Probabilistic Human Health Risk Assessment of Trace Elements in Ballast Water Treated by Reverse Osmosis Desalination Plants

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Abstract:

Very few studies have paid attention to the transport of heavy and toxic metals via ballast water coming from different countries of the world. In the present study, ballast water samples (n=83) were collected from ships, tankers and vessels of 21 different origins arriving at the two main ports of Qatar. Besides the basic physical parameters of pH, electrical conductivity (EC), and total organic carbon (TOC), concentrations of 24 elements (As, Sb, Al, Cd, Pb, Si, V, Ag, Zn, Cr, Mn, Ba, Co, Ni, Sr, Be, Cu, Tl, B, Fe, Se, Sn, Mo and U) were determined. In addition, the potential human health risks of drinking water treated by reverse osmosis (RO) were assessed using Monte Carlo simulations. Two scenarios were used to assess the risks to the general population, namely, seawater (baseline) and ballast water (worst-case scenario). Our results show significant differences among the tested elements, depending on the origin of the ballast water. The human health assessment showed that all hazardous quotients (HQs) were below the safety limits. However, for the ballast water scenario, thallium (Tl) HQs were 10% above the safety level. Ballast water in Qatar does not pose risks for human health through drinking water, but ballast water discharges should take into consideration seawater catchments and potential toxic elements, especially Tl. Regular monitoring campaigns need to be performed.

Keywords: Arabian Gulf; Ballast water; Health risks; Monte Carlo simulation; Reverse osmosis desalination.

1. Introduction:

Almost two thirds of all goods traded worldwide are transported by ships (Kumar and Hoffmann, 2002), and to ensure ship buoyancy, stability and maneuverability, oceangoing ships need to have ballast water. At a global scale, water transfer by maritime traffic is estimated to be about five billion tons annually (Tsolaki and Diamadopoulos, 2010). Consequently, thousands of species get transported through ballast water each day and unwanted introduction of approximately 450 marine and estuarine non-invasive species (NIS) have been documented in North America (Bailey, 2015; Ruiz et al., 2000; Tsolaki and Diamadopoulos, 2010).

The ongoing war between Russia and the Ukraine has created many global challenges, including the crises of food (FAO, 2022), energy (UN, 2022), industry (OECD, 2022) and trade, in general (World Economic Forum, 2022). The European countries, in particular, have sought to quickly search for alternatives to energy (European Council, 2022) from many markets (European Commission, 2022), the most important of which are the Arab Gulf states (European Union, 2022).

The movement of ships has doubled during 2022 (IMO, 2022), bringing thousands of tons of ballast water to the already extremely fragile ecosystem in the Arabian Gulf (Tolian et al., 2020), as it constitutes the stage for all industrial and commercial operations in the region. The Gulf is the only source of water for desalination plants (Rashkeev and Shomar, 2020) in most of the Arabian Gulf countries, considered to be among the poorest in fresh water resources in the world (Shomar et al., 2014). Therefore, they resort to desalination operations as a strategic basis to meet the increasing need for fresh water in all sectors, especially agriculture, industry and daily human use (Ajjur and Al-Ghamdi, 2022).

Ships coming to the Gulf with ballast water bring along a cocktail of pollutants (Sayinli et al., 2022), including heavy metals (Tjahjono et al., 2016). These metals accumulate in the water of the Gulf (Tolian et al., 2020), and end up in the sediments (Amoresano et al., 2022) or in the food chain (Lim et al., 2022).

Although many studies on ballast water concentrate on the microbial invasions and their effects (Bailey, 2015; Tsolaki and Diamadopoulos, 2010; Ruiz et al., 2000), a few authors have paid attention to the transport of heavy and toxic metals through ballast water coming from different countries of the world (Tolian et al., 2020; Tjahjono et al., 2016), where laws, control systems and treatment technologies vary (Sayinli et al., 2022). Also, very few studies have covered the negative impact of heavy metals in ballast water discharge on the marine ecosystem (Tolian et al., 2020), especially the effect of heavy metals on water, sediment and the food chain (Lim et al., 2022).

Recognition of the ecological, socio-economic and public health impact of invasive species and toxic metals has led to the establishment of various guidelines and legal instruments aimed at mitigating the risks posed by global vessel traffic. Among such instruments, the most widely recognized and used is the Ballast Water Management Convention (BWMC) adopted by the International Maritime Organization (IMO) in 2004. This framework was ratified by ~75% of the global fleet by tonnage by September 2017 (IMO, 2022). In the United States, similar numerical standards were adopted in 2012 in the final ballast water (BW) rules of the US Coast Guard (USCG) and in the US Environmental Protection Agency's (USEPA) 2013 Vessel General Permit. Qatar became a full member of the International Maritime Organization (IMO) Convention in May 2018.

Strategic decisions have been made in all of the Arabian Gulf countries to transition to Reverse Osmosis (RO) desalination technology (MEDRC, 2020). Pretreatment of seawater for RO represents more than 60% of the total cost of water production (Darwish et al., 2016). Design, operation and maintenance of RO desalination plants depend mainly on the quality of feed water (Darwish et al., 2013). Pollution load of feed water is very critical in the fragile marine system. Adding loads of contaminants, including trace metals, may result in long-term challenges for the existing RO plants and the ones to come (Ahmed et al., 2022). Additionally, the rejection rate of trace metals by membranes used in the RO technology may deteriorate the membranes faster through the escalating loads coming from several sources such as ballast water (Yousif et al., 2022).

Through this study, we provide, for the first time, a two-year monitoring of ballast water for many commercial vessels arriving at Qatar's ports; especially those specialized in transporting oil, gas and petrochemicals, among other goods. We focus on 21 origins sourcing these vessels. As the human population continues to grow, increased arrival of non-indigenous species and toxic chemicals is anticipated, due to increased eutrophication, shipping traffic, and stressors associated with ocean warming. In spite of the regional vulnerabilities, there have been very few assessments of ballast water going into the region for toxic metals and metalloids that potentially pose a threat to local ecosystems. A goal of this study is to fill this critical gap in knowledge. This study is also important from the public-health perspective. The study addresses the chemistry of ballast water from ships sailing to Qatar from different parts of the world. It investigates the chemistry of ballast water disposed of in the Qatari marine system compared to other locations in the world. Additionally, it describes the scenarios of ballast water affecting reverse osmosis desalination plants, over the short and long terms, and finally it introduces data that can be used by scientists and decision makers on effective measures recommended by the IMO in reducing the seawater pollution and monitoring strategies.

