Heavy metals in seawater, sediments and marine organisms in the Gulf of Chabahar, Oman Sea

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Levels of the heavy metals Copper (Cu), Zinc (Zn), Lead (Pb), Cadmium (Cd), Chromium (Cr), Nickel (Ni), Iron (Fe) and Manganese (Mn) were determined in coastal water, sediments and soft tissues of the Saccostrea cucullata, from the intertidal zone at five stations in the Gulf of Chabahar on the Iranian coasts along the Oman Sea. The concentrations of heavy metals in seawater ranged between 3.37 to 5.74, 18.01 to 22.62, 4.24 to 4.52, 0.15 to 0.19, 20.16 to 21.46, 16.42 to 17.14, 15.43 to 24.76 µg L\(^{-1}\) and 7.06 to 8.67 µg L\(^{-1}\) for Cu, Zn, Pb, Cd, Cr, Ni, Mn and Fe, respectively. The corresponding concentration values in the sediments were 46.79 to 54.76, 40.14 to 43.12, 25.63 to 28.23, 0.53 to 0.63, 47.16 to 51.43, 26.45 to 28.68, 52.13 to 53.46 and 84.42 to 89.14 µg g\(^{-1}\) for Cu, Zn, Pb, Cd, Cr, Ni, Fe and Mn, respectively. The highest accumulated metals were Zn, Cu and Mn in, S. cucullata while the lowest one was Cd. The highest concentrations of all metals in seawater, sediments and Oyster were recorded at Tiss harbour eastern parts of the Gulf, while the lowest concentrations were recorded at Damagheh. Based on this research, land based activities; shipping activity and the sewage disposal from vessels and residential area close to these harbors are the main source of metal pollution in the Gulf of Chabahar.

Key words: Heavy metals, seawater, sediment, Saccostrea cucullata, Oman Sea, Gulf of Chabahar.

INTRODUCTION

Marine pollution is a global environmental problem; human activities in the coastal area and marine water contribute to the discharge of various kinds of pollutants such as heavy metals into the marine ecosystems (Censi et al., 2006; Pote et al., 2008). The main reason for the metal contamination is considered as persistent and due to their toxic properties, could create several problems for different kinds of marine ecosystems and could be accumulating in marine organisms (Wen et al., 2007; Wcislo et al., 2008). Many marine organisms have the potential to bioconcentrate high levels of metals from their environment (Fowler, 1990; Phillips and Rainbow, 1993; Szeffer et al., 1999). Metal bioaccumulation by marine organisms has been the subject of considerable interest in recent years because of serious concern that high levels of metals may have detrimental effects on the marine organisms and may create problems in relation to
their suitability as food for humans. The pollution levels of the aquatic environment by heavy metals can be estimated by analysing water, sediments and marine organisms. The levels of heavy metals in Oysters and other invertebrates are often considerably higher than in other constituents of the marine environment. Compared to sediments, marine organisms exhibit greater spatial sensitivity and therefore, are the most reliable tool for identifying sources of biologically available heavy metal contamination (Goldberg et al., 1978; Koide et al., 1982; Thomson et al., 1984; Szefer, 1986). As a result, biomonitoring process has been widely used to monitoring metals in the last two decades (Zelika et al., 2003; Nicholson and Lam, 2005; Stanly et al., 2008).

Different types of organisms may be used for biomonitoring, such as marine algae (Topcu et al., 2003; Besada et al., 2009) and filter-feeding mollusks (Mashinchian Moradi, 2001; Zorita et al., 2006; Hamed and Emara, 2006). Many studied showed, bivalves do not regulate the level of some metals within their body (Stanly et al., 2008) and they can deflect the metals contamination from surrounding area. Thus, bivalves to be considered as good biomonitor agents for heavy metal monitoring in aquatic ecosystems (Elfving and Tedengreen, 2002; Yap et al., 2003; Zelika et al., 2003; Nicholson and Lam, 2005; Zorita et al., 2006; Vlahogianni et al., 2007; Maanan, 2008). Gulf of Chabahar on the Iranian coasts along the Oman Sea is a developing area; traditionally this area is used for fisheries and aquaculture activities of the local people.

