BW (Supplementary material 1 Fig. 6A and B). Based on S2 and S3 an average GW:BW proportion of 80:20 was recorded. The present result matches the Cleanship study (Madjidian and Rantanen, 2011) which reported GW:BW distribution between 70:30 and 90:10, constituting an average of 80:20. Therefore, GW discharge volumes on RoPax ships can be as much as four times the volume of BW discharged.

3.3. Greywater characterization

The dissolved concentration of metals in the eight GW streams were variable (Fig. 7A). Zn, Mn, and Cu had the highest average concentrations, but with a huge variation from approximately 1.00 μg/l in S3G to 300 μg/l in S2G for Zn; 7.10 μg/l in S3A/L to 170 μg/l in S2G for Mn and 2.20 μg/l in S3G to 96.3 μg/l in S1 for Cu (Fig. 7B). The average concentrations of these metals in the eight GW streams were 78.2 μg/l for Zn, 40.3 μg/l for Mn and 32.1 μg/l for Cu. S2 and S3 are "sister" ships with the same sizes, configurations, and similar onboard activities. Therefore, the huge variation in Zn concentrations recorded in the galley GW streams from both ships (1.00 μg/l and 300 μg/l, respectively) was surprising. Regarding the five GW categories, highest average concentrations of Zn and Mn (about 151 μg/l and 110 μg/l, respectively) were recorded in galley GW ($n = 2$), while highest average concentration of Cu (52.5 μg/l) was in mixed A/L. Zn, Cu and Mn have been measured in

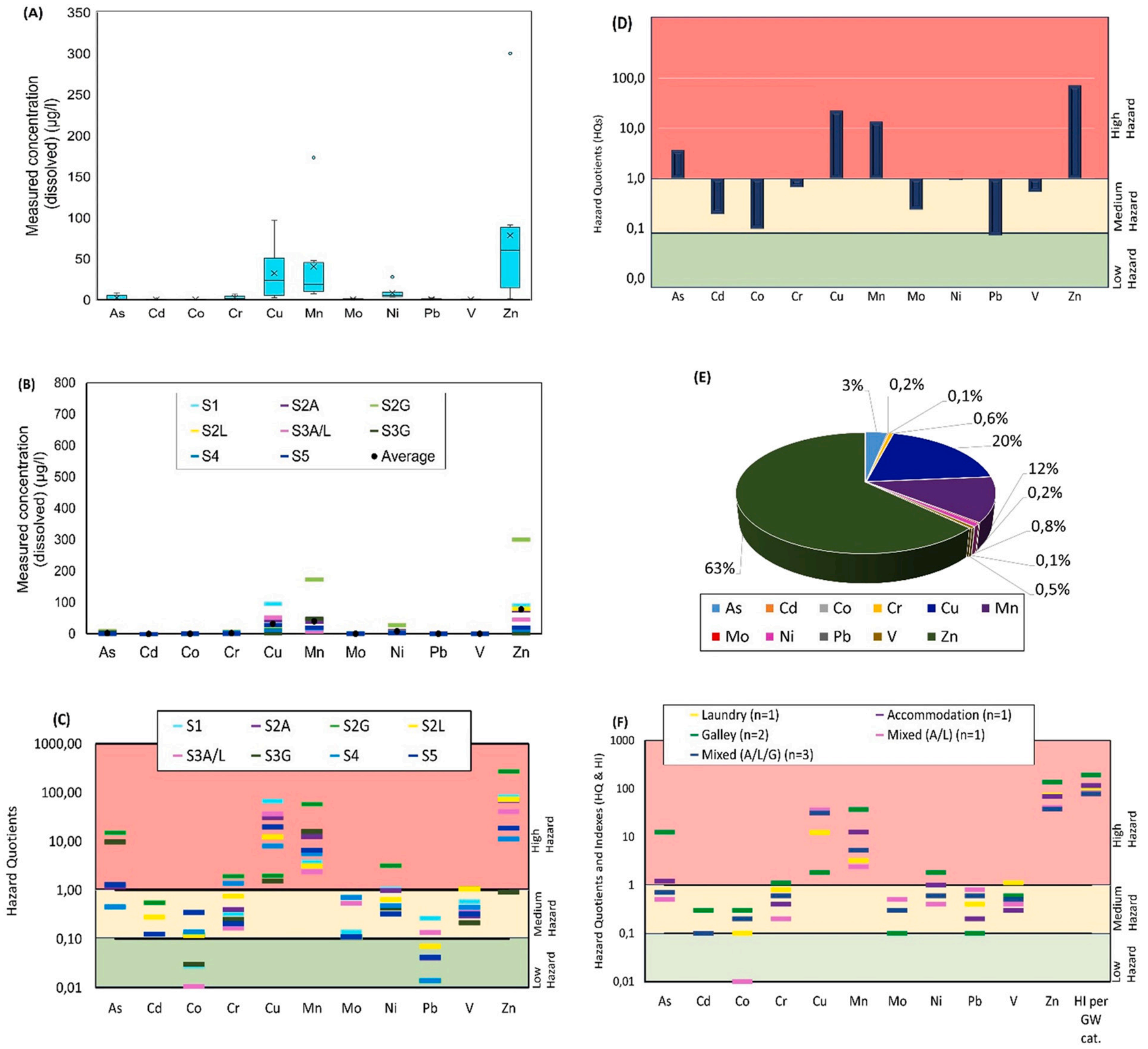

Fig. 7. (A) Variation in the concentrations of metals in ship-generated GW (B) Concentrations of metals in 8 ship-generated GW streams (C) HQs of metals in eight ship-generated GW streams (D) Hazard ranks of metals in ship-generated GW (E) Relative contribution of metals to the HI of eight ship-generated GW streams. Zn, Cu, Mn and As collectively contributed 98 % to the HI (F) Hazard ranks of five categories of ship-generated GW streams.

ship-generated GW and BW, as well as in domestic wastewater samples within the Baltic Region and beyond. A comparison of the average concentrations of these metals and other conventional pollutants across several studies, and in various sanitary wastewater streams from ship-based and land-based sources, has been summarized in Supplementary material 1 Table 6 and Supplementary material 2 Table 19.

