Source and risk assessment of heavy metals and microplastics in bivalves and coastal sediments of the Northern Persian Gulf, Hormogzan Province

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Abstract

The objectives of the current study are to investigate the concentration, biological risks, chemical speciation, and mobility potential of heavy metals and also the determination of spatial distribution, physicochemical characteristics, and abundance of microplastics in coastal sediments and edible bivalves of the Persian Gulf, the coastal area of Hormozgan Province. Sampling points were selected considering the location of industrial, urban and Hara forest protected areas. In November 2017, a total of 18 sediment samples from coastal sediments (top 0-10 cm) and the most consumed bivalve species in the region were collected from two stations of Lengeh and Bandar Abbas Ports. The average concentration of heavy metals (except for Ni and Cd) in the sediments were lower than their average shale and the upper continental crust. Enrichment factors revealed significant enrichment of Ni, Mn, Cr, Cd and As. The fractionation of heavy metals using the Community Bureau of Reference (BCR) sequential extraction scheme indicated the high bioavailability potential of Zn, As, Mn, and Co. In general, the highest concentration of Mo, Cd, Pb, Zn, Cr, Cu, Mn, Hg, and Sb was detected in areas with frequent human activities including Shahid Rajaee Port, Shahid Bahonar Port, and Tavanir station, respectively. Shahid Rajaee and Shahid Bahonar Ports are the most important ports on the coast of Hormozgan province. The Risk Assessment Code calculated for the study elements indicates that As, Co, Zn, and Cu pose a moderate environmental risk and a threat to aquatic biota. Health risks of most heavy metals exposed from bivalves consumption were safe, except for As which is associated with the high target cancer risk values. With reference to the type of microplastics found, they were mainly fibers with lengths ranging between 100 and 250 μm in sediments and bivalves. Most of the microfibers found in the sediments were made of polyethylene terephthalate (PET) and polypropylene (PP), and the fibers found in the bivalves were made of PP.

Keywords: Heavy metals; Microplastics; Bivalve and Sediment; Risk assessment; Persian Gulf

Highlights

•This is the first study finding microplastics in bivalves in the Persian Gulf.

•Diverse sources of heavy metals: geogenic, industrial, wastewaters and marine traffic.

•BCR sequential extraction results revealed higher Zn, As, Mn, and Co concentration in the mobile phase.

•Arsenic poses cancer risk to humans in the Northern Persian Gulf (THQ).

•The predominant microplastics in both sediment and bivalve samples were fibers <250 μm.

1. Introduction

Coastal environments faced rapid urbanization and industrialization over the past century. Marine sediment is a valuable indicator for monitoring pollution since it acts both as a sink and as a secondary source of pollutants in the aquatic environment (Crawford and Quinn, 2016; Rocha-Santos and Duarte, 2017). Biota can also be used to biomonitor pollution, and itself can be a source of exposure to pollutants such as microplastics (fragments or original plastics < 5mm) and heavy metals (de Sá et al., 2015; Maulvault et al., 2015; Arulkumar et al., 2017; Ward et al., 2019). The status of the coastal environment can be assessed through heavy metals contamination in the marine organisms and coastal sediments (Frias et al., 2016; Pellini et al., 2018a) and microplastics pollution. Microplastics have not been regularly monitored in recent years, however, they were first reported in coastal sites in the early 1970s (Di Cesare et al., 2020; Tien et al., 2020).

Heavy metals tend to bioaccumulate in marine organisms, therefore this contamination is recognized as a threat to ecosystems and may also cause a health risk to humans through the intake of contaminated seafood or fish (Uysal et al., 2009; Soltani et al., 2019). Heavy metals' uptake by aquatic organisms occurs mostly through water and contact with superficial sediment via benthic and pelagic biota (Soltani et al., 2019). In contrast, the effect of microplastics on ecosystems is still unclear. Numerous marine organisms, including bivalves, fish and oysters pick up microplastics from the water column or sediments as they can be mistaken for food (Bessa et al., 2018; Pellini et al., 2018a, 2018b). When ingested, depending on their size and shape, microplastics can cause blockage of the digestive track of organism (Akhbarizadeh et al., 2019a, Akhbarizadeh et al., 2019b) and they could exert toxicity when entered the bloodstream (microplastics <20µm) (Rothen-Rutishauser et al., 2006). Microplastics can also act as carriers of organic and inorganic contaminants present in water (Zhou et al., 2020; Abbasi et al., 2020; 2021) and could cause enhanced exposure to these. Microplastics, depending on their size, are also excreted (Xu et al., 2020) and then they do not exert apparent impact on amphipods or plants (Fang et al., 2021). The microplastics accumulated in organs of aquatic species (i.e. muscle, gut, gills, and liver) often tend to be very variable in terms of size, roughness, and shape (Abbasi et al., 2018) and finally can reach humans via indirect and direct pathways (Abbasi and Turner, 2021).

Heavy metals presence in coastal sediments can be related to atmospheric deposition, geological weathering, soil erosion, airborne dust, and human activities, including waste disposal. Recently, anthropogenic sources have dominated heavy metals contamination in sediments (Qiao et al., 2020). The most important anthropogenic sources of heavy metals include: fertilizers, pesticides, wastes from smelting ores, leachates from mining sites, leakage of brake fluids, vehicular traffic, sewage sludge, industrial wastes, and partially treated or untreated industrial wastewater (Wang et al., 2020). The total heavy metals concentration is not sufficient to evaluate the adverse effects of contaminated sediment, because heavy metals exist in various chemical species in sediment, which have different mobility, bioavailability and potential toxicity (Delshab et al., 2017). With reference to microplastics, their known main sources are the degradation of plastics released from industry, uncontrolled litter disposal, and inputs from wastewater, gray water and urban run-off (Li et al., 2020). It is important to identify heavy metals and microplastics in environmental studies because, although these are very different type of contaminants, they could have a synergistic effect.

Mobility/bioavailability monitoring studies of pollutants are often restricted by the numerous steps needed for sample treatment. The harmonized and most common procedure used for heavy metals fractionation is the European Community Bureau of Reference sequential extraction, BCR (Davidson et al., 1998; Quevauviller et al., 1994; Rauret et al., 2000). There are no harmonized methods for the monitoring of microplastics and current methods involve their separation from the environmental matrix by filtration or flotation; the characterization of their sizes and shapes with microscopy, and composition with IR or Raman spectroscopy (Li et al., 2020).

The Hormozgan Province coast is an important Iranian port and hosts a wide variety of marine organisms. In recent decades, the fishery industry has co-existed with the oil industry and the area has seen an expansion or new installation of power plants, zinc and steel processing plants, desalination plants, food-processing units, and cement factories. These developments and activities can result in the release of relevant pollutants in the marine ecosystem (Nozar et al., 2014). Since 2010, the presence and impact of heavy metals and microplastics in sediments and selected aquatic organisms from the Persian Gulf has been investigated (Abdolahpur Monikh et al., 2013; Abbasi et al., 2018, 2019; Delshab et al., 2017). However, there is no comprehensive study on the levels and distribution of microplastics and heavy metals in the Hormozgan Province coast. The objectives of the current study were: i) to investigate the concentration and associated biological risks of heavy metals in the sediments of the Persian Gulf, ii) to identify chemical speciation and mobility potential of heavy metals in sediment, iii) to assess health risks of heavy metals via bivalve consumption, iv) to determine the spatial distribution and abundance of microplastics in coastal sediments and selected bivalves in the

Persian Gulf. The possible sources of heavy metals was investigated using multiple linear regression with principal component analysis (MLR/PCA).

2. Material and Methods

2.1.Study area

Being located in an optimal geographic situation and having numerous natural, economic, fisheries, maritime, agricultural as well as livestock resources, Hormozgan is a significant province in Iran which has always been of high economic importance (Amanizadeh, 2018). From recent decades, the obvious developments have occurred in Hormozgan Province as one of the most important Iranian ports and led to high industrial progress such as expansion or establishment of food processing units, steel and zinc plants, desalination plants, fisheries, cement units, and electricity-generation plants. It is well-known nationally and internationally for its strategic position in oil production and export. Furthermore, Hormozgan Province is bordered by the Gulf of Oman and the Persian Gulf, which is at risk from different types of contamination sources such as the discharge of ballast water from ships, contamination sources related to petroleum, saline and hot wastewaters from desalinization plants, agricultural, industrial discharges, and municipal waste. Moreover, Hormozgan is rich in oil and mineral resources which together with its access to High Seas and international maritime trade routes create a chain of unique economic, energy, and socio-cultural potentials in this province (Nozar et al., 2014).

2.2.Sampling, sample preparation, and analysis of heavy metals

Sampling points were selected considering the location of industrial, urban and Hara forest protected areas. In November 2017, a total of 18 sediment samples from coastal sediments (top 0–10 cm) of Hormozgan Province were collected by Van Veen grab in a boat (Fig. 1). Details of sediment sampling stations are given in Table S1 in the Supplementary Information. In the laboratory, samples were first air-dried at room temperature (25 °C) and then sieved (0.63 μm sieve). Heavy metals concentration (Al, As, Cd, Co, Cr, Cu, Fe, Hg, Mo, Mn, Ni, Pb, Sb, Sc and Zn) in the <63 μm fraction were determined using inductively coupled plasma-mass spectrometry (ICP-MS) at the accredited ACME Analytical Laboratory in Canada. Briefly, 0.5 g of each dried sediment sample was heated in a concentrated $HF–HNO_3–HClO_4$ mixture to fuming and taken to dryness. The residue was dissolved in mixture of three volumes of HCl (37%) and one volume of HNO_3 (67%), and the digested samples were subsequently analyzed by ICP-MS. The detection limits were 5 μg kg^{-1} for Hg, 0.01 mg kg^{-1} for Mo, Cd, Pb and Cu, 0.02 mg kg^{-1} for Sb, 0.1 mg kg^{-1} for As, Co, Ni, Zn and Sc, 0.5 mg kg^{-1} for Cr, 1 mg kg^{-1} for Mn, and 0.01% for Fe and Al.

After preliminary research on the natural habitat of bivalves along the coastline of Hormozgan Province, the most consumed bivalves in the region were considered. This allowed us to target the following bivalve species for this work: *Saccostrea cucullata, Circenita callipyga, Barbatia helblingii Solen brevis, Amiantis umbonella,* and *Telescopium telescopium* , which were collected from two stations of Lengeh Port and Bandar Abbas in December 2017 (Table S2). About 30 individual samples of each species in similar sizes from each sampling site were taken by stainless steel hammer and rod and immediately placed in an icebox and transferred to the laboratory for further analysis. The soft body of the bivalves was dissected using a clean scalpel and rinsed with Milli-Q water to remove impurities. Then, seven pooled samples for each species were prepared by mixing the edible parts. Finally, the homogenized samples were sent to the laboratory of Zarazma Mineral Studies Company, Iran. Concentrations of Al, As, Cd, Co, Cr, Cu, Fe, Hg, Mo, Mn, Ni, Pb, Sb, and Zn were determined using ICP-MS. Detection limits were 0.01 mg kg^{-1} for As, Cd, Co, Cr, Hg, Mo, Ni, Pb, and Sb and 1 mg kg^{-1} for Al, Cu, Fe, Mn, and Zn. The method of bivalves digestion for heavy metals analysis is also given in detail in the supplementary information.

2.3.The modified BCR sequential extraction procedure

The modified BCR sequential extraction method was performed on five sediment samples (namely S4, S6, S10, S12, and S17) to evaluate heavy metals operationally defined fractions F1 (exchangeable, carbonate-associated fractions), F2 (reducible, fraction associated with Fe and Mn oxides), F3 (oxidizable, fraction bound to organic matter and sulfide), and F4 (residual fractions). The samples were selected based on the geographical location of the sampling stations and the metal content. The chemical extraction reagents and experimental procedure (Yan et al., 2010) are summarized in Table S3. The method was performed to 1.00 g of dry sediment samples. The heavy metals (As, Zn, Pb, Cu, Co, Cr, Ni, Fe, Al, and Mn) extracted in the above-mentioned fractions were determined by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) (Agilent model 735, United States) in the laboratory of Zarazma Mineral Studies Company, Iran. The detection limits were 0.05 mg l^{-1} for As, Zn, Pb, Cu, Co, Cr, and Ni and 0.1 mg l^{-1} for Fe, Al, and Mn.

2.4.Microplastics extraction and identification

For the extraction of microplastics, sediment samples were pre-treated to remove organic material that could interfere with microplastics counting. For this purpose, 200 g of each sediment sample (<5 mm, previously sieved) mixed with 100 ml of 35% H_2O_2 for 7 days. Then, the digested sediment samples were vacuum-filtered through S&S filter paper (Trade mark grade 589/3, 2 μm pore size) and washed with deionized water. After drying samples on a sand bath at 60 °C for 6 hours, 200 ml of NaI solution with a density of 1.6 mg/cm^3 was added to each sample stirred at 350 rpm for 5 min to separate agglomerated particles and finally allowed to settle for 1.5 hours. The remaining solution at the top of the sample was centrifuged at 4000 rpm for 5 min and filtered using a vacuum filtration unit, onto S&S filter paper (grade 589/3 blue ribbon, pore size <2 μm). The procedure of using the NaI solution, centrifuging, and filtering was repeated three times. Microplastics were separated and identified considering their elastic properties, unexpected forms, homogeneous colours, structures, shininess and hardness under a Carl-Zeiss binocular microscope with up to ×200 magnification (Abbasi et al., 2019). Microplastics were characterized with fluorescence microscopy (Olympus CX31), SEM-EDS (TESCAN-VEGA3, Czech Republic, and an Oxford Instruments X-Max 50 silicon drift detector with AZtec and INCA software) and Micro-Raman spectroscope (μ-Raman-532-Ci, Avantes, the Netherlands).

