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Distribution and Accumulation of Heavy Metals in Sediments of the Northern Part of Mangrove in Hara Biosphere Reserve, Qeshm Island (Persian Gulf)

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Abstract

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The mangrove of Hara Biosphere Reserve, stretching over 100 thousand hectares in the southern coast of Iran and in the northwest of Qeshm Island, belongs to the most important and largest mangroves in the Middle East. Twenty sedimentary samples were collected and concentrations of seven heavy metals were investigated in order to assess the extent of pollution distribution in this area and to discuss the origin of these contaminants in sediments. The mean heavy metal concentrations followed the scheme: Fe > Cr > Ni > Zn > Cu > Pb > Cd. Based on the geo-accumulation index, the Fe, Pb, Zn, and Cu levels were graded as non-contamination, the levels of Cr and Ni as non-contamination to moderate contamination, while those of Cd as moderate contamination to moderate to heavy contamination. According to the enrichment factor and quantification of contamination calculations, Cu, Pb, and Zn were derived mainly from natural processes and exposure of material from the Earth's crust, while the increased values of Cd, Ni, and Cr were ascribed to anthropogenic activities. The ecological risk of heavy metals was moderate, largely due to Cd contamination. The elevated values identified for Cd, Ni, and Cr are supposingly associated with activities including human refuse, shipping, transportation, fuel smuggling, and industrial wastewater discharges from factories located around Hara Biosphere Reserve (e.g. Al-Mahdi aluminum factory, lead and zinc Qeshm factory, and Hormozgan cement factory).

Keywords: anthropogenic; contamination; ecological risk; pollution; sediments

Coastal and marine ecosystems are potentially at risk due to a high concentration of heavy metals in sediments (Kumar *et al.* 2015). Heavy metals are introduced to the aquatic environment and accumulate in sediments by several pathways via natural and anthropogenic processes (Akoto *et al.* 2008). Geomorphological setup are important natural factors which affect the concentration of heavy metals in the sediments within estuaries (Kumar *et al.* 2015). Mangrove plants comprise a group of intertidal plants that dominate the coastlines of many tropical and subtropical regions. These plants are highly productive and play a vital role as the major primary producers in estuarine ecosystems (MacFarlane *et al.* 2007). Some of the potentially most serious anthropogenic pollutants in mangrove ecosystems are heavy metals (Wang *et al.* 2013). Elevated concentrations of heavy metals have been recorded in mangrove sediments from many parts of the world (Wang *et al.* 2013). The geo-accumulation index (I_{geo}), enrichment factor (*EF*) (Ghrefat *et al.* 2011), and quantification of contamination index (*QoC*) are different statistical indices used to establish the source and the magnitude of metal pollution in the

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environment (Ghrefat *et al.* 2011; Khuzestani & Souri 2013; Zarei *et al.* 2014). Hara Biosphere Reserve is one of the most important coastal areas located in the south of Iran. The area is internationally recognized and important because of its vast biological diversity and valuable coastal resources (Neinavaz *et al.* 2011). It is situated near the city of Bandar Abbas (the largest southern port of Iran in the Persian Gulf) and stretches along the northern coast of Qeshm Island (the largest island and commercial-industrial free zone of the country in the Persian Gulf) (Nowrouzi *et al.* 2012). This region has a complex and interesting ecosystem and is influenced by anthropogenic activities including shipping and transport, oil and petrochemical industry, fishing, harbour, as well as residential and commercial wastewater (Safahieh *et al.* 2011; Kazemi *et al.* 2012). The present study aimed to determine the concentrations, distribution, and sources of heavy metals (Cd, Cu, Ni, Pb, Zn, Cr, and Fe) in the sediments of Hara Biosphere Reserve to assess the pollution status as well as the possible influence of anthropogenic activities and to compare the heavy metals contamination levels with the international sediment quality criteria. The following contamination indices were employed: enrichment factor (*EF*), geo-accumulation index (I_{geo}), ecological risk factor (E_{ri}), and quantification of contamination index (*QoC*) (Paul 2001).

MATERIAL AND METHODS

Study area. Hara Biosphere Reserve is one of the most important coastal areas located in the south of Iran in the Khuran straits between northwest of Qeshm Island and the mainland of Iran (Hormozgan province) (Figure 1). The study area (26°45' to 26°58'N; 55°30' to 55°50'E) is situated in the Mehran River delta (Dehghani *et al.* 2010). Hara Biosphere Reserve has vast biological diversity and extremely important coastal resources, which are vital to Iranian socioeconomic development. A vast majority of human population lives in the coastal area, and most communities depend on local resources for their livelihood. This region is internationally recognized and known as Ramsar International Wetland sites, and has also been added to UNESCO's Man and Biosphere Program (MAB) convention list. The area belongs to the most important Iranian protected areas (Dehghani *et al.* 2010).

