Seasonal variation of heavy metal in seawater, sediment and hooded oyster, *Saccostreacucullata*, in Iranian southern waters (Chabahar coast)

M. Reza Mirzaei[1,*], Mahmoud Reza Azini[1], and Teymour Amin Rad[1]

[1]Offshore Fisheries Research Center, Chabahar, Iran

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Abstract

Seasonal variation of different heavy metal concentrations; iron (Fe), copper (Cu), cadmium (Cd), lead (Pb), zinc (Zn), chromium (Cr), manganese (Mn), and nickel (Ni) were determined in water, sediment, and Oyster tissue of *Saccostreacucullata* from three stations along Chabahar coast, north coast of Oman Sea. The concentration order of heavy metals was as follows: Fe > Cr > Zn > Ni > Mn > Cu > Pb > Cd (seawater), Zn > Cu > Mn > Ni > Cr > Pb > Fe> Cd (sediment) and Zn > Cu > Mn > Ni > Cr > Pb > Fe > Cd (Oyster tissue). There was significant difference ($p<0.05$) in metals levels between different seasons and stations. Generally, the highest values of heavy metals were detected during summer and the lowest values during winter. The highest concentrations of heavy metal in seawater, sediment and oyster tissue were recorded at site II where is near the harbour and industrial activities. The highest concentration of Mn, Fe, Cr, Cd, and Pb was detected in sediment and the highest concentration of Ni, Zn and Cu was identified in Oyster tissue. This study showed that there was a positive significant correlation between accumulation of metals in oysters, seawater, and sediment. The results showed that the sediment is a suitable indicator of heavy metal pollution, because it can work as a hunter agent for heavy metals and an adsorptive sink in aquatic environments. Furthermore, oysters accumulate certain heavy metals in their tissue and can be considered as a bio-indicator to determine heavy metals pollution on ecosystems.

Keywords: Heavy metals; Sediment; Seawater; Oyster; Bio indicator; Oman Sea.

1. Introduction

The presence of heavy metals in marine environments and their deposition in fish and bivalve tissue were completely investigated throughout the past few years due to their harmful influences in living organisms and human health. Marine organisms can deposited heavy metals and provide a feature for measure concentration of heavy metals in their tissue depending on the species (Fang *et al.*, 2003). These types of toxins could cause extensive influences on living organisms in sea and ocean, even in noticeable impact compared to other sources of pollution (Govind and Madhuri, 2014). A wide range

* Corresponding Author: mirzaei.mr@gmail.com

of heavy metals have been known that is essential for living organisms at low concentrations and can be harmful in high concentrations (e.g., Cu, Fe, Mn, and Zn). Järup (2003) stated long-term exposure to arsenic in water is mainly related to increased risks of skin cancer. On the other hand, a group of heavy metal are classified in toxic categories (e.g., Cd, Hg, and Pb). Jaishankar *et al.* (2014) described toxicity of cadmium (Cd) and lead (Pb) along their health effects. Govind and Madhuri (2014) stated sometimes the action of essential elements can be changed by the toxic metals, resulting into toxicity by interfering with the metabolic process.

Most bivalves are filter feeders of phytoplankton, zooplankton, benthic food organism, and inorganic matter from the bottom of water (Cugier *et al.*, 2010). Therefore, they can absorb heavy metals from water, sediment and ingested phytoplankton (Wang, 2001). Fish and bivalve tissues were chosen for heavy metal investigation due to the human consumption and health risk (de Mora *et al.*, 2004; Yi *et al.*, 2011; Copat *et al.*, 2012). This feature along with long life span, living in sediment and sea floor and from the shallow waters near shore to great depths in the ocean makes them remarkable bio-indicators of the pollution level of the marine ecosystem (Boening, 1999; Cosson, 2000).

A number of authors have considered the effects of heavy metals on seawater, sediment and marine organism. El-Moselhy and Gabal (2004) showed spatial distribution patterns for the highest mean values of the different heavy metals on seawater. Pempkowiak *et al*. (1999) used metal contents in sediments to explain the different contents of metals in marine organisms in the corresponding sites. Furthermore, Dural et al (2007) stated bio-concentration factors calculated for marine species, correlated with metals concentrations in water/sediments. There is little published data on heavy metals from the southern water of Iran, and Chabahar coast. Most studies in heavy metals have only been carried out in a small areas such as heavy metal contents in water and sediment in Gulf of Chabahar (Bazzi, 2014); and distribution and origin of heavy metals in the sand sediments in a sector of the Oman Sea (Pakzad *et al.*, 2014). Therefore, this study attempts to make a major contribution to research on heavy metals by demonstrating seawater, sediment, and living organisms; and suggests a number of future research directions. The primary purpose of this study was to obtain quantitative information on the concentration of heavy metals in *Saccostreacucullata* species from southern water of Iran. The results reported here will provide valuable information on heavy metal pollution along the Chabahar coast. Furthermore, this study tries to show the relationship between metal contents in the oyster tissue, water and sediments in where they inhabit.