For microplastics extraction from bivalves, 15g of their soft tissues were put into a series of 100 mL glass beakers (15g/ beaker) to which 30 mL of 10% KOH (99.99% purity, Merck) was added. The tissue was digested at 60 ºC for 72h. The solutions were filtered under vacuum using S&S grade 589/3 filters, dried (at room temperature), and kept in individual glass Petri dishes for microplastic identification as above mentioned methods (Abbasi et al., 2019). Morphological, chemical characteristics and polymeric construction of microplastic particles were investigated using a Scanning Electron Microscopy (SEM) with Energy Dispersive X-Ray Analysis (EDX) and Raman spectrometer. The high vacuum SEM (TESCAN Vega 3, Czech Republic) was run with a resolution of 2 nm at 20 kV. The polymeric construction was identified using a μ-Raman spectrometer (LabRAM HR, Horiba, Japan) with a Raman shift of 400-1800 cm^{-1} and a laser of 785 nm.

2.5 Quality assurance (QA) and quality control (QC)

The analysis included blank reagents, certified reference material (GXR-1, GXR-4, GXR-6, OREAS 45d and SdAR-M2 (U.S.G.S) for sediments and DOLT-3, DORM-2, GXR-4 and GXR-6 for bivalves) and duplicates/replicates were used to QA/QC of the experiment.

Standard reference materials of each heavy metal were measured with a coefficient of variation below 15%. The recovery rates of the total heavy metals concentration were 95–111% indicating a good agreement between the certified and measured values. Blank samples for digestion and analysis methods were evaluated in duplicate with each set of samples. The relative deviation of the duplicate samples was <5%. Furthermore, to confirm the reliability of the modified BCR sequential extraction results, certified reference material (GBW 7312) and reagent blank were used. The concentrations measured for each element indicated recoveries higher than 85%, which indicate satisfactory performance for such type of analyses. In the case of microplastics, the QA/QC was reported in the Supplementary Information.

2.5. Assessment methods

Enrichment Factor (EF), Contamination Factor (CF), Modified Pollution Index (MPI), Modified Potential Ecological Risk Index (MRI), and Risk Assessment Code (RAC) were used to assess the risk of heavy metals contamination.

2.5.1. Enrichment Factor

Enrichment factor (EF) is applied to estimate whether heavy metals are enriched over uncontaminated background concentrations:

$$EF = (C_i/C_{ref})_{sample}/(C_i/C_{ref})_{background} \quad (1)$$

where C_i and C_{ref} are the concentration of heavy metal in sediment and reference element, respectively. In the current study, values of average shale and aluminum (Al) were used as background and geochemical normalization. The normalization against a conservative element accounts for the lithogenic and sedimentary inputs of the element of interest enhancing the prediction of anthropogenic pollution with an enrichment factor (Duodu et al., 2016). Generally, the seven EF classes used to explain the degree of heavy metals pollution. $EF < 2$ indicates deficiency to minimal enrichment, $2 \leq EF < 5$ is moderate enrichment, $5 \leq EF < 20$ is significant enrichment, $20 \leq EF < 40$ is very high enrichment, $EF \geq 40$ is extremely severe enrichment (Yongming et al., 2006).

2.5.2. Contamination factor and modified pollution index

The contamination factor (CF) quantifies the degree of contamination with a specific element (Hakanson, 1980). This gives information about how an element has been concentrated at the site of interest relative to a background site. The calculation of CF does not take into account the lithogenic and sedimentary inputs of the element of interest, which is a limitation considering sedimentation and metal input from the terrestrial environment in waterways (Brady et al., 2015). The limitations of the single element indices have led to the development of multielement pollution indices for the assessment of sediment quality. More recently, Brady et al (2015) proposed a modified pollution index (MPI), which is an improvement of the pollution index (PI) developed by Hakanson (1980) and uses enrichment factors instead of contamination factors in its calculation. This takes into account the background concentrations and the complex, non-conservative behaviour of sediments. Therefor, the MPI was applied to evaluate the overall contamination of the sediment samples with heavy metals (Brady et al., 2015). Since various heavy metals may have effects on one sediment sample, the MPI can help to interpret the heavy metals pollution at each site as a whole (Ranjbar Jafarabadi et al., 2017). Equations (2) and (3) were used to calculate the CF and the MPI:

$$\mathrm{CF} = \mathrm{C_s}/\mathrm{C_b} \tag{2}$$

$$\mathrm{MPI} = \sqrt{(\mathrm{EF_{average}})^2 + (\mathrm{EF_{max}})^2/2} \tag{3}$$

where, C_s, C_b, EF_{max} and $EF_{average}$ refer to heavy metal concentration in sediment, reference background concentration, maximum enrichment factors and average enrichment factor, respectively. Table S4 indicates threshold levels for description of the two integrated indices in sediment samples and the sediment quality categorization.

2.5.3. Modified ecological risk index

The potential ecological risk index (RI) is widely used and universally accepted for quantitative ecological risk assessment (Hakanson, 1980). However, the lithogenic and sedimentary inputs of the element of interest were not considered because the RI used the contamination factor (CF) as a basic calculation unit. The enrichment factor (Eq.(1)) was developed to account for the effects of terrestrial sedimentary input (Duodu et al., 2016). Therefore, the Modified ecological Risk Index (MRI) was used to assess the degree of the heavy metals contamination at a specific station, which has been used frequently in the aquatic ecosystem (Brady et al., 2015). The MRI value was calculated as follows:

$$\mathrm{MRI} = \sum_{i=1}^{n} \mathrm{Er^i} = \sum_{i=1}^{n} \mathrm{Tr^i} \times \mathrm{EF^i} \tag{4}$$

where Er^i and EF^i are the potential ecological risk index and enrichment factor of every single heavy metal, respectively. Tr^i refers to the response coefficient for the toxicity of each element, which indicates the ecological sensitivity to heavy metals contamination and the level of element toxicity (Yavar Ashayeri and Keshavarzi, 2019). The response coefficients (Tr^i) used for Hg, Cd, As, Cu, Pb, Ni, Cr, and Zn were 40, 30, 10, 5, 5, 5, 2, and 1, respectively (Hakanson 1980). Er^i and MRI classifications used here are those proposed by Hakanson (1980) and Zhang et al. (2017), respectively (Table S4).

2.5.4. Risk assessment code

The RAC estimates the availability of heavy metals using the percent of heavy metal in the exchangeable phase (% F1 for BCR) (Marrugo-Negrete et al., 2017). It is important because this is the fraction impacted by human activities and is specified by the adsorptive, exchangeable and bound to carbonate phases, which are weakly bonded heavy metals that can equilibrate with the aqueous phase and hence become bioavailable (Liu et al., 2008). Based on the RAC guideline, for any element, there is no hazard when the percentage RAC is lower than 1%; a low hazard from 1-10%; a medium hazard from 11-30% and a high hazard from 31-50 % (Jain, 2004). Furthermore, RAC > 50% indicates that the sediments pose a very high hazard and are considered very dangerous for the aquatic biota (Sundaray et al., 2011)

2.5.5. Human Health risk assessment of bivalve consumption

The human health risk posed by heavy metals in edible portions of bivalves was assessed using target hazard quotients (THQ), target carcinogens risk (TR) and the estimated daily intake (EDI). The EDI considers daily exposure doses of heavy metals for the human population via the consumption of edible bivalve tissues (Yap et al., 2016). The THQ is applied to assess non-cancer health risk and presents an indication of the risk value because of heavy metal exposure (Wang et al., 2018). THQ > 1 indicates that the exposure level is higher than the reference dose, which suggests that a daily exposure at this level probably poses adverse health impacts during a lifetime in a human population (Jović and Stanković, 2014). The TR value estimates the incremental possibility of an individual developing cancer over a lifetime exposure to potential carcinogens (Liu et al., 2019). These methods were available in US EPA Region III Risk-based Concentration table (USEPA, 1989) and they are calculated by the following equations:

$$\mathrm{EDI} = \mathrm{C} \times \mathrm{IR}/\mathrm{BW} \tag{5}$$

$$\mathrm{THQ} = (\mathrm{EF} \times \mathrm{ED} \times \mathrm{IR} \times \mathrm{Cf} \times \mathrm{C}/\mathrm{BW} \times \mathrm{AT} \times \mathrm{RfD}) \times 10^{-3} \tag{6}$$

$$\mathrm{TR} = (\mathrm{CSF} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{IR} \times \mathrm{C} \times \mathrm{Cf}/\mathrm{BW} \times \mathrm{AT}) \times 10^{-3} \tag{7}$$

where C represents the heavy metal concentration in the edible bivalve tissues (mg/kg, w/w), BW represents the average body weight (16 kg for children and 70 kg for adults), IR represents the bivalve ingestion rate. According to local consumption, and as reported by the Hormozgan fisheries organization, the average consumption rate for bivalve was considered to be 0.2 mg day^{-1} (Mohebbi Nozar et al., 2013). EF is the exposure frequency (365 days/year), ED is the exposure duration (6 years for children and 70 years for adults), Cf is the conversion factor (0.208) for converting wet weight (w/w) to dry weight (dw), The average exposure time (EF×ED), RfD represent the oral reference dose (mg kg^{-1} day^{-1}), and CSF is the oral carcinogenic slope factor (mg kg^{-1} day^{-1}). In this study, RfD and CSF of the investigated elements were adopted from the regional screening level summary table (USEPA, 2017), with the exception of Pb (Liu et al., 2017), to evaluate the EDIs element risk in benthic bivalves. The target carcinogen risks for Pb, Ni, Cr and As were measured on the basis of available CSFs. Based on US EPA methods, carcinogenic risk less than 10^{-6} is considered to be negligible, > 10^{-4} is unacceptable, and in the range between 10^{-6} and 10^{-4} represents the acceptable risk (USEPA, 2004). Humans are mostly exposed to more than one heavy metal and suffer interactive or combined impacts. Consequently, the total THQ (TTHQ) was utilized to evaluate the total non-cancer health risks THQ amounts (Bogdanović et al., 2014). If the TTHQ value is lower than 1, there is no significant risk. If the value of TTHQ is >1, there is a chance that negative impacts can occur (Wang et al., 2018). In addition, quantitative analysis was also used to compare the levels of heavy metals in the edible bivalve tissue on the basis of the provisional maximum tolerable daily intake (PMTDI) (Jović and Stanković, 2014; Mok et al., 2015; WHO, 2009).

2.6. Data analysis

To evaluate the possible sources of heavy metals in sediments, principal component analysis (PCA) and multiple linear regression (MLR) were applied using the Statistical Package SPSS 22.0 (Yavar Ashayeri et al., 2018). Heavy metals concentration below the detection limit (LOD) were considered to be 0.75 of the LOD for statistical analyses (Hornung and Reed, 1990).

3. Results and discussion

3.1. Heavy metals in sediments

Heavy metals in sediments and bivalves were instigated by their elevated levels reported previously in fish, prawn, and crab (Soltani et al., 2019; Keshavarzi et al., 2018). The heavy metals concentration in sediments and bivalves are summarized in Table 1. The highest concentration of Mo, Pb, Zn, Cr, Cu, and Mn, Hg, and Sb was detected in areas with frequent human activities including Shahid Rajaee Port (S3), Shahid Bahonar Port (S1), and Tavanir station (S4), respectively. Shahid Rajaee Port and Shahid Bahonar Port are the most important ports in the coast of Hormozgan province. It can be observed that the maximum concentrations of Ni and Co, and As occurred at the S11 and S14 stations, respectively, that located in the mangrove forest. The highest level of Cd was detected in Geshm water desalinization (S8). Furthermore, high concentrations of Fe and Al were found at the station S16 (East of Hormozgan Province). The lowest amounts for Cu, Zn, Ni, Co, Cr, and Al were detected at the station S14 (Mangrove Forest) while those for Co and Mn, Mo and Pb, Fe, and As, Cd and Sb were found at the stations S4, S7 (Velayat station), S13 (Mangrove Forest), and S16, respectively. Also, Hg concentration was not detected at the S4 and S13 stations due to values below the instrumental detection limits.

The average concentration of all heavy metals (except for Ni and Cd) in the sediments were lower than their average shale (Turekian and Wedepohl, 1961) and the upper continental crust (Rudnick and Gao, 2003). A comparison of the mean and median heavy metal values revealed that the distribution of Cd, Hg, Sb, and Mn varied slightly across the sampling locations. The mean concentrations of Cu, Zn, Ni, Cr, Fe, and Al were above their median content, which indicates that their extreme concentrations considerably increased the mean value. Median levels of Hg, As, and Cd were close to their upper continental crust values, therefore there is limited contamination of these heavy metals in the coastal sediments. Nevertheless, median levels of Ni and Cd were considerably above the upper continental crust elemental abundances; this could be due to their enrichment in the sediments (Table 1).