Sampling and laboratory procedures. Sediment samples were collected in the northern area of Hara Biosphere Reserve during December 2015. They were taken from twenty sampling sites in three replicates based on ecological conditions, characteristics of sediments, and human activities (Figure 1). Bed sediment samples were taken from each site using stainless steel Van Veen grab and superficial sediments from 10–20 cm depths from approximately 5 × 5 cm plots close to each sampled plant. The samples were placed on ice, immediately transported to the laboratory on the same day, and stored at –20°C until analysis. The sediments were dried at 105°C for 24 h and passed through a 63-µm mesh stainless screen because metals generally are associated with sediment particles sizing less than 63 µm (Rae 1997; Monikh *et al.* 2013). The sieved sediment was

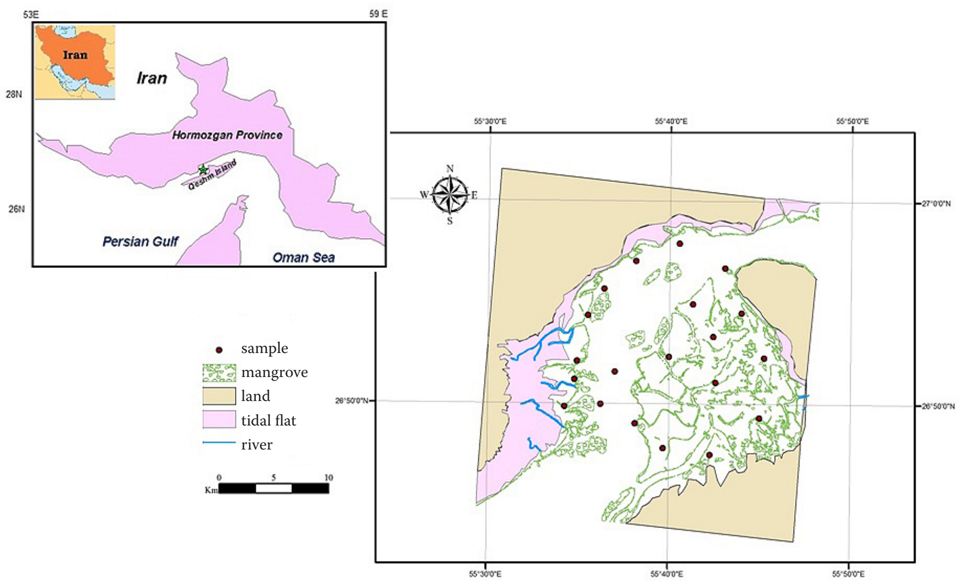

Figure 1. Map of the study area and sampling sites in Hara Biosphere Reserve of Persian Gulf

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powdered using an agate mortar and pestle. About 0.5 g of the powdered sample was placed in a Teflon beaker containing 10 ml aqua regia (HNO_3 + HCl, 1:3 v/v). The mixture was heated until most of the liquid evaporated and allowed to cool before 5 ml of hydrogen fluoride (HF) were added. The samples were further cooled to room temperature for 1 h before being filtered (Karbassi *et al.* 2008). The residue was filtered through a Whatman filter paper (No. 42) and diluted to 50 ml with distilled deionized water. For each digestion program, a blank was also prepared in the same manner as that employed for sediment samples with equal amounts of acid (Moopam 1999). The blank was also run at the same time. Blanks were used for correction of background and other sources of error (Zarei *et al.* 2014). A standard sample (NCS DC 73014 and NCS DC 73313a) was analyzed using the same methods as an accuracy check. Then, the concentrations of metals including Cr, Pb, Cu, Zn, Ni, Cd, and Fe in the final solutions were determined by an atomic absorption device (Thermo-solar) (AAS: Flame/Furnace, Thermo Scientific, Waltham, USA). The obtained data were initially analyzed with Kolmogorov-Smirnov test to commit the normal distribution and if needed the transformation of the data for normal distribution was performed before analysis (Khuzestani & Souri 2013). Graph outputs were created using MS Excel 2016.

