

Effect of fishing vessels on trace metal contamination in sediments of three harbors along Iranian Oman Sea coast

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Abstract In this study, the levels of natural and anthropogenic metal contamination (aluminum (Al), iron (Fe), manganese (Mn), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn)) in surface sediments of three harbors along the Iranian coast of Gulf of Oman were examined and reported for the first time. Effect of grain size, mineralogy, normalization technique using Fe concentrations, and different sediment quality guidelines were discussed. Data from the harbors were compared with other harbors worldwide. Sediments inside the harbors are characterized by moderate and high levels of pollution by trace metals Cu (12–287 $\mu\text{g/g}$), Pb (11–1,780 $\mu\text{g/g}$), Zn (38–547 $\mu\text{g/g}$), Cr (70–2,370 $\mu\text{g/g}$), and Ni (31–116 $\mu\text{g/g}$). However, As and Cd did not show considerable pollution inside and outside the harbors. Considering that there is no industrial activity around the study harbors, the major sources of contamination in the harbors are repairing, fueling, greasing, and painting of fishing ships and boats. Mineralogy of sediments in the study area as well as trace metal concentration in reference samples taken from onshore geological units

confirmed that natural inputs of Cd, Cu, Pb, and Zn in the harbors are very low while most of Ni and Cr contamination in the study area comes from erosion of ophiolitic mélange units at the North of Makran mountains.

Keywords Fishing vessels · Harbors · Metals · Gulf of Oman · Pollution

Introduction

Marine sediments are sensitive indicators for monitoring contaminants in aquatic environments (Pekey et al. 2004). The bottom sediments serve as a reservoir for heavy metals and therefore, deserve special consideration in the planning and design of aquatic pollution researches (Ahdy and Khaled 2009). Once contaminants are released into the aquatic environment, they may be removed from the water column by interacting with suspended solids and subsequently be deposited as bottom sediments (Tang et al. 2008). Although most adsorbed pollutants on the sediments are not readily available for aquatic organisms, the variation of some physical and chemical characteristics (pH, salinity, redox potential, and the content of organic chelators) of the overlying water may provoke the release of the metals back to the aqueous phase, hence under changing environmental conditions sediments may become themselves important pollution sources (Soares et al. 1999).

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Harbors, because of the associated shipping activities, are often sites of serious marine pollution (Denton et al. 2005). As a consequence, there is a rapid decline in the quality of the water and bottom sediments in the harbor, as anthropogenic contaminants such as metals, PCBs and dioxin-like compounds, PAHs, organochloride pesticides (OCPs), oil, radio-nuclides, trace metals, and other pollutants seriously affected fish and other marine life (Arslan et al. 2009; Stronkhorst 2003; PIANC 1999). Heavy metals, unlike other pollutants, are not biodegradable and can accumulate in sediments over time. The contamination of metals in sediments can reach a level that is toxic to aquatic life (Zulkifli et al. 2009).

The objective of this study is to assess the origin, pollution levels (using Sediment Quality Guidelines), and geochemical behavior of metals in the three harbors of Ramin, Beris, and Pasabandar, along the Iranian coast of Gulf of Oman. These harbors are the major fishing centers (especially tuna fish) in Iran; however, there is no previous study of the anthropogenic pollution on this coastal environment. In this paper, we report the first results for trace metal contamination in sediments of these three harbors.

There is no direct river discharges to the harbors. Industrial activity adjacent to the harbors and neighboring cities is ignorable. Therefore, the potential contamination should be mainly derived from the shipping activities and the sewage disposal from vessels and residential area close to these harbors. During the recent

years, control of contamination by shipping operations has not been effective and fishing vessel-related activities are still polluting the harbors.

Setting

The harbors of Ramin, Beris, and Pasabandar are located along the Northern coast of the Gulf Oman in Sistan and Baluchestan province, Southeast extreme of Iran (Fig. 1). The coastal area belongs to the Western part of Makran area. The Makran coast is part of an accretionary wedge which accumulated near Makran subduction margin, it mainly consists of turbidites and ophiolitic mélanges (Farhoudi and Craig 1977; Jacob and Quittmeyer 1979). Along the coast, outcrops are mainly sequences of calcareous Mudstones. The coastal geomorphology characterized by a series of prominent headlands separated by low areas (Page et al. 1979; Snead 1993; Reyss et al. 1998).

The coastline of Iranian Makran is very hot and humid. Mean temperature in January is $>20^{\circ}\text{C}$, and the maximum temperature in August is $>40^{\circ}\text{C}$. Mean annual rainfall is <100 mm, and the rainfall variability is extremely high (Uchupi et al. 2002). The tidal range is generally between 2 and 4 m. Winds and currents are in accordance with the monsoon circulation. The NE monsoon with moderate winds begins in October but is mainly characteristic between November and March, with a maximum in December and January. The SW

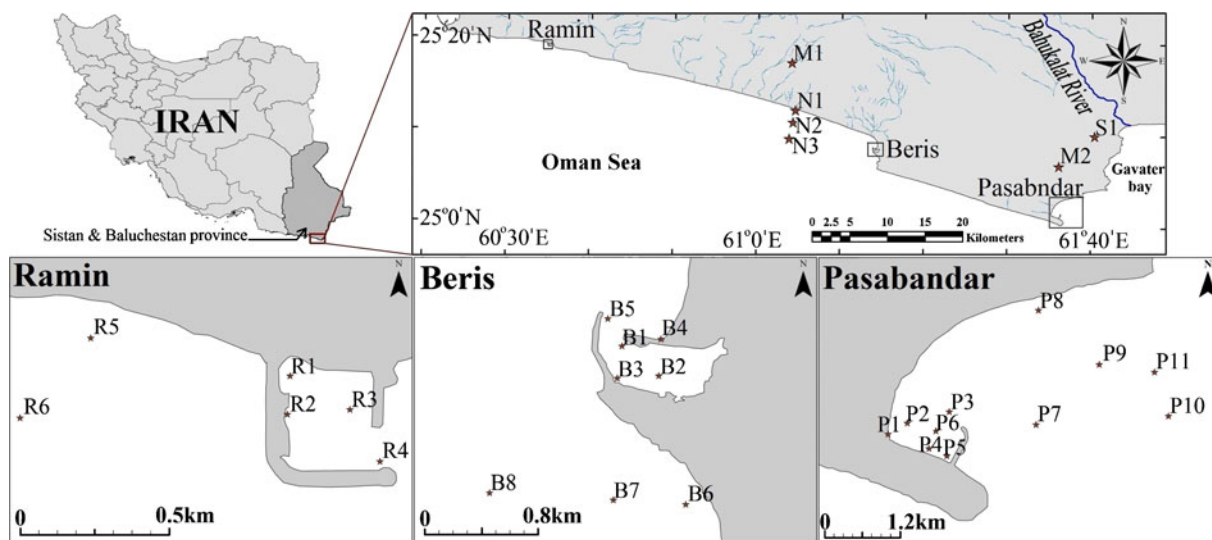


Fig. 1 Location map of studied harbors and sediment sampling sites

monsoon is longer and stronger. It begins in April and is very strong from June to September (Wyrski 1973). During summer monsoon, the sea is very rough and navigation is dangerous, so the fishers stay in harbors and repair their boats and vessels.

Harbors of Sothern Sistan and Baluchestan province, altogether, provide about 80 % of total tuna fish supply of the country. The fishing boats occupy most area of the harbors where fish discharge, boat building and repairs take place. About 1,000 particular small-to medium-sized wooden vessels and boats (mostly small) are busy of fishing (Table 1). Because the harbors are semi-closed basins with restricted water circulation, it may serve as an entrapment of the wastes from land sources as well as shipping activities in the harbors. The sedimentation rate in the Makran shelf is about 34 cm/ka (Prins et al. 2000) while this rate in the studied harbors is lesser.