The mean concentration of heavy metals in sediments were compared with other similar studies worldwide (Table S5). The concentration of Cu in samples is lower than that of Nemrut Bay (Turkey), Thermaikos Gulf (Greece), Eastern coast of Thailand Gulf Shandong Peninsula (China), Gorgan Bay (Iran), and Assaluyeh coast (Persian Gulf). The Pb concentration from the Nemrut Bay (Turkey), Thermaikos Gulf (Greece), Eastern coast of Thailand Gulf, and Shandong Peninsula (China), Zn concentration from the Nemrut Bay (Turkey), Thermaikos Gulf (Greece), and Eastern coast of Thailand Gulf, As concentration from the Nemrut Bay (Turkey), Shandong Peninsula (China), and Gorgan Bay (Iran) is higher than those obtained in this study. The concentration of Ni, Co, Cd (except for Nellore coast (India) and the Gulf of Oman), and Cr (except for Nemrut Bay (Turkey) and the Gulf of Oman) in the present study are higher than all other presented sediments. Also, Hg concentration in the Assaluyeh coast (Persian Gulf), Eastern coast of Thailand Gulf, and Shandong Peninsula (China) show a higher mean compared with those obtained in the Northern Persian Gulf (Hormogzan Province). The concentrations of Al and Mn in the present study are lower than Gorgan Bay (Iran) and the Eastern coast of Thailand Gulf, respectively, while Fe value in the current study is higher than those for the Nellore coast (India) and the Gulf of Oman.

3.2. Environmental implications

The average EF for heavy metals revealed the following decreasing trend Ni (8.27) > Mn (5.72) > Cr (5.70) > As (5.34) > Cd (4.48) > Co (4.35) > Fe (3.43) > Pb (3.42) > Mo (3.07) > Zn (2.68) > Cu (2.51) > Sb (0.82) > Hg (0.35). Hence, Ni, Mn, Cr, and As displayed significant enrichment while Cd, Co, Fe, Zn, Mo, Cu, and Pb indicated moderate enrichment. The minimal degree of enrichment was obtained for Hg and Sb. The box plot of the enrichment factor in the sediments of the study area is represented in Fig. 2. and shows the distribution of EF in stations. The highest EF for Mo, Co, As, Cd and Mn occurred at the S14 sampling location (see Fig. 1), while the highest Pb, Cr and Fe contents were found at the S4 site (Tavanir station). S4 site being close to Tavanir Power Plant and industrial wastewater discharge to coastal sediment may be an important factor in increasing these elements concentration. The S14 site may also be exposed to heavy metals released from urban and agricultural wastewater discharge from adjacent coastal cities. This supports the fact that human factors can play an important role in sediment heavy metals enrichment.

In order to obtain a better insight into the Persian Gulf sediment contamination and related adverse effects, CFs and MPI were calculated. According to the CFs classification (Hakanson, 1980), all heavy metals show low pollution except for Ni (at S1, S3, S8-S9, S11-S12, S16, and S18), As (at S14), and Mo (at S3), which revealed moderate contamination (Table S4 and S6), and therefore these elements should be prioritized. The calculated contamination factors indicate that anthropogenic pollution is likely to a source of As, Ni, and Mo at specific sites. Generally, there are high levels of Ni, Zn, Cu, As, Pb and Cd in coastal sediments close to cities (Amin et al., 2009). Pb was utilized as an anti-knock additive in the combustion of gasoline and could persist in soil and sediment for a long time (Gao et al., 2016). Cu and Zn could reach the aquatic ecosystems from industrial effluents, urban stormwater, metal processing units, distillery units, fly ash from coal-powered plants, agricultural and domestic waste disposal, copper and zinc plating, and dry and wet deposition (Lahijanzadeh et al., 2019). Other elements are commonly used in nickel-cadmium batteries, plastic stabilizers, pigments, and electroplating and enter the living ecosystem through metalworking industries, heating systems, urban traffic, and power stations. High Ni alloys are also utilized in oil refining, electrical, marine, chemical, and other industrial processes (Jumbe and Nandini, 2009). In general, the results of MPI showed that stations S16 and S17 were “moderately to heavily polluted”, S4 and S14 were “severely polluted” while other sites were “heavily polluted” (S1-S3, S5-S13, S15, and S18). Thus, high MPI is probably the result of high EF of some heavy metals (Fig. S1).

3.3. Source apportionment of heavy metals

PCA was performed to unravel quantitative assessments of heavy metals sources and analyze the relationships among heavy metals and their geochemical associations. PCA was carried out with Varimax rotation and four factors were identified with 90.67 % of the variance and eigenvalues >1 (Table 2). The first principal component (PC1) accounting for 52.06% of the variance has high loading on Ni, Zn, Co, Cu, Fe, Cr, Pb and Al. The grouping of heavy metals reflects common related characteristics or sources of pollution (Keshavarzi et al., 2015). This component demonstrates elements with CF and little enrichment, and thus less influenced by anthropogenic activities. The concentration of Fe, Mn, Pb, Ni, Co, Sb, Cr in the sediments was higher than in bivalve samples. Among these, Mn and Fe were the most abundant metals in all sediment samples, probably due to the fact that they could be fundamental of the geogenic source (Yavar Ashayeri and Keshavarzi, 2019). Indeed, according to previous investigations, some heavy metals such as Cr and Ni occur in high concentrations naturally in soil of Southern Iran (Abbasi et al., 2019). Thus, PC1 is presumed to demonstrate the percent of geogenic origins in the current study. The second component (PC2) includes Mn and Hg with 15.98% of the total variance while Mo, Sb and As with 14.5% of the variance are dominant in the third component (PC3). The grouping of Mn and Hg in PC2 is either related to their high content in some stations or associated with similar anthropogenic origins. The concentration of Mn and Hg in this study was lower than the reference values (Table 1). However, the high EF value for Mn suggests that human activities are the major sources of this element. In addition, Hg concentration in all stations was low, indicating geogenic origin. However, the highest level of Hg (0.03 mg kg^{-1}) found in Shahid Bahonar Port (S1) could be due to the high maritime traffic of Shahid Bahonar Port load associated with transportation of ores and crude oil (Nozar et al., 2014). Hg could be naturally present in crude oil and gas, mostly in the volatile elemental form and inorganic forms, such as selenide, sulfide, chloride, and mercuric oxide that can dissolve in the production water employed in the petroleum extraction (Saint’Pierre et al., 2013). It is also noted that anthropogenic sources such as mining and fossil fuel extraction are major sources of much of the Hg that exists within the ecosystems of the earth (Hadden and Moss, 2010). As a result, these elements have a mixed geogenic and anthropogenic origin. Also, the occurrence of Mo, As and Sb in the PC3 is due to high enrichment factors at the S14 site. Moreover, the S14 site is exposed to pollutants released from urban and agricultural wastewater discharges from adjacent coastal cities. Therefore, the presence of As, Mo, and Sb in this factor suggests that sediment is polluted by agricultural practices and discharge of effluents from adjacent coastal cities (Varol and Sünbül, 2018). The fourth component (PC4) comprising of Cd with 8.13% of the total variance. However, in general, it can be said that the cause of Cd separating from other elements is perhaps reflective of its source is different. Cadmium is most likely released from ship and boat engine emissions (Yavar Ashayeri and Keshavarzi, 2019). Furthermore, the contribution of these four factors was assessed using multiple linear regression (MLR) model. The major aim of performing MLR is evaluation of the percentage contribution of various heavy metals origin for a specified sediment sample. Regression coefficients for sediment samples ($R^2 = 0.964$, $n = 18$) proved to be significant at $p < 0.05$ (95% confidence level). The PCA-MLR data are represented in Table 2, demonstrating that most heavy metals were dominant in sediment samples and that 61.35% of the metal contribution comes from geogenic sources. The anthropogenic sources such as crude oil transporting tankers or industrial effluents contribute up to 12.79% of the sediments heavy metal pollutants. Urban and agricultural wastewaters and emissions form ship traffics and boats engines contributed up to 18.76% and 7.11% of heavy metals, respectively. Nevertheless, these estimates represent possible sources of heavy metals in sediment samples, and there may also be other sources for them in the study area. Therefore, further studies are needed to accurately estimate heavy metal sources. Besides the wastewater discharge, which are the major sources of contamination in the coastal sediment of Hormozgan province, as mentioned in section 2.1, anthropogenic emissions from incinerators, industrial chimneys, and flares might be considered as contamination sources in coastal sediments of Hormozgan province. Since there is high road traffic in the Hormozgan province coastline, vehicles and traffic load could be considered as a probable origin for heavy metals contamination. Furthermore, stations located close to Bandar Abbas city can influence by runoff of Bandar Abbas street dust particles to the coastal sediments of the sea (Keshavarzi et al., 2018).

3.4. Chemical partitioning of heavy metals

Distribution of heavy metals in the sediment needs to be considered to understand their fate, bioavailability, environmental behaviors, and origins. The geochemical phases of heavy metals

(Zn, Ni, Al, Cu, As, Pb, Cr, Fe, Mn, and Co) obtained by the modified BCR sequential chemical extraction are presented in Table S7 and Fig. S2. Cu, Ni, Zn, Pb, Fe, Al, and Cr were mainly detected in the residual fraction (F4), averaging 98.1%, 94.2%, 90.3%, 77.2, 66.6%, 61.5%, and 41.6%, respectively of their total amounts. This suggests that these heavy metals could be associated to crystalline structures of the minerals and resistant components of the solid matrix and thus being geogenic in the source (Li et al., 2013). The contribution of the acetic acid exchangeable fraction (F1) of these heavy metals were also significantly smaller compared with the other heavy metals. The oxidizable fraction (F3) was the dominant phase for As (45.8%) and Cu (20.37%), indicating its affinity for combining with organic matter and sulfides in sediment samples.

Under oxidizing conditions, the dissolution of organic matter and sulfides could lead to the release of heavy metals bound to these components (Filgueiras et al., 2002). The majority of Mn (39.5%) occurred in the residual fraction (F2) and indicates its high affinity to amorphous Fe and Mn oxides and hydroxides in sediment samples (Gao et al., 2018a). Among the four extracted fractions in the BCR sequential extraction, the acetic acid extractable (exchangeable) fraction is the most mobile and hence bioavailable phase. In general, the mobility and bioavailability of heavy metals decrease in the order of sequentially extracted fractions (Davidson et al., 1998). Heavy metals extracted from F1 indicate high mobility and have the potential to be taken up by biota and pose potential risks to marine ecosystems (Gao et al., 2018b). The proportions of heavy metals in this phase revealed the following decreasing order Co (21.6%) > As (21.5%) > Mn (20.4%) > Zn (19.9%) > Cu (13.0%) > Ni (8.0%) > Cr (1.9%) > Pb (1.5%) > Fe = Al (0.15%). In general, although maximum percentages of elements occur in phases with limited mobility, care must be taken, since the sudden change of the environmental conditions of the sediments can result in their mobilization, increasing their bioavailability. The mobility sequence of elements according to the sum of the first three fractions (F1+F2+F3) for all the sediments (Table S7) was in the order: As (75.5%) > Mn (61.6%) > Zn (58.4%) > Co (53.7%) > Cu (38.5%) > Pb (33.4%) > Ni (22.8%) > Cr (9.7%) > Fe (5.8%) > Al (1.9%). This demonstrates that As, Zn, Mn, and Co are most probably assimilated by benthic biota. Other heavy metals are considered to be the most stable elements due to their low mobility. Generally, in terms of the spatial distribution of the geochemical fractions of heavy metals in sediment samples, the contributions of the non-residual fractions (F1 + F2 + F3) were higher in the residual fraction (F4), indicating predominance of anthropogenic sources. Also, to survey the risk level arising from the mobility of heavy metals in the sediment, the risk assessment code (RAC) values were computed for heavy metals (Zn, Ni, Al, Cu, As, Pb, Cr, Fe, Mn, and Co). Since the RAC is calculated according to the percent of F1, Cr, Ni and Pb revealed low risk, while Zn, Mn, Cu, Co, and As should have medium environmental risks. Al and Fe besides having low environmental risk occur mainly in F4 phase, from which their dissolution/leaching is very low and hence are grouped under the no-risk class (<1%) (Fig. S3).

3.5. Ecological risk assessment of sediments

The Modified Ecological Risk Index (MRI) uses the enrichment factor (EF) as a basic calculation unit that can detect significant pollution (regarding both stations and metal). Therefore, the potential ecological risk index (Er^i) with (EF) was applied. The Er^i of eight heavy metals (Ni, Zn, Pb, Hg, Cu, Cr, Cd, and As) in the sediment samples are presented in Table 3. On average, low ecological risk (Er_i) was observed for Cr (11.4), Cu (12.54), Pb (17.12), Hg (13.8) and Zn (2.68) elements and the elements of As (53.37) and Ni (41.33) fall in the range of moderately ecological risk. Among these, Cd (145.92) element shows a considerable ecological risk. The results indicated that Er^i of Zn, Pb, Hg, Cr, Cu and As (except in S4, S7, S13-S15) display low potential ecological risk. Er^i also revealed that As poses a moderate risk at S7 and S13 sampling stations, and a considerable risk at S15 and a high risk at S4 and S14. The S14 site indicated the highest EF for Mo, Co, As, Cd and Mn, and the S4 site demonstrated the highest EF for Pb, Cr, and Fe. In general, the results are consistent with the information found from the calculated EF values in Section 3.1.