Contamination indices

Geo-accumulation index. The index of geo-accumulation (I_{geo}) introduced by Muller (1969) is used to assess metal pollution in sediments. It enables us to assess the contamination by comparing the current concentrations with the pre-industrial ones using original bottom sediments (Muller 1969). It can also be applied to assess the contamination of different environments. The index is calculated as follows:

$$I_{geo} = \log_2(Cn)/1.5\ Bn \qquad (1)$$

where:
Cn – measured concentration of a metal in sediments
Bn – background value for a metal

The factor 1.5 is used because of possible variations of the background data due to lithological variations. The world average shale and the world average soil are among the materials often used to provide background metal levels. The geochemical background values for the studied heavy metals are not available in this region. Thus the quantity I_{geo} is calculated using the global average shale data (Turekian & Wedepohl 1961). According to Gonzalez-Macias *et al.* (2006), the I_{geo} for a metal is classified into seven grades: uncontaminated ($I_{geo} \leq 0$); uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$); moderately contaminated ($1 < I_{geo} \leq 2$); moderately to heavily contaminated ($2 < I_{geo} \leq 3$); heavily contaminated ($3 < I_{geo} \leq 4$); heavily to extremely contaminated ($4 < I_{geo} \leq 5$); and extremely contaminated ($I_{geo} \geq 5$) (Muller 1969).

Ecological risk factor. The ecological risk factor (E_{ri}) quantitatively expressing the potential ecological risk of a given contaminant suggested by Hakanson (1980) is calculated as

$$E_{ri} = T_{ri} \times C_i/C_o \qquad (2)$$

where:
T_{ri} – toxic-response factor for a given substance (i.e. Hg = 40, Cd = 30, As = 10, Pb = Cu = Ni = 5, Cr = 2, Zn = 1, Cr = 2)
C_i – contamination factor
C_o – regional background value of heavy metals in the sediments

The regional background value of heavy metals in the sediments is based on values from relatively non polluted bottom sediments (Adami *et al.* 2000; Adamo *et al.* 2005). The T_{ri} values of heavy metals (including As) by Hakanson (1980) are also given in Table 1. The following terminologies are used to describe the risk factor: $E_{ri} < 40$, low potential ecological risk; $40 \leq E_{ri} < 80$, moderate potential ecological risk; $80 \leq E_{ri} < 160$, considerable potential ecological risk; $160 \leq E_{ri} < 320$, high potential ecological risk; and $E_{ri} \geq 320$, very high ecological risk (Khuzestani & Souri 2013).

Enrichment factor. The enrichment factor (*EF*) (Ghrefat *et al.* 2011) has been widely reported as an important tool to differentiate between anthropogenic and naturally occurring metal sources in the sediment (Selvaraj *et al.* 2004; Ismail & Naji 2011). Aluminum (Al) and iron (Fe) are two main elements used as normalizers for *EF* computation. However, Fe is used as a normalizer in this study because it is the fourth major element in the Earth's crust and most often has little or no adverse environmental concerns. The *EF* for Fe-normalized data was calculated using the following equation:

$$EF = (M_{sample}/Fe_{sample})/(M_{shale}/Fe_{shale}) \qquad (3)$$

where $(M/Fe)_{sample}$ and $(M/Fe)_{shale}$ values, respectively, are the metal concentrations (mg/kg) dry weight

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Table 1. Means ± standard deviation of metal concentrations in sediment samples (mg/kg) from twenty sampling sites in Hara Biosphere Reserve

Sample	Fe (%)	Pb	Ni	Zn	Cu	Cr	Cd
		(mg/kg)					
Average	2.63 ± 0.04	7.94 ± 0.04	101.48 ± 0.05	49.39 ± 0.04	20.98 ± 0.04	194.29 ± 0.0	2.63 ± 0.04
MEC[a]	4.1	14	80	75	50	100	0.02
MWS[a]	4.1	19	52	95	33	70	–
MCS[a]	4.7	20	68	95	45	90	0.3
ERL[a]	34	46.7	20.9	150	34	81	1.2
ERM[a]	270	21.8	51.6	410	270	370	9.6
TEL[a]	–	30.2	–	124	18.7	25.3	0.7

[a](Adami *et al.* 2000; MacDonald *et al.* 2000); – not detected; MEC – Mean Earth's crust; MWS – mean world sediment; MCS – mean continental shale; TEL – threshold effect level; ERL – effect range low; ERM – effect range median retrieved

in relation to Fe levels (dry weight) in sediment samples and average shale values taken from Krauskopf and Bird (1967). The *EF* values were interpreted as suggested by Wang *et al.* (2008). If $0.5 \leq EF \leq 1.5$, then it indicates that a metal could be mainly from natural weathering process, and $EF > 1.5$ indicates that the metal is from anthropogenic sources or a greater percentage of the metal is from non-natural weathering process (Wang *et al.* 2008). However, the degree of enrichment was interpreted as proposed by Birch (2003): $EF < 1$ indicates no enrichment, $1 < EF < 3$ indicates minor enrichment, $3 \leq EF \leq 5$ indicates moderate enrichment, $5 \leq EF \leq 10$ indicates moderate to severe enrichment, $10 \leq EF \leq 25$ indicates severe enrichment, $25 \leq EF \leq 50$ indicates very severe enrichment, and $EF > 50$ suggests extremely severe enrichment.