However, it has developed as an important free zone for Iran import and export, economy, urbanization and industries have also grown in the area parallel to economic development (Amini-Ranjbar and Miraki, 2006). Although, the area is still growing and especial weather seems to greatly influence on distribution and redistribution of metals in coastal ecosystems, there is a lack of information concerning trace metals contamination in Chabahar. Such information could be used to establish baselines in the area. The study of De Mora et al. (2004) in southern coasts of the Oman Sea showed some metals like the Ni that have natural source in this area. Meanwhile, the human activity including shipping, fishing and canny industry can increase the Pb concentration in coastal area. These activities are conducted in Iranian coasts of the Oman Sea. These metals also can be accumulated in tissues of organisms (Gochfeld, 2003; Yi et al., 2008). Therefore, for the first time, the concentrations of trace metals (Cd, Pb, Cu, and Zn) in seawater, sediments and its accumulation in the tissues of Oysters (Saccostrea cucullata) and green algae have been determined and compared in different marine environmental compartments in the area of the coast of India and Southern coast of Oman sea. As there is no quantitative data available on the concentration of these metals in seawater, sediment and marine organisms in this coastal part of the northern Oman Sea, the results of this study could serve as a baseline for future assessments of anthropogenic effects in this part of the Oman Sea.

**MATERIALS AND METHODS**

**Study area**

The Gulf of Chabahar is located in the south-east of Iran and north-east of the Oman Sea (25° 17' 28" N 60° 38' 15" E). This Gulf is the largest embayment in the Oman Sea and extends for about 350 km from the Konarak city in the west to Chabahar city in the east (Figure 1). The width of the Gulf varies between 14 and 17 km, and its depth throughout its axis is fairly constant with a mean of 16 m. Historically, due to its antiquity as a port near the Oman Sea, it assumed importance in trade and was the centre of business and navigation. Shahid Kalantary and Shahid–Beheshhti ports are two important ports in Gulf of Chabahar. The executive operation of this port started in 1981 and it became operational with the completion of four jetties in 1983. This area has a subtropical weather with two distinct seasons, dry season in summer and rainy season in winter. It also experience monsoon (seasonal western winds) during summer and winter. The seawater is completely disturbed by huge waves during monsoon period (Zareii, 1995). Therefore, considerable amount of sediment is displaced by the power of waves. This condition could result in redistribution of metals in the coastal environment.

**Sampling and analytical methods**

Samples of seawater (0.5 m depth from the seawater surface) and sediments (about 5 to 10 cm thickness of the surface sediment with a stainless steel grab sampler during low tide) were collected three times, representing different seasons, during winter and summer, 2013 from the intertidal zone of five stations in the western to eastern side of the Gulf of Chabahar (Figure 1). Sampling location was detected using GPS localization. The stations were as follows: Station I: infront of the Shahid–Beheshhti Port, Station II: near to the industrial activity of Chabahar free zone and in-front of the Tiss Harbour, Station III: in-front of the old sewage drain, near to the desalinization unit and industrial activity, Station IV: north of the Konarak Port, and Station V: in-front of the Damaghe area and away from industrial activity. All stations are situated in the proximity of different geochemical, hydrological and human impacts. Total metals in seawater samples were extracted by using the APDC–MIBK procedure (Brewer et al., 1969; APHA, 1989). The pH of seawater sample was adjusted to about pH 3 to 4, add 5 ml ammonium pyrrolidine dithiocarbamate (APDC) solution, and shake to mix. Add 50 ml methyl isobutyl ketone (MIBK) and shake vigorously for 30 s. The extracted organic layer was aspirated directly to the flame-atomic absorption spectrophotometer to determine the metals concentration. Sediments were dried in the oven at 70°C then kept in polyethylene until analysis (Amini-Ranjbar, 1998). Sediment samples of 0.5 g were digested in Teflon vessels for 2 h with a mixture of 3: 2:1 HNO3, HClO4 and HF acids, respectively, according to the method described by Origioni and Aston (1984). The two species of the most common marine organisms were examined for trace metals levels in four stations in the study area. Samples were stored after collection in plastic bags in a freezer. The soft part of live oysters was dissected to separate the different organs. Digestive glands, headfoot in addition to whole animal were digested in concentrated nitric acid (FAO, 1976). The metals, Fe, Cr, Ni, Cd, Pb, Cu, Mn and Zn were analyzed by using flame-atomic absorption spectrophotometer. The precision of analysis was checked by replicate measurements of target metals.
in a marine biota sample. Good precision (7.1 to 8.9%) was observed for all metals.