For Ni, a total of 13 sites (72.2%) exhibited a moderate ecological risk. Approximately 27.8% of the sites showed a low ecological risk. The Er^i values of Cd in the sediments varied from 26.8 to 410.8, suggesting a very broad range of risk, from low to very high ecological risk. The highest Er^i for Cd (Er^i =410.81) occurred in the Hara forest (S14) due to its high enrichment factor value. The Er^i for Cd recorded the greatest potential ecological risk among the elements because of its high toxicity. The sampling sites can be classified into four categories on the basis of their MRI (Table 3). The first category including S16 suggests low ecological risk (MRI <150) due to low values of EF and Er_i for most heavy metals, while the second category (S1-S3, S5-S6, S8-S9, S11-S12, and S17-S18) exhibited moderate ecological risk ($150 \leq$ MRI <300), and the third category (S4, S7, S10, S13, and S15) revealed considerable ecological risk ($300 \leq$ MRI < 600). The fourth category (S14) demonstrated very high ecological risk (MRI $\geq$ 600). In general, a moderate ecological risk (average MRI = 298.17) was observed in the sampled sediments.

3.6. Heavy metals in bivalves

Heavy metals in edible portions of the sampled bivalves (*Saccostrea cucullata*, *Circenita callipygo*, *Barbatia helblingii*, *Solen brevis*, *Amiantis umbonella*, and *Telescopium*) are presented in Table 1. A wide range of heavy metals concentrations in edible parts of bivalves was found ranging from below the LOD for Mo, Pb, Ni, Co, Hg, As, Sb to 5457 mg kg^{-1} for Al. Sb content in seven studied bivalve samples were below the detection limit (LOD, 0.01 mg kg^{-1}), indicating little or no risk of Sb accumulation in the sampled bivalves. The maximum concentrations of Ni (4.27 mg kg^{-1}), Mo (2.84 mg kg^{-1}), and Co (3.74 mg kg^{-1}) were observed in *Circenita callipygo* while minimum concentrations were detected in *Solen brevis*, *Saccostrea cucullata* and *Telescopium*, *Saccostrea cucullata* and *Telescopium*, and in *Saccostrea cucullata* (were all below the LOD (0.01 mg kg^{-1})), respectively. The highest Zn (4314.5 mg kg^{-1}), Cu (793 mg kg^{-1}), As (173.3 mg kg^{-1}), and Cr (4.89 mg kg^{-1}) contents were measured in *Saccostrea cucullata*, and the lowest concentrations were found for Zn (60.5 mg kg^{-1}) in *Amiantis umbonella*, Cu (9 mg kg^{-1}) in *Circenita callipygo*, As (<LOD, 0.01 mg kg^{-1}) in *Saccostrea cucullata* and *Telescopium,* and Cr (0.3 mg kg^{-1}) in *Telescopium*. The highest values of Mn and Pb (166 and 2.75 mg kg^{-1}), and Hg and Fe (0.34 and 4461 mg kg^{-1}, respectively) were measured in *Amiantis umbonella* and *Telescopium*, respectively. The lowest Mn, Pb, Hg, and Fe concentrations were detected in *Saccostrea cucullata*. The maximum (0.95 mg kg^{-1}) and minimum (0.07 mg kg^{-1}) concentrations of Cd were also detected in *Barbatia helblingii* and *Saccostrea cucullata*, respectively. In general, the average concentration of heavy metals in edible tissues of bivalves decreased in the following order: Al > Fe > Zn > Cu > Mn > As > Cr > Ni > Co > Mo > Pb > Cd > Hg > Sb. Compared to other studies (Table S5), the mean heavy metals concentrations were comparable to those from the other Coast. For example, most worldwide studies have reported considerably lower concentrations of Fe and Co (except for Musa Estuary (Persian Gulf)), As, Mn, Al, Hg (except for Musa Estuary (Persian Gulf) and the Gulf of Oman), Cr (except for Musa Estuary (Persian Gulf) and Laizhou Bay (China)), and Pb (except for Musa Estuary (Persian Gulf) and Sydney estuary (Australia)) in edible tissues of bivalves than the current study. Also, the mean Cd concentration in the Northern Persian Gulf (Hormogzan Province) is lower than those of other regions, except for Shandong Peninsula (China). The mean Cu value in the present study is lower compared to that found in the Gulf of Oman, Assaluyeh port coasts (Persian Gulf), Sydney estuary (Australia), and Musa Estuary (Persian Gulf). The mean Zn concentration in the current study is lower than the mean Zn levels found in the Gulf of Oman, Assaluyeh port coasts (Persian Gulf), and Sydney estuary (Australia), and higher than in some other localities. Also, the Ni amounts in the Gulf of Oman, Sydney estuary (Australia), and Assaluyeh port coasts (Persian Gulf) was higher than those in the Northern Persian Gulf (Hormogzan Province).

The concentrations of heavy metals in sediment and bivalves were compared and displayed in Fig. S4. The results indicated that the concentration of Fe, Mn, Pb, Ni, Co, Sb, and Cr in the sediments was higher than in the bivalve samples. However, the concentrations of Mo, Cu, Zn, Hg, Cd, and As were greater in bivalves, suggesting that benthic organisms are enriched because of direct contact with bottom sediment Mo, Cu, Zn, As, Cd, and Hg (Khoshnood et al., 2010). It is well accepted that aquatic organisms accumulate different levels of heavy metals depending upon several factors including physiological mechanisms of the involved organism, metabolic activity, dietary routes, growth rate, habitat, trophic levels, longevity as well as chemical characteristic and levels of heavy metals (Adel et al., 2016; Soltani et al., 2019).

3.7. Human Health risk assessment of bivalve consumption

Quantitative and qualitative analyses were used to assess the risk of heavy metals to human health via the consumption of bivalves. The THQ of each heavy metal from the consumption of various bivalves are given in Table 4. The THQ for all of the investigated heavy metals in bivalve samples were below the safe level (THQ=1) for children and adults, suggesting that there is no potential non-cancer risk from consumption of the six investigated bivalve species. In general, the mean THQ of heavy metals in both children and adults displayed the following descending order: As > Co> Cu > Zn > Hg > Cr > Mn > Cd > Pb > Mo > Ni. It should be noted that bivalve consumption involves exposure to multiple elements, which can cause interactive and/or additive impacts. Therefore, cumulative health risks were estimated using the total target hazard quotient (TTHQ). However, TTHQ values in all bivalves ranged from 0.01 to 0.81 which is significantly lower than the safe level (=1) for all sampled species. A TTHQ < 1 indicates that the exposure level is less than the RfD (reference dose), which signifies that a daily exposure at this level does not probably lead to any adverse impacts during the lifespan of a human population (Bogdanović et al., 2014).

The TR of Pb, As, Ni and Cr in all the analyzed bivalves (Table 4) indicates that the cancer risk of arsenic in children is within the acceptable limit (10^{-4} to 10^{-6}) in all investigated species (except *Telescopium*). The risk for As in the *Telescopium* specie was lower than the negligible value (TR < 10^{-6}), whereas for Pb (except *Barbatia helblingii*, *Solen brevis* and *Amiantis umbonella*), Ni (except *Circenita callipygo* and *Amiantis umbonella*) and Cr were lower than the negligible level in all species. In the case of adults, Pb (except *Amiantis umbonella*), Ni and

Cr presented negligible risk values (TR < 10^{-6}) in all species and cancer risk for As was within considered acceptable value (10^{-4} to 10^{-6}) in all species (except *Telescopium*). In this study, consumption of the six bivalve species posed carcinogenic risk mainly from arsenic. Arsenic, particularly inorganic arsenic, is a carcinogen and can lead to various cancers (Liu et al., 2019). However, marine organisms mainly contained large numbers of organic As, which exhibited hypotoxicity and were considered safe for human ingestion (Liu et al., 2019). Also, the accumulation of high levels of arsenic by marine organisms is a well-known natural phenomenon, and there is no suggestion of anthropogenic contamination being a factor (Krishnakumar et al., 2016). However, the total TR of the all heavy metals for bivalves was relatively high, ranging from 6.0E-06 to 3.9E-04. The highest total TR was also found in children. In fact, all risk estimates (except *Telescopium*) in children were higher than 1×10^{-4}, a value used as the acceptable risk ceiling (USEPA, 2004).

Table S8 includes the EDI values of selected heavy metals from bivalve consumption by local consumers (children and adults). EDI data is compared with the provisional maximum tolerable daily intake (PMTDI) for heavy metals summarized by Jović and Stanković (2014) and Mok et al. (2015) from the data of Joint FAO/WHO Expert Committee on Food Additives and U.S. Environmental Protection Agency are presented as one of the references points to estimate the health risks arising from seafood consumption (Table S8). Compared with PMTDI values, As (> 0.0003 mg/kg/day) and Pb (> 0.0036 mg/kg/day) in muscles of the five investigated bivalve species (except *Telescopium*), Cd (> 0.00083 mg/kg/day), Co (> 0.0016 mg/kg/day), Mn (> 0.06 mg/kg/day) and Cr (> 0.0003 mg/kg/day) in all species, Zn (> 1 mg/kg/day) and Hg (> 0.00057 mg/kg/day) in *S. cucullata*, *B. helblingii* and *Telescopium*, Cu (> 0.5 mg/kg/day) in *S. cucullata* and *Telescopium*, Ni (> 0.02 mg/kg/day) in *A. umbonella* and *C. callipygo*, and Mo (> 0.01 mg/kg/day) in *C. callipygo, B. helblingii* and *A. umbonella* in children may cause health problems via bivalve consumption. Also, the EDI values of As in the five bivalve species (except *Telescopium*), Co in *, B. helblingii* and *S. brevis*, Pb and Hg in *B. helblingii*, Mn in *B. helblingii*, *Telescopium* and *A. umbonella*, Zn and Cu in *S. cucullata*, and Cr in all species can lead to health problems in adults. Nevertheless, the results suggest that the mean EDI of all selected heavy metals were higher for children than in adults.

3.8. Microplastics in sediments and bivalves

Examples of the microplastics found in the study sediments and bivalves are shown in Fig. S5. Primarily microplastics were detected visually and then their organic nature and morphology were observed with fluorescent microscopy and SEM/EDX (Fig. 3). The fluorescence properties of fibrous microplastics are visible in Fig. 3A. Based on the fluorescence properties of the most common microplastic particles, this method can be used for microplastic identification without a visual pre-sorting process (Abbasi et al., 2017). However, the operator must also pay attention to other similar particles such as wood and paper (Abbasi et al., 2019).The results showed that fibers mostly displayed a smooth surface. Furthermore, some materials, inorganic and/or organic were detected on the microplastics'surfaces. The results of chemical composition analysis revealed that most microplastics included O, S, Si, Cl, and C. The high percentage of carbon and oxygen may indicate that the detected particle is plastic. However, this can also be confused with wood particles. In fact, EDX elemental analysis provides indirect information of found particles. For example, on the surface of polypropylene, polyethylene, and polystyrene particles, strong nitrogen peaks can be a biomass proxy (Pan et al., 2019). Also, the existence and direct observation of the surface environmental weathering are easily confirmed by chlorinated microplastics, for example, PCV (Gniadek and Dąbrowska, 2019).

Fig. S7 to S10 present the abundance of collected microplastics. Based on the shape, the majority of the identified microplastics in the sediments and bivalves samples were fibrous (Fig. S6) similar to the result already obtained in the Persian Gulf's fish and prawns (Abbasi et al., 2018). A large number of fibers in sediments could result from various kinds of ropes and nets used in the fishing nets, close to urban areas. Also, the literature perusal indicates that more than 1900 fibers are released per every single wash of a synthetic garment into the marine environment (Duis and Coors, 2016; Frias et al., 2016). The low density of microplastics can be transported from urban areas to the sea (Abbasi et al., 2019), eventually settling in the sediments. For instance, this research shows that in S13 station located in Hara forest the density of microplastic fibers is comparable with stations that are located near urban areas indicating that such fibers can be transported long distances. Considering the fact that microplastics can also adsorb organic and inorganic pollutants and translocate them over long distances (Bakir et al., 2012), thus in stations near petrochemical complexes, wharf, and urban areas pollutants may be adsorbed to microplastic fibers and be transferred to protected areas of Hara forest. Generally, the most common microplastics were fibrous (71%), followed by a polyhedron, planar and spherical, which accounted for 14, 11 and 4% of the total microplastics in sediments, respectively. In the present study, there was no predominance of a particular microplastic shape across sampling sites and a great variety of shapes of plastic debris were uniformly distributed.

The results showed that the dominant color of the detected microplastics in the sediments and bivalves was red (34%) and blue (52%), respectively (Figs. S7 and S8). The high diversity of microplastics color in both, sediment and bivalves demonstrates that the particles come from a wide range of origins (Gallagher et al., 2016). The color of microplastics in sediments was black/gray (33%), blue/green (7%), red/pink (34%), yellow/orange (13%) and white/transparent (14%), and in bivalves it was black/gray (32%), blue/green (53%), red/pink (12%) (Figs. S7 and S8). These colored and small plastics can be consumed by marine fauna and birds and can affect their health (Costa et al., 2010).

The size distributions of microplastics are represented in Fig. S9 for sediment and in Fig. S10 for bivalve. The most common size of microplastics found in sediment and bivalve were between 100 and 250 μm. It should be noted that microplastics smaller than100 μm were not visible in the optic microscope used, and thus their abundance in the samples may be more than what is reported here. Nonetheless, the results revealed that the proportion of microplastics < 250 μm in both sediment and bivalve samples were more abundant. Indeed, only particles smaller than 250 μm were detected in the bivalves, confirming the high possibility of fine particles ingestion by aquatic organisms (Costa et al., 2010). In general, large microplastics are less abundant in sediments (Martins and Sobral, 2011). Plastic fragments ranging in size between 250 to 500 μm made 20% of the total microplastics found, followed by fragments with 500 to 1000 μm and 1000 to 5000 μm making 8% and 3% occurrence of the total plastics in sediment samples.