The main objectives of the study are: (i) to assess ballast water quality for physical and chemical parameters; (ii) to investigate variation in the characterization of ballast water based on origin; and (iii) to assess the potential human health risks of trace elements after treatment with RO desalination plants where ballast water is the source of feed water.

2. Methodology:

2.1. Sampling campaigns and data collection

Collection of ballast water samples was facilitated by trained personnel from Qatar Energy who provided the project with sterilized and acid-washed containers. Samples were collected from ships at the two main ports of Qatar, namely, Mesaieed and Ras Laffan. During sample collection, the physical-chemical parameters were measured using portable kits. Basic information for each ship has been recorded as well as date and time. The team collected ballast water samples from 86 ships over two years. Sampling campaigns were designed to include: (i) ships and tankers from different parts of the world taking different travel times to reach Qatar; (ii) vessels that have treated their ballast water and those that have not because they rely on ballast water exchange (BWE) or they are from areas determined to be low-risk; (iii) different treatment methods of ballast water; and (iv) treated and untreated water from the same ship.

According to the information provided by the port authorities, the total number of the collected samples was distributed according to the following origins: 8 samples from Singapore, 8 samples from India, 4 samples from Taiwan, 8 samples from Japan, 2 samples from South Korea, samples from China, 2 samples from Egypt, 3 samples from Oman, 10 samples from the UAE, 19 samples from the Arabian Sea, 5 samples from open sea, 2 samples from The Mediterranean Sea, 2 samples from the Indian Ocean, 1 sample from Kenya, 1 sample from Bahrain, 1 sample from Kuwait, 1 sample from Iran, 1 sample from Iraq, 1 sample from Sri Lanka, 1 sample from Australia, and finally, 1 sample from Philippines.

Seawater samples were collected at a depth of 20-40 cm in the accessible territories opposite the industrial areas and desalination plants. Samples were treated in the same way as ballast water samples. The following aliquots have been collected at each sampling outlet: (1) sample for trace metals, which was acidified after collection; (2) sample for total organic carbon; (3) sample for measurement of physical-chemical parameters, which were taken on site, and (4) sample for quality assurance/quality control (QA/QC) needs.

Samples were labeled and submitted to the lab immediately after collection, to be properly processed and/or preserved (Kuiper et al., 2014; Rowell et al., 2014). The following parameters have been measured on site using portable instruments: pH, total dissolved solids (TDS), electrical conductivity (EC), and chloride concentration (with specific electrode).

The following additional information collected in the routine monitoring program were obtained from Qatar Energy: the origin of ballast water, the type of tanker/vessel, the date of ballast water upload, the volume of ballast water onboard, and the specific location of ballast water collection from each ship.

2.2. Analysis of trace metals in ballast water

For analysis of the trace elements, we followed the established procedure used in our previous studies utilizing state-of-the-art laboratory infrastructure, including an ICP-MS, and the highest level of Quality Control and Quality Assurance. The protocols highlighted the use of metal-free plastic ware, metal-free acids, certified reference materials and chemicals of ultrapure grade (Karami et al., 2021; Nriagu et al., 2018; Shomar et al., 2020; Shraim et al., 2013).

Instrumentation, reagents and quality control

Glassware and plastic ware were soaked in 10% HNO_3 overnight, rinsed with deionized water (dH_2O) (resistivity 18.2 Ω.cm) obtained from "Elga PURELAB Chorus, High Wycombe UK" purification system. This dH_2O was used in the preparation of blanks, calibration standards, and sample dilution. Nitric acid (67% analytical grade for trace metal analysis) was obtained from VWR Chemicals International (Karami et al., 2021).

The inductively coupled plasma mass spectrometry (ICP-MS) (PerkinElmer NexION 300D) was used to determine the total concentrations of As, Sb, Al, Cd, Pb, Si, V, Ag, Zn, Cr, Mn, Ba, Co, Ni, Sr, Be, Cu, Tl, B, Fe, Se, Sn, Mo and U. The instrument was operated using the Kinetic Energy Discrimination (KED) collision mode with He as a non-reactive gas. The ICP-MS operating parameters are summarized in (Table 2.S). Details of the ICP-MS operating conditions are given in Shraim et al. (2022). To go below the normal detection limit of the instrument, the Ultrasonic Nebulizer unit was connected to it. For ICP-MS, the QC protocol of Saghafi et al. (2021); Shraim et al. (2022) was used.

Analytical blanks and four samples with known concentrations of trace metals were prepared and analyzed using the same procedures and reagents (Kiani et al., 2022). Standard reference materials (SRM) from the National Institute of Standards and Technology-USA (NIST: 1643c and 1643d) were used in addition to SRM from the Spectra Pure-Norway (SPS: SW2 Batch 122 and NUTR Batch 106) for the determination of trace elements (NIST, 2010, SPS, 2010). The ICP-MS calibration solutions (0.01 - 200 µg/L) were prepared using serial dilutions of a 10 mg/L multi-element stock standard solution (ICP-MS Calibration Standard 2, AccuStandard, New Haven, USA). The remaining calibration solutions were prepared from the stock solution using simple dilutions. All calibration solutions were made using dH_2O water containing 5% HNO_3.

The above-mentioned characteristics were preceded by initial calibration validation through multi-point calibration curves, initial calibration verification (ICV), and continuous calibration verification (CCV). Out of the 14 calibration solutions prepared, seven calibration points were considered for each element, taking into account the variations in elemental concentrations among samples (Shraim et al., 2022). The results of all method performance characteristics were found to be within acceptable limits. The Linearity of the calibration curves was assessed based on the coefficient of determination (R^2). Highly linear curves were obtained for all elements with R^2 values greater than 0.999.

For assessing the quality of the analytical method, the two reference materials from NIST and SPS were used. Additionally, both the ICV and CCV were assessed using a 10.0 µg/L standard solution that was analyzed 7 times. Analysis of the CCV solution was repeated after every ten ICP-MS runs and used as an indicator for the stability of the ICP-MS (Shraim et al., 2022). The obtained average concentrations for all elements were within a narrow range of 0.1-0.5 µg/L with a percent relative standard deviation (RSD(%)) between 2.1 and 5%. These results are within the acceptable criteria for field samples (+/-20%) (Granato et al., 1998).