Mixed (A/L/G) greywater in the present study (n = 3) showed average dissolved concentrations of Zn:41.3 ± 43.2 µg/l, Cu:45.5 ± 44.8 µg/l and Mn:15.9 ± 4.2 µg/l. This is low compared with the concentrations of the same metals (Zn: 358 ± 217 µg/l, Cu: 43.4 ± 31.5 µg/l, and Mn: 62.4 ± 52.2 µg/l) in mixed GW samples (n = 5) from two cruise ships, two RoPax and a RoRo ship in Stockholm (STA, 2014). Higher and lower dissolved concentrations of Zn and Cu, respectively (Zn: 229 ± 870 and Cu: 43.3 ± 66.9) were obtained from an analysis of 27 ADEC reports of mixed GW sampling (n = 149) from cruise ships in the Alaska Region, USA (Supplementary material 2 Table 19). All these results are likely influenced by the number of samples analyzed as well as the types and sizes of the ships involved. The average dissolved concentrations from all the cited studies (n = 167) were estimated as Zn: 233 ± 859 µg/l, Cu: 43.7 ± 66.0 µg/l and Mn: 44.9 ± 46.3 µg/l. On average, these metals were higher in mixed GW from land-based sources than in the current study (Supplementary material 1 Table 6).

Compared with ship-based BW, Gryaab (pers. comm.) reported dissolved concentrations of Zn, Cu and Mn from four cruise ships (n = 4) as Zn: 200 ± 122 µg/l, Cu: 18.1 ± 19.6 µg/l and Mn: 40.2 ± 11.6 µg/l, while the Swedish Transport Agency (2014) recorded dissolved concentrations (n = 3) from two passenger ships and a RoRo ships in Stockholm as Zn: 101 ± 41.9 µg/l, Cu: 17.0 ± 7.8 µg/l and Mn: 26.0 ± 19.0 µg/l. These show higher Zn and Mn but lower Cu concentrations than the mixed GW in the present study. An analysis of blackwater (n = 3) from cargo ships in seaports in Nigeria by Onwuegbuchunam et al. (2017) show substantially lower concentrations (Zn: 0.040 µg/l and Cu:1.20 µg/l) than in the mixed GW from S1 (Zn: 90.9 µg/l and Cu: 96.3 µg/l) which was considered a proxy for cargo ships. Results from 7 US-EPA reports (n = 38) recorded Zn: 130 ± 230 µg/l and Cu: 45.6 ± 77.6 µg/l, showing the higher Zn but the same Cu concentration as in mixed GW in the present study. The average dissolved concentrations in BW based on these cited studies (n = 47) were obtained as Zn: 131 ± 212 µg/l, Cu: 40.2 ± 71.4 µg/l and Mn: 34.2 ± 15.7 µg/l. Relative to land-based BW from households, Palmquist and Hanaeus (2005) reported considerably average concentrations (Zn: 525 µg/l, Cu: 126 µg/l, and Mn: 130 µg/l) from three households in Stockholm (n = 3), higher than the average concentrations in mixed GW from ship-based sources.

In relation to mixed BW/GW (mixed wastewater) average dissolved concentrations of Zn, Cu and Mn reported by the Swedish Transport Agency (2014) were (Zn: 156 ± 159 μg/l, Cu: 17.7 ± 17.5 μg/l and Mn: 46.0 ± 77.6 μg/l), the port of Trelleborg (pers. comm.) (Zn: 1255 ± 5565 μg/l, Cu:146 ± 388 μg/l), 8 US-EPA reports (Zn:170 ± 534 μg/l, Cu: 72.3 ± 67.8 μg/l) and Özkaynak et al. (2022) (Zn:4635 ± 7.00 μg/l, Cu: 2075 ± 559 μg/l), show great variability likely due to differences in ship types and sizes. Up to 86,000 μg/l and 5900 μg/l representing Zn and Cu concentrations, respectively, were recorded in the mixed wastewater from a ship at the Trelleborg port. Agglomerating all these studies ($n = 324$), the average concentrations were obtained as Zn: 1027 ± 4971 μg/l, Cu: 126 ± 344 μg/l, Mn: 46.0 ± 77.7 μg/l. Analysis of these metals in domestic wastewater ("wastewater from residential settlements and services which originates predominantly from the human metabolism and from household activities" (European Commission, 1991)) from three municipalities in Gothenburg, reported by Gryaab (Press et al., 2020) showed dissolved concentrations as Zn:114 ± 44.5 μg/l, Cu:73.8 ± 19.90 μg/l, Mn:87.2 ± 30.0 μg/l. This result revealed lower Zn, but higher Cu and Mn concentrations, than the average concentration in mixed GW from ship-based sources.

Regarding the GW sub flows on ships (accommodation, laundry, galley and mix accommodation/laundry), the average dissolved metal concentrations agglomerated from various studies were Zn: 302 ± 178 μg/l, Cu: 231 ± 194 μg/l, Mn:13.4 ± 12.8 μg/l in accommodation GW, Zn: 323 ± 214 μg/l, Cu: 304 ± 225 μg/l, Mn: 6.19 ± 1.43 μg/l in laundry GW, Zn: 551 ± 596 μg/l, Cu: 299 ± 411 μg/l, Mn: 31.5 ± 41.8 μg/l in galley GW and Zn:148 ± 120 μg/l, Cu:127.5 ± 85.1 μg/l, Mn: 7.08 μg/l in mixed accommodation/laundry GW. This shows that galley GW is the highest contributor of Zn and Mn while laundry GW is the highest contributor of Cu in mixed GW. The analysis of two galley GW samples by the Port of Trelleborg presented relatively higher average concentrations of Zn and Cu than the average concentration from the same sub flow type in the current study. Both results are from the same ship, sampled during different periods by different institutions. This difference is likely due to seasonal changes in the activities on this ship. Similar differences were observed among the mixed A/L sub flows in which the dissolved concentrations of the same metals in the Trelleborg port analyses showed relatively higher concentrations than in the same sub flow in the current study.