The chemical structure analysis of microplastic particles by μ-Raman indicated that most of them in the sediments are made of polyethylene terephthalate (PET) and polypropylene (PP), similar to other Persian Gulf studies (Kor and Mehdinia, 2020). Nabizadeh et al. (2019) also reported that the dominant chemical structure of microplastics in the Bandar Abbas coastline are expanded polystyrene (EPS), PE, PET, and PP. The existence of PE, PET, and PP particles in the samples can be due to the storage container, plastic bags, ropes, drinks bottles, fishing gears, and bottle caps (Kershaw et al., 2015). A similar study in the region revealed that PET and polyamide (nylon) particles are dominant (Naji et al., 2017). The sampling location, sampling strategy, and microplastics extraction methods, however, can result in different outcomes. Also, the density difference of PET, nylon, and PP can influence the distance of their transportation and settling (Duis and Coors, 2016). Microplastics with low specific density allow them easily to be transported by tides, waves, currents or rivers (Zhang, 2017). Finally, PE and PP have been reported previously as the two most common polymer types with widespread distribution in aquatic environments and our results are not surprising considering their widespread usage (Erni-Cassola et al., 2019; Horton et al., 2017).

It is worth mentioning that a wide range of marine organisms, including bivalves, zooplankton, fish, invertebrates, birds, and cetaceans, incidentally take up microplastics from sediment or the water column because they mistake them for food (Andrady, 2017; Cole et al., 2013). In the current study, all of the microplastics in the bivalve are made up of PP. This indicates that environmental microplastics can enter (mostly by ingestion pathway) and accumulate in bivalves. Hwang et al. (2019) investigated the cellular responses of secondary polypropylene (PP) particles and found that PP particles have low cytotoxicity effects in size and concentration manner, however, at high concentration (>500 μg/mL), small size (< 20 μm) of PP particles can increase that effect. While other studies indicated the high adsorption capacity of microplastic particles for pollutants (Daugherty, 2016; Yu et al., 2019; Abbasi et al., 2020).

4. Conclusion

This study has evaluated the concentrations of heavy metals and microplastics in the edible tissues of bivalves and coastal sediments in the Northern Persian Gulf. The mean EF values are more than 5 for As, Mn, Cr, and Ni, which indicates the anthropogenic contribution to pollution in the sediment samples. The MPI revealed that sediments are moderate to severely polluted, mostly from anthropogenic sources and this would justify policies to regenerate the area and reduce anthropogenic pollution. Moreover, the MRI results indicated considerable to very high ecological risk at some stations. This clearly supports the assumption that human impacts play an important role in the enrichment of heavy metals in sediment samples in the study area. Geochemical fractionation data indicated that >50% of As, Mn, Zn, and Co can be regarded as easily mobilizable and bioavailable. The results of the PCA-MLR model revealed the high influence of geogenic and anthropogenic sources such as crude oil transporting tankers, urban and agricultural wastewaters, and emissions form ship traffics and boats engines, on sediments chemistry. The potential health risk of heavy metals from investigated bivalve samples evaluated by the THQ and TR suggested that the exposure doses of most elements for human consumption were safe for non-carcinogenic and carcinogenic risk, except for As with a high TR value. The current study shows the presence of microplastics in bivalves and their color, size, and shape in the sediments and bivalves are similar. Microplastics in sediments can be ingested by aquatic organisms, and they could be a vector for transport and enhance exposure to heavy metals, among other pollutants. A general conclusion can be inferred: contaminants are moving among different environments including sediments, water, aquatic organisms, and air. The similarity of observed microplastics and heavy metals in sediments and aquatic organisms in the current study confirms this issue. The produced plastics and any other contaminants such as heavy metals will eventually reach the top of the food chain, humans, and endanger or health.

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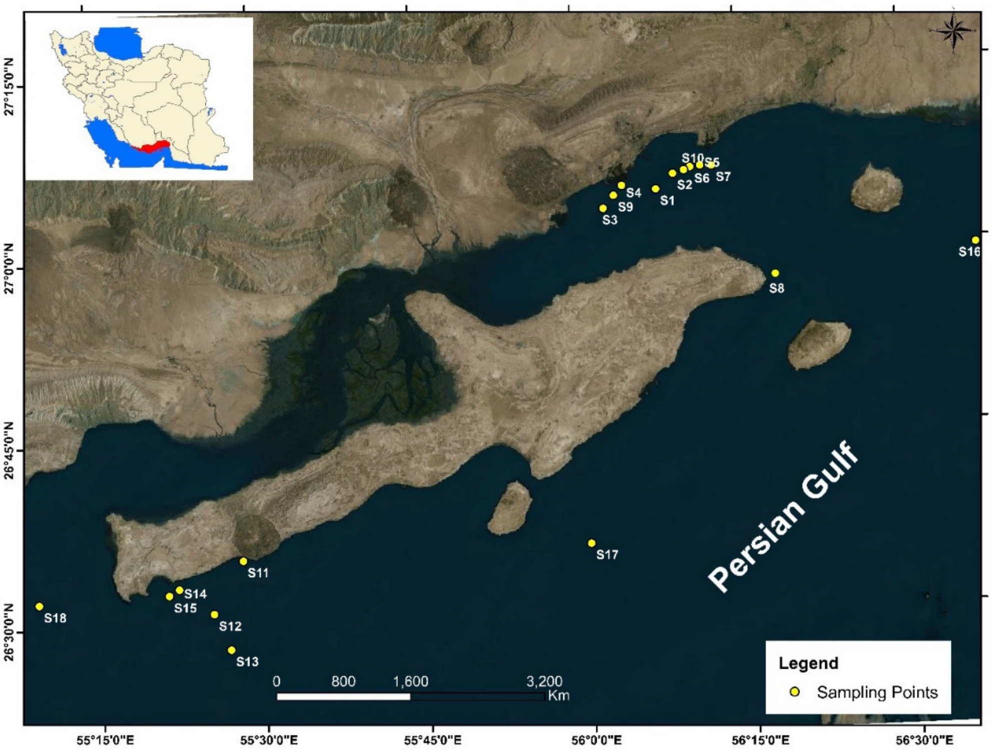
Figure captions:

Fig. 1. The sediment sampling stations in the coastal line of Hormozgan Province

Fig. 2. Box-plot of EF for heavy metals in the sediments of Persian Gulf

Fig. 3. Images of microplastics using (a) fluorescence microscopy and (b) Scanning electron microscopy coupled with an energy dispersive X-ray (SEM/EDX)



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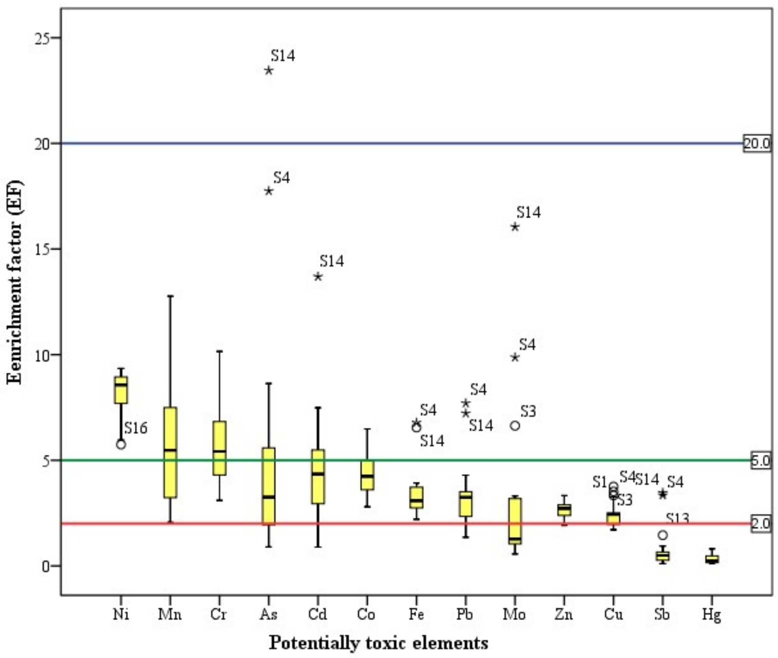

Fig. 2. Box-plot of EF for heavy metals in the sediments of Persian Gulf

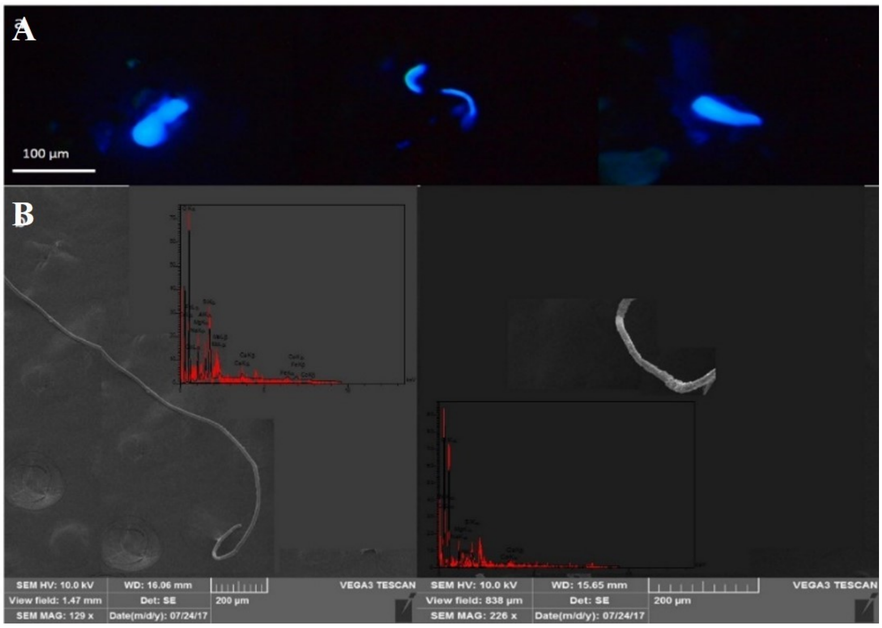

Fig. 3. Images of microplastics using (a) fluorescence microscopy and (b) Scanning electron microscopy coupled with an energy dispersive X-ray (SEM/EDX).

Table 1 Summary of heavy metals concentration in sediments and bivalves from Persian Gulf and comparison with average shale and upper continental crust (mg kg^{-1}).

Element	**Sediment (n=18)**					**Avg. Shale** [b]	**UCC** [c]	**Bivalve (n=7)**				
	Min	Max	Mean	Median	CV [a]			Min	Max	Mean	Median	CV [a]
Mo	0.21	3.86	0.79	0.48	1.12	2.6	1.1	ND [d]	2.84	1.07	1.03	1.06
Cu	5.05	37.86	14.87	11.01	0.59	45	28	9.00	793	153	31	1.86
Pb	5.58	12.36	7.42	6.69	0.27	20	17	ND	2.75	0.90	0.73	1.02
Zn	12.70	61.90	32.51	26.20	0.49	95	67	60.53	4314.51	787.11	122.17	2.00
Ni	25.80	130	71.38	59.25	0.45	68	47	ND	4.28	1.61	1.58	0.92
Co	4.30	16	9.89	8.75	0.35	19	17.3	ND	3.74	1.36	0.74	1.14
Hg	ND	0.03	0.02	0.01	0.52	0.4	0.05	ND	0.34	0.12	0.04	1.05
As	3.50	14.10	5.92	4.55	0.50	13	4.8	ND	173.30	55.51	37.82	1.09
Cd	0.08	0.22	0.15	0.15	0.24	0.3	0.09	0.07	0.95	0.40	0.31	0.82
Sb	0.05	0.27	0.10	0.08	0.60	1.5	0.4	ND	0.29	0.05	0.01	2.28
Cr	28.30	86.60	59.22	57.65	0.28	90	92	0.30	4.89	2.05	1.80	0.69
Al	3700	23900	10522.22	9600	0.52	80000	154000	161	5457	1141.29	329	1.68
Fe	12500	32300	18488.89	17400	0.30	47200	50400	287	4461	1095	545	1.36
Mn	382	615	505.44	516	0.13	850	1000	12	166	57	21	1.14

[a] CV, coefficient of variation

[b] Average shale (mg kg^{-1}) refer to Turekian and Wedepohl, (1961)

[c] UCC, data of elemental abundance in upper continental crust is from Rudnick and Gao, (2003)

[d] ND, represents not-detected

Table 2 Loadings of heavy metals on component matrices and source contribution (in %) calculated using MLR model.

	Component			
	PC1	PC2	PC3	PC4
Al	**0.98**	0.03	-0.08	-0.08
Zn	**0.95**	0.25	0.06	0.05
Ni	**0.94**	0.23	-0.11	0.15
Fe	**0.94**	0.02	0.24	-0.15
Co	**0.92**	0.31	-0.08	0.08
Cr	**0.91**	0.30	-0.03	-0.08
Cu	**0.91**	0.28	0.26	0.10
Pb	**0.67**	0.25	0.58	0.21
Mn	0.34	**0.81**	0.14	0.10
Hg	0.08	**0.80**	-0.23	-0.09
Mo	0.23	-0.06	**0.87**	-0.02
As	-0.52	-0.47	**0.60**	0.14
Sb	-0.52	-0.51	**0.59**	0.04
Cd	0.01	-0.03	0.05	**0.99**
% of Variance	52.06	15.98	14.50	8.13
MLR (%)	61.35	12.79	18.76	7.11

Bold signifies good correlations (>0.5).