2.3. Human health risk assessment of ballast water

Non-occupational exposure through direct consumption and dermal contact with ballast water is improbable. However, seawater intake territories of reverse osmosis desalination plants include the inputs of ballast water mixed with seawater. Two different scenarios were built depending on the source of water intake of the RO plan and finished (desalinated) drinking water. First, seawater quality with trace elements determined in 2019 as a baseline scenario, and second, a worst-case scenario for trace elements in ballast water of this study (Loganathan et al., 2019). Due to the high number of ships operating in the area, the low removal rate of seawater in the Arabian Gulf and the unknown distance between water catchment and ballast water discharge points, the worst-case scenario, considering all inputs of RO desalination plants, the ballast water discharge was assumed.

Metal intake through drinking desalinated water and hazardous quotient (HQ) were assessed according to equations (1) and (2), respectively. Statistical analysis were performed using Monte Carlo simulation (Oracle Crystal Ball© software, version 11.1.2.4.850). Calculations were based on the propagation of parameters' variability and uncertainty given by each parameter probability function up to 100,000 iterations (Kiani et al., 2022; Shariatifar et al., 2020; Martinez et al., 2018; Rovira et al., 2016).

$$Intake_i = C_i \cdot Dr \cdot (1 - Rr_i)/BW \qquad (1)$$

$$HQ_i = Intake_i/RfD_{o,i} \qquad (2)$$

where C_i is the concentration of element *i*, *Dr* is the daily rate of drinking water consumption; Rr_i is the removal rate of the element *i* in *RO* desalination plants; *BW* is the adult body weight and $RfD_{o,i}$ is the oral reference dose of element *i*. Parameters used were shown in Table (1). Elements intake above their respective oral reference dose (RfD) means an HQ above unity and implies a health impact for population.

Table 1. Parameters, values and distribution used in probabilistic human health risk assessment through drinking

	Parameter	**Value**	**Distribution**	**Reference**
Ci	Concentration of element in water	-	Log Normal	Present study
Dr	Daily rate consumption of drinking water	0.5-2 L	Uniform	Assumed
Rri	Removal rate of element in RO desalination plants	0.990-0.999	Uniform	Bolisetty et al., 2019
BW	Body weight	70 (107) Kg	Log-Normal	US EPA, 2011
RfDoi	Oral reference dose		Punctual	US EPA, 2022a

Median (Percentile 95th) Log Normal distribution; Minim-maximum for uniform distribution

3. Results and Discussion

3.1. Ballast water quality and origin

Table (2) shows the results of seawater concentrations, to be compared with Table (3) showing comprehensive description of the quality of ballast water of the collected samples from the 21 origins. In brief, ballast water of all samples tends to be alkaline, where the average pH is 8, while that of seawater in Qatar is 7. The average electrical conductivity of ballast water of collected samples is 56 mS/Cm. However, many samples showed results above 60 mS/Cm, which is higher than the current EC values of the seawater of Qatar. Such high salinity input may affect the whole marine system on the long term. pH values of ballast water show the same trend as EC; where more alkalinity of seawater on the long term may affect the ecosystem.

Table 2. Seawater quality in the areas of major ports of Qatar

Seawater of Qatar		**2019**	**2019**	**Average 2022**
		Ras Laffan (n=5)	**Mesaieed (n=5)**	**(n=2)**
Temp	(°C)	28.5	26	26
pH		7-8.1	7.2	7
EC	(mS/Cm)	55-65	57-66	55
TOC	**(mg/L)**	1.56	2.03	1.52
As	(µg/L)	4.4	3.5	3.5
Sb	(µg/L)	0.001	7.6	0.001
Al	(µg/L)	0.001	3.7	0.001
Cd	(µg/L)	0.1	0.001	0.001
Pb	(µg/L)	1.42	4.01	1.44
Si	(µg/L)	60	195	61
V	(µg/L)	0.001	1.31	0.001
Ag	(µg/L)	0.601	0.21	0.21
Zn	(µg/L)	16	6.01	6.11
Cr	(µg/L)	0.301	0.21	0.2
Mn	(µg/L)	0.92	0.61	0.63
Ba	(µg/L)	9.321	12.51	9.33
Co	(µg/L)	1.71	1.51	1.52
Ni	(µg/L)	0.001	0.001	0.001
Sr	(µg/L)	9800	12050	9820
Be	(µg/L)	0.112	0.101	0.102
Cu	(µg/L)	3.21	4.21	3.23
Tl	(µg/L)	0.52	2.21	0.55
B	**(mg/L)**	5.52	6.54	5.51
Fe	**(mg/L)**	0.001	0.001	0.001
Se	(µg/L)	12.3	11	10
Sn	(µg/L)	0.001	1.7	0.001
Mo	(µg/L)	7.2	9.1	7.4
U	(µg/L)	1.1	2.3	1.4

Table 3. Chemical characteristics of ballast water samples collected from 83 ships and tankers coming from 21 origins