The exact sources of Zn and Cu on board the studied ships were unclear and require further investigation. However, a study by ADEC (2010) revealed that onboard piping systems, cookware and some disinfectants could be potential sources of Cu in wastewater from ships. Some parts of evaporators used in making potable water onboard, as well as Cu refrigeration and air conditioning systems also contain Cu which could leach into GW through AC condensate (ADEC, 2010). There are claims that the presence of Zn and Cu in GW could originate from the materials used in ship plumbing (Friends of the Earth, 2023), however, S4 and S5 which are sister ships had a great variation in the concentration of Cu, with S5 being more than double S4. Both ships have the same manufacturer, hence the same materials could have been used for plumbing and piping, therefore, there may be other sources of Cu on board these vessels, yet to be identified. In a land-based study, significant contribution of Cu was measured in tap water (109 μg/l), bathroom sink (1220 μg/l) and dishwasher (434 μg/l), however, as these appliances were low volume, relatively small Cu loads were obtained for bathroom sink (2440 μg/wash) and dishwasher (7000 μg/wash). Product analysis from the said study also detected relatively high concentrations of Cu in laundry and dishwasher products and soaps (Diaper et al., 2008). Zn in GW is also believed to originate from sacrificial anodes usually used in GW tanks to prevent corrosion (Swedish Transport Agency, 2014). Another probable account could be that the ship environment is corrosive and rust from zinc-galvanized surfaces on board may contribute to Zn concentration in the ship's wastewater (Swedish Transport Agency, 2014). Finally, an assessment of the products used on board four of the studied ships shows that some products such as surface coating, glue/adhesive/tape, metal cleaner, and lubricants contain Zn and Cu, which may leach into GW. In Tjandraatmadja et al. (2010), Zn concentrations in GW from household appliances outlets varied widely, hence, further investigation of Zn contributions from household appliances as opposed to products used was recommended.

The average dissolved concentration of Mn in the mixed GW samples was higher than Cu. In S2G, the concentration of Mn was the next highest (178 μg/l) after Zn (300 μg/l). This shows that Mn could be another metal of concern in ship's GW, whose origin has not yet been established. In Tjandraatmadja et al. (2010), the chemical analysis of GW samples from household appliances (kitchen sinks, vanity units, showers, dishwashers, tap water, front loader and top loader washing machines) all showed concentrations of Mn below the LOD. This may not rule out the possibility of Mn originating from washing and cleaning products used on board ships, as its presence in paint and cosmetic products has been recognized (Rudi et al., 2020). Approximately 90 % of the total Mn demand is accounted for by the iron and steel industry where it is used as an oxidant for cleaning, bleaching and disinfection during production (van Zyl et al., 2016; Rudi et al., 2020) and to improve the strength, hardness, and stiffness (Bull, 2010), therefore, Mn in GW could likely originate from iron and steel products used on board ship's galley. The presence of Mn in stock food additives, milk, nuts, oats, spinach, and tea (Marsidi et al., 2018) could also explain its high average concentration in galley GW in the form of food waste additions.

The highest concentrations of Fe and Al in the present study, approximately 22.1 mg/l in S2G and 195 μg/l in S4, respectively, were comparable to concentrations in riverine inputs of Fe and Al from some Swedish river mouths into the Baltic Sea (Swedish University of Agricultural Sciences (SLU) Environmental data MVM version 2.17.12).

Additionally, since 80 % of the studied ships discharged GW to PRFs, the results of the present study were compared with the Swedish Water's limit values, and ABVA requirements from two municipalities in Sweden (Table 7). Apart from Cd and Hg which, according to ABVA Trelleborg, should not exist, all other pollutants were below the Swedish Water's and the ABVAs' limit values, suggesting that the discharge of ship-generated GW ashore should not constitute a threat to the biological processes in the MWTPs.

However, the discharge of galley GW individually could be discouraged due to the presence of Zn. Zn concentration in one sample (300 μg/l) was above the Swedish Water's and ABVA Trelleborg's limits, but lower than ABVA Helsingborg's limit. In comparison, Zn concentration in Swedish Transport Agency (2014) showed higher values than the Swedish Water's limit values. Although these local regulations are implemented together, according to Swedish Water (2019), where there is a doubt on which rules to implement, the stricter requirements always take precedence.

3.4. Hazard potential assessment of metals in greywater from ships

The HQs of Zn, Cu and Mn were >1 in all eight GW streams while the HQ of As was >1 in four GW streams (Fig. 7C). Although the average MC of As was relatively low (2.01 μg/l), its HQ represented a high potential hazard due to its low PNEC value (0.55 μg/l) (Fig. 7D). The cumulative HI based on the average metal concentrations from the eight GW streams was obtained as 113. Zn, Cu, Mn and As with HQs of 71.0, 22.1, 13.4 and 3.65, respectively, drive the hazard potential of GW. Individually, they contribute 63 %, 20 %, 12 %, and 3 %, respectively (Fig. 7E), and 98 % collectively to the HI, as shown in Table 8. The contribution of Zn and Cu to the HI, is in line with Ytreberg et al. (2020), which showed that Zn and Cu had the largest contribution to the cumulative risk of GW discharge to the Baltic Sea (67 % and 27 %, respectively).

Some metals (Cr and Ni) and the metalloid (As) presented a high hazard potential only with regards to some GW streams, while Zn, Cu and Mn presented a high hazard potential with regards to all eight GW streams (see Fig. 7C) implying that all eight streams are potentially

Table 8
Hazard Quotients (HQ) of selected metals in ship-generated GW, their relative contributions (RC) to the Hazard Index (HI) and their hazard ranks.

