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


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 supposedly 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...
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 (Igeo), ecological risk factor (Er), 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
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 ($I_{\text{geo}}$) 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:

$$I_{\text{geo}} = \log_2 (Cn)/1.5 \times Bn$$

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 $I_{\text{geo}}$ is calculated using the global average shale data (TUREKIAN & WEDERFOHL 1961). According to GONZALEZ-MACIAS et al. (2006), the $I_{\text{geo}}$ for a metal is classified into seven grades: uncontaminated ($I_{\text{geo}} \leq 0$); uncontaminated to moderately contaminated ($0 < I_{\text{geo}} \leq 1$); moderately contaminated ($1 < I_{\text{geo}} \leq 2$); moderately to heavily contaminated ($2 < I_{\text{geo}} \leq 3$); heavily contaminated ($3 < I_{\text{geo}} \leq 4$); heavily to extremely contaminated ($4 < I_{\text{geo}} \leq 5$); and extremely contaminated ($I_{\text{geo}} \geq 5$) (MULLER 1969).

Ecological risk factor. The ecological risk factor ($E_r$) quantitatively expressing the potential ecological risk of a given contaminant suggested by HAKANSON (1980) is calculated as

$$E_r = T_r \times C/C_o$$

where:
- $T_r$ – 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)
- $C$ – 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 $T_r$ 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: $E_r < 40$, low potential ecological risk; $40 \leq E_r < 80$, moderate potential ecological risk; $80 \leq E_r < 160$, considerable potential ecological risk; $160 \leq E_r < 320$, high potential ecological risk; and $E_r \geq 320$, 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:

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

where $(M/Fe)_{\text{sample}}$ and $(M/Fe)_{\text{shale}}$ values, respectively, are the metal concentrations (mg/kg) dry weight.
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 ≤ EF ≤ 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 ≤ EF ≤ 5 indicates moderate enrichment, 5 ≤ EF ≤ 10 indicates moderate to severe enrichment, 10 ≤ EF ≤ 25 indicates severe enrichment, 25 ≤ EF ≤ 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):

\[
QoC \% = \left( X - Xe/X \right) \times 100
\]  

(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 < \( I_{geo} \) ≤ 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 heavily contaminated (\( I_{geo} = 1.37 \))

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

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

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

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fe</th>
<th>Pb</th>
<th>Ni</th>
<th>Zn</th>
<th>Cu</th>
<th>Cr</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max</td>
<td>−0.8</td>
<td>−1.61</td>
<td>0.52</td>
<td>−1.0</td>
<td>−0.001</td>
<td>0.89</td>
<td>3.16</td>
</tr>
<tr>
<td>Min</td>
<td>−2.6</td>
<td>−2.2</td>
<td>−0.9</td>
<td>−2.23</td>
<td>−2.58</td>
<td>0.26</td>
<td>1.37</td>
</tr>
<tr>
<td>Average</td>
<td>−1.47^a</td>
<td>−1.91^a</td>
<td>−0.07^a</td>
<td>−1.56^a</td>
<td>−1.63^a</td>
<td>0.5^b</td>
<td>2.46^c</td>
</tr>
</tbody>
</table>

^a uncontaminated; ^b uncontaminated/moderately contaminated; ^c moderately to heavily contaminated
to 3.16). According to Figure 2 the variation trends of the $I_{\text{geo}}$ 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 ($I_{\text{geo}} > 0$) 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...
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

<table>
<thead>
<tr>
<th>Sample</th>
<th>Pb</th>
<th>Ni</th>
<th>Zn</th>
<th>Cu</th>
<th>Cr</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average</td>
<td>0.77&lt;sup&gt;a&lt;/sup&gt;</td>
<td>2.76&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.98&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.891&lt;sup&gt;a&lt;/sup&gt;</td>
<td>4.24&lt;sup&gt;c&lt;/sup&gt;</td>
<td>14.35&lt;sup&gt;d&lt;/sup&gt;</td>
</tr>
<tr>
<td>Max</td>
<td>1.4</td>
<td>5.5</td>
<td>1.78</td>
<td>2.1</td>
<td>10.47</td>
<td>36.84</td>
</tr>
<tr>
<td>Min</td>
<td>0.47</td>
<td>1.53</td>
<td>0.48</td>
<td>0.31</td>
<td>2.39</td>
<td>1.54</td>
</tr>
</tbody>
</table>

<sup>a</sup>no enrichment; <sup>b</sup>minor enrichment; <sup>c</sup>moderate enrichment; <sup>d</sup>severe enrichment

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

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

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fe</th>
<th>P</th>
<th>Ni</th>
<th>Zn</th>
<th>Cu</th>
<th>Cr</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average</td>
<td>-193.85&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-246.00&lt;sup&gt;b&lt;/sup&gt;</td>
<td>27.12&lt;sup&gt;a&lt;/sup&gt;</td>
<td>-161.14&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-207.51&lt;sup&gt;b&lt;/sup&gt;</td>
<td>144.96&lt;sup&gt;a&lt;/sup&gt;</td>
<td>76.18&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Max</td>
<td>-112.21</td>
<td>-194.28</td>
<td>53.74</td>
<td>-62.11</td>
<td>-116.69</td>
<td>215.14</td>
<td>95.89</td>
</tr>
<tr>
<td>Min</td>
<td>-399.71</td>
<td>-301.19</td>
<td>-25.23</td>
<td>-284.37</td>
<td>-387.65</td>
<td>77.17</td>
<td>-50.00</td>
</tr>
</tbody>
</table>

<sup>a</sup>anthropogenic magnitude; <sup>b</sup>geogenic source

Table 4. Enrichment factor values of metal concentrations in the sediments of Hara Biosphere Reserve
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) values are related to sediment quality guidelines (SQGs).

Figur 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)
other literature values. Cd values are considerably higher than the literature values \cite{Wang2013, Udechukwu2015}, 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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Received for publication January 1, 2016
Accepted after corrections July 26, 2016
Published online February 3, 2017