Materials and methods

Sediment sampling

Sediment sampling took place on February 2010 using the research boat of Chabahar Oceanography Center. A total of 31 stations were chosen. Sampling consists of 28 bottom sediment samples (13 inside the harbors, 12 outside the harbors, and 3 reference samples). Two mudstone and one sandstone samples were taken from onshore outcrops (Fig. 1). Sampling depth varied between zero (beach) to depth of 11 m. The bottom sediments were taken with a stainless steel Van–Veen grab with a sampling surface of 0.05 m² and a depth of 10 cm. The beach samples were collected by a plastic snapper. All samples were carefully inspected to ensure that undisturbed sediments were collected. Three samples were taken from each site and mixed carefully. Samples were then stored in polyethylene bags at –18°C prior to undergoing laboratory analyses. The physical–chemical parameters, including the salinity

and pH of the harbor waters, were measured in situ using portable conductivity meter (WTW Cond315i) and pH meter (WTW pH330i), respectively.

Sediment characterization

Subsamples of the air-dried sediments were homogenized in order to normalize for variations in grain size distribution. Wet sieving analysis separated five size fractions: >2 (gravel), 2–0.5 (coarse–very coarse sand), 0.5–0.25 (medium sand), 0.25–0.063 (fine–very fine sand), <0.063 mm (silt+clay). Silt and clay subsamples were analyzed using a laser diffraction particle sizer. The organic matter content measured by heating to 500°C (loss in ignition method) using a muffle furnace. The dried homogenized sediment samples were sieved through a 63-µm screen and kept in clean plastic containers ready for metal analysis; 0.3 g of prepared samples were digested with HNO₃, HF, Aqua regia (HNO₃/HCl), and HClO₄ (Loring and Rantala 1992). The determination of metal content was carried out by applying inductively coupled plasma–atomic emission spectrometry. Accuracy of the determinations was checked by the analysis of the international standard reference material PACS-2 (National Research Council Canada). The analytical precision was generally better than 5 %. All reagents used were of analytical grade, and all glassware was acid-washed and deionized water rinsed prior to use. The results of trace metals concentrations were determined on a dry weight basis in micrograms per gram. The mineral composition of seven samples was studied by X-ray powder diffraction (XRD).

Results and discussion

Sediment granulometry

Figure 2 shows the granulometric characteristics of the samples of each station. The most of samples taken from inside the fishing harbors of Pasabandar (P2 to P6) and

Table 1 General characteristics of Ramin, Beris, and Pasabandar harbors

	Latitude	Longitude	Wave breaker (m)	Jetty (m)	Area (km ²)	Strait (m)	Boats	Vessels	Fishermen
Ramin	25°16' N	60°44' E	788	100	250	135	93	118	4,378
Beris	25°08' N	61°10' E	594	160	250	190	274	152	5,486
Pasabandar	25°04' N	61°24' E	660	160	500	880	130	120	5,708

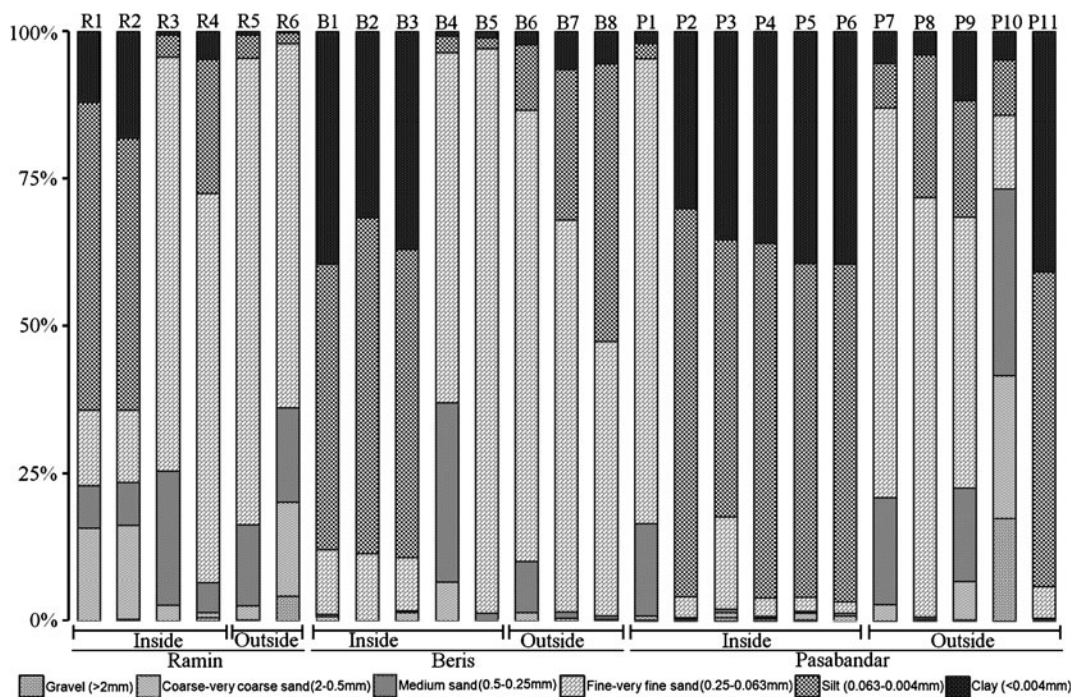


Fig. 2 Sediment granulometry of sediment samples in the harbors

Beris harbors (B1, B2, and B3) are mostly composed of silt and clay size particles. Sample P1 taken from sandy beach in Pasabandar harbor composed of fine and very fine sand. Clay content in sediments of Ramin harbor is lower than that of Beris and Pasabandar. Mud content (silt+clay) in R1 and R2 are more than 60 % (mostly silt) while in R3 and R4 is very low.

Stations outside the fishing harbors are typified as very fine sands. Station P10 has considerable gravel as well as coarse–very coarse sands. This station is close to a small rocky island, and its grains were derived from physical erosion of this island. Based on textural classification of clastic sediments (Folk 1980), sediments inside Beris and Pasabandar harbors (except P1, B4, and B5) as well as P11 are named clayey silt, samples R1 and R2 (inside Ramin harbor) are sandy clayey silt, and other samples (mostly outside harbors) are classified as silty sand. From Ramin to Pasabandar (West to East), the clay content of sediments in the harbors (and the sediment's ability to absorb trace metals) increases.

Concentrations of pollutants

Results of the chemical analysis for sediments of the selected fishing harbors are summarized in Table 2.

There is no information available on background values for trace metal concentrations in sediments of Iranian Oman Sea coasts. Therefore, these data were compared with the background concentrations in natural sediments sampled far from human activities in the area (samples N1, N2, and N3) (Fig. 1). In addition, some rock samples were collected from onshore natural environment to assess the natural origin of metals in the study coasts (Table 3). The chemical analysis data indicate that the concentration aluminum (Al), iron (Fe), and manganese (Mn) in the harbors and background sediments in the study area is less than the concentrations in the Earth's crust. In addition, the natural values of trace metals in this area is less than or equal to the mean values of the Earth's crust (except for arsenic and chromium (Cr)). This confirms that the natural input of these elements in the study area is low.