No.	Metals	Average dissolved MC (μg/l)	PNEC (μg/l)	HQs	RC to the HI (%)	Hazard rank
1	As	2.01	0.55[b]	3.65	3.21	High
2	Cd	0.04	0.2[a]	0.198	0.174	Medium
3	Co	0.24	2.36[d]	0.101	0.089	Medium
4	Cr	2.30	3.4[b]	0.678	0.596	Medium
5	Cu	32.1	1.45[b]	22.1	19.5	High
6	Mn	40.3	3.0[d]	13.4	11.8	High
7	Mo	0.55	2.28[d]	0.242	0.213	Medium
8	Ni	8.14	8.6[a]	0.946	0.833	Medium
9	Pb	0.54	1.3[a]	0.412	0.363	Medium
10	V	0.31	0.57[c]	0.545	0.480	Medium
11	Zn	78.2	1.1[b]	71,0	62.53	High
Hazard Index (HI): 113						

[a] According to Directive 2013/39/EU (European Commission, 2013).
[b] According to the SwAM regulation (HVMFS, 2012:18; HVMFS, 2013:19, cited by Ytreberg et al., 2020).
[c] According to Tulcan et al. (2021).
[d] According to the European Chemical Agency (ECHA).

highly hazardous. In a land-based study in a single person household (Diaper et al., 2008), relatively high average concentrations of As were measured in greywater from a dishwasher using a leading brand detergent (145 μg/l), and from a washing machine using both a leading brand (35 μg/l) and a no-name brand laundry detergent (27.5 μg/l).

In the current study, the highest average concentration of As was in galley GW (8.32 μg/l), and below LOD in laundry GW therefore, dishwashing detergents may likely be the source. However, the relatively low average concentration of As in the current study may likely be due to dilution factor from the use of relatively higher volumes of water by the number of persons on the ships, compared to the single person household or due to the use of ecolabel products on board the ships which may have lower As content. The HI obtained in the current hazard potential assessment is more than two orders of magnitude higher than 1, showing a potential high hazard. However, to assess the risk posed by GW if discharged into the Baltic Sea, a site-specific risk assessment should be performed and operational procedures such as discharge flow rates and dilution factors need to be considered.

Regarding the five GW categories, the HIs in decreasing order were galley (192) > accommodation (115) > laundry (92.2) > mixed A/L (82.2) > mixed A/L/G (77.9) GW streams, all of which are classified as high hazards (Fig. 5F). Zn, Cu and Mn collectively contributed to >95 % to the HIs of laundry, accommodation, mixed A/L and mixed A/L/G GW streams. However, the contribution of Cu to the HI of galley GW was relatively low, suggesting that accommodation and laundry GW sources could be the main contributors of Cu in the mixed GW stream. Moreover, the relative contribution of Zn to the HI of laundry and galley GW streams was utmost (>70 %) showing that Zn is the main driver of the hazard potential of these GW streams (Supplementary material 1, Table 9). Comparing the HI differences between selected sanitary wastewater streams from ship-based and land-based sources using one-way ANOVA showed no significant differences in HI between any of the groups tested, i.e. greywater, blackwater and mixed grey- and blackwater from ships as well as domestic wastewater (mixed grey- and blackwater from houses) (p-value = 0.101) (Supplementary material 2, Tables 3, 6, 9 and 10). This shows that although the generated volumes of these streams are different, their impact on the environment may be the same, if discharged untreated. Therefore, municipal wastewater treatment plants in Sweden should receive and treat GW from ships in the same way as domestic wastewater.

The HQs of three priority substances in the field of water policy according to the EU EQS Directive 2013/39/EC of 2013 (European Commission, 2013), were Cd:0.2, Ni:0.9, and Pb:0.4, indicating a medium hazard potential. All three collectively contributed 1.4 % to the HI (Table 8). The HQs of Cd, Ni, and Pb presented a medium hazard potential for all five GW categories, while Ni presented a high hazard for accommodation and galley GW streams, and a medium hazard for the rest (Fig. 7F). This result could be considered in the development and implementation of GW management strategies.

Although the Concentration Addition (CA) concept has been used in estimating metals' cumulative effect in mixtures, it is established that there's a tendency of metals to interact with each other in the mixture, generating effects which may be antagonistic, strictly additive, or synergistic with the possibility of influencing their toxic effects on the aquatic organisms (Norwood et al., 2003). For instance, investigating the toxicity of the binary metal mixtures on the survival of zebrafish larvae, Gao et al. (2018) revealed synergistic interactions with the Cu – Cd and Cu – Pb mixtures, antagonistic interaction for Cu – Zn mixtures and no interactions for the Cd – Pb mixtures, which signifies that toxic action mode may depend on the combination and concentrations of the metal mixtures. This implies that the presence of Cu, Cd and Pb in the GW mixture may give rise to both synergistic and/or antagonistic effects. The current study recognizes the uncertainties related to the use of the CA approach such as incomplete knowledge of the mixture composition in terms of all the compounds and their concentrations, as well as the hazard posed by all the mixture components (Norwood et al., 2003). However, it is beyond the scope of the current study to do a complete risk assessment, therefore the assumption that the metals interaction in the GW is strictly additive, is applied.

3.5. Potential negative impacts of other contaminants in greywater from ships

3.5.1. Concentration of nutrients

Total N and P concentrations show a wide variation across the eight GW streams (Fig. 8A). The average N concentration was 17.1 ± 8.80 mg/l, with the lowest contribution of 9.23 mg/l from S1 and the highest of about 33.8 mg/l from S2G (Fig. 8B). In terms of the five GW categories, galley GW (n = 2) had the highest average concentration of N (approx. 30.2 mg/l), and mixed A/L had the lowest (approx.10.2 mg/l) (Fig. 8C). The geomean concentration of N (15.4 mg/l) was lower than the IMO MEPC 227(64) sewage effluent standards for N (20 mg/l). Among the five GW categories, only the geomean concentration of N in galley GW (29.9 mg/l) was above the MEPC 227(64) standard for N (Fig. 8D). This shows that although N from galley GW has a relatively high influence on the nutrient pollution potential of GW, the geomean concentration of N obtained from the eight streams shows acceptable GW discharge level like adequately treated sewage from AWTP.