RESULTS

Heavy metals in seawater

The variations of heavy metals concentrations in seawater samples in summer and winter 2013 from the five stations of the Gulf of Chabahar are summarized in Table 1. Concerning the sites variation, the highest concentrations for all metals were recorded at Station II (Tiss Port) in the eastern part of the Gulf of Chabahar (Figure 2). These high concentrations may be attributed to sewage and wastes discharged from the Industrial activities related to Chabahar Free Trade-Industrial Zone, shipping activity (such as: repairing, fueling, greasing and painting of fishing ships) and boats. The remarkably high Cu concentration at Station II in the eastern part of the Gulf may be due to corrosion of ships’ hulls coatings and anti-fouling paints at Tiss port. Similarly, Zn increased at Station II during winter and decreased during summer; this may be due to its consumption by phytoplankton, which increases in spring and summer as stated by El-Samra et al. (1995). On the other hand, Abou-El-Sherbini and Hamed (2000) reported that the removal of zinc from seawater could be aided by phytoplankton, which spread in spring and summer. Also, the center parts (Station III) of the Gulf of Chabahar could be heavily contaminated by Cr and Ni and high levels of Fe and Mn. This may be due to the natural origins. The geology of southeastern of Iran (Makoran zone) is rich in ophiolites and metalliferrous sediments of marine origin. The Makoran ophiolites contain chromite and various nickel sulfide minerals (Jacob and Quittmeyer, 1979; Mcall, 1997; Farhoudi and Craig, 1977). Thus, the high metal concentrations are most likely due to the local mineralogy, and are natural, rather than pollution.

On a seasonal scale, Cu and Zn showed their highest values during winter (5.74 and 22.62 µg L⁻¹) where the lowest values were exhibited during summer (1.08 and 5.32 µg L⁻¹). The levels of Pb and Cd varied from
Table 1. Heavy metal concentrations (µg L⁻¹) in seawater samples collected from the Gulf of Chabahar during winter and summer, 2013.

| Station | Cu   | Zn   | Pb   | Cd   | Cr   | Ni   | Fe   | Mn   |
|---------|------|------|------|------|------|------|------|------|
| Winter  |      |      |      |      |      |      |      |      |
| Ι       | 3.22 | 9.76 | 2.08 | 0.16 | 12.76| 9.69 | 22.18| 4.75 |
| ΙΙ      | 5.74 | 22.62| 4.52 | 0.19 | 15.59| 10.08| 19.70| 2.12 |
| ΙΙΙ     | 2.68 | 8.43 | 1.91 | 0.1  | 21.46| 17.14| 15.43| 7.06 |
| ΙV      | 2.90 | 11.57| 1.68 | 0.13 | 15.12| 10.85| 25.36| 5.18 |
| V       | 1.16 | 6.72 | 1.16 | 0.12 | 9.63 | 5.63 | 11.52| 3.32 |
| Summer  |      |      |      |      |      |      |      |      |
| Ι       | 3.03 | 8.83 | 2.75 | 0.13 | 9.34 | 12.96| 23.82| 4.56 |
| ΙΙ      | 3.37 | 18.01| 4.24 | 0.15 | 17.67| 14.89| 16.48| 2.46 |
| ΙΙΙ     | 2.42 | 13.36| 0.98 | 0.1  | 20.16| 16.42| 24.76| 8.67 |
| ΙV      | 2.08 | 12.58| 1.79 | 0.16 | 17.34| 8.46 | 19.18| 3.29 |
| V       | 1.06 | 5.32 | 1.13 | 0.1  | 15.99| 3.20 | 13.82| 3.66 |