Quantification of contamination. The quantification of contamination index (*QoC*) mainly quantifies the anthropogenic concentration of a metal employing the concentration of the background metal to represent the lithogenic metal (Asaah *et al.* 2006). This is calculated in accordance with Eq. (4):

$$QoC\ (\%) = (X - Xe/X) \times 100 \quad (4)$$

where:

X – average concentration of the metal in the sample under investigation

Xe – average concentration of the metal in background (Asaah *et al.* 2006)

The values of this index are mainly expressed as percentage, demonstrating the magnitude of lithogenic and anthropogenic impacts (Zarei *et al.* 2014).

RESULTS AND DISCUSSION

The concentration of metals in the mangrove sediments of Hara Biosphere Reserve and global baseline values are presented in Table 1. In summary, mean heavy metal concentrations were: Fe (2.63 ± 0.043%) > Cr (194.29 ± 0.042 mg/kg) > Ni (101.48 ± 0.049 mg/kg) > Zn (49.39 ± 0.044 mg/kg) > Cu (20.98 ± 0.039 mg/kg) > Pb (7.94 ± 0.04 mg/kg) > Cd (2.63 ± 0.044 mg/kg). The I_{geo} values for selected metals at each sampling site are listed in Table 2. The results for Cu, Zn, Pb, and Fe based on the geo-accumulation index showed that greater part of the samples could be considered as uncontaminated ($I_{geo} < 0$) (variations of I_{geo} were –2.58 to –0.001 for Cu, –1 to –2.23 for Zn, –1.61 to –2.2 for Pb, and –0.80 to –2.6 for Fe). The result for Cr and Ni based on I_{geo} values was uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$) (variations of I_{geo} were 0.26 to 0.72 for Cr and –0.90 to 0.52 for Ni), while the result for Cd was moderately contaminated to moderately to heavily contaminated (I_{geo} = 1.37

Table 2. Geo-accumulation index (I_{geo}) for concentration of metals in the sediments of Hara Biosphere Reserve

Sample	Fe	Pb	Ni	Zn	Cu	Cr	Cd
Max	–0.8	–1.61	0.52	–1.0	–0.001	0.89	3.16
Min	–2.6	–2.2	–0.9	–2.23	–2.58	0.26	1.37
Average	–1.47[a]	–1.91[a]	–0.07[a]	–1.56[a]	–1.63[a]	0.5[b]	2.46[c]

[a]uncontaminated; [b]uncontaminated/moderately contaminated; [c]moderately to heavily contaminated

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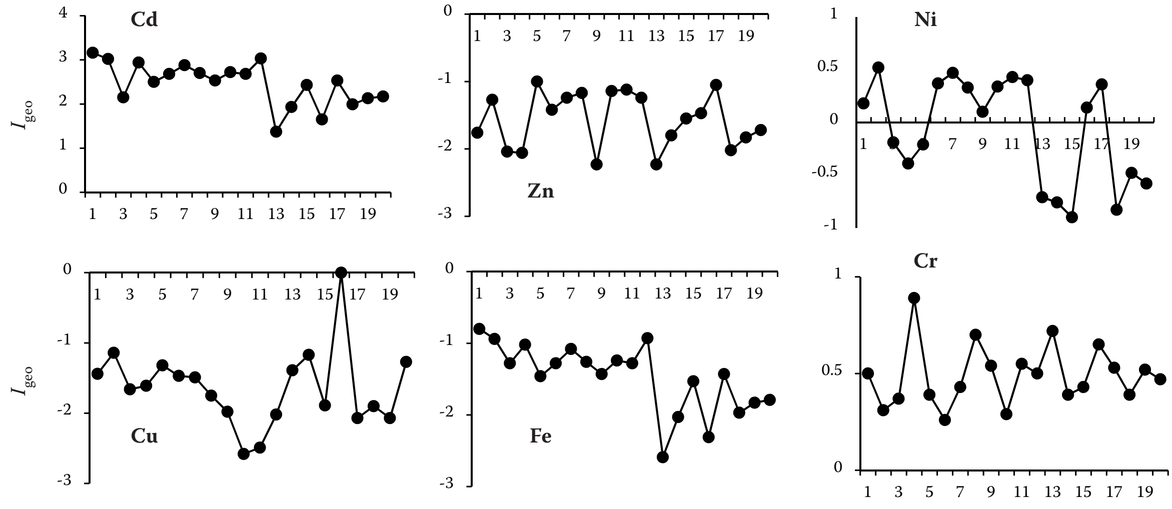