The average concentration of P was 12.53 ± 14.5 mg/l, with the highest contribution from the lone laundry GW stream, S2L (42.0 mg/l) and the lowest contribution of 0.531 mg/l from S1 (Fig. 8B). Among the five GW categories mixed A/L/G had the lowest average concentration of P (1.23 mg/l), while accommodation and galley GW streams had similar average concentrations of P (approx. 17.3 mg/l and 16.8 mg/l, respectively) (Fig. 8C). The geomean concentration of P (5.53 mg/l) was more than five times higher than the IMO MEPC 227(64) requirement (1 mg/l). Regarding the five categories, all except the mixed A/L/G stream were above the MEPC 227(664) requirements (Fig. 8D). This shows that most of these GW streams can pollute the marine environment with P as much as inadequately treated sewage if discharged individually. This result was somewhat surprising, because all the ships reported the use of ecolabel products for handwashing, general cleaning, dishwashing, and laundry (pers. comm.). Ecolabel products are regulated and contain low P content (Supplementary material 1, Table 10). Therefore, further investigation on the dosage and chemical analysis of the P concentration in the products used on board the ships may clarify the origin of high P content in the GW streams.

Moreover, the average concentrations of N and P (11.7 mg/l and 1.23 mg/l, respectively) in the mixed A/L/G stream (n = 3) are lower

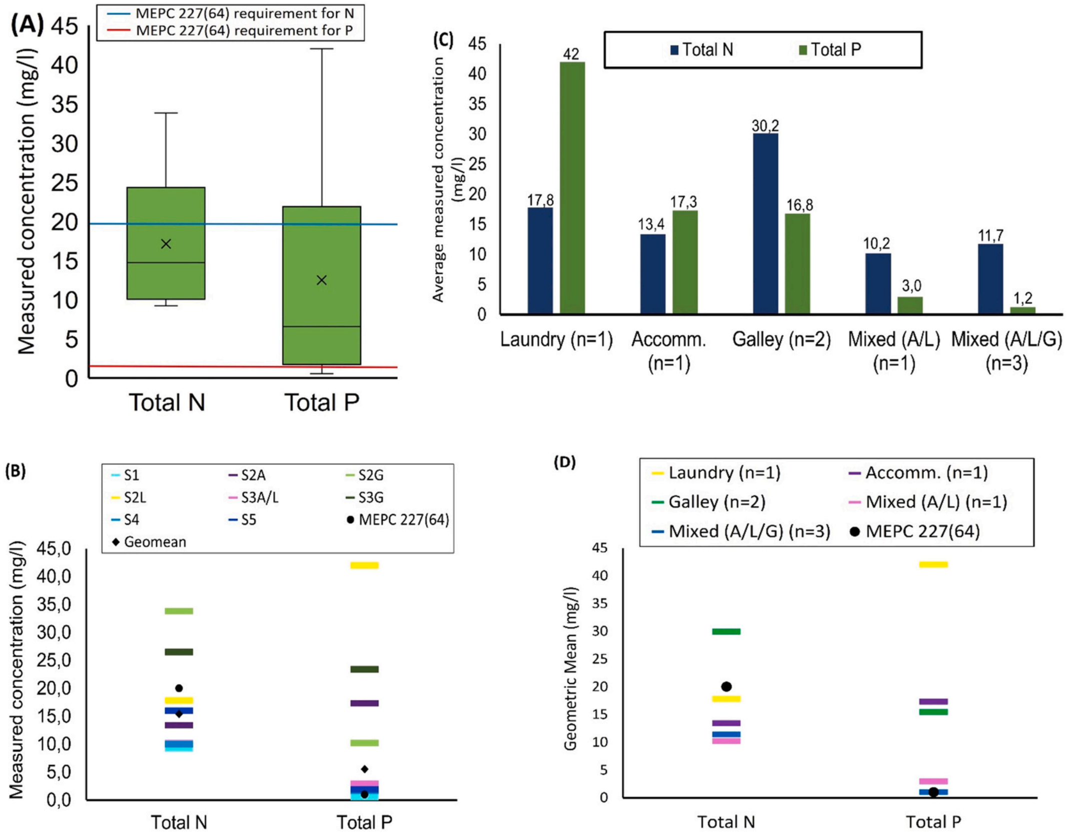

Fig. 8. (A) Variation in the concentrations of nutrients in ship-generated GW streams (B) Concentrations and geometric mean concentrations of nutrients in eight ship-generated GW streams (C) Average concentrations of nutrients in ship-generated five categories of ship-generated GW streams (D) Geometric mean concentrations of five categories of GW streams compared with MEPC 227(64) sewage effluent requirements for nutrients from advanced water treatment plants (AWTPs).