Cr, Ni, and Mn values in reference samples are higher than that of marls and sandstone. It suggests that these elements are derived from erosion of ophiolite mélange units far from coastal area as well as coastal rock unit erosion. However, the concentration of other metals in reference samples is comparable with that of coastal rock units (Table 3). Element concentrations in marls

Table 2 Summary table providing a selection of statistical parameters to describe selected variables of the determined inside and around harbors

Harbors	Statistic	Depth (m)	Organic matter content (%)	Al (%)	Fe (%)	Mn (µg/g)	As (µg/g)	Cd (µg/g)	Cr (µg/g)	Cu (µg/g)	Ni (µg/g)	Pb (µg/g)	Zn (µg/g)
Ramin	Min	1.5	2.8	2.28	2.08	423	4.0	0.04	70	17.5	34.5	12.6	42.0
	Max	7.4	6.7	5.79	3.63	778	14.0	1.50	964	287.0	85.2	475.0	547.0
	Average	4.6	4.7	3.75	2.58	545	7.8	0.58	347	99.7	51.9	99.0	203.7
	SD	2.1	1.9	1.19	0.60	151	3.7	0.61	376	97.5	17.8	184.5	198.9
Beris	Min	0.0	2.8	2.90	1.77	483	5.0	0.01	103	12.5	31.6	11.3	38.0
	Max	11.0	7.2	7.02	3.91	853	12.0	0.16	849	50.4	103.0	1,780.0	99.0
	Average	4.8	4.2	4.90	2.71	596	7.4	0.12	322	30.4	63.9	240.0	67.0
	SD	3.6	1.9	1.65	0.86	147	2.9	0.05	296	16.4	27.3	622.3	25.7
Pasabandar	Min	0.0	3.1	3.20	1.89	394	5.0	0.08	105	15.4	35.4	11.5	39.0
	Max	8.0	16.3	7.92	4.46	1,150	7.0	0.33	2,370	101.0	116.0	29.5	151.0
	Average	4.2	6.2	5.54	3.25	599	6.4	0.13	420	35.2	75.9	16.3	79.5
	SD	2.7	4.3	1.46	0.83	198	0.7	0.07	679	24.8	26.4	5.1	31.5
Background in Earth's crust ^a		–		8.13	5.00	950	1.8	0.20	100	55.0	75.0	13.0	70.0
Background in study area			2.5	3.44	1.84	579	8.0	0.11	592	15.5	36.8	13.2	39.0

^a Turekian and Wedepohl (1961)

are lower than the concentrations in the sandstone-forming coastal cliffs, because the trace elements do not tend to enrich in calcareous rocks (Faure 1992). These two rock types are the only rock types in the onshore areas.

Sediment quality

It is accepted that without defensible Sediment Quality Guideline (SQG), it would be difficult to assess the extent of sediment contamination (Casado-Martinez et al. 2006). To evaluate the sediment contamination and potential ecotoxicological effects associated with the

observed concentrations of contaminants, three published SQGs have been used. These guidelines provide interpretative tools for assessing the biological significance of chemical pollution (see Nipper 1998; Mucha et al. 2003; Guerra-Garcia and Garcia-Gomez 2005). The most used approaches are based on the values reported by Long et al. (1995), Persuad et al. (1992), and CEDEX (1994).

Recently, the Division of Fish, Wildlife and Marine Resources from the Department of Environmental Conservation, New York State, USA, advises the use of Long et al.'s (1995) guidance values for evaluation of risk from contaminants in marine and estuarine

Table 3 Values obtained for sediment variables in the reference samples and onshore rocks

Samples	TOC (%)	Al (%)	Fe (%)	Mn (µg/g)	As (µg/g)	Cd (µg/g)	Cr (µg/g)	Cu (µg/g)	Ni (µg/g)	Pb (µg/g)	Zn (µg/g)
Reference samples											
N1	2.3	2.92	1.93	702	10	0.11	822	18.1	31.1	15.4	40
N2	2.2	3.38	1.77	565	7	0.10	776	14.2	36.4	12.4	41
N3	3.0	4.03	1.83	470	7	0.11	179	14.2	43.0	11.7	36
Onshore geological units											
M1	3.0	1.62	1.42	319	4	0.16	17	13.1	19.7	10.8	22
M2	3.1	1.17	0.91	283	3	0.15	15	6.2	17.9	7.3	13
S1	1.8	7.18	4.35	565	5	0.13	96	25.7	28.0	14.7	83

sediments. The data proposed by Long et al. (1995) are in agreement with the guidelines of Persuad et al. (1992) suggested by the Ontario Ministry of Environment, Canada. According to these classifications, two levels of risk established for metal contamination in sediments are considered, including lowest effect level (LEL) and the severe effect level (SEL). The LEL and the SEL of Persuad et al. (1992) correspond to the effects range low (ERL) and effect range moderate (ERM) of Long et al. (1995), respectively.

Concentrations below the LEL value are rarely associated with biological effects. Concentrations above the LEL but below the SEL indicate a possible range in which effects would occur occasionally. Concentrations above SEL are associated with frequent negative biological effects (Guerra-Garcia and Garcia-Gomez 2005). Consequently, sediment with concentrations below LEL is considered not contaminated. If the concentration ranges between LEL and SEL, the sediment is moderately contaminated, and if the values are higher than SEL, the sediment can be considered very much contaminated.

According to these proposed guidelines, several internal stations of the selected harbors are moderately contaminated with heavy metals (Fig. 3). In most cases, arsenic and cadmium (Cd) contents are lower than LEL. However, Cd concentration in Ramin harbor is relatively high. Copper (Cu) values in all stations of Ramin (except R5), internal stations of Beris (except B2), and in P5 and P6 from internal stations of Pasabandar are moderately contaminated. Sample R1 in external station of Ramin is very much contaminated with Cu. The scenario of zinc (Zn) is relatively similar with that of Cu. Lead (Pb) content in most cases is lower than LEL. Sample B3 from internal station of Beris shows a dramatic contamination that is about nine times more than SEL level. The distribution of Ni and Cr is different with other trace metals. In most cases, Cr tends to increase in external stations, especially in samples taken from sandy beach. Approximately, Cr content in all samples in the study area is higher than LEL, and most of external samples have Cr values more than SEL. Sample P1 taken from sandy beach in the Fishing harbor of Pasabandar has the highest Cr values. Ni concentration inside harbors is obvious. However its concentration in external samples is above LEL. Ni concentration variability is lower than that of Cr. In most of the internal samples, Ni value is higher than SEL. Its concentration increases from West to East.

In several studies (e.g., Carrasco et al. 2003; Guerra-Garcia and Garcia-Gomez 2005), the analyses for sediment physical and chemical characterization are made according to Spanish recommendations for dredged materials and following the recommended protocols (CEDEX 1994). According to these recommendations, two action levels are defined on the basis of the concentrations of metals in sediments (Belzunce et al. 2001). Sediments with metal concentrations below the lower action level are considered to exert no potential risk to the environment. These sediments (classified as category I) can be disposed in the sea after dredging since they are not contaminated. Sediments with metal concentrations between the two action levels are classified as category II and are believed to exert potential risk. Their disposal to the sea must be controlled, and detailed investigations of the disposal area must be completed. Sediments with metal concentrations above the upper action level but less than eightfold higher than this level (category IIIa) should be contained, and sediments with metal concentrations more than eightfold above the upper action level must be contained and are classified as category IIIb. All sediments of category III are very much polluted and dangerous for the biota. In Table 4, these action levels are used to evaluate the degree of trace metal pollution in the area.

All these SQGs can be used to assess individual chemicals by comparing the chemical concentration with the limit concentrations or to estimate the probability of acute sediment toxicity and to determine the possible biological effect of combined toxicant groups by calculating mean quotients for a large range of contaminants (Casado-Martinez et al. 2006). This mean ERM quotient (m-ERM-Q) has been calculated according to Long and MacDonald (1998):

$$m - ERM - Q = \sum (C_i / ERM_i) / n$$

where C_i is the sediment concentration of compound i , ERM_i is the ERM for compound i , and n is the number of compounds. Mean ERM quotients have been related to the probability of toxicity (Long and MacDonald 1998; Long et al. 2000) based on the analyses of matching chemical and toxicity data from 1,068 samples from the USA estuaries. The mean ERM quotient of <0.1 has a 9 % probability of being toxic; a mean ERM quotient of 0.11–0.5 has a 21 % probability of toxicity; a mean ERM quotient of 0.51–1.5 has a 49 % of being toxic; and mean ERM quotient of >1.50 has a 76 % of toxicity.