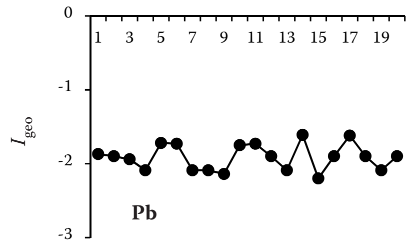

Figure 2. The schematic state of metal pollution in Hara Biosphere Reserve based on geo-accumulation index (I_{geo}) of Cd, Zn, Ni, Cu, Fe, Cr and Pb calculations

to 3.16). According to Figure 2 the variation trends of the I_{geo} index for Cu, Zn, Pb, and Fe are below the baseline contaminated zone (uncontaminated), while for Cr, Cd, and Ni the variation trends are illustrated to exceed the contaminated zone ($I_{geo} > 0$) for some sampling sites of the study. The pattern of E_{ri} of each heavy metal was more or less the same as its concentration (Figure 3). The ecological risk for Cd was high ($160 \leq E_{ri} < 320$) for most sampling sites. The ecological risk for the rest (i.e. Cr, Cu, Ni, Pb, and Zn) was low ($E_{ri} < 40$), for all sampling points overall. *QoC* analysis of metals studied is shown in Table 3 and Figure 4. This factor was used to describe the geogenic and anthropogenic sources of metal contamination in sediments samples (Zarei *et al.* 2014). The results showed that the concentrations of Fe, Cu, Pb, and Zn for all the sampling sites were mainly derived from geogenic sources with no evidence of anthropogenic impacts. Cr and Cd concentrations (except sampling site 20) were shown to be associated with anthropogenic source of contamination. The values of Ni also varied between the geogenic and anthropogenic sources. The *QoC* values of Ni exceeded the geogenic sources in sampling sites 13, 14, 15, 18, and 20. These might be related to the human activities including shipping and transport, urban and domestic wastewater, agriculture, industrial wastewater at shipbuilding

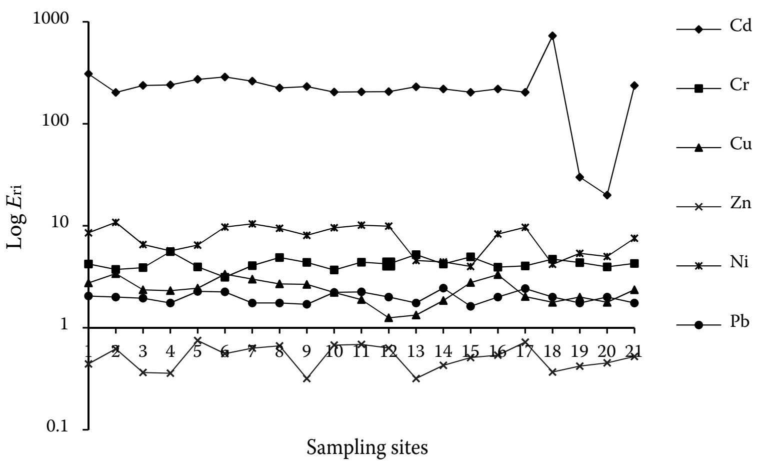

Figure 3. The potential ecological risk coefficients (E_{ri}) of the sediment in Mangrove of Hara Biosphere Reserve at sampling site

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Table 3. Quantification of contamination (*QoC*, %) values of metals in the sediments of Hara Biosphere Reserve

Sample	Fe	P	Ni	Zn	Cu	Cr	Cd
Average	−193.85[b]	−246.00[b]	27.12[a]	−161.14[b]	−207.51[b]	144.96[a]	76.18[a]
Max	−112.21	−194.28	53.74	−62.11	−116.69	215.14	95.89
Min	−399.71	−301.19	−25.23	−284.37	−387.65	77.17	−50.00

[a]anthropogenic magnitude; [b]geogenic source

Table 4. Enrichment factor values of metal concentrations in the sediments of Hara Biosphere Reserve