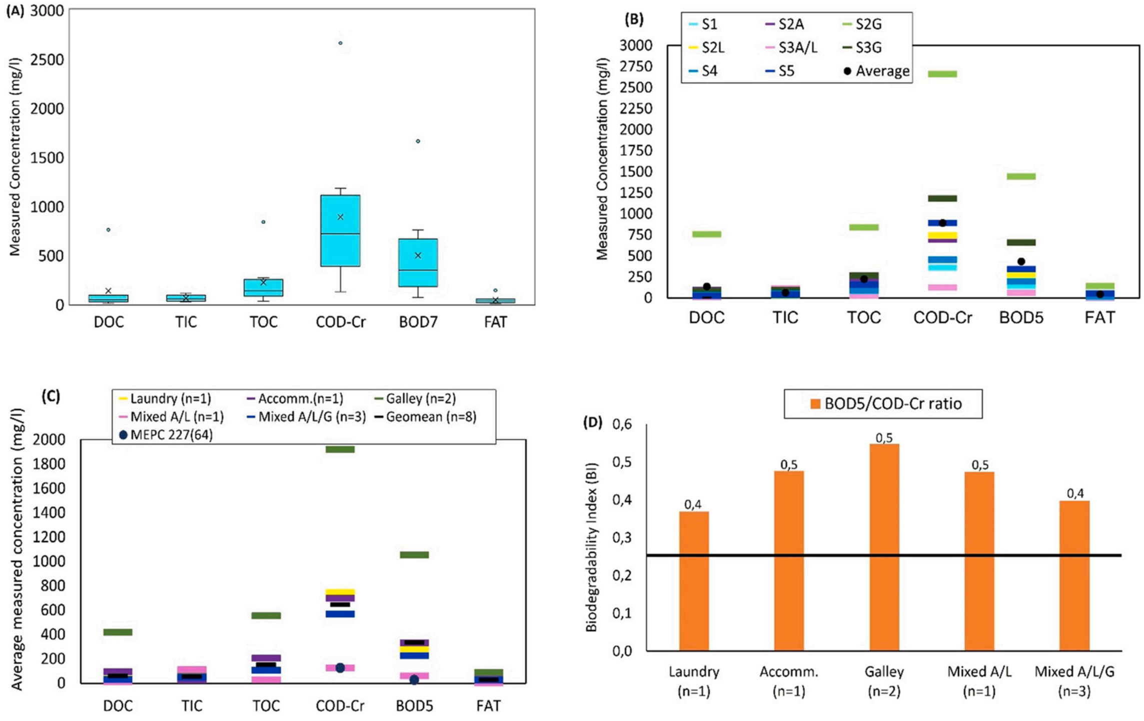

Fig. 9. (A) Variation in the OMOCS in ship-generated GW streams (B) OMOCS in eight ship-generated GW streams (C) Average OMOCS in eight ship-generated GW streams, including the geometric mean of COD-Cr and BOD_7 compared with the MEPC 227(64) sewage effluent requirements (C) Biodegradability Indexes of five categories of ship-generated GW streams expressed as BOD_5/COD-Cr ratios.

than the study by Ytreberg et al. (2020) (29 ± 100 mg/l and 4.8 ± 12 mg/l, respectively), which could possibly be explained by the difference in sample size ($n = 143$) and the ship types (cruise ships) in the latter. A comparison of N and P concentrations with other ship-based and land-based sanitary wastewater streams are presented in Supplementary material 2, Table 6. Per these comparisons, N and P were lower in mixed GW than in BW and mixed BW/GW, likely due to the absence of faeces and urine from where most nutrients in sanitary wastewater originate. For instance, approx.. 88 % of the N and 67 % of P in excreta (urine + faeces) originate from urine and the rest is from faeces (Jönsson et al., 2004). However, the variation across ship-based studies may likely be due to differences in sample sizes, ship types, ship sizes and GW management strategies such as the mixing of food waste with GW before discharge.

3.5.2. *Concentration of OMOCS*

The analyses of OMOCS showed wide variation in the concentrations of COD-Cr and BOD_5 and, to a lesser extent, TOC in the eight GW streams (Fig. 9A). Average concentrations of all OMOCS analyzed were approximately COD-Cr: 890 mg/l, BOD_5:430 mg/l, TOC:221, DOC: 140 mg/l, Fat: 42.7 mg/l and TIC: 60.3 mg/l. S2G had the highest concentrations of OMOCS, except for TIC, while S3A/L had the highest TIC but the lowest concentrations of COD-Cr, BOD_5 and FAT. COD-Cr and BOD_5 were the most dominant OMOCS, with the highest concentrations accounted for by the galley GW streams from ships S2 and S3 (approx. 2660 mg/l and 1180 mg/l for COD-Cr and approx. 1440 mg/l and 660 mg/l for BOD_5, respectively) (Fig. 9B). The geomeans of COD-Cr (approx. 640 mg/l) and BOD_5 (approx. 290 mg/l) were several times higher than the MEPC 227(64) sewage effluent requirement for COD-Cr:125 mg/l and BOD_5:25 mg/l (IMO, 2012). Average COD-Cr and BOD_5 concentrations in each of the five GW categories were several times higher than the MEPC 227(64) requirement, except for mixed A/L, whose COD-Cr concentration was the same as the MEPC 227(64) requirement (125 mg/l). (Fig. 9C). The two GW streams, S2G and S3G had the highest TOC (approx. 840 mg/l and 270 mg/l, respectively), therefore, the galley GW stream had the highest average TOC (553 mg/l) while S3A/L had the lowest (29 mg/l). Mixed A/L/G stream (n = 3) had average COD-Cr, BOD_5 and TOC concentrations of 568 ± 283 mg/l mg/l, 226 ± 103 mg/l and 107 ± 44 mg/l. In previous investigations of OMOCs in ship's mixed GW, BW and mixed BW/GW streams (Swedish Transport Agency, 2014; Holmberg, 2021; Press et al., 2020, Trelleborg port (pers. comm.) and ADEC reports) showed highest concentrations of COD-Cr and BOD_5 in mixed BW/GW (ship-based). Up to 67,000 mg/l, 44,000 mg/l and 13,000 mg/l for COD-Cr, BOD_5 and TOC, respectively, was reported on one passenger ship (Swedish Transport Agency, 2014). Comparatively, lower average concentrations (548 ± 183 mg/l, 234 ± 78.7 mg/l and 52.0 ± 21.3 mg/l for the same OMOCs were obtained in domestic wastewater from three residential areas (n = 3) in Sweden (Press et al., 2020). Supplementary material 1, Table 6 and Supplementary material 2 show average concentrations of OMOCs in various sanitary wastewater streams.