		Singapore (n=8)			**India (n=8)**			**Taiwan (n=4)**			**Japan (n=8)**			**South Korea (n=2)**		
		Min	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**
pH	**Unit**	7.9	8.4	8.3	8.1	8.4	8.2	8.1	8.4	8.2	8.0	8.2	8.2	7.4	8.4	7.9
EC	(mS/Cm)	46.7	59.7	52.9	50.7	61.6	54.3	48.5	63.8	56.9	47.1	53.9	50.4	49.8	57.6	53.7
TDS	(g/L)	33.8	43.2	38.3	36.7	44.6	39.3	35.1	46.2	41.2	34.1	39.0	36.5	36.1	41.7	38.9
TOC	(mg/L)	8.6	10.5	9.6	6.5	10.7	9.4	8.5	11.0	10.1	6.1	9.8	8.2	9.0	12.1	10.5
TC	(mg/L)	26.9	30.7	28.9	25.3	31.6	29.2	26.3	31.9	29.8	23.8	28.7	26.7	27.3	32.5	29.9
IC	(mg/L)	18.1	20.3	19.3	18.8	20.9	19.8	17.8	20.9	19.7	17.7	19.5	18.5	18.3	20.4	19.4
As	(µg/L)	0.5	1.9	1.4	1.1	4.2	2.9	0.9	3.1	1.7	0.3	3.7	1.8	1.3	2.2	1.8
Sb	(µg/L)	2.6	25.0	8.8	4.0	26.0	13.2	12.0	20.0	15.8	8.0	48.0	21.4	8.0	24.0	16.0
Al	(µg/L)	8.0	953	231	11	608	163	15	36	24	5	42	22	71	171	121
Cd	(µg/L)	1.0	3.1	2.2	1.0	14.0	3.9	1.8	2.0	1.9	2.0	3.0	2.2	1.0	2.1	1.6
Pb	(µg/L)	8.0	445	84	5	109	40	6	57	25	13	391	77	47	55	51
Si	(µg/L)	6.0	250	63	3	694	209	4	157	62	5	99	55	10	67	39
V	(µg/L)	3.0	26	9	4	23	13	4	27	13	2	24	18	3	14	9
Ag	(µg/L)	8.0	20	12	8	21	14	9	12	10	7	20	14	11	17	14
Zn	(µg/L)	2.5	291	71	3	2017	687	31	602	262	8	1129	547	293	688	491
Cr	(µg/L)	1.7	6.0	3.3	1.9	15.0	5.9	2.1	7.0	4.3	1.7	9.0	5.0	2.5	6.0	4.2
Mn	(µg/L)	0.4	5.0	1.6	0.9	5.0	3.0	0.5	1.9	1.1	1.0	2.9	1.7	1.3	1.3	1.3
Ba	(µg/L)	1.0	6.9	4.0	1.0	27.0	8.8	2.0	6.2	4.6	2.0	3.5	2.2	1.0	5.0	3.0
Co	(µg/L)	3.0	10.3	7.8	5.0	11.0	8.0	7.0	11.0	9.1	6.0	10.0	8.4	7.0	12.0	9.5
Ni	(µg/L)	0.4	43	9	1	44	16	2	53	16	5	50	21	2	10	6
Sr	(µg/L)	4132	7594	5850	4494	7512	5474	5323	6826	6068	4120	5338	4875	4412	5006	4709
Be	(µg/L)	0.2	0.4	0.3	0.2	0.4	0.3	0.2	0.4	0.3	0.2	0.4	0.3	0.2	0.3	0.3
Cu	(µg/L)	0.9	219	62	1	138	85	9	441	168	21	870	204	120	192	156
Tl	(µg/L)	11.0	26	19	7	110	30	13	32	22	0	43	18	17	19	18
B	**(mg/L)**	1.4	4.0	2.4	1.9	3.0	2.5	2.9	3.0	2.9	1.6	2.5	2.3	2.1	2.4	2.3

Fe	**(mg/L)**	2.3	4.7	3.5	2.3	3.9	3.0	3.2	4.7	3.9	2.3	3.6	2.8	3.2	3.2	3.2
Se	(µg/L)	0.4	0.8	0.6	0.2	1.0	0.6	0.5	0.9	0.6	0.4	0.8	0.5	0.2	0.5	0.3
Sn	(µg/L)	0.6	20.0	5.9	0.8	10.0	3.7	5.0	12.0	7.3	2.0	15.0	6.4	0.2	0.7	0.4
Mo	(µg/L)	5.7	14.8	9.9	6.0	22.4	14.6	7.3	28.7	15.3	5.5	24.0	14.5	6.6	12.4	9.5
U	(µg/L)	0.9	4.0	1.9	0.8	5.5	2.6	0.8	6.9	2.6	0.1	3.8	1.7	0.8	1.6	1.2

TC: Total carbon; IC: Inorganic carbon.

Table 3. Continued

		China (n=2)			Egypt (n=2)			Oman (n=3)			UAE (n=10)			Arabian Sea (n=19)		
		Min	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**
pH	**Unit**	8.2	8.3	8.2	8.2	8.4	8.3	7.7	8.4	8.1	7.7	8.8	8.2	7.6	8.3	8.1
EC	(mS/Cm)	48	50	49	60.1	61.1	60.6	52.6	60.5	55.7	49.8	61.1	56	45.5	64.6	55.2
TDS	(g/L)	34.9	36.2	35.5	43.5	44.2	43.9	38.1	43.8	40.3	36.1	44.2	40.5	32.9	46.8	40.0
TOC	(mg/L)	8.7	8.8	8.7	6.5	10.3	8.4	9.5	11.8	10.6	6.7	12.2	9.1	6.2	30.5	10.1
TC	(mg/L)	26.6	26.8	26.7	28.6	30.7	29.7	29.3	31.4	30.5	16.2	34.2	27.8	24.9	51.0	29.8
IC	(mg/L)	17.9	18.0	18.0	20.4	22.1	21.3	19.7	20.2	19.9	9.5	22.0	18.7	18.1	20.9	19.6
As	(µg/L)	1.4	2.5	2.0	1.1	1.6	1.3	1.5	3.1	2.6	1.2	2.9	2.3	0.2	4.4	2.4
Sb	(µg/L)	12.0	12.0	12.0	3.3	10.9	7.1	9.4	17.0	14.1	3.0	27	14.9	5.0	33.0	17.3
Al	(µg/L)	69	69	69	32.0	88	60	5.0	268	97.3	5.0	608	114.0	7.0	953	101
Cd	(µg/L)	2.0	2.0	2.0	1.3	2.0	1.6	1.8	3.0	2.3	2.0	7.0	2.9	1.0	3.0	2.1
Pb	(µg/L)	11	48	30	19.0	23.0	21.0	17.0	17.0	17.0	5.0	134	39.7	5.0	66.0	33.8
Si	(µg/L)	52	121	87	10.0	10.0	10.0	13.0	35.0	24.3	13.0	265	106.8	6.0	220.0	77.6
V	(µg/L)	11	16	14	3.2	4.1	3.7	4.7	25.0	15.9	10.0	27.0	18.4	13.0	25.0	19.6
Ag	(µg/L)	13	19	16	5.0	7.0	6.0	6.0	18.0	13.0	7.0	18.0	12.3	5.0	20.0	15.1
Zn	(µg/L)	148	417	283	0.8	1.9	1.3	2.0	1052	527	34	2574	601	6	5803	598
Cr	(µg/L)	6.0	7.0	6.5	1.8	2.1	1.9	2.0	6.0	4.0	1.0	9.0	5.6	2.0	11.0	6.1
Mn	(µg/L)	3.0	3.0	3.0	0.5	72.6	36.5	1.0	5.0	3.0	1.0	24.0	7.1	1.0	12.0	4.4
Ba	(µg/L)	2.0	2.0	2.0	5.2	6.9	6.0	2.0	6.6	4.9	1.0	69.0	11.3	1.0	12.0	4.1
Co	(µg/L)	9.0	11.0	10.0	11.3	13.2	12.3	6.4	9.0	7.8	6.0	12.0	9.4	7.0	30.0	11.1
Ni	(µg/L)	12	43	28	0.3	12.8	6.5	0.4	17.0	9.8	11.0	62.0	38.6	9.0	111.0	31.2
Sr	(µg/L)	4861	5003	4932	6600	7260	6930	5306	7016	5922	3727	6198	5481	4829	6378	5472
Be	(µg/L)	0.2	0.3	0.3	0.4	0.4	0.4	0.4	0.4	0.4	0.2	0.4	0.3	0.2	0.4	0.3
Cu	(µg/L)	91	116	104	0.3	1.7	1.0	0.8	127.0	81.3	95.0	262.0	146.6	83.0	178.0	126.5
Tl	(µg/L)	9	15	12	19.0	28.0	23.5	26.0	33.0	29.3	5.0	57.0	27.5	8.0	71.0	26.3