Table 3 Investigated pollution indices based on ecological risk (Er^i) with enrichment factor (EF) and modified potential ecological risks (MRI) for sediment assessment in Persian Gulf.

Sample	**Er^i**								**MRI**	**Ecological risk**
	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn		
S1	19.48	109.35	9.89	16.68	16.49	18.99	46.59	3.33	240.81	Moderate
S2	38.91	141.18	13.67	12.24	17.56	14.12	44.72	2.89	285.28	Moderate
S3	15.13	62.57	8.60	18.80	13.81	5.81	37.56	2.87	165.15	Moderate
S4	177.46	223.26	20.30	17.61	38.51	6.98	38.44	3.17	525.73	Considerable
S5	35.54	135.21	13.30	12.11	16.65	18.03	44.74	2.68	278.25	Moderate
S6	31.60	118.92	14.51	12.42	15.30	32.43	45.47	2.82	273.47	Moderate
S7	55.86	160.00	15.02	11.50	17.17	23.38	42.44	2.72	328.09	Considerable
S8	25.03	149.15	9.34	12.53	11.36	5.42	41.72	2.60	257.16	Moderate
S9	22.89	125.62	10.11	16.36	15.97	7.93	43.22	3.22	245.32	Moderate
S10	33.48	164.71	13.86	12.35	17.15	24.71	45.24	2.93	314.43	Considerable
S11	16.46	83.72	8.30	11.88	10.09	8.84	44.46	2.27	186.01	Moderate
S12	28.25	85.25	9.22	12.31	13.00	7.21	41.95	2.46	199.65	Moderate
S13	74.97	218.18	11.57	9.83	21.13	5.45	44.71	2.39	388.22	Considerable
S14	234.51	410.81	13.60	12.13	36.11	17.30	41.02	2.89	768.37	Very High
S15	86.37	224.56	11.57	9.70	21.47	29.47	46.75	2.72	432.62	Considerable
S16	9.01	26.78	6.21	8.97	6.76	5.69	28.72	2.18	94.33	Low
S17	36.62	99.17	7.93	8.58	11.70	10.58	29.80	1.92	206.31	Moderate
S18	19.10	88.28	8.12	9.78	8.01	6.07	36.39	2.12	177.87	Moderate

Table 4 Target hazard quotient (THQ), total THQ (TTHQ), and target cancer risk (TR) of six bivalve species consumption in two age classes.

Species	Age class	Target hazard quotient (THQ)											TTHQ
		Mo	Cu	Pb [a]	Zn	Ni	Co	Hg	As	Cd	Cr	Mn	
S. cucullata	Children	2.7E-04	2.9E-02	2.7E-04	2.2E-02	1.1E-04	2.7E-03	3.2E-03	7.5E-01	1.1E-03	2.8E-03	3.0E-04	0.81
C. callipygo		1.5E-03	5.9E-04	2.9E-04	5.9E-04	5.6E-04	3.2E-02	9.4E-04	4.6E-01	8.0E-04	1.1E-03	3.9E-04	0.50
B. helblingii		1.1E-03	8.5E-04	9.4E-04	1.7E-03	2.0E-04	6.5E-03	5.3E-03	7.9E-01	2.5E-03	1.9E-03	4.8E-04	0.81
S. brevis		3.9E-06	2.0E-03	6.2E-04	5.2E-04	1.6E-04	6.4E-03	6.5E-04	2.9E-01	4.7E-04	2.0E-03	3.2E-04	0.30
A. umbonella		8.0E-04	1.1E-03	2.0E-03	5.2E-04	3.1E-04	3.0E-02	7.8E-04	3.3E-01	5.2E-04	1.6E-03	3.1E-03	0.37
Telescopium		3.9E-06	6.2E-03	1.8E-04	1.1E-03	9.8E-07	2.0E-03	8.7E-03	6.5E-05	8.6E-04	2.6E-04	2.5E-03	0.02
Mean		6.1E-04	6.7E-03	7.1E-04	4.3E-03	2.2E-04	1.3E-02	3.3E-03	4.4E-01	1.0E-03	1.6E-03	1.2E-03	0.47
S. cucullata	Adults	6.2E-05	6.7E-03	6.1E-05	5.0E-03	2.6E-05	6.1E-04	7.2E-04	1.7E-01	2.5E-04	6.4E-04	6.8E-05	0.19
C. callipygo		3.4E-04	1.3E-04	6.5E-05	1.4E-04	1.3E-04	7.4E-03	2.1E-04	1.0E-01	1.8E-04	2.6E-04	8.9E-05	0.11
B. helblingii		2.5E-04	1.9E-04	2.1E-04	3.9E-04	4.7E-05	1.5E-03	1.2E-03	1.8E-01	5.7E-04	4.2E-04	1.1E-04	0.19
S. brevis		8.9E-07	4.6E-04	1.4E-04	1.2E-04	3.7E-05	1.5E-03	1.5E-04	6.6E-02	1.1E-04	4.7E-04	7.2E-05	0.07
A. umbonella		1.8E-04	2.5E-04	4.5E-04	1.2E-04	7.1E-05	6.9E-03	1.8E-04	7.5E-02	1.2E-04	3.6E-04	7.0E-04	0.08
Telescopium		8.9E-07	1.4E-03	4.1E-05	2.4E-04	2.2E-07	4.7E-04	2.0E-03	1.5E-05	2.0E-04	6.0E-05	5.8E-04	0.01
Mean		1.4E-04	1.5E-03	1.6E-04	9.9E-04	5.1E-05	3.1E-03	7.5E-04	1.0E-01	2.4E-04	3.7E-04	2.7E-04	0.11
		Target carcinogens risk (TR)											**Total TR**
S. cucullata	Children	-	-	8.2E-06	-	3.9E-06	-	-	3.4E-04	-	4.2E-06	-	3.5E-04
C. callipygo		-	-	8.8E-06	-	1.9E-05	-	-	2.1E-04	-	1.7E-06	-	2.4E-04
B. helblingii		-	-	2.9E-05	-	7.0E-06	-	-	3.6E-04	-	2.8E-06	-	3.9E-04
S. brevis		-	-	1.9E-05	-	5.5E-06	-	-	1.3E-04	-	3.1E-06	-	1.6E-04

A. umbonella		-	-	6.1E-05	-	1.1E-05	-	-	1.5E-04	-	2.3E-06	-	2.2E-04
Telescopium		-	-	5.5E-06	-	3.3E-08	-	-	2.9E-08	-	4.0E-07	-	6.0E-06
Mean		-	-	2.2E-05		7.6E-06	-	-	2.0E-04	-	2.4E-06	-	2.3E-04
S. cucullata	Adults	-	-	1.9E-06	-	8.9E-07	-	-	7.7E-05	-	9.6E-07	-	8.1E-05
C. callipygo		-	-	2.0E-06	-	4.3E-06	-	-	4.7E-05	-	3.9E-07	-	5.4E-05
B. helblingii		-	-	6.6E-06	-	1.6E-06	-	-	8.1E-05	-	6.4E-07	-	9.0E-05
S. brevis		-	-	4.4E-06	-	1.3E-06	-	-	3.0E-05	-	7.0E-07	-	3.6E-05
A. umbonella		-	-	1.4E-05	-	2.4E-06	-	-	3.4E-05	-	5.4E-07	-	5.1E-05
Telescopium		-	-	1.3E-06	-	7.6E-09	-	-	6.7E-09	-	9.1E-08	-	1.4E-06
Mean		-	-	5.0E-06		1.7E-06			4.5E-05	-	5.5E-07	-	5.2E-05
	RfD [b]	0.005	0.04	0.0036	0.30	0.02	0.0003	0.0001	0.0003	0.0010	0.003	0.14	
	CSF [c]	NA	NA	8.5	NA	1.7	NA	NA	1.5	NA	0.5	NA	

[a] RfD (Reference dose, mg/kg/day)of Pb obtained by Liu et al. (2017).
[b and c] RfD and CSF (Oral carcinogenic slope factor, mg/kg/day) of heavy metals as published by USEPA (2017).
NA, not available

Supplementary Information

Source and risk assessment of heavy metals and microplastics in bivalves and coastal sediments of the Northern Persian Gulf, Hormogzan Province

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1. Material and Methods

Table S1 Details of sediment sampling stations

Number	Name of station	X	Y
S1	Shahid Bahorar Port	2707771	5612267
S2	Suru	2709810	5614966
S3	Shahid Rajaei Port	2705386	5604082
S4	Tavanir station	2708437	5607051
S5	Fishery station	2710810	5619191
S6	Gursuzan	2710631	5617638
S7	Velayat station	2710726	5620965
S8	Geshm water desalinization	2695082	5628424
S9	Oil refinery	2707115	5605741
S10	Hotel Amin	2710252	5616665
S11	Mangrove Forest	2658758	5546957
S12	Mangrove Forest	2651583	5542241
S13	Mangrove Forest	2648033	5544674
S14	Mangrove Forest	2655090	5537027
S15	Mangrove Forest	2656260	5535469
S16	East of Hormozgan Province	2698858	5661002
S17	South of Geshm	2659482	5600314
S18	West of Hormozgan Province	2653488	5515507

Table S2 The characteristics of the bivalves.

No	Bivales species	Family	Location
B1	Saccostrea cucullata	Ostreidae	Bandar Lengeh
B2	Circenita callipyga	Veneridae	Bandar Lengeh
B3	Barbatia helblingii	Arcidae	Bandar Lengeh
B4	Solen brevis	Solenidae	Bandar Abbas
B5	Amiantis umbonella	Veneridae	Bandar Abbas
B6	Telescopium telescopium	Potamididae	Bandar Abbas
B7	Saccostrea cucullate	Ostreidae	Bandar Abbas

Table S3 Four-step sequential extraction and pseudo-total digestion protocols.

Step	Fraction	Chemical reagents and experimental protocols
F1	Exchangeable	A total of 40 mL of 0.11 mol L^{-1} of acetic acid (CH_3COOH, pH 2.85) was added to 1.0 g dry weight of sediment samples and shaken for 16 h and the extract was separated by centrifugation at 3500 rpm for 15 min
F2	Reducible	A total of 40 mL of 0.1 mol L^{-1} of hydroxylammonium chloride ($NH_2OH \cdot HCl$, pH 2) was added to the residue from step 1 and the extraction performed as above
F3	Oxidisable	A total of 10 mL of hydrogen peroxide (H_2O_2, 30%) was added to the residue from step 2 at room temperature for 1 h, and then evaporated at 85 ± 2 °C until the volume was reduced to near dryness. Additional 25 mL of 1 mol L^{-1} of ammonium acetate (NH_4CH_3COO, pH 2) was added and shaken for 16 h. The extraction performed as above
F4	Residual	The residue from Step 3 was digested in aqua regia (3:1 conc. $HCl:HNO_3$)

1.1. Chemical analysis of bivalves

Approximately 5.0 g (± 0.1 g) of wet composite tissue samples were freeze-dried by the Freeze-drying instrument (model Zirbus technology GmbH VaCo5, Germany) at -50°C for 24 hours. The samples were then ground and homogenized with a mortar and pestle. Finally, the homogenized samples were sent to the laboratory of Zarazma Mineral Studies Company, Iran. Concentrations of Al, As, Cd, Co, Cr, Cu, Fe, Hg, Mo, Mn, Ni, Pb, Sb, and Zn were determined using ICP-MS.

From each freeze-dried homogenized sample, 0.5 g was microwave digested with 2.5 mL of ultrapure H_2SO_4, 4.0 mL of ultrapure HNO_3 and 1.5 mL of H_2O_2. The ramp and hold time were 10 (from 30 to 90 ºC) and 20 min, respectively. The final volume of each digested sample solution was made to 50 ml with deionized distilled water and analyzed for metals (except for Hg). Hg was extracted by Aqua Regia at 90°C and measured by the cold vapor flow injection technique (Perkin Elmer FIMS 100 cold vapor Hg analyzer). Hg in the resulting solution was oxidized to the stable divalent form. Since the Hg concentration was determined via the absorption of light at 253.7 nm by Hg vapor, Hg (II) was reduced to the volatile free atomic state using stannous chloride ($SnCl_2$). Argon was applied for the Hg vapor release from the liquid mixture of sample and reductant solution in a closed reaction system to liberate and to transport the Hg atoms into an absorption cell.

1.2 Quality assurance (QA) and quality control (QC)

To avoid fiber and plastic pollution during the extraction step of the samples in the laboratory, all reagents and distilled water were filtered through S&S blue band filters. Moreover, streel devices were used to collect and keep the samples and, when necessary, samples and containers were protected by Al foil. Two samples were done in duplicate. All steps were carried out without bivalve and sediment samples (blank samples) to determine likely pollution arising from devices. Also, two wide dishes full of filtered water were fixed on the laboratory bench at the step as a control for the microplastics to detect airborne pollution during the extractions. The results revealed no contamination in the blank samples and control dishes.

Table S4. Threshold values for sediment quality classification base on the contamination factor (CF), the modified pollution index (MPI), modified ecological risk index (MRI) and the potential ecological risk index (Er^i).