COD-Cr, BOD_5 and TOC are important parameters used to measure the biodegradability of wastewater. The effluent's biodegradability could be an issue for treatment plants that use biological treatment, and the Biodegradability Index (BI) can be used to measure the magnitude or toxicity of water pollution (Bader et al., 2022). The BI could be expressed as the COD/BOD_5 (Morel and Diener, 2006), BOD_5/COD (AikHeng and Nikraz, 2015; Papadopoulos et al., 2001) or BOD_5/TOC ratios (Akcin et al., n.d.). To achieve good biodegradability, various studies suggest that BOD_5/COD should be >0.3 (Saravanathamizhan et al., 2021), ≥0.4 (AikHeng and Nikraz, 2015) and >0.5 (Akcin et al., n.d.; Swedish Water, 2019; Samudro and Mangkoedihardjo, 2020). Akcin et al. (n.d.) suggested typical BOD_5/TOC range for municipal wastewater to be 0.3 to 0.8 but that easily biodegradable wastewater should be >0.5. Ratios below 0.3 means the wastewater may have some toxic components and its stabilization may require the use of adapted microorganisms (Akcin et al., n.d.). The BOD_5/COD ratios varied among the five GW categories and were within the range 0.4–0.5 allowing for good biodegradability, therefore should not negatively affect the operation of MWTP. The highest was seen in the galley GW, and the lowest in the laundry GW (Fig. 9E). This shows that the galley GW is more biodegradable than laundry GW and can be easily treated in the treatment plants. Similar results, BOD_5/COD: 0.34–0.63, with minimum rates in laundry and kitchen wastewater from land-based GW systems in low and middle-income countries, were obtained by Morel and Diener (2006). The corresponding BOD_5/TOC ratio for untreated wastewater is equally important and varies from 1.2 to 2.0 (Akcin et al., n.d.). In the current study, the BOD_5/TOC was between 1.6 and 2.5, indicating good biodegradability. The presence of Xenobiotic Organic Compounds (XOCs) in greywater causes COD-Cr to be dominant over BOD_5 (Oteng-Peprah et al., 2018; Noman et al., 2019). XOCs are organic compounds occurring in chemicals, pharmaceuticals, household detergents and personal care products, and are resistant to conventional treatment due to their non-biodegradable nature, hence they can leach into the marine environment through wastewater treatment plants (Noman et al., 2019). Therefore, high COD-Cr concentration in greywater is usually associated, in addition to biodegradable food waste, with the presence of surfactants from detergents and dishwashing soap which are difficult to biodegrade. High BOD_5 concentration is associated with biodegradable food particles from the kitchen (Khanam and Patidar, 2021).

3.5.3. *Concentrations of total suspended solids*

The concentrations of TSS varied widely across the eight GW streams (Fig. 10A). It ranged from 20.0 mg/l in S3A/L to 270 mg/l in S2G (Fig. 10B). The TSS geomean concentration was 102 mg/l which was several times higher than the MEPC 227(64) requirement for TSS (35 mg/l). All the five GW categories, except A/L, had TSS several times higher than the IMO requirement (Fig. 10C and D) with the galley GW stream having the highest average TSS concentration (230 ± 57.0 mg/l), followed by laundry GW stream (130 mg/l). Mixed A/L/G stream had an average TSS concentration of 114 ± 84.0 mg/l. Compared with other studies; TSS concentration in mixed GW samples from Swedish Transport Agency (2014) and Holmberg (2021) were more than twice higher, than in the mixed A/L/G in the present study. Relative to BW, Swedish Transport Agency (2014) reported average TSS concentrations of more than an order of magnitude higher, from two passenger ships and one RoRo ship. Up to 2800 mg/l was reported in one of the passenger ships. In relation to mixed BW/GW, the Trelleborg port reported average TSS concentrations from 20 RoPax ships ($n = 242$) which was close to five times higher than mixed GW in the present study. Finally, in relation to domestic wastewater which has the same sources as mixed BW/GW from ships, Gryaab (Press et al., 2020) registered TSS concentrations about thrice higher than in the mixed A/L/G in the current study. These comparisons (Supplementary material 2) show that TSS in mixed GW differ across studies likely due to the number of passengers involved, the volume of water used, the food waste management strategy on board, the sampling technique used and the pumping speed. For instance, the suspended solids in the GW may leave the tanks in beats during pumping, such that the concentration of solids may fluctuate during different sampling episodes (Madjidian and Rantanen, 2011). The differences across sanitary wastewater types are likely due to the presence or absence of faeces, or sanitary habits such as the use or non-use of tissue paper. This justifies the higher TSS in BW, mixed GW/BW and domestic wastewater than mixed GW.

The geomean of TSS higher than the MEPC 227(64) requirement reveals that GW has suspended solids in higher concentrations than authorized limits in treated sewage effluent discharged from AWTP. Given that GW is generated on board passenger ships in volumes about four times higher than BW, GW constitutes a potential hazard and its discharge into the marine environment may be perilous. Hence, practices such as the mixing of food waste with GW before discharging, a common practice on board ships operating in the Baltic Region (Kalnina