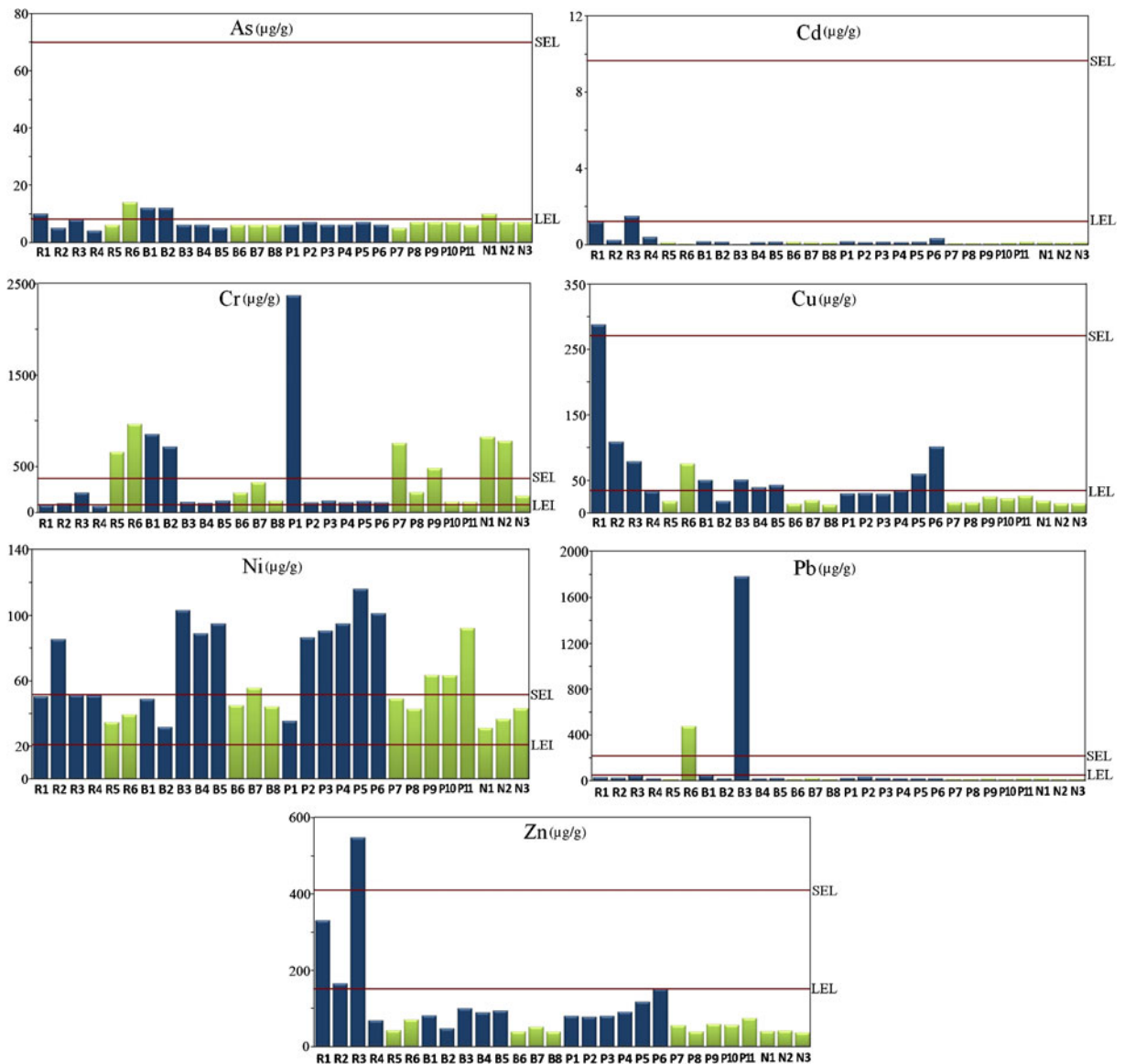


Fig. 3 Distribution of As, Cd, Cr, Cu, Ni, Pb, and Zn in internal (dark columns) and external (light columns) stations. Lines mark two levels or risk considered (Long et al. 1995; Persuad et al.

1992). *LEL* (=ERL) low effect level (=effects range low), *SEL* (=ERM) severe effect level (=effects range moderate)

Pollution levels are compared with the different SQGs (Table 4). Two approaches of comparison have been applied: (1) the number of single-species limit values exceeded and (2) the mean quotient calculated for the empirically derived set of SQG, using the ERM value reported by Long et al. (1995). About one third of the sediments do not fail any of the AL1 values: one of them is in the harbor of Ramin, three samples in the Beris, five samples in the Pasabandar, and one background sample. All the rest of the samples failed any

of the AL1 values, and only two samples that account for about the 7 % of the sediments failed at least one AL2 value, including B3 (Pb value, 1,780 $\mu\text{g/g}$) and P1 (Cr value, 2,370 $\mu\text{g/g}$).

It has been suggested that sediment toxicity is unlikely when bulk concentrations in sediment of all listed chemicals are below the ERL value. In contrast, toxicity is probable when at least one chemical concentration exceeds an ERM value (Long et al. 1995). Among 28 studied samples, 23 failed at least one of

Table 4 Number of exceeded SQGs (AL1 and AL2 from CEDEX 1994 and ERL and ERM from Long et al. 1995), mean quotient using the ERM values and anomalies.

Anomaly	m-ERM-q	ERM	ERL	AL2	AL1	Samples	
Cu>>Ni>Zn>As>Cd	0.49	1	4	0	2	R1	Internal
Ni>Cu>Cr>Zn	0.42	1	4	0	1	R2	
Zn>Ni>Cr>Cu>Cd	0.53	1	5	0	3	R3	
Ni	0.23	0	1	0	0	R4	
Cr>Ni>As>Cu>Pb	0.58	1	5	0	1	B1	
Cr>As>Ni	0.43	1	3	0	1	B2	
Pb>Ni>Cr>Cu	1.57	2	4	1	2	B3	
Ni>Cr>Cu	0.36	1	3	0	0	B4	
Ni>Cr>Cu	0.39	1	3	0	0	B5	
Cr>>Ni	1.08	1	2	1	1	P1	
Ni>Cr	0.36	1	2	0	0	P2	
Ni>Cr	0.37	1	2	0	0	P3	
Ni>Cr>Cu	0.38	1	3	0	0	P4	
Ni>Cu>Cr	0.47	1	3	0	1	P5	
Ni>Cu>Cr>Zn	0.46	1	4	0	2	P6	
Cr>Ni	0.40	1	2	0	1	R5	External
Pb>Cu>Ni>As	0.89	2	5	0	2	R6	
Cr>Ni	0.25	0	2	0	1	B6	
Cr>Ni	0.34	1	2	0	1	B7	
Ni>Cr	0.21	0	2	0	0	B8	
Cr>Ni	0.48	1	2	0	1	P7	
Cr>Ni	0.25	0	2	0	1	P8	
Cr>Ni	0.42	2	2	0	1	P9	
Ni>Cr	0.28	1	2	0	0	P10	
Ni>Cr	0.36	1	2	0	0	P11	
Cr>As>Ni	0.46	1	3	0	1	N1	Reference
Cr>Ni	0.45	1	2	0	1	N2	
Ni>Cr	0.23	0	2	0	0	N3	

the ERM values, and thus, toxicity can be considered probable for more than the 80 % of the samples. All of the samples (even reference samples), exceed any ERL value.

Mean ERM quotients range from 0.21 to 1.57 (values reported for B8 and B3, respectively). Only two samples

show values higher than 1. Table 5 shows the probability of toxicity for the samples based on mean ERM quotient. Most of the samples are medium to low contaminated. In most cases, Ni and Cr are responsible for this pollution. These two elements show anomaly in all of the samples (Table 4). In internal stations, beside Ni

Table 5 Classification of probabilities of toxicity for each sample according to the calculated mean ERM quotients

Samples	Priority area	Probability of toxicity (%)	Mean ERM quotient
B3	Highest	76	>1.5
R3, R6, B1, P1	Medium-high	49	0.51–1.5
R1, R2, R4, R5, B2, B4, B5, B6, B7, B8, P2, P3, P4, P5, P6, P7, P8, P9, P10, P11, N1, N2, N3	Medium-low	21	0.11–0.5
–	Lowest	9	<0.1

and Cr, other trace elements are also enriched. However, in external stations, only Ni and Cr are anomalous.