Sample	Pb	Ni	Zn	Cu	Cr	Cd
Average	0.77[a]	2.76[b]	0.98[a]	0.891[a]	4.24[c]	14.35[d]
Max	1.4	5.5	1.78	2.1	10.47	36.84
Min	0.47	1.53	0.48	0.31	2.39	1.54

[a]no enrichment; [b]minor enrichment; [c]moderate enrichment; [d]severe enrichment

plants and desalination facilities, coastal activities (for example marinas, jetties, ports, and harbours), and fishing boats (Kazemi *et al.* 2012). The results of *EF* calculated for the metals in the sediment are presented in Table 4. The results from the present investigation showed that *EF* for Cd ranged from 2.37 to 36.84, from 0.48 to 1.78 for Zn, from 0.47 to 1.40 for Pb, from 0.62 to 2.01 for Cu, from 1.53 to 5.50 for Ni, and from 2.39 to 10.47 for Cr. The analysis of *EF* also reveals minor to moderate enrichment for Pb, Zn, and Cu, minor to moderately severe enrichment for Cr and Ni, and moderately severe to severe enrichment for Cd. Fe was excluded from the analysis of *EF* mainly because of its selection as the background metal (normalization) in the calculations. *EF* values greater than 1.5 suggest that the sources are more likely to be anthropogenic. The *EF* average values for Cu, Pb, and Zn were below 1.5 for all sampling sites, indicating that these metals in most sediments were derived mainly from

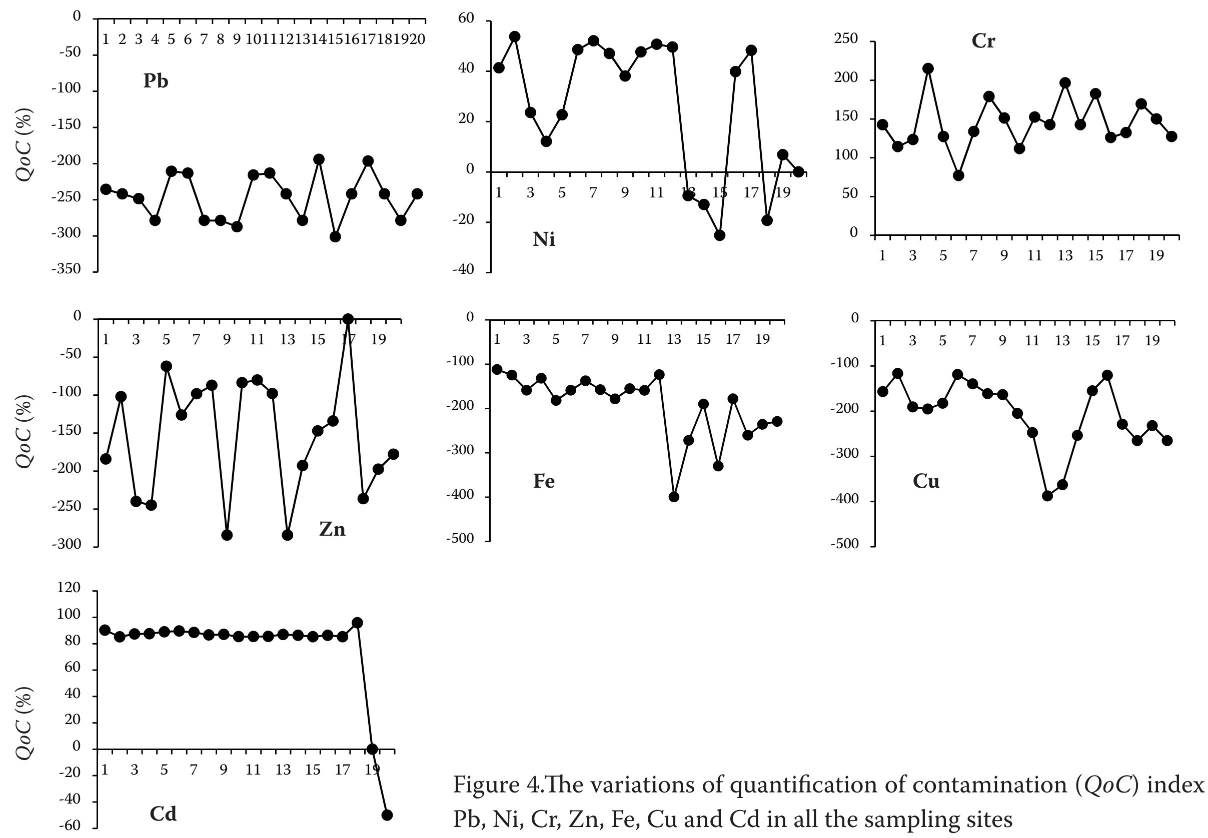

Figure 4.The variations of quantification of contamination (*QoC*) index of Pb, Ni, Cr, Zn, Fe, Cu and Cd in all the sampling sites