1.13 and 0.1 µg L⁻¹ during summer to 4.52 and 0.19 µg L⁻¹ during winter, respectively. The maximum values of Cr and Ni recorded 21.46 and 17.14 µg L⁻¹ during winter and their minimum values (9.34 and 3.20 µg L⁻¹) during summer, while Fe recorded the maximum (25.36 µg L⁻¹) and the minimum (11.52 µg L⁻¹) during winter. In contrast, Mn recorded its maximum value (8.67 µg L⁻¹) during summer and the minimum value during winter (2.2812 µg L⁻¹).

Generally, the highest values of heavy metals were observed during summer and the lowest during winter except Mn. Moreover, the concentrations of heavy metals in the coastal water of the Gulf of Chabahar could be arranged in the following sequence: Fe>Cr>Zn>Ni>Cu>Fe>Mn>Cd. From these results (Table 1), the local distribution of metals in water at different stations also varied during winter and summer 2013. The maximum value of Cu, Zn, Pb, and Cd were found at Station II, Ni at Station III and Fe at Station IV during winter, while the highest value of Cr and Mn was recorded at Station III during summer. The minimum values of Cu, Zn, Pb, Ni and Cd were found at Station V but Cr were found at Station I during summer, while Fe and Mn recorded the minimum values at Stations V and II during winter (Figure 2).

Heavy metals in sediments

Table 2 shows the concentrations of heavy metals (Cu, Zn, Pb, Cd, Cr, Ni, Fe and Mn) in sediment samples collected from intertidal zone from the five stations at the Gulf of Chabahar during winter and summer 2013. The highest values for metals in the Gulf of Chabahar were recorded at the eastern part (Tiss Port), while the lowest ones were recorded at Station V in the western part of the Gulf (Figure 3). This means that anthropogenic contribution to the heavy metal concentrations at the eastern part of the Gulf was clearly noticed in sediments. The local distribution of metals in sediments gave similar pattern with that found in seawater, with highest concentrations of Cu, Zn, Pb and Cd levels in Station II, while highest Cr, Fe and Mn in Station III. This means that the factors affecting the levels of metals in seawater and sediments are similar.

Although, the results indicated that the accumulation of heavy metals are predominant in sediments rather than seawater. This can be interpreted as sediments act as reservoir for all the contaminants and dead organic matter descending from the ecosystem above. The relatively high concentrations of heavy metals during winter coincide principally with decreasing rate of organic matter decomposition, due to low water temperature. The highest concentrations of Cu, Zn, Pb, Cd and Ni were found at Station II (54.76, 43.12, 28.23, 0.63 and 28.68 µg g⁻¹) and its lowest values at Station V (10.97, 16.21, 10.72, 0.21 and 8.32 µg g⁻¹), respectively. Cu ranged from 10.97 µg g⁻¹ (Station V) to 54.76 µg g⁻¹ (Station I). Also, Ni, Cd, Pb and Zn gave a similar pattern for their lowest and highest concentrations, where their lowest values were found at Station V respectively, during summer. The local distribution of Cu in sediment at Station II showed higher concentration than the other stations. This may be due to the anti-fouling paints. The seasonal concentration of sedimentary zinc showed higher level at Station II (43.12 and 40.14 µg g⁻¹) during winter and summer, respectively, while the lowest levels recorded 16.21 µg g⁻¹ during winter and 18.77 µg g⁻¹ during summer at Station V. This may be due to the high amount of suspended organic matter coming from different industrial wastes that precipitate to the bottom and also due to the decrease uptake of zinc by
phytoplankton at lower temperature. In the present study, Pb content of sediments ranged from 13.88 to 28.23 µg g⁻¹ and 18.77 to 40.14 µg g⁻¹ at Stations V and II during winter and summer, respectively. The high sedimentary Pb content may be due to the precipitation of decomposed organic matter. The maximum value of Mn (89.14 µg g⁻¹) was observed at Station III probably due to natural origins at the northern part of the Gulf. Fe and Cr accompanied Mn, which recorded its maximum value (53.46 and 51.43 µg g⁻¹) at the same Station III. This can be explained by common origin of the ophiolitic rocks, fact that their hydroxides precipitated together in the bottom (Price and Calvert, 1973), and the common geochemistry of both (Beltagy et al., 1983).