Generally, organic matter content of sediments increase as the sediment texture becomes finer (Bianchi 2007). Figure 4 indicates high value of organic materials inside harbors where mud content is higher than outside harbors. The presence of organic matter can potentially increase metal concentrations in sediment by adsorption of metals from surrounding environment onto organic material (Luoma 2000). It seems that wastewater discharge (containing fish body wastes and sewage of vessels) into the harbors is the main origin of organic carbon inside the harbors.

As indicated in Table 6, the concentration levels of Cr and Ni observed in the surface sediments of selected harbors are higher than the concentrations traced in harbors in other parts of the world. Arsenic and Cd contents are relatively lower than these values in other harbors. The values for the rest of the trace metals are comparable with that of other locations.

Normalization of trace metal contamination

Natural occurrence of trace metals in the environment, due to different particle size for example, complicates assessments of contaminated marine sediments. Measurable quantities of metals do not automatically imply anthropogenic enrichment. In most sedimentary environments, there is a linear relationship between inert elements (conservative elements or normalisers such as Fe and Al) and the clay content of the samples, which allows the use of a normalizer as a substitute for the granulometric variability of the sediments. Geochemical normalization can compensate for both the granulometric and mineralogical variability of metal concentrations in

sediments (Liu et al. 2003). Consequently, in order to assess the possible anthropogenic impact, several authors (Schiff and Weisberg 1999; Mucha et al. 2003; Guerra-Garcia and Garcia-Gomez 2005) have successfully used Fe to normalize the trace metal contaminants.

We have used Fe as a conservative tracer to differentiate natural and anthropogenic metal concentrations. Fe is abundant in the Earth’s crust and scarcely influenced by anthropogenic inputs, especially compared with the naturally occurring high levels of this element (Eby 2004).

Figure 2 shows a spatial distribution of normalized trace elements using Fe (obtained by Arcinfo GIS 9.1 software, graduated symbols). The normalization of the concentrations using Fe as a conservative element confirmed that the majority of the internal stations of Ramin, Beris, and Pasabandar harbors are enriched with Cu, Ni, Pb, and Zn. To be as restrictive as possible, we considered the highest metal/Fe values measured in the reference stations (N1, N2, and N3) as baseline values (Fig. 5). The concentration range of arsenic is relatively limited. Cd and Zn content in Ramin harbor is higher than that of the other harbors. Cr shows anomaly only in sample P1. However, its concentration in most of samples is higher than mean concentration in crust. It confirms that the natural environment have the most important role in its pollution in the study area.

Sediment mineralogy and geochemistry

Once trace metals are deposited in coastal sediments, they undergo a series of physical, biological, and chemical processes (Lee and Cundy 2001). Trace metals occur in a number of different forms, mainly in the dissolved and the solid state (adsorbed onto surfaces

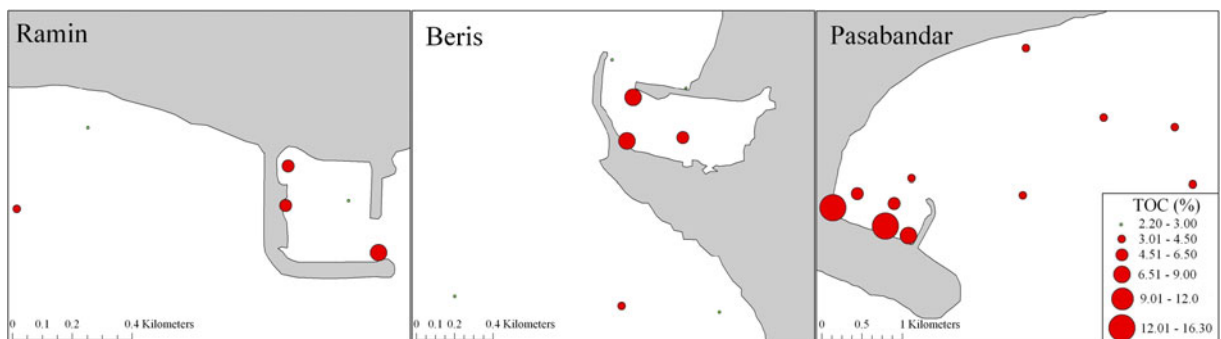


Fig. 4 GIS-based geochemical maps showing distribution of total organic carbon values in the study area. Values more than baseline values. The highest values obtained in reference

stations are shown with *dark (red) circles* and others are shown with *light (green) circles*

Table 6 Comparison of metallic contamination levels in harbors of Ramin, Beris, and Pasabandar and reference harbors worldwide

References	Zn	Pb	Ni	Cu	Cr	Cd	As	Location
Present study	68–547	15.9–42.3	50.3–85.2	32.6–287	70–216	0.23–1.5	4–14	Ramin harbor, Iran
Present study	47–99	13.8–1780	31.6–103	17.7–50.4	103–849	0.01–0.16	5–12	Beris harbor, Iran
Present study	77–151	15–29.5	35.4–116	28.7–101	105–2370	0.11–0.33	6–7	Pasabandar harbor, Iran
Mostafa et al. (2004)	58.5–382	38–1070	13–53	33–649	39–207	0.61–2.44	–	Western harbor, Alexandria, Egypt
Muller- Karulis et al. (2003)	17–254	3–44	5–35	3–29	12–71	–	<3	Ventspils harbor, Latvia
Belt Collins Hawaii (1993)	82–3524	53–129	9.3–21.5	3–123	–	2.0–4.3	–	Outer Apra harbor, Guam
Guerra-Garcia and Garcia-Gomez (2005)	29–695	10–516	8–671	5–865	13–381	–	4–42	Ceuta harbor, Spain
Fatoki and Mathabatha (2001)	19–126	9–62	–	9–82	–	–	–	Elizabeth harbor, South Africa
Poulton et al. (1996)	338–5930	18–1250	8–61	8–135	5–140	–	–	Hamilton harbor, Canada
Bothner et al. (1998)	39–414	18–263	8–191	7–142	42–292	–	–	Boston Harbor, USA
Chen and Wu (1995)	–	34.3–138	–	37.9–505	–	0.1–4.64	–	Kaohsiung Harbor, Taiwan
Niadu and Morrison (1994)	88–670	19.3–272	17–38	59–306	16–106	0.74–3.04	0.7–45	Suva harbor, Fiji