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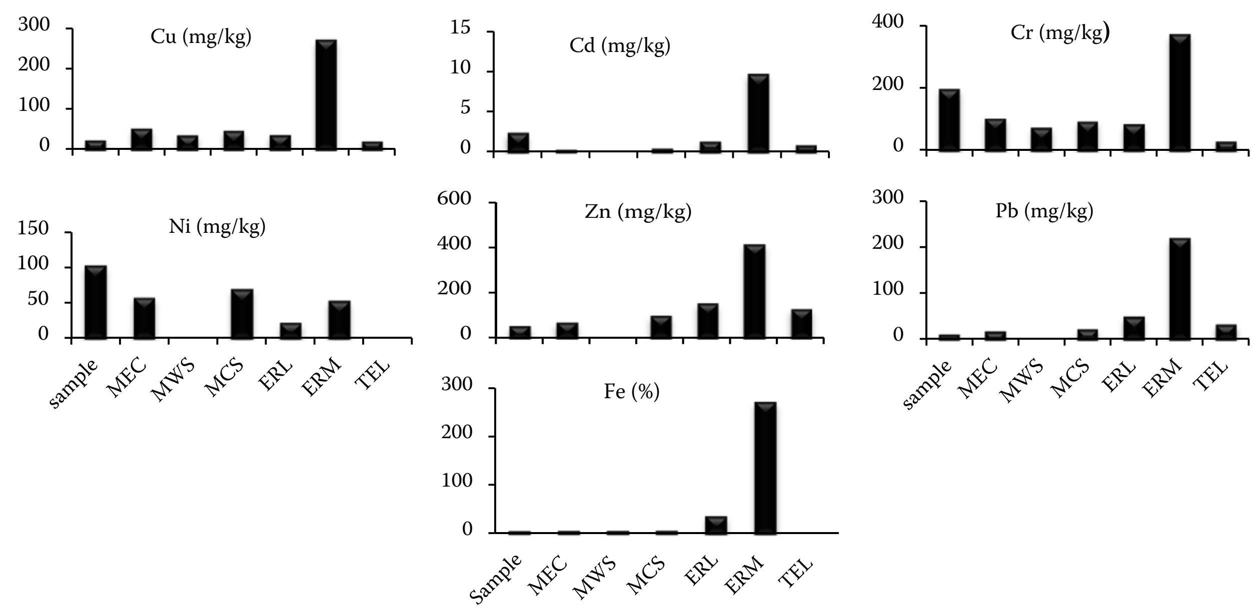

Figuer 5. Comparison of concentrations of metals in Hara Biosphere Reserve with the corresponding global baseline a sediment quality guideline (SQG) values Mean earth's Crust (MEC), Mean World Sediment (MWS), and Mean Continental Shale (MCS) are related to global baseline values, while threshold effect level (TEL), effects range Medium (ERM) and effect range low (ERL) (Adami *et al.* 2000; MacDonald *et al.* 2000) values are related to sediment quality guidelines (SQGs)

natural processes and were related to the exposure of the material from the Earth's crust (García *et al.* 2008; Zarei *et al.* 2014). *EF* values demonstrate that Ni, Cr, and Cd in the surface sediments of Hara Biosphere Reserve were enriched by anthropogenic activities. This is in agreement with the observation of González and Brügmann 1991 that municipal and/or industrial waste water discharges into coastal areas are the most important sources of contamination of water and sediment with heavy metals. The wastewater of most factories located around Hara Biosphere Reserve, such as Al-Mahdi aluminum factory, lead and zinc Qeshm factory, and Hormozgan cement factory, was discharged to the Persian Gulf and Hara Biosphere Reserve directly, without any remediation; only a simple physical screening has been performed. Lots of toxic metals like Pb, Cd, Ni, and Cr have been detected. Oil tankers, commercial ships and recreational boats traffic within the study area have released large amounts of Pb, Cd, and Ni-containing compounds into the water and sediments as well (Nowrouzi *et al.* 2012). In this investigation, we compared the total metal concentrations with the Canadian Sediment Quality Guidelines (SQG), Mean Earth's Crust (MEC), Mean World Sediment (MWS), and Mean Continental Shale (MCS) and threshold effect level (TEL), effects range median (ERM) and effects range low (ERL) (Adami *et al.* 2000; MacDonald *et al.* 2000). A comparison of the Cr and Cd concentration in sediments with the corresponding values of these metals in MEC, MWS, MCS, and SQG showed that the levels of Cr and Cd are higher than those in TEL, MWS, MCS, ERL, and MEC and lower than in ERM. Pb and Zn are at lower concentrations in comparison with TEL, MWS, MCS, ERM, ERL, and MEC values. The comparison of Ni concentrations with the studied standard values showed that the level of Ni exceeds all standard values, while the content of Cu is lower than that of ERL, ERM, MCS, MWS, MEC and higher than that of TEL and the level of Fe is lower than all standard values (Figure 5). Table 5 shows a comparison of metal concentrations in Hara Biosphere Reserve with results from similar studies in the world. According to these data the Fe, Zn, and Pb concentrations appear to be considerably lower than those reported from the other contaminated mangrove environments. However, the ranges of Cr concentration in the present study are higher than those reported previously. Our Ni values are lower than those observed for the South China mangrove sediments (Wang *et al.* 2013). The Cu values obtained are considerably higher than the literature values (El-Said & Youssef 2013; Wang *et al.* 2013; Zarei *et al.* 2014) and lower than the