B	**(mg/L)**	2.1	2.3	2.2	1.5	2.1	1.8	2.5	2.6	2.5	1.9	3.0	2.5	2.0	3.0	2.5
Fe	**(mg/L)**	2.3	2.4	2.3	2.9	3.2	3.1	2.3	3.1	2.7	2.4	3.9	3.1	2.3	4.7	3.4
Se	(µg/L)	0.7	0.9	0.8	0.6	0.7	0.7	0.3	0.7	0.6	0.2	1.1	0.7	0.3	1.3	0.7
Sn	(µg/L)	2.0	2.0	2.0	5.0	6.0	5.5	2.0	6.0	3.8	0.3	30.0	10.4	1.0	21.0	7.6
Mo	(µg/L)	12.1	21.1	16.6	7.9	8.5	8.2	8.4	11.0	10.1	10.3	29.8	22.1	9.5	25.9	17.9
U	(µg/L)	3.4	4.1	3.7	1.6	1.8	1.7	1.7	3.8	2.5	0.0	3.7	1.8	0.7	4.1	2.4

Table 3. Continued

		Open SEA (n=5)			**Mediterranean Sea (n=2)**			**Indian Ocean (n=2)**			**Kenya**	**Bahrain**	**Kuwait**	**Iran**	**Iraq**	**Sri Lanka**	**Australia**	**Philippines**
		Min	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**n=1**	**n=1**	**n=1**	**n=1**	**n=1**	**n=1**	**n=1**	**n=1**
pH	**Unit**	8.1	8.3	8.2	7.9	8.0	7.9	8.0	8.2	8.1	8.4	7.2	8.0	8.1	8.2	8.3	8.1	8.2
EC	(mS/Cm)	49.1	65.7	53.9	25.5	53.0	39.3	54.0	65.6	59.8	59.4	58.8	60.2	58.4	66.2	52.3	51.8	61.7
TDS	(g/L)	35.5	47.6	39.0	18.5	38.4	28.4	39.1	47.5	43.3	43.0	42.6	43.6	42.3	47.9	37.9	37.5	44.7
TOC	(mg/L)	8.4	10.7	9.4	7.6	8.9	8.2	9.3	10.6	10.0	10.6	1.9	9.8	77.1	11.5	6.9	8.8	9.4
TC	(mg/L)	26.1	30.7	28.2	26.7	29.2	28.0	29.0	30.0	29.5	30.7	13.6	30.0	97.6	28.9	25.5	27.8	27.7
IC	(mg/L)	17.6	20.1	18.8	19.2	20.3	19.7	19.4	19.7	19.6	20.1	11.7	20.3	20.5	17.4	18.6	19.0	18.3
As	(µg/L)	0.7	4.2	2.4	1.6	2.4	2.0	2.3	3.1	2.7	1.9	1.9	1.4	3.8	0.6	3.2	1.2	3.7
Sb	(µg/L)	1.0	28.0	9.0	9.0	21.0	15.0	4.0	9.0	6.5	3.8	<0.005	14.0	18.0	12.0	23.0	12.0	14.0
Al	(µg/L)	22.0	62.0	36.0	22.0	22.0	22.0	8.0	463.0	235.5	62.0	<0.005	<0.005	13.0	48.0	18.0	227.0	<0.005
Cd	(µg/L)	1.0	3.0	1.6	2.0	3.0	2.5	2.0	2.0	2.0	1.6	3.0	2.0	3.0	5.0	4.0	3.0	3.0
Pb	(µg/L)	7.0	50.0	27.3	19.0	74.0	46.5	21.0	37.0	29.0	9.0	12.0	101.0	63.0	40.0	34.0	77.0	60.0
Si	(µg/L)	8.0	157	60	37	1542	790	24.0	61.0	42.5	59.0	68	27	<0.005	269	199	78	15
V	(µg/L)	16.0	23.0	19.4	14.0	19.0	16.5	19.0	58.0	38.5	4.8	13.0	20.0	28.0	22.0	28.0	23.0	20.0
Ag	(µg/L)	8.0	15.0	11.2	11.0	25.0	18.0	9.0	17.0	13.0	7.0	10.0	14.0	7.0	12.0	15.0	9.0	19.0
Zn	(µg/L)	6	2085	444	17	17	17	3.0	499.0	251.0	9.7	5146	<0.005	10	7135	416	377	28
Cr	(µg/L)	4.0	8.0	5.6	2.0	7.0	4.5	7.0	8.0	7.5	2.4	5.0	8.0	7.0	6.0	4.0	5.0	6.0
Mn	(µg/L)	1.0	321.0	83.5	1.0	1.0	1.0	6.0	11.0	8.5	1.1	590.0	57.0	2.0	100.0	2.0	2.0	1.0
Ba	(µg/L)	2.0	5.0	2.8	24.0	24.0	24.0	2.0	6.0	4.0	6.9	20.0	6.0	4.0	32.0	4.0	<0.005	4.0
Co	(µg/L)	10.0	16.0	11.6	8.0	12.0	10.0	8.0	11.0	9.5	9.1	13.0	9.0	10.0	8.0	9.0	11.0	9.0
Ni	(µg/L)	39.0	1615.0	358.2	18.0	20.0	19.0	45.0	47.0	46.0	2.6	49	9	20	25	16	20	45
Sr	(µg/L)	4553	6283	5135	2513	5103	3808	5593	6271	5932	7390	5103	5913	5963	6355	5354	5504	5703
Be	(µg/L)	0.2	0.4	0.3	0.4	0.4	0.4	0.2	0.3	0.3	0.4	0.4	0.4	0.3	0.4	0.3	0.2	0.4
Cu	(µg/L)	122.0	165.0	136.6	112.0	116.0	114.0	116.0	136.0	126.0	2.1	121.0	125	130	140	125	141	148