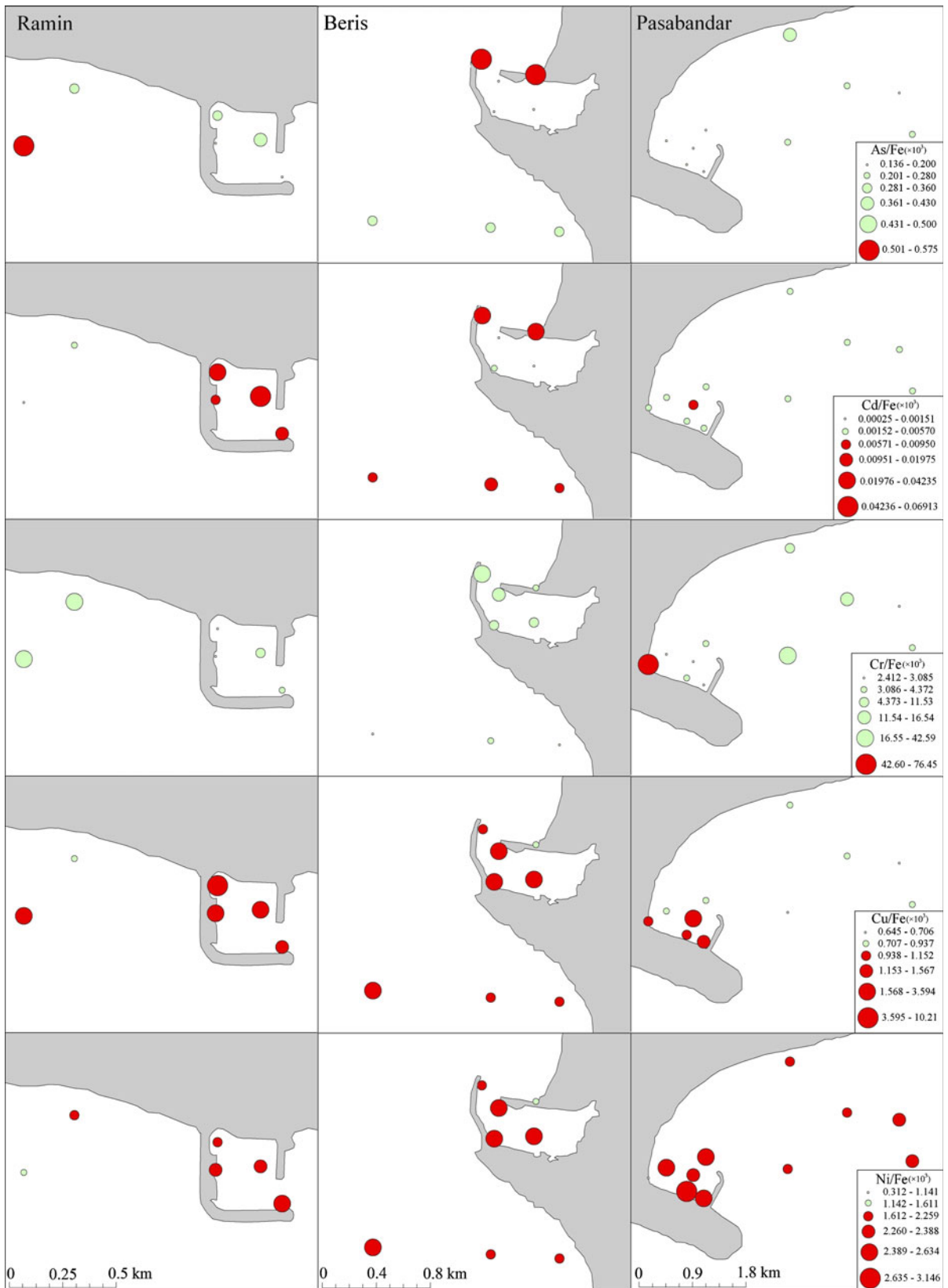
of clay minerals, element oxides, organic material, co-precipitated with sediment phases, and incorporated into organic matter). The highest proportion is usually in the solid phase (Berkowitz et al. 2008). Understanding the geochemical processes affecting trace metals concentration and their fate in sediments is important in gauging appropriate designs for sampling and monitoring programs and planning for appropriate remediation options (Fergusson 1990).

The mineralogy of sediment samples using XRD is presented in Table 7. Most frequent minerals includes: quartz, calcite (with or without aragonite and dolomite), albite, and muscovite. These minerals are common in the beach sediments (Libes 2009). The important point is abundance of clinocllore and chlorite in the samples. Clinocllore ((Mg,Fe²⁺,Cr)₅Al₂Si₃O₁₀(OH)₈) is one of the most common members of the chlorite group of minerals. These minerals are all difficult to differentiate by ordinary means and often the general mineral name chlorite is given to specimens that lack distinguishing characteristics. Clinocllore is an alteration mineral and commonly occurs in metamorphic environments, especially in serpentine deposits (Worden and Morad 2003). Therefore, its origin in the study area is erosion of ophiolite mélange units at the Northern parts of the Makran mountains. It seems this mineral is an important natural

source for Ni and Cr in the area. Heavy mineral chromite derived from ophiolites is also responsible for high Cr content in sand beaches. In addition to chlorite group of clay minerals, illite and smectite are also present in some sediment samples. Structurally, illite is quite similar to muscovite with slightly more silicon, magnesium (Mg), Fe, and water and slightly less tetrahedral Al and interlayer potassium. The cation exchange capacity of illite is smaller than that of smectite (>100 meq/100 g) but higher than that of kaolinite, typically around 20–30 meq/100 g (Eby 2004). Comparing with Ramin, occurrence of smectite in sediments of Beris and Pasabandar can increase trace metal adsorption in these harbors. Two species of Fe oxides, goethite and hematite, occurring in Ramin and Beris harbors are introduced by anthropogenic pollution.

The particle-size distribution and organic-matter content may be two critical factors that influence heavy metal distribution in sediments (Schulz and Zabel 2006). The Fe and Al content of sediments increase from West to East. It suggests that clay minerals content is higher in Eastern harbors (Beris and

Fig. 5 GIS-based geochemical maps showing normalised ratios of heavy metals using Fe as conservative element. Values more than baseline values. The highest values of the metal/Fe obtained in reference stations are shown with *dark (red) circles* and others are shown with *light (green) circles*



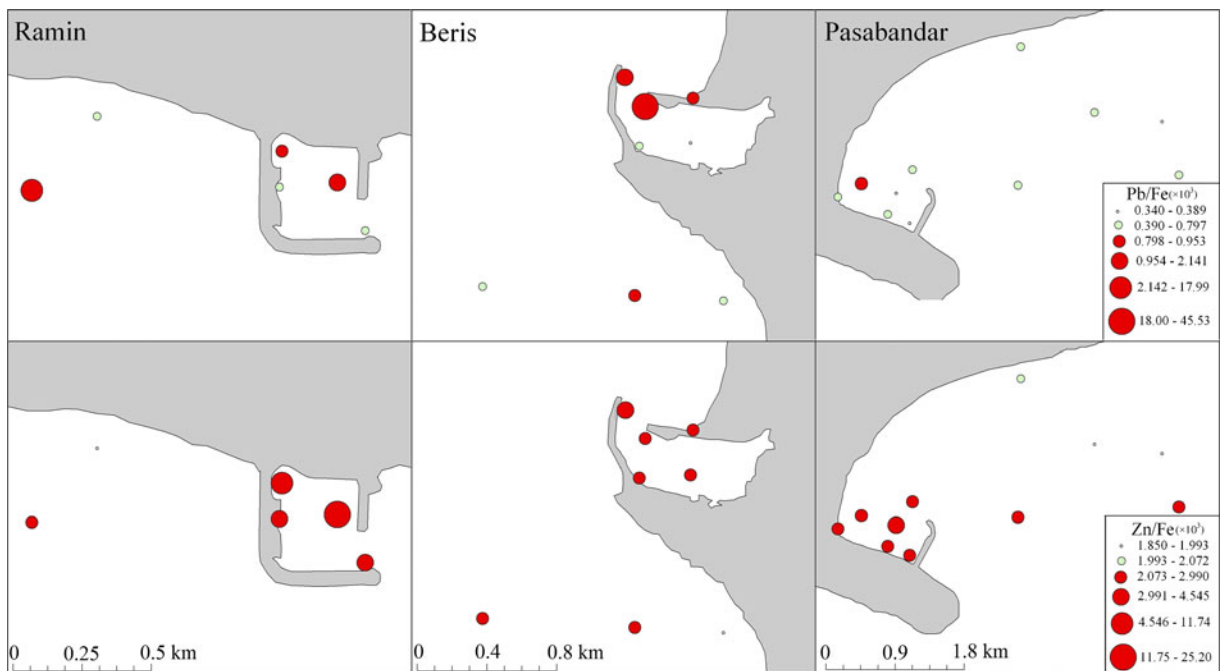


Fig. 5 continued.

Pasabandar). Therefore, the sediments of Beris and Pasabandar harbor are more capable to retain trace metals comparing to Ramin harbor.