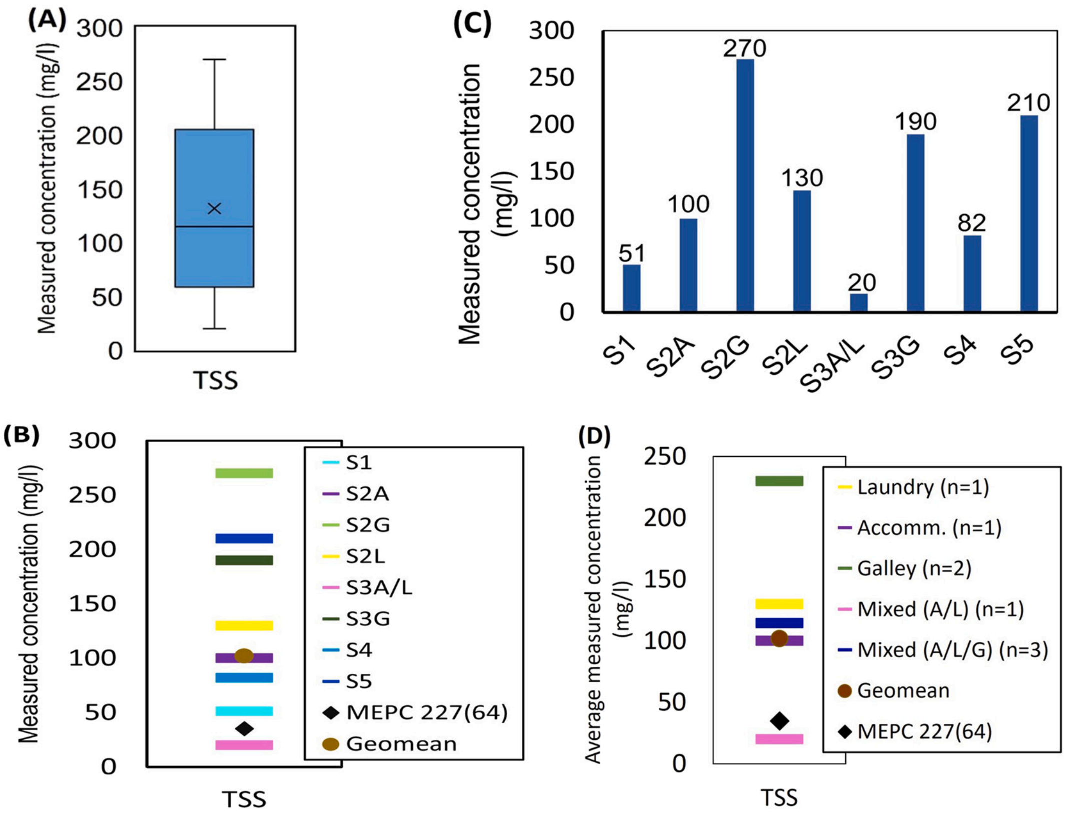

Fig. 10. (A) Variation in the TSS in ship-generated GW streams (B) TSS in eight ship-generated GW streams showing the highest levels in S2G and the lowest in S3A/L (C) TSS in eight ship-generated GW streams and the geometric mean compared with MEPC 227(64) requirement for TSS in sewage effluent (D) Average TSS in five categories of ship-generated GW and the geometric mean compared with MEPC 227(64) requirement for TSS in sewage effluent.

et al., 2021; Vaneeckhaute and Fazli, 2020) should be discouraged.

It could be deduced that N, COD-Cr, BOD_5, TOC, FATs and TSS occur in the highest concentrations in galley GW streams due to the presence of food waste. The practice of diverting food pulper and some dishwasher GW to the black water tank may have significantly reduced the OMOCS, nitrogen and TSS content of the mixed GW on ship S1 which was the lowest among the three mixed GW streams. The separation of galley GW from accommodation and laundry GW on ship S3 resulted in the lowest concentrations of N, TSS and OMOCS in S3A/L. This further confirms that food waste separation from GW plays a decisive role in reducing the organic matter, N and TSS content of GW, as such, this GW management strategy should be encouraged on board passenger ships. The impact of food waste on N and OMOCS content has been reported in similar studies. For instance, Juneau (2021) reported the results from US-EPA's 2004 studies in which food pulper and galley GW contained the highest concentrations of N, COD-Cr, BOD_5, and TSS compared with accommodation, and laundry GW. The average BOD_5 occurred, in decreasing order, in food pulper GW (30,490 mg/l), galley GW (9078 mg/l), accommodation GW (177.4 mg/l), and laundry GW (90.3 mg/l). Other measures used on some ships to reduce OMOCs include the use of grease traps (Nellesen et al., 2019). However, on ships S4 and S5, grease traps were present but faulty and non-functional resulting in these streams having the second highest average TSS concentration (146 mg/l) after galley GW.

4. Conclusion

This study developed a sampling strategy, quantified, characterized, and assessed the hazard potential of GW generated on board ships operating in the Baltic Sea. Even though the sample size was small, it is the largest available data set, for the first time resolving sub flows, for contaminant in GW in the Baltic Region. Passenger ships GW generation rates were between 49 and 138 L/person/day, and the total GW volumes generated were up to four times the volume of BW. The metals Zn, Cu, Mn and the metalloid As, collectively contributed 98 % to the HI of GW and the geomeans of COD-Cr, BOD_5, TSS and P were above the limits set by IMO resolution MEPC 227(64) guidelines. In terms of metals, there were no significant differences in HI between greywater, blackwater and mixed wastewater from ships, and domestic wastewater. All pollutants were below the limit values set by Swedish Water and local ABVAs, hence, ship-generated GW does not constitute a threat to MWTP operations. This study contributes to increased understanding about the contaminant content of ship-generated GW and showed that GW from ships presents a potential hazard to the marine environment in comparable magnitude to inadequately treated sewage. The lack of international legislation governing GW management may lead to further pollution of the Baltic marine ecosystem if GW is continuously being discharged into the sea. Moreover, the variability of pollutant types and concentrations across the GW sub-flows warrants further investigation into the feasibility of incorporating innovative technologies like source-separation systems into future ship-generated GW management strategies, which have shown great potential to recover resources while protecting the environment.

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CRediT authorship contribution statement

J.T. Mujingni: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **E. Ytreberg:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Formal analysis. **G.B.M. Rathnamali:** Writing – review & editing, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **M. Hassellöv:** Writing – review & editing, Validation, Supervision, Project administration, Funding acquisition, Formal analysis. **K. Salo:** Writing – review & editing, Validation, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Kent Salo reports financial support was provided by Swedish Agency for Marine and Water Management. If there are other authors, they 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

Data will be made available on request.

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Appendix A. Supplementary data

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