Tl	(µg/L)	11.0	46.0	27.8	26.0	42.0	34.0	3.0	14.0	8.5	30.0	39.0	0.0	50.0	36.0	0.0	40.0	18.0
B	**(mg/L)**	2.0	2.9	2.3	1.1	2.4	1.8	2.5	2.8	2.6	2.9	2.3	2.7	2.7	3.0	2.4	2.5	2.6
Fe	**(mg/L)**	3.1	4.0	3.5	2.7	2.8	2.8	2.7	4.7	3.7	3.6	2.8	4.0	3.2	3.1	2.9	3.7	2.3
Se	(µg/L)	0.3	1.1	0.7	0.5	0.9	0.7	0.4	0.9	0.6	0.8	0.8	1.0	0.5	0.7	0.7	0.7	0.6
Sn	(µg/L)	3.0	6.0	4.2	2.0	16.0	9.0	1.0	5.0	3.0	9.0	8.0	4.0	4.0	40.0	1.0	5.0	6.0
Mo	(µg/L)	20.3	29.3	23.3	6.4	9.3	7.8	24.9	27.7	26.3	9.1	22.5	13.7	13.4	16.0	17.6	11.0	25.9
U	(µg/L)	0.5	7.2	3.3	1.5	6.5	4.0	0.0	4.9	2.4	1.7	2.7	0.0	3.8	2.8	1.9	1.8	1.6

Because thallium (Tl) shows the higher levels of HQ, it was selected to show the difference among the 21 origins. Figure (1) shows the variation of Tl average concentration. The lowest concentration was found in samples originating in Kuwait and Sri Lanka (0.5 µg/L) and the highest was found in the samples coming from Iran (50 µg/L). More importantly, the average concentration of Tl in the samples of Qatar seawater was 4 µg/L, of the same level as those found in several locations in the Atlantic and Pacific Oceans (Flegal and Patterson, 1985). In the comprehensive study of Rehkämper and Nielsen (2004), five main sources are behind the occurrence of Tl in seawater: rivers, hydrothermal fluids, eolian contributions from volcanic eruptions, mineral aerosols, and benthic fluxes from continental margin sediments. The potential sources of Tl in the seawater of Qatar may need further investigation.

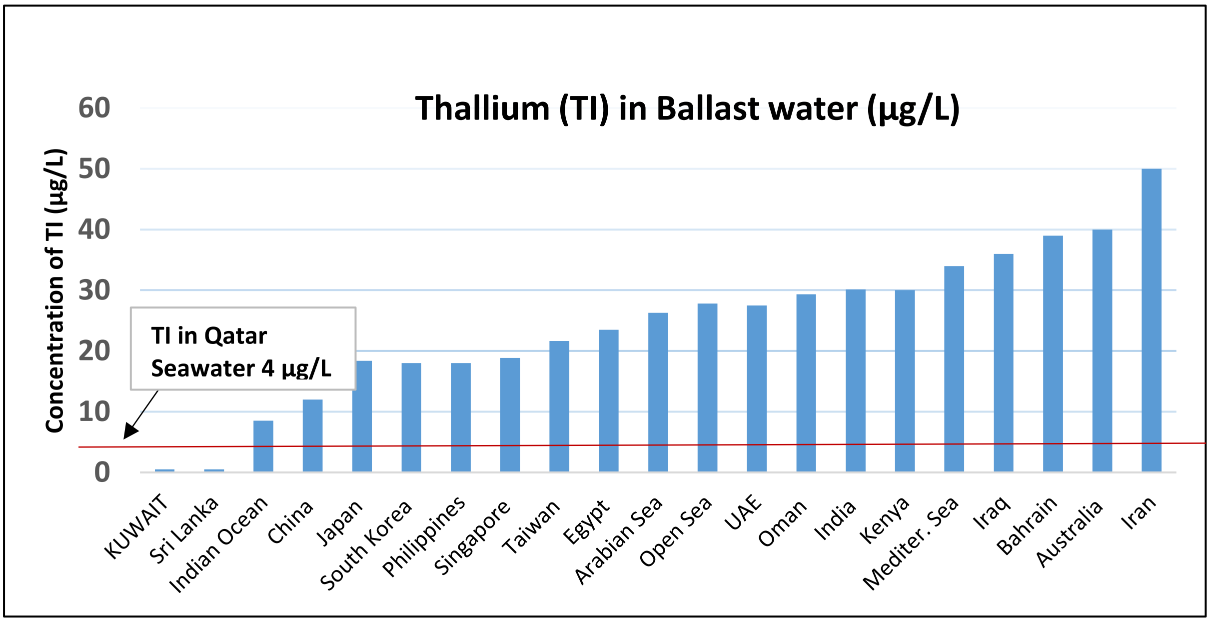

Figure 1. Thallium average concentration in Ballast water of 21 origins

The chemistry of ballast water arriving at Qatar ports is very similar to that of ballast water in many developed and developing countries. Regardless of the treatment technologies and the existing regulations of these countries, the concentrations of the tested parameters are very similar to the ones found in several countries such as Germany (Werschkun et al., 2012), the USA (Oemcke and van Leeuwen, 2003), Canada (Scriven et al., 2015), Denmark (MUDP, 2019) Croatia (Hasanspahić et al., 2022), and Taiwan (Chen et al., 2022). The basic statistical data (minimum, maximum, median, average, and standard deviation) of all ballast water samples in this study are given in the supplementary material (Table 1S).

3.2. Human health risk assessment

Results of element intake through drinking water are summarized in Table (4). Generally, exposure to elements is between one and two orders of magnitude higher in ballast water than in a seawater scenario. These differences are even higher for Fe and Ni.