Heavy metals in *S. cucullata*

Table 4 shows the concentrations (µg g⁻¹ dry weight) of heavy metals in whole animal of oysters (*S. cucullata*) collected from five stations at the Gulf of Chabahar during winter and summer 2013, respectively. The highest mean values of the heavy metals were found at Station II, for Cu (145.07), Zn (191.25), Pb (17.48), Cd (0.45) and Ni (Table 3). This is due to the fact that seawater of the eastern parts of the Gulf (near to the industrial activity of
Chabahar free zone) has higher levels of metals than other parts. In other words, spatial data show the agreement between the concentration levels of heavy metals in the seawater as well as in S. Cucullata (Figure 4). On the other hand, the lowest mean values of the studied heavy metals were away from industrial activity and human impacts (Station v). There have been many suggestions that molluscs can be used as monitors of contamination by trace metals in aquatic environments (Shuster and Pringle, 1968; Boyden, 1974; Darracott and Watling, 1975).

Comparisons of metal levels in oysters should be made with caution because of factors such as, differences in the sampling times, size (age) of oysters, genetic differences, individual variability in metal uptake ability, gonadal maturation of organisms, and induction of metalbinding proteins can also influence the result (Schuhmacher and Domingo, 1996). Although,
comparison between different stations is open to criticism, the mean concentrations of all studied metals in *S. cucullata* were of the same range with exception for the extreme values. For example, the highest mean value of Pb (17.48 µg g⁻¹) was higher 7.4 times than the lowest mean value (2.36 µg g⁻¹), while other values, which varied between 145.07 and 59.25 µg g⁻¹ have insignificant range (Table 3). In the present study, the highest mean value of Zn (191.25 µg g⁻¹) at Station II was higher 2.18 times than the lowest ones (87.56 µg g⁻¹) at Station V, while other values, varied between 93.67 and 127.53 µg g⁻¹ with insignificant range (Table 3).

**DISCUSSION**

In this study, the levels of natural and anthropogenic heavy metal contamination metals (Copper (Cu), Zinc...
(Zn), Lead (Pb), Cadmium (Cd), Chromium (Cr), Nickel (Ni), Iron (Fe) and Manganese (Mn) in seawater, sediments and soft tissues of the S. cucullata, from the intertidal zone at five stations in the Gulf of Chabahar on the Iranian coasts along the Oman Sea were examined and reported for the first time. In summary, the order of Cu, Zn, Pb, Cd, Cr, Ni, Fe and Mn concentrations in seawater, sediments and oysters (S. cucullata) from the Gulf of Chabahar was Fe>Cr>Zn>Ni>Mn>Cu>Pb>Cd; Mn>Cu=Fe>Cr>Zn>Pb>Ni>Cd and Zn>Cu>Mn>Ni>Cr>Pb>Fe>Cd, respectively.