Class	MPI [a]	CF [b]	Sediment qualification	MRI [c]	Er^i [d]	Ecological risk
0	MPI < 1	-	Unpolluted	-	-	-
1	1 < MPI < 2	CF < 1	Slightly polluted	MRI <150	$Er^i < 40$	low ecological risk
2	2 < MPI <3	1 ≤ CF < 3	Moderately polluted	150 ≤ MRI < 300	$40 \leq Er^i < 80$	Moderately ecological risk
3	3 < MPI < 5	3 ≤ CF < 6	Moderately-heavily polluted	300 ≤ MRI < 600	$80 \leq Er^i < 160$	Considerable ecological risk
4	5 < MPI < 10	-	Heavily polluted	-	$160 \leq Er^i < 320$	High ecological risk
5	MPI >10	CF ≥ 6	Severely polluted	MRI ≥ 600	$Er^i \geq 320$	Very high ecological risk

[a] adapted from Brady et al. (2015)

[b and d] adapted from Hakanson (1980)

[c] adapted from Zhang et al. (2017)

2. Results and discussion

Table S5 Concentrations of heavy metals in sediments and bivalves of the Northern Persian Gulf, Hormogzan Province and comparison with those of other regions (mg kg^{-1}).

Sediment	Cu	Pb	Zn	Ni	Co	Hg	As	Cd	Cr	Al	Fe	Mn	References
Northern Persian Gulf (Hormogzan Province)	14.9	7.4	32.5	71.4	9.9	0.02	5.9	0.15	59.2	10522.2	18488.9	505.4	This study
Nemrut Bay (Turkey)	29.5	60.4	182.9	50.4	-	0.006	17.5	0.14	69.6	-	28497.9	298.3	Esen et al. 2010
Thermaikos Gulf (Greece)	80	77	184	-	-	-	-	-	47	-	-	-	Christophoridis et al. 2009
Qatar coast (Persian Gulf)	4.4	2.4	-	11.2	1.3	0.002	4.3	-	27.1	-	-	70.1	De Mora et al. 2004
Assaluyeh coast (Persian Gulf)	15.4	3.4	21.1	19	2.2	0.12	3.7	-	16.1	-	-	168.7	Delshab et al. 2016
Meiliang Bay (China)	0.70	0.6	-	-	-	-	-	0.10	1.10	-	-	-	Rajeshkumar et al. 2018
Eastern coast of Thailand Gulf	39.4	18.8	43.6	-	-	0.04	-	0.04	-	-	230000	4500	Thongra-Ar et al. 2008
Shandong Peninsula (China)	19.5	15.5	26.07	-	-	0.03	7.84	0.15	22.61	-	-	-	Liu et al. 2021
Gorgan Bay (Iran)	16.8	7.4	29.5	16.6	-	-	8.1	-	17.9	13000	-	-	Gholizadeh and Patimar 2018
Nellore coast (India)	3.6	2.5	3.9	2.9	3.1	-	-	1.8	6.1	854	2189	83	Jha et al. 2019
Gulf of Oman	2.8	1.1	5.68	37.54	3.07	0.002	2.51	0.16	70.96	7072	5429.83	122.4	De Mora et al. 2004
Bivalve													
Northern Persian Gulf (Hormogzan Province)	136.75	1.5	704	1.41	1.44	0.11	48.97	0.37	2.46	1033.4	1042.88	55.25	This study
Nellore coast (India)	2.8	0.6	18.1	0.6	0.29	-	-	0.79	0.66	39.8	141.4	11.64	Jha et al. 2019
Shandong Peninsula (China)	0.27	0.08	4.36	-	-	0.005	1.13	0.15	0.13	-	-	-	Liu et al. 2021
Gulf of Oman	159.6	0.51	1084	1.57	0.36	0.12	14.66	14.24	1.42	-	199.8	4.86	De Mora et al. 2004
Laizhou Bay (China)	10.06	1.40	116.04	-	-	0.06	16.65	2.88	2.74	-	-	-	Liu et al. 2017
Assaluyeh port coasts (Persian Gulf)	283.79	0.46	1777.35	0.26	-	0.018	1.30	1.54	0.06	-	-	-	Delshab et al. 2017
Sydney estuary (Australia)	1419	8.9	6518	2.8	-	-	-	2.5	1.2	-	-	-	Birch et al. 2014
Parangipettai (India)	12.45	1.31	44.14	0.40	0.24	-	-	0.79	0.48	37.21	91	7.34	Satheeswaran et al. 2019
Southern Coast of Korea	32.48	0.15	154.38	0.15		0.009	2.69	0.59	0.22				Mok et al. 2015
Musa Estuary (Persian Gulf)	378.62	1.75	506.05	9.71	3.87	12	4.18	7.51	3.52	926.1	1063.75	23.24	Lahijanzadeh et al. 2019

Table S6 The contamination factor (CF) and modified pollution index (MPI) of sediments in the study area.

Sample	Contamination factor (CF)													MPI	Sediment qualification
	Mo	Cu	Pb	Zn	Ni	Co	Cr	Hg	As	Cd	Fe	Sb	Mn		
S1	0.20	0.58	0.57	0.58	1.62	0.73	0.86	0.08	0.34	0.63	0.48	0.04	0.67	7.0	Heavily polluted
S2	0.11	0.21	0.30	0.25	0.76	0.42	0.58	0.03	0.33	0.40	0.28	0.05	0.61	6.9	Heavily polluted
S3	1.48	0.84	0.62	0.64	1.68	0.76	0.96	0.03	0.34	0.47	0.61	0.05	0.72	5.8	Heavily polluted
S4	0.53	0.19	0.41	0.17	0.41	0.23	0.55	0.01	0.95	0.40	0.36	0.18	0.45	13.5	Severely polluted
S5	0.10	0.21	0.30	0.24	0.79	0.46	0.59	0.04	0.32	0.40	0.30	0.05	0.63	6.9	Heavily polluted
S6	0.13	0.23	0.28	0.26	0.84	0.44	0.67	0.08	0.29	0.37	0.32	0.05	0.61	7.0	Heavily polluted
S7	0.08	0.19	0.28	0.22	0.69	0.42	0.61	0.05	0.45	0.43	0.30	0.05	0.66	6.7	Heavily polluted
S8	0.16	0.37	0.34	0.38	1.23	0.53	0.69	0.02	0.37	0.73	0.37	0.04	0.58	6.3	Heavily polluted
S9	0.16	0.49	0.48	0.49	1.31	0.65	0.76	0.03	0.35	0.63	0.43	0.07	0.67	6.5	Heavily polluted
S10	0.12	0.21	0.29	0.25	0.77	0.42	0.59	0.05	0.28	0.47	0.29	0.05	0.62	7.0	Heavily polluted
S11	0.20	0.51	0.43	0.49	1.91	0.84	0.89	0.05	0.35	0.60	0.51	0.05	0.67	6.6	Heavily polluted
S12	0.14	0.38	0.40	0.38	1.28	0.64	0.70	0.03	0.43	0.43	0.42	0.05	0.61	6.3	Heavily polluted
S13	0.22	0.14	0.29	0.16	0.61	0.34	0.40	0.01	0.52	0.50	0.26	0.10	0.52	7.1	Heavily polluted
S14	0.74	0.11	0.33	0.13	0.38	0.30	0.31	0.02	1.08	0.63	0.30	0.16	0.59	17.6	Severely polluted
S15	0.22	0.14	0.31	0.19	0.67	0.38	0.41	0.05	0.62	0.53	0.28	0.07	0.55	7.4	Heavily polluted
S16	0.17	0.54	0.40	0.65	1.72	0.84	0.93	0.04	0.27	0.27	0.68	0.03	0.62	4.3	Moderately-heavily polluted
S17	0.50	0.26	0.35	0.29	0.90	0.46	0.60	0.04	0.55	0.50	0.43	0.07	0.47	4.6	Moderately-heavily polluted
S18	0.23	0.35	0.29	0.38	1.32	0.53	0.74	0.03	0.35	0.53	0.40	0.05	0.47	5.4	Heavily polluted

Table S7 Fractionation of heavy metals in different operationally defined phases (%)

Element	Al	As	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
F1 (%)	0.15	21.52	21.63	1.87	13.04	0.15	20.41	8.03	1.56	19.88
F2 (%)	1.73	8.18	14.67	3.05	4.69	5.64	39.54	10.29	29.91	37.01
F3 (%)	0.06	45.77	17.42	4.81	20.73	0.03	1.64	4.51	1.96	1.46
Mobile phases (%)	1.94	75.46	53.71	9.73	38.46	5.83	61.58	22.83	33.43	58.35
F4 (%)	98.06	24.54	46.29	90.27	61.54	94.17	38.42	77.17	66.57	41.65

F1: exchangeable and associated with carbonates

F2: associated with easily and moderately reducible Fe and Mn oxyhydroxides

F3: associated with soil organic matter and sulfides

F4: residual fraction

Table S8 Comparison between estimated daily intake (EDI) of heavy metals via the consumption of bivalves (based on heavy metals concentration in wet weight) collected from the Persian Gulf and recommended PTDI (provisional tolerable daily intake) and PTWI (provisional tolerable weekly intake) values.

Species	**Age class**	**EDI (Estimated daily intake,** mg/kg/day**)**										
		Mo	Cu	Pb	Zn	Ni	Co	Hg	As	Cd	Cr	Mn
S. cucullata	Children	0.006	5.7	0.005	31.3	0.011	0.004	0.0015	1.1	0.0053	0.040	6.7
C. callipygo		0.035	0.11	0.005	0.9	0.05	0.047	0.00045	0.7	0.0039	0.017	3.3
B. helblingii		0.026	0.16	0.016	2.5	0.0197	0.009	0.0026	1.1	0.012	0.027	3.3
S. brevis		0.0001	0.39	0.011	0.8	0.016	0.009	0.0003	0.4	0.0023	0.030	7.6
A. umbonella		0.019	0.21	0.034	0.8	0.030	0.04	0.00038	0.5	0.0025	0.023	4.1
Telescopium		0.0001	1.19	0.0031	1.5	0.0001	0.003	0.0042	0.0001	0.0041	0.0038	68.2
Mean		0.015	1.29	0.012	6.3	0.022	0.019	0.0016	0.6	0.005	0.02	15.5
S. cucullata	Adults	0.001	1.29	0.0011	7.15	0.003	0.0009	0.00035	0.25	0.0012	0.009	0.046
C. callipygo		0.008	0.03	0.0011	0.20	0.012	0.011	0.00010	0.15	0.0009	0.004	0.06
B. helblingii		0.006	0.04	0.0037	0.56	0.005	0.002	0.00059	0.26	0.0027	0.006	0.07
S. brevis		0.00002	0.09	0.0025	0.17	0.004	0.002	0.00007	0.10	0.0005	0.007	0.049
A. umbonella		0.004	0.05	0.0078	0.17	0.007	0.010	0.00009	0.11	0.0006	0.005	0.47
Telescopium		0.00002	0.27	0.0007	0.35	0.00002	0.0007	0.00096	0.00002	0.0009	0.0009	0.39
Mean		0.003	0.29	0.0028	1.43	0.005	0.0044	0.00036	0.14	0.001	0.005	0.18
PMTDI [a]		0.01 [c]	0.5	0.0036	1	0.02	0.0016	0.00057	0.0003	0.00083	0.0003	0.06
PTWI [b]		NA	3.5	0.025	7	2.1	0.67	0.0016	0.015	0.007	NA	25.2

[a] PTWI, the provisional tolerable weekly intake (mg/kg/day)[b] PMTDI, the provisional maximum tolerable daily intake (mg/kg/day). PMTDI and PTWI values were summarized by Jović and Stanković (2014) and Mok et al. (2015) from Joint FAO/WHO Expert Committee on Food Additives and U.S. Environmental Protection Agency.[c] The tolerable daily intake (TDI) (µg/kg/day) value for Mo were based on the oral reference doses established by Baars et al. (2001).NA, not available

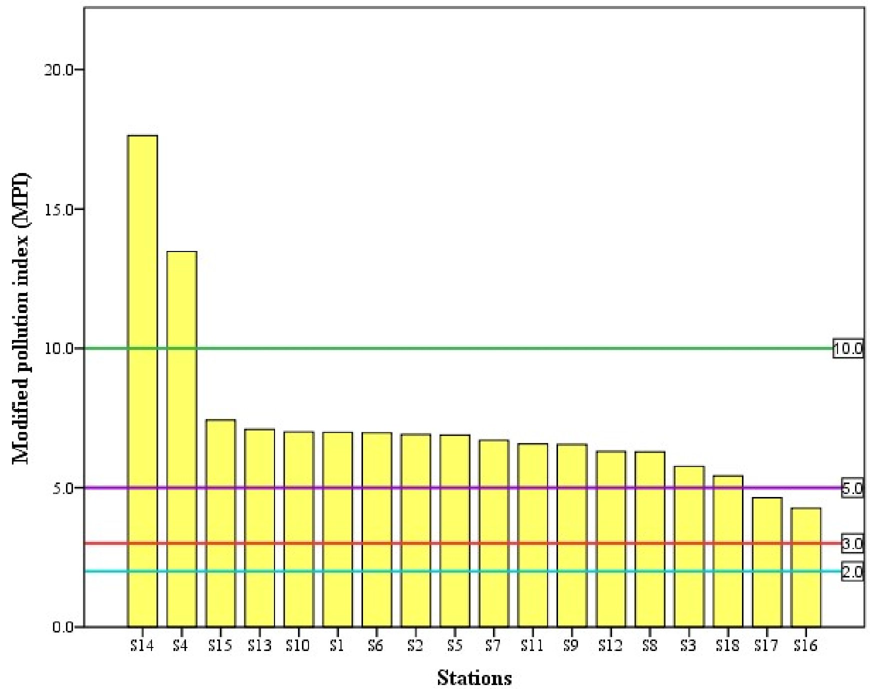

Fig. S1. Sediment quality evaluation using the modified pollution index (MPI) in sediments samples in the study area.