Table 4. Element intake through drinking water (mg/kg/day) after RO desalination for both scenarios; seawater trace elements of 2019 and same trace elements in ballast water of this study

	Seawater					**Ballast water**				
	Mean	**SD**	**P50**	**P75**	**0.95**	**Mean**	**SD**	**P50**	**P75**	**P95**
Ag	4.7E-08	4.5E-08	3.4E-08	6.4E-08	1.4E-07	1.4E-06	1.1E-06	1.1E-06	1.9E-06	3.6E-06
Al	3.1E-07	3.3E-07	2.0E-07	4.2E-07	9.6E-07	8.7E-06	2.3E-05	2.8E-06	7.9E-06	3.4E-05
As	4.2E-07	3.0E-07	3.4E-07	5.7E-07	1.0E-06	2.3E-07	2.1E-07	1.7E-07	3.0E-07	6.2E-07
B	6.4E-07	4.6E-07	5.3E-07	8.8E-07	1.5E-06	2.6E-04	1.9E-04	2.1E-04	3.5E-04	6.3E-04
Ba	1.2E-06	8.6E-07	9.4E-07	1.6E-06	2.8E-06	6.1E-07	1.2E-06	2.6E-07	6.5E-07	2.3E-06
Be	1.1E-08	7.9E-09	9.5E-09	1.5E-08	2.7E-08	3.3E-08	2.6E-08	2.6E-08	4.5E-08	8.4E-08
Cd	8.4E-09	8.9E-09	5.5E-09	1.1E-08	2.6E-08	2.5E-07	2.8E-07	1.7E-07	3.2E-07	7.5E-07
Co	1.7E-07	1.2E-07	1.4E-07	2.3E-07	4.1E-07	1.0E-06	8.2E-07	8.0E-07	1.4E-06	2.6E-06
Cr	2.7E-08	2.1E-08	2.2E-08	3.7E-08	6.8E-08	5.6E-07	5.1E-07	4.1E-07	7.3E-07	1.5E-06
Cu	3.9E-07	2.9E-07	3.2E-07	5.3E-07	9.6E-07	1.3E-05	1.6E-05	8.0E-06	1.6E-05	4.1E-05
Fe	1.1E-10	7.5E-11	8.8E-11	1.4E-10	2.5E-10	3.4E-04	2.5E-04	2.8E-04	4.6E-04	8.4E-04
Mn	8.1E-08	6.3E-08	6.4E-08	1.1E-07	2.1E-07	2.0E-06	6.9E-05	2.9E-07	1.0E-06	6.5E-06
Mo	8.7E-07	6.3E-07	7.1E-07	1.2E-06	2.1E-06	1.7E-06	1.5E-06	1.3E-06	2.3E-06	4.6E-06
Ni	1.1E-10	7.4E-11	8.8E-11	1.5E-10	2.5E-10	4.7E-06	2.2E-05	8.8E-07	3.1E-06	1.8E-05
Pb	3.2E-07	3.0E-07	2.2E-07	4.3E-07	9.2E-07	4.3E-06	8.9E-06	1.8E-06	4.4E-06	1.6E-05
Sb	6.4E-07	6.8E-07	4.2E-07	8.6E-07	2.0E-06	1.4E-06	1.6E-06	8.9E-07	1.7E-06	4.2E-06
Se	1.2E-06	8.7E-07	1.0E-06	1.7E-06	2.9E-06	6.9E-08	5.7E-08	5.3E-08	9.2E-08	1.8E-07
Si	1.5E-05	1.5E-05	1.1E-05	2.1E-05	4.5E-05	1.0E-05	2.5E-05	3.5E-06	9.6E-06	4.0E-05
Sn	1.4E-07	1.5E-07	9.4E-08	1.9E-07	4.4E-07	7.1E-07	9.9E-07	4.0E-07	8.4E-07	2.4E-06
Sr	1.2E-03	8.3E-04	9.5E-04	1.6E-03	2.8E-03	5.8E-04	4.2E-04	4.8E-04	7.9E-04	1.4E-03
Tl	1.8E-07	1.8E-07	1.2E-07	2.4E-07	5.3E-07	2.6E-06	2.8E-06	1.7E-06	3.3E-06	7.7E-06
U	1.9E-07	1.6E-07	1.4E-07	2.5E-07	5.1E-07	2.4E-07	2.6E-07	1.6E-07	3.1E-07	7.3E-07
V	1.1E-07	1.2E-07	7.2E-08	1.5E-07	3.4E-07	1.8E-06	1.7E-06	1.3E-06	2.3E-06	5.0E-06
Zn	1.3E-06	1.2E-06	9.0E-07	1.7E-06	3.6E-06	5.7E-05	1.8E-04	1.8E-05	5.1E-05	2.2E-04

SD: Standard deviation; P50, P75 and P95: Percentile 50th, 75th and 95th, respectively

Several studies have made the assumption that reverse osmosis can remove 99-99.9% of minerals from the feed water (El-Dessouky and Ettouney, 2002). Other studies have found different results in real RO desalination plants (Kaya et al., 2020; Indika et al., 2021). Our hypothesis is based on several practical experiments conducted at different locations. The study of Indika et al. (2021) found that rejection rates (equal to the removal rates) of Ca, Mg, K, Na, Ba, Sr, Cl, F, and SO_4 in the RO drinking water plants in the North Central Province of Sri Lanka were 93.5%, 97.4%, 86.6%, 90.8%, 95.4%, 96.3%, 95.7%, 96.6%, and 99.0%, respectively, while the best scenario for boron rejection by RO is up to 60% (Kaya et al., 2020). Table (5) shows the rejection rates of several elements and ions by RO desalination plants.

Table 5. Rejection rates of several elements and ions by RO system

Element	Rejection Rate (%)	Element	Rejection Rate (%)
Sodium	90-95	Chromium (VI)	94-99
Calcium	93-98	Chromium (III)	94-99
Magnesium	93-98	Lead	94-99
Potassium	90-95	Chloride	90-95
Iron (II)	93-98	Bicarbonate	90-95
Manganese (II)	93-98	Nitrate	85-90
Aluminum	93-98	Fluoride	90-97
Copper	94-99	Phosphate	93-98
Nickel	93-98	Chromate	90-95
Zinc	93-98	Cyanide	90-95
Strontium	93-98	Sulfate	93-98
Cadmium	93-98	Boron	55-60
Silver	93-98	Arsenic (III)	70-80
Mercury	93-98	Arsenic (V)	94-99
Barium	93-98	Selenium	93-98

Source: https://www.freedrinkingwater.com/rejection-wqa.htm

The health risk of drinking water, mean and 95th percentile HQ, is far below the safety limit set at 1. HQ values are below 0.01, with the exception of Tl, in both scenarios. Thallium HQ levels presented mean values of 0.018 and 0.256, and 95th percentile levels of 0.024 and 0.767, in seawater and ballast water scenarios, respectively (Fig. 2). While the HQ values were below 1 for around 97% of the 100,000 trials performed, more than 50% of the iterations present HQ values above 0.1, and these are for only one exposure source, drinking water. Thus, drinking water could be also used for cooking, bathing or irrigation. Other potential exposure sources for Tl include food ingestion and air inhalation. Due to the findings above, total hazard index (THI), the sum of individual HQ for each exposure pathway could surpass the safety value.