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Table 5. Comparison of metal concentrations in sediments of Hara Biosphere Reserve with similar studies from other world mangroves (± standard deviation)

Location	Fe (%)	Cd	Cr	Zn	Pb	Ni	Cu
		(mg/kg)					
Hara Biosphere Reserve, Iran[a]	2.63 ± 0.04	2.63 ± 0.04	194.29 ± 0.04	49.39 ± 0.04	7.94 ± 0.04	101.48 ± 0.05	20.98 ± 0.04
Sg. Puloh mangrove estuary, Malaysia[b]	7.14	0.94	–	1023.68	78.84	35.54	46.89
Gulf of Kachchh, western coast of India[c]	5.6	4.8	163	82	20	69	86
Hara Biosphere Reserve, Iran[d]	3.74 ± 0.52	–	73.09 ± 1.20	39.54 ± 7.24	37.08 ± 6.61	–	16.09 ± 4.18
Hara Biosphere Reserve, Iran[e]	–	3.55 ± 0.54	–	–	36.65 ± 3.07	79.86 ± 5.72	–
Sirik Azini Creek, Iran[f]	–	31.06 ± 041	–	77.03 ± 6.01	68.27 ± 08	101.5 ± 3.72	26.89 ± 3.5
South China[g]	–	0.28–0.49	0.089–3.49	78.0–235.4	0.28–73.2	15.2–37.4	17.4–95.8
Red Sea, Egypt[h]	–	0.65–5.75	–	–	5.0–56	5.5–52.4	6.8–190.2
Eastern Fujian, China[i]	–	1.35 ± 0.96	–	105.14 ± 39.65	104.32 ± 51.12	–	30.46 ± 15.83

[a]The present study; [b]Udechukwu *et al.* (2015); [c]Kumar *et al.* (2015); [d]Zarei *et al.* (2014); [e]Nowrouzi *et al.* (2012); [f]Parvaresh *et al.* (2011); [g]Wang *et al.* (2013); [h]El-Said and Youssef (2013); [i]Zhao *et al.* (2015)

other literature values. Cd values are considerably higher than the literature values (Wang Yutao *et al.* 2013; Udechukwu *et al.* 2015), and lower than in other studies.

CONCLUSION

In this study, the mean concentrations of metals in Hara Biosphere Reserve decreased in the following order: Fe > Cr > Ni > Zn > Cu > Pb > Cd. Based on the geo-accumulation index, the Fe, Pb, Zn, and Cu levels in the sediment samples were graded as non-contamination, the levels of Cr and Ni as non-contamination to moderate contamination, while those of Cd as moderate contamination to moderate to heavy contamination. The *EF* results demonstrated that the metals in the study area have been enriched. The *EF* and *QoC* average values for Cu, Pb, and Zn showed that in most sediments these metals were derived mainly from natural processes and geogenic sources and were related to the exposure of the Earth's crust material, with no evidence of anthropogenic impacts. *EF* and *QoC* values demonstrated that Ni, Cr, and Cd were in the surface sediments of Hara Biosphere Reserve enriched by anthropogenic activities. The elevated values identified for Cd, Ni, and Cr might be related to human activities including human refuse, shipping, transportation, fuel smuggling, and industrial wastewater discharges from factories located around Hara Biosphere Reserve, such as Al-Mahdi aluminum factory, lead and zinc Qeshm factory and Hormozgan cement factory.

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