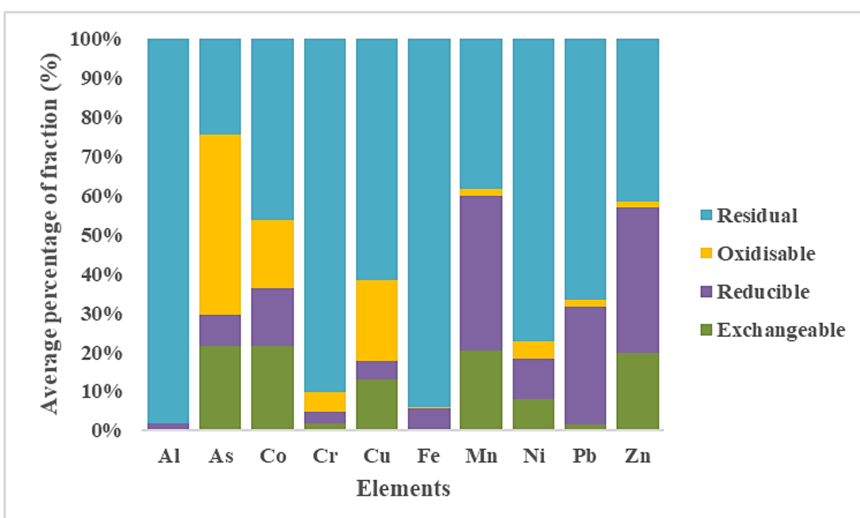

Fig. S2. Fractionation of heavy metals in different operationally phases (%)

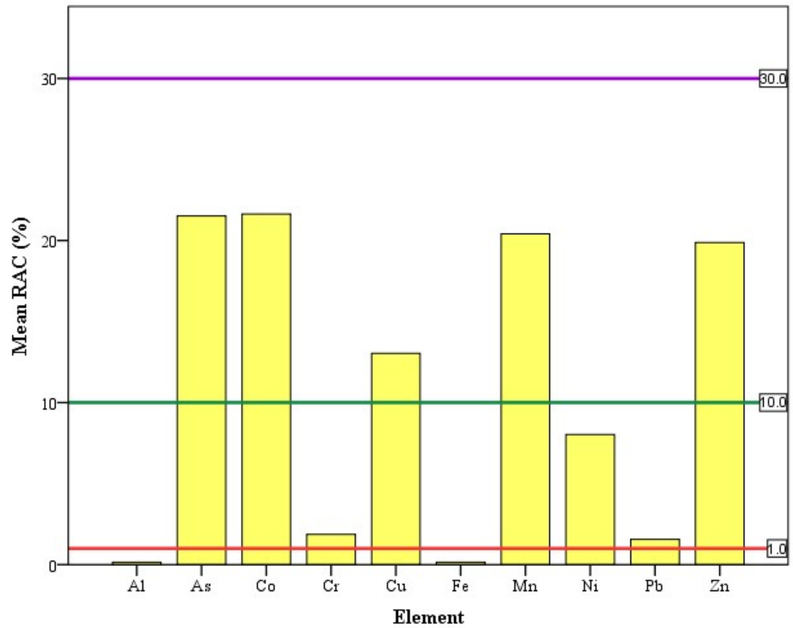

Fig. S3. Risk assessment code (RAC) of heavy metals in the sediments of Persian Gulf

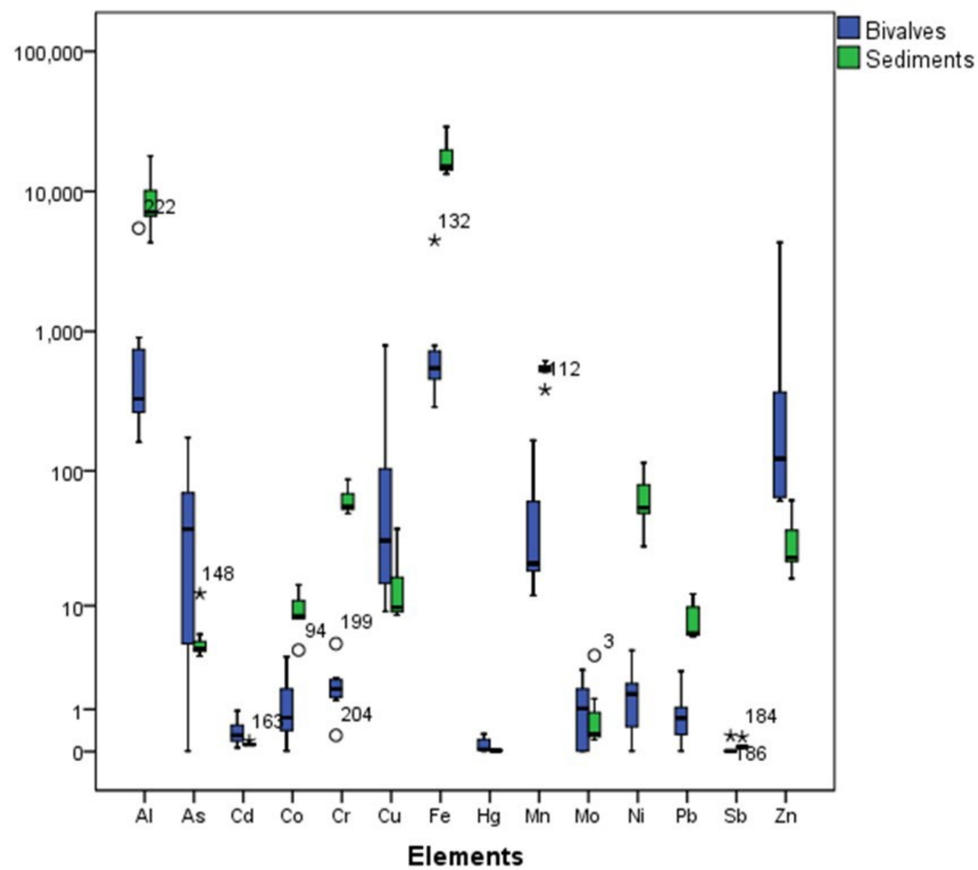

Fig. S4. Comparison of the concentration of heavy metals in sediments and bivalves.

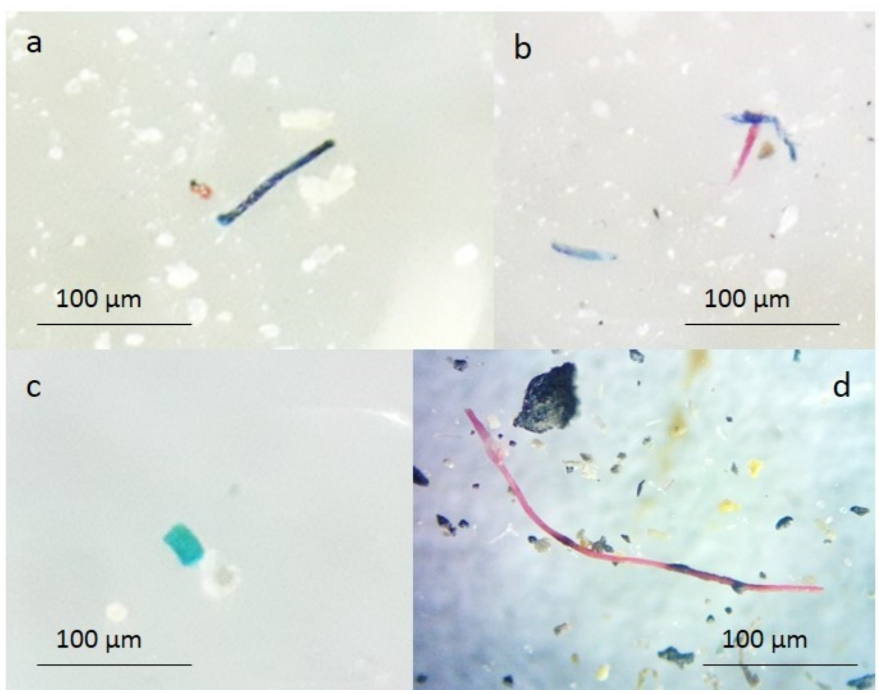

Fig. S5. Optical microscope image of microplastics in sediments and bivalves.

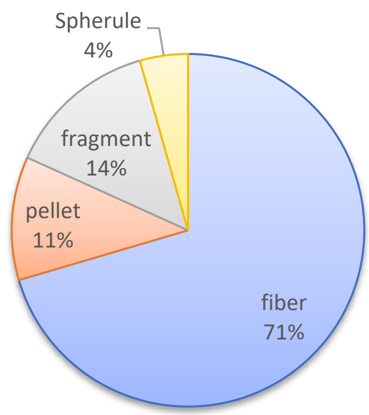

Fig. S6. The shapes of some collected microplastics in sediments.

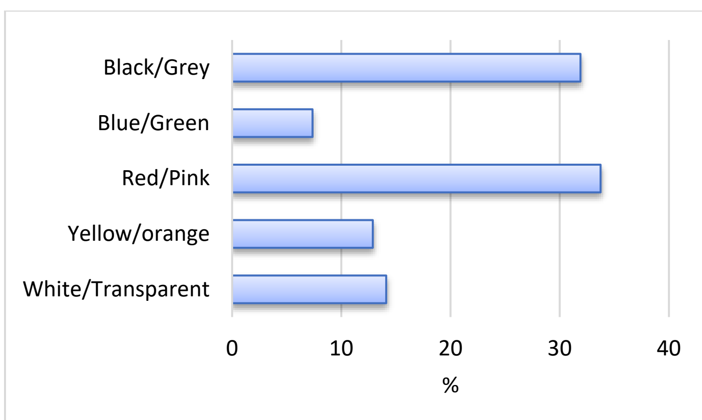

Fig. S7. The overall color distribution of the microplastics observed in the sediments.

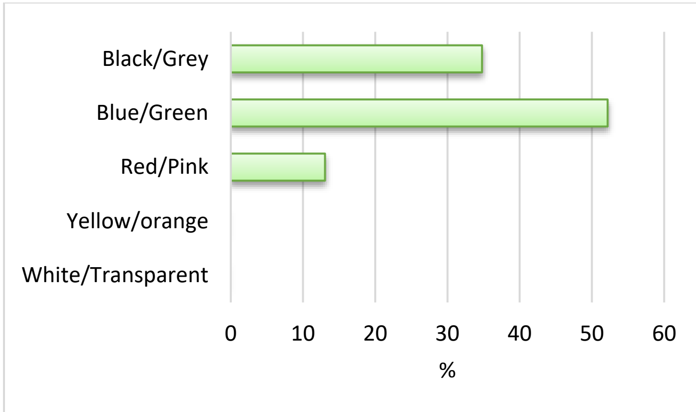

Fig. S8. The overall color distribution of the microplastics observed in the bivalves.

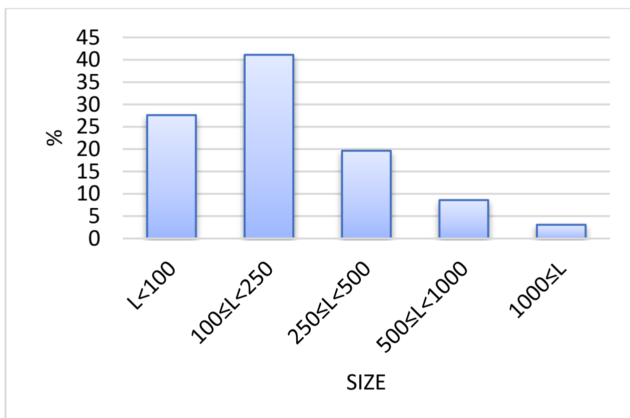

Fig. S9. Abundance percentage of microplastics among different size categories in the sediment.

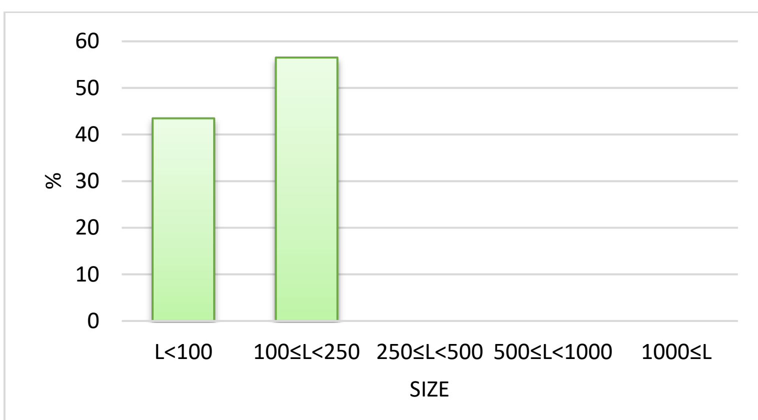

Fig. S10. Abundance percentage of microplastics among different size categories in bivalves.

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