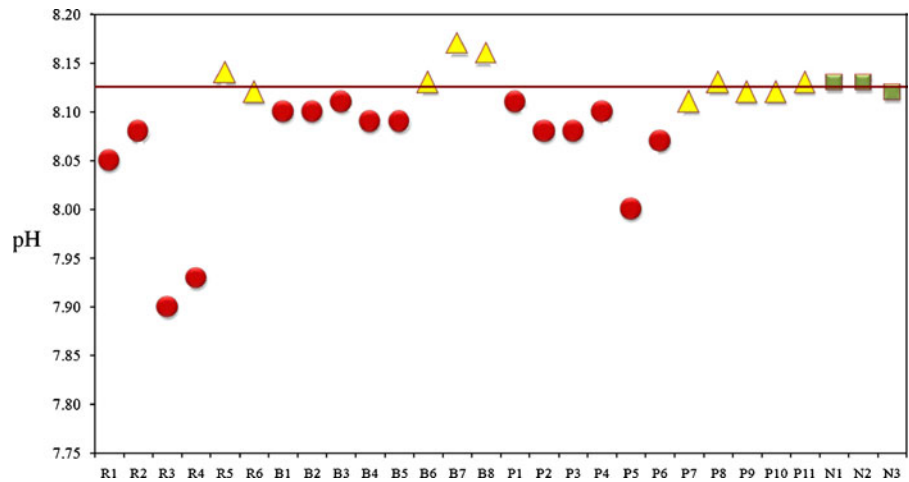
Although it accounts for only a small proportion of the total dissolved metals in sea or interstitial waters, the free ions are the most readily available as well as the most toxic inorganic species of trace metals. However, the free ions are very sensitive to complexation in the presence of natural organic chelators or high salinity (Eby 2004). If the contamination of the water column remains at high levels, then the related sediments will also be high in metal concentration (Ibhadon et al.

2004). Since toxicity is a function of bioaccumulated metal burden, the environmental parameters influencing bioavailability also influence biological effects. Temperature and salinity, for example, are principal components affecting the coastal environments and both parameters may significantly modify the acute toxicity of a number of metals because the combined effect of low salinity and high temperature act synergistically to increase metal toxicity (Luoma 2000). The maximum impact at low salinities for metals such as Cd and Cu arise from the increase in the more bioavailable free ions, Cu^{2+} and Cd^{2+} coupled with the reduced

Table 7 Mineralogy of some selected sediment samples based on XRD analysis

Minerals	Sample	
Quartz–calcite–albite–clinochlore–muscovite–halite–alkali feldspar–dolomite	N1	Reference samples
Quartz–calcite–albite–clinochlore–dolomite–halite–muscovite	N2	
Calcite–quartz–aragonite–albite–halite–chlorite–gypsum–illite	N3	
Calcite–quartz–aragonite–halite–dolomite–albite–muscovite–mhlorite–hematite	R3	Ramin harbor
Quartz–calcite–albite–clinochlore–muscovite–halite–goethite–dolomite–smectite	B3	Beris harbor
Quartz–calcite–albite–clinochlore–halite–muscovite–dolomite–smectite	P4	Pasabandar harbor
Quartz–calcite–albite–clinochlore–muscovite–dolomite–halite–alkali feldspar	M1	Mudstone

Fig. 6 pH variations in the selected harbors. The *line* indicates the mean pH value obtained in reference stations. *Triangles*, outside harbors; *circles*, inside harbors; and *rectangular*, reference



competition for uptake sites from elements including Ca and Mg. Bioavailability and toxicity are then ultimately linked for the majority of metal–organism interactions. The salinities measured in this study are relatively high (36,200–39,100 mg/l), and this may increase the possibility of complexation of the free metal ions and reduce their bioavailability. High organic matter content in harbors can act as a strong colloid to adsorb trace metals in the bottom sediments. Decomposition of organic matter releases carbon dioxide into the water column and this phenomenon in turn, reduce pH value of water inside the harbors (see Fig. 6). Even in such a relatively low pH, trace metals are insoluble and may be absorbed by clay minerals, organic matters, and Fe oxides that are frequent in the harbors.

The pattern obtained for Mn is exactly the opposite to that of the remaining trace metals, especially when normalized with Fe (Fig. 7). The ratio Mn/Fe is lower in the most polluted stations such as R1, R3, B3, and

P1. The depletion of Mn in anoxic and polluted sediments has been reported in previous studies (Jung et al. 1996; Guerra-Garcia and Garcia-Gomez 2005). When sediments are contaminated and usually anoxic, there is a precipitation of Cu sulphide and a dissolution of Mn oxide which are responsible for the enrichment of Cu and the depletion of Mn, respectively. The existence of these processes in the selected harbors are corroborated by the high values obtained for the Cu/Mn ratio measured in the most contaminated samples (internal stations) (Fig. 8).

The strong correlation between decreasing grain size and increasing heavy metal concentration is well documented and strongly suggests that adsorption is the main mechanism by which trace metals accumulate on particles, because small particles have a much higher surface area relative to their volume than large particles (Matthai and Birch 2001; Berkowitz et al. 2008).

Because (1) Al and Fe (to a lesser extent) are tightly associated with clay fraction that is the dominant metal-

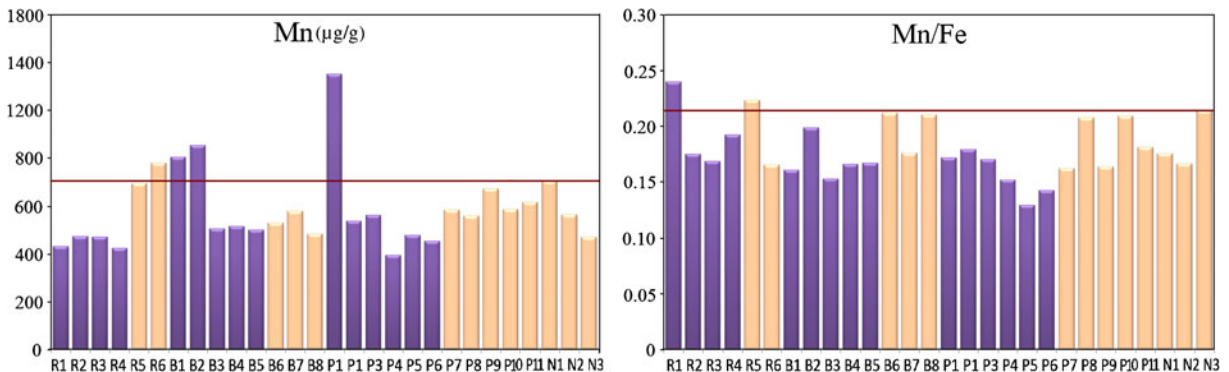
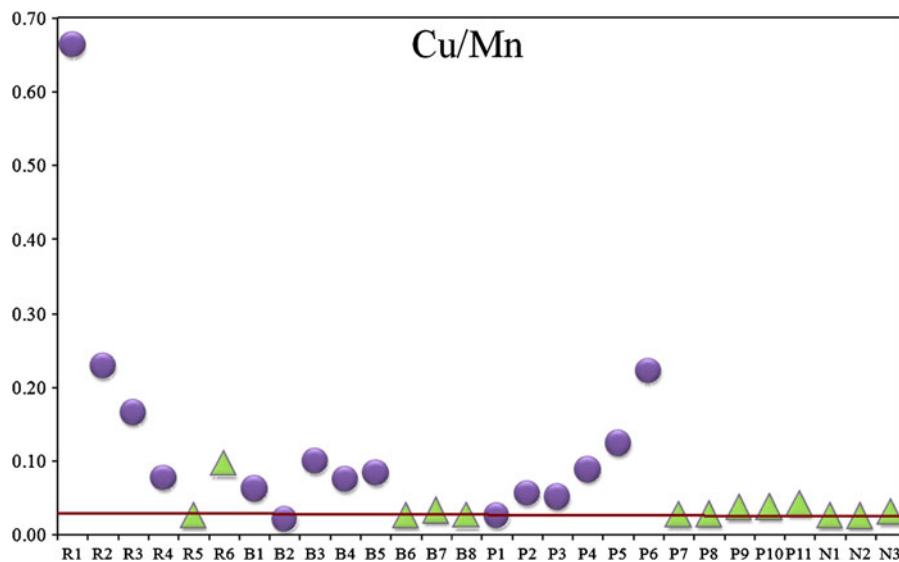


Fig. 7 Distribution of Mn in internal (*dark columns*) and external (*light columns*) stations in the harbors. The ratio Mn/Fe is also indicated, together with the baseline value (maximum value of the ratio obtained in reference stations)

Fig. 8 Variation of the Cu/Mn ratio in the internal (circles) and external (triangles) stations harbors. The line indicates the maximum value of the ratio obtained in reference stations



bearing phase of the sediment (Faure 1992), (2) Fe in relatively oxic and low pH values (above 2–3) forms insoluble Fe oxyhydroxides and adsorbs and coprecipitates with trace metals (Eby 2004), and (3) TOC content in sediments plays a major role to adsorb trace metals, therefore, concentration of these parameters were compared with that of trace metals (Fig. 9).