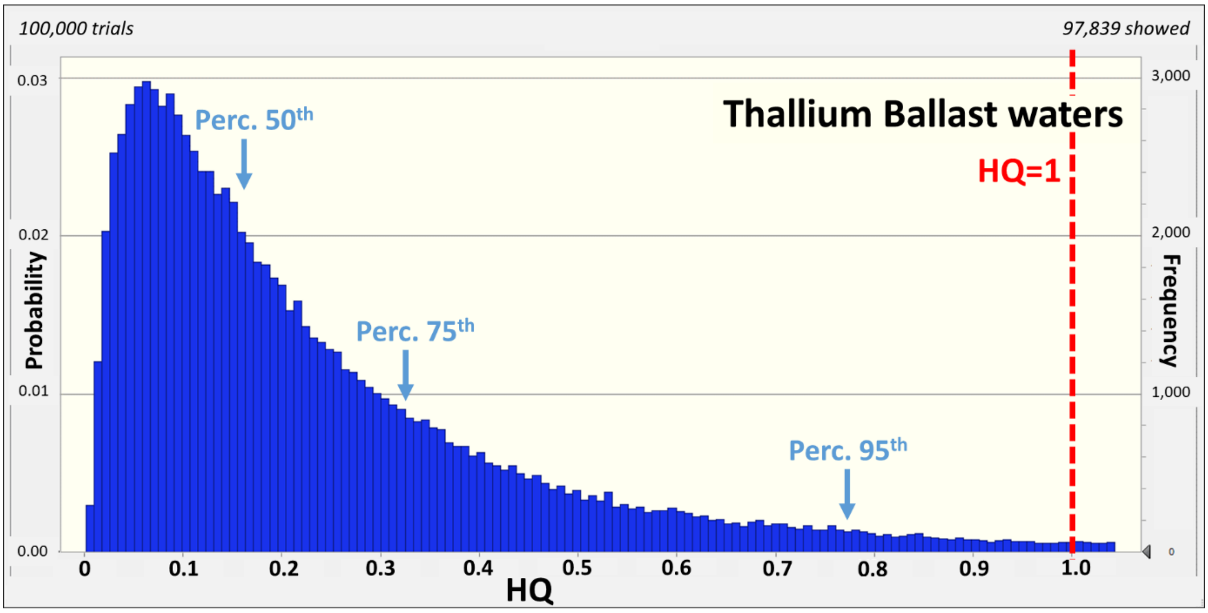

Figure 2. Probability distribution of hazardous quotient (HQ) of thallium exposure through desalinated drinking water coming from ballast water. (Perc.: Percentile).

Levels of Tl found in ballast water (24±16 µg/L) were around ten times higher than its concentration in seawater (1.4±1.2 µg/L). If Tl were not completely removed in RO desalination plants, it could reach the limit of maximum contaminant level set at 2 µg/L, with the aim to lowering it at 0.5 µg/L, by US EPA (US EPA, 2022b). By contrast, European Union legislation does not include thallium as a parameter to be monitored in drinking waters. In Italy, near abandoned mining sites, levels of Tl up to 79.5 µg/L were detected in drinking water (Nuvolone et al., 2021). Although there are no increase in morbidity or mortality rates found in the population living in the area, this chronic exposure may increase the risks for low birth weight and pre-term birth (Nuvolone et al., 2021).

4. Conclusions

An initial fingerprint for the major chemicals present in the ballast water of 21 origins has been established. However, more research is needed to highlight the correlation among chemicals of each ship/tanker and the source of the ballast water. The study has resulted in the generation of fundamental information needed for policy and decision makers to establish ballast water monitoring strategies and best practices for treatment technologies and safe disposal.

Results indicate that there is no human health risk (HQ<1) associated with drinking desalinated water from reverse osmosis plants, regardless of the source of feed water (seawater or ballast water). However, regulators should be aware of the increase in ingestion of potentially toxic elements that may be found in desalinated water, especially in the areas of ballast water discharges. Although dilution may need to be considered, special care should be given to Tl which show the highest HQ (HQ>0.1) levels in the scenario of ballast water as a feed water source. Ballast water discharge strategies should take into account the catchment of desalination plants especially in a semi-closed sea such as the Arabian Gulf. Moreover, it would be necessary to establish a periodically monitoring strategy for potentially toxic elements, including Tl, in seawater near desalination plants and drinking finished water to detect potential problems early on.

Finally, regular monitoring programs are essential to avoid long-term risks.

Although we succeeded to collect a significant and representative number of ballast water samples from the two harbors of Qatar, there were two levels of limitations. The first concerns the logistics and sampling strategy, where regular and periodic samples are needed for the comprehensive investigations, in order to establish a database and actual fingerprints for chemicals of ballast water. The second level concerns the data associated with each ship and tanker, including detailed description of filling and emptying ballast water, the existing treatment facilities and technologies of ballast water within these ships.

We believe that a long -term monitoring strategy which would involve active coordination between relevant governmental entities and research institutions, is urgently needed. Besides the monitoring programs of the chemicals in ballast water, several related questions need to be answered in the future. Examples include fate of the chemicals in the marine system of Qatar and the surrounding regions; climate change and occurrence of the chemicals in total and at the species levels; behavior of chemicals within the sediment-water interaction inside the tanks of ballast water; establishment of digital and smart monitoring systems and action plans; chemicals enhancing the presence, growth and behavior of microorganisms in ballast water and, finally, the best and safest disposal strategies including treatment technologies.

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Supplementary material

Supplementary Table **1.S.** Basic Statistical Parameters of all Ballast Water Samples.
Supplementary Table **2.S** ICP-MS operating parameters

Ethical approval

This article does not contain any studies with human or animals performed by any of the authors.

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Data availability

The dataset produced for this study is available within this article.

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.

Authorship Contribution

Basem Shomar is leading the research project, he coordinated with the relevant authorities, collected and analyzed the samples, drafted the manuscript and he is the corresponding author.
Joaquim Rovira Solano made the risk calculations, wrote the relevant sections, and reviewed the manuscript.

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