The local distribution of metals in sediments gave similar pattern with that found in seawater. Although, the results indicated that the accumulation of heavy metals are predominant in sediments rather than of seawater. This can be interpreted as sediments act as reservoir for all the contaminants and dead organic matter descending from the ecosystem above. The relatively high concentrations of heavy metals during winter coincide principally with decreasing rate of organic matter decomposition, due to low water temperature. The highest values for metals in the Gulf of Chabahar were recorded at the eastern part, while the lowest ones were recorded at western part of the Gulf. This means that anthropogenic contribution to the heavy metal concentrations at the eastern part of the Gulf was clearly noticed in sediments. Comparison of the heavy metal levels in the present study with background concentrations of heavy metals in seawater and sediments are presented in Table 2. As the table shows, Cu, Pb, Zn, Cr and Ni levels generally exceeded the background levels in Open Ocean. While, Zn concentration is only less than the coastal water level but Cu higher than the coastal water concentration. Zn and Cd concentration lies in the range of coastal water concentration (0.30 to 70.0 and 0.01 to 0.17 µg/l) reported by Bryan and Langston (1992), UNEP (1993) and Sadiq (1992). Ni levels in sediment samples found in this study are higher than the background levels. Pb, Cd, Cu, Zn and Cr lies in the range of background level of sediment.

The order of heavy metals concentrations in seawater, sediments and Marine Organism (S. cucullata) from the Gulf of Chabahar was Marine Organism>sediments>seawater, respectively. It can therefore be concluded that marine organism is a good indicator for essential and toxic metals, Zn and Cu. Ayling (1974) suggested that different mechanisms exist for the uptake of Cu, Cr, Zn and Cd within the oyster Crassostrea gigas. These mechanisms may also vary with physiological and environmental factors (Bryan, 1973) or even with the sexual state of the animal (Alexander and Young, 1976). Thus, the situation described above could arise as a result of both environmental and physiological factors. So the observed variation in metal levels in S. cucullata at different stations may be related to one or both of the following mechanisms: 1) the availability of different metals to the animal varies with different stations, and 2) the animal involves different uptake and retention mechanisms for the same metal at different stations. Therefore, the order of metal accumulation in the animal was: Fe>Zn>Cu>Mn>Ni>Cr>Pb>Cd, where, Fe was the highest followed by Zn, Cu, Mn, Ni and Pb then Cd. This order may be due to variation in the levels of discharged metals at different stations and also the chemical changes of metal before being taken up by the tested limpet.

The second mechanism could operate for one or two reasons, either the different animal populations have various abilities to accumulate metals at the different stations or the uptake of other metals. The second alternative has been extensively studied with other molluscs (Romeril, 1971; Martin and Felgal, 1975; Howard, 1975; Coughtrey and Martin, 1977). It is also suggested by the results of the present data that a significant correlation exists between Mn, Cr and Ni at all stations. Coughtrey and Martin (1977) showed that the correlation between Cd, Pb, Zn and Cu for Helix aspersa was significant at the less contaminant site and non-significant and marginally significant at the more contaminant site.

Conclusion

Generally, the land based activities and the shipping activity (such as: repairing, fueling, greasing and painting of fishing ships) and boats in the studied area are the main source of heavy metals in the Gulf of Chabahar. The level of Cr, Mn and Ni did not significantly change among different station suggesting that these three elements originate from natural sources and due to the Regional Geology of Makoran zone. The concentration of Pb, Zn and Cu varied among different station with maximum levels in Station II (Tiss harbour). Human activities such as shipping, marine transportation and fisheries may be associated in heavy metals enrichment in the sediment. During summer heavy metals concentration in Chabahar sediment increased markedly. It is suggested that increasing of nutrient availability due to upwelling during monsoon season (summer) enhanced phytoplankton growth followed by increasing of suspended organic matter which should be involved in heavy metals enrichment of sediment. This study showed the pattern of heavy metals accumulation in different S. cucullata in five stations. It could be related to the biological role of those metals in the body. The stronger correlation coefficients were obtained for metals concentration between sediment and soft tissue of oyster, it is generally more sensitive and precise biomonitoring material for heavy metals than the soft tissues of S. cucullata. Soft tissue of S. cucullata for heavy metals was
useful tool for biomonitoring of total metals studied in Iranian coasts of the sea of Oman.

Conflict of interests

The author has not declared any conflict of interests.

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