In general, the correlation diagram shown in Fig. 9 indicates significant correlation between selected colloids (consist of clay minerals, Fe oxides, and organic matter) and trace metals. It is attributed to adsorption of trace metals by these colloids. The highest correlation is related to Ni and Al values ($r^2=0.92$). It confirms that most of Ni occurs in crystal lattice of clay mineral clinocllore. Negative correlation between Cr and clay minerals is seen in Fig. 9. Cr mostly occurs in heavy mineral chromite and tends to enrich in >63 μm fraction of sediments in sandy beaches.

Conclusions

The presence of a semi-closed environment in the harbors of Ramin, Beris, and Pasabandar increases the homogeneity of sediments, which consist mostly of mud in the harbors. However in external stations due to effect of waves and erosion of coastal cliffs, the majority of sediments are silty sand. The high values of organic matter measured inside the harbors indicated influence of sewage from fishing vessels. There is no urban effluents discharge into the harbors. However,

discharge water from some of the buildings inside harbor environment is transported in these harbors. The most important source of the organic matter pollutants is effluents produced by cleaning the vessels, anthropogenic sewages produced by sailors and fish body wastes, which are released into the semi-closed environment of these harbors.

Taking into account the fact that industrial activity around these harbors is almost absent, we must resort to other sources to explain the high abundance of heavy metals, especially Ni, Cr, Cu, Pb, and Zn. It seem shipping operations such as fish landing, ship repairing, fueling, greasing, and painting are responsible for high trace metals in the selected harbors. In addition to the organic and heavy metal pollution derived from the vessel effluents, another likely source of heavy metal pollution in the harbors is the antifouling paints. Such paints are used to cover the underwater parts of the ferries and vessels to protect them from the development of fouling organisms such as algae and barnacles and are based on organic solvents mixed with highly concentrated toxic metals such as Cu and Zn (Orlic and Tang 1999). The analysis of several samples of antifouling paints shows the concentrations of Cu and Zn are typically between 15 and 30 % (Orlic and Tang 1999). Therefore, elevated concentrations of Cu and Zn are expected due to the vessel body repairing and washout of the antifouling paints used for ships in the harbors of Ramin, Beris, and Pasabandar during summer monsoon time. During June, July, and August, all vessels stay in the harbors and are being

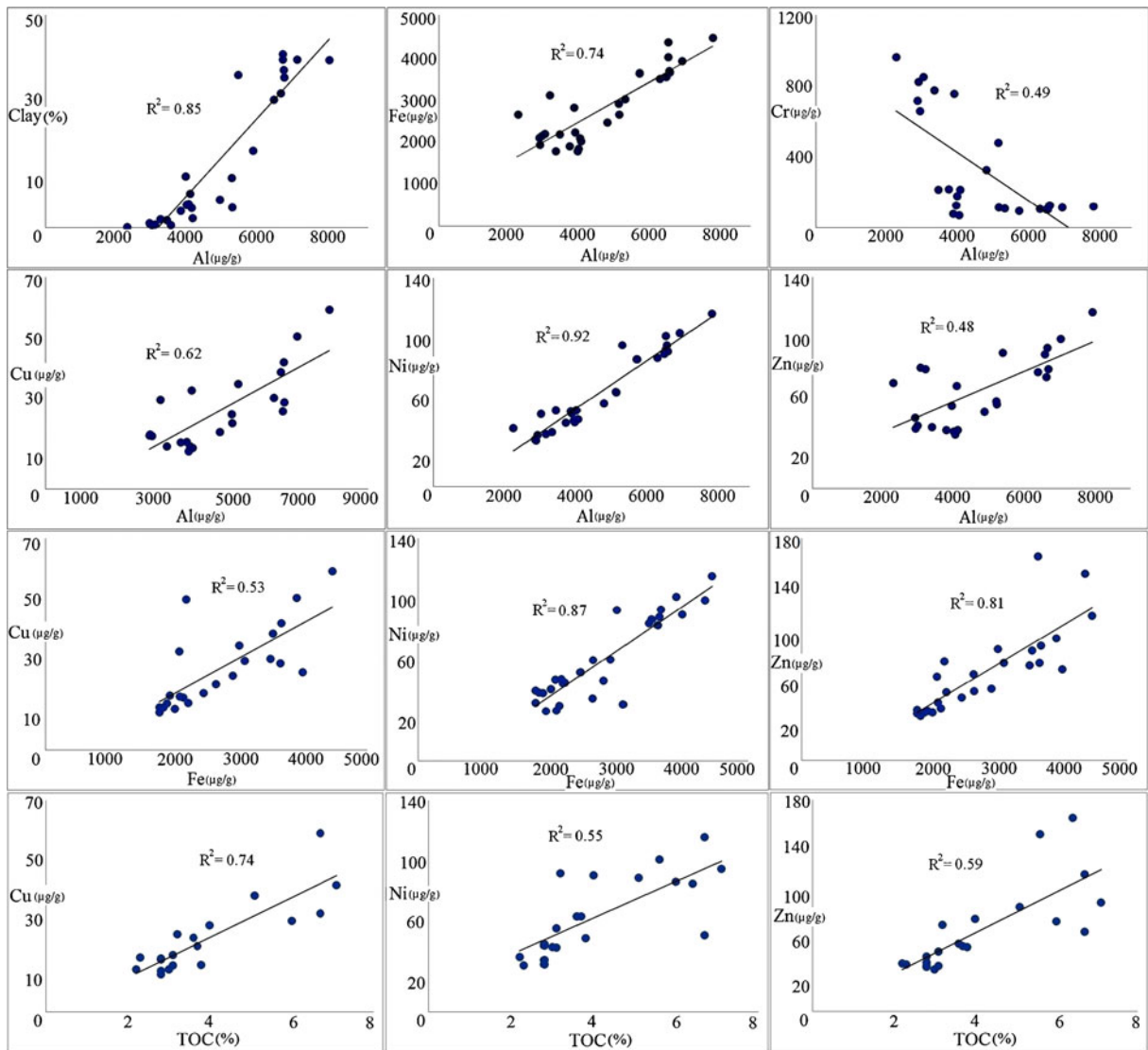


Fig. 9 Correlation coefficient between Al, Fe, and TOC as colloids and trace metals

repaired. The leaching of the toxic metals from the matrix of the paint causes toxic effects not only to fouling organisms but also to other “non-target” biota. Recent studies (Katranitsas et al. 2003) have demonstrated a negative effect of a Cu-based antifouling paint on mortality and enzymatic activity of non-target marine organisms. Discarded engine greases also contain deteriorated mechanical parts of engine that can elevate Fe, Ni, Zn, Cu, Zn, and Pb and concentration in bottom sediments.

High concentrations of Cr and Ni were also measured in all stations (internal, external, and even reference stations). These heavy metals are usually associated with industrial activity. Nevertheless, industrial activity

around the study harbors is absent. So, the origin of these metals should be independent from industrial influence and other anthropogenic sources. As stated before, the most of Cr and Ni pollution in the study area is natural pollution. These two elements are common elements in the ophiolitic units found in the northern Makran mountains and are transported to the coastal area of Oman Sea by several seasonal streams and the only perennial river of Bahukalat. The Bahukalat River reaches to the sea in the middle of Gavater bay at the East of Pasabandar harbor. Increasing Ni and Cr values from West to East indicates that Bahukalat River carries more Ni- and Cr-bearing compounds and suspended particles

than that of other seasonal rivers into the study area. However, Ni and Cr are also released in the harbors by human activities. The most Cr concentration in the area is related to sample P1, from the sand beach of Pasa-bandar harbor. Its tendency to enrich in sand beach is due to its high specific gravity (7.18 to 7.20 in 20°C).

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