2. Materials and methods

2.1. Sample collection

Samples of water, bottom sediments, hooded oyster, *Saccostreacucullata*, were prepared according to the procedure used by Bazzi (2014) and Hung *et al.* (2001) at three locations along the Oman Seacoast, Chabahar coastal waters, during January 2014 (winter) and July 2014 (summer) (Figure 1). Stations were selected based on their expected contamination status as follows: Station I (Beris) was located at sewage drain area and near to harbour, Station II (Tis) near harbour and industrial activity, and station III (Lippar) in an open area and away from pollution sources.

Replicate sediment samples were collected using a Wildco Hand core sediment' sampler from the top layer of bottom sediment (5 cm depth). All sediment samples were placed in plastic bags and were immediately transferred to the laboratory. Oyster specimens were collected randomly by hand picking during low tide from 0.5 m to 1.0 m depth. Elimination of undigested matter was performed by flush out filtered seawater for 24 hours. The external surface of collected oysters was thoroughly cleaned to remove adhering on shell layer. Oysters

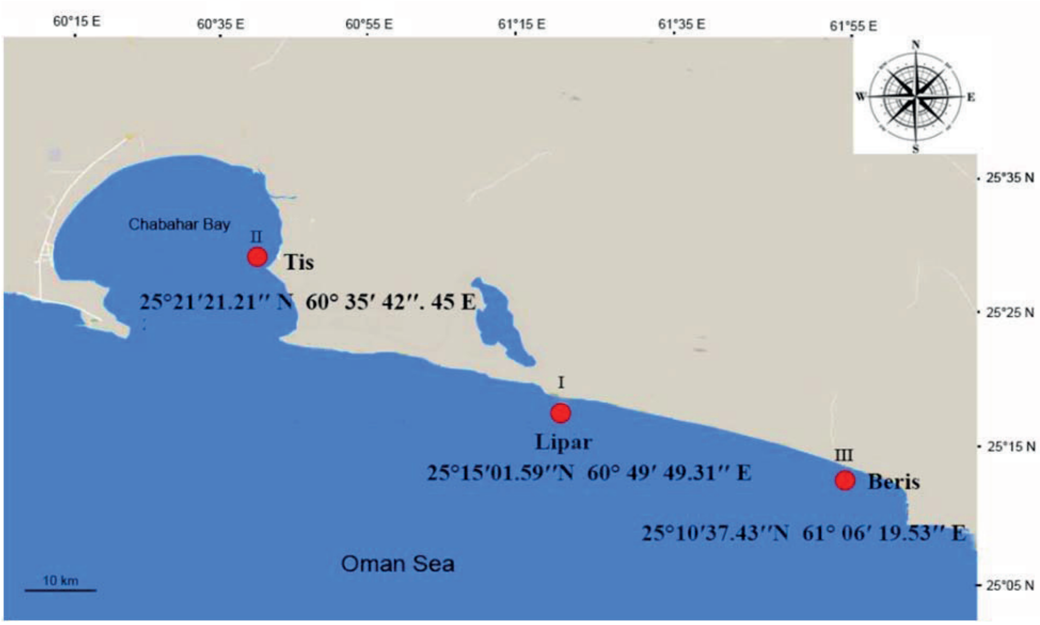

Figure 1. Location of study site, Chabahar coast, North Coast of Oman Sea

were dissected and soft tissues of individuals were removed from the shells with a plastic knife. Surface and bottom seawater samples were collected at a depth of 0.5 m from the same three selected sites by in polypropylene bottles. Seawater samples were filtered on study sites. The filtered seawater stored in a polycarbonate (P C) bottle and transferred to the laboratory and keep at a temperature of 5 °C for further analysis.

2.2. Sample preparation

Seawater, sediment and oyster tissue were prepared based on standard method described by Hung *et al.* (2001). The sediment samples and shells were dried for 2 hours at 105 °C, and pooled samples were prepared separately by ground to a fine powder and homogenizing the < 63 µm of the samples for further analysis. The soft tissues of bivalve placed in ten pools to reduce individual variations in heavy metal concentrations. The soft tissues were dried for 24 hours at 70 °C. Dried samples homogenized by grinding and sieved with a 63-µm nylon mesh. Regarding sample preparation for metal analysis, sediment, soft tissues, and shells powder digested with HNO3 by High-pressure digestion bombs for 24 hours in an oven at 120 °C. Then, the samples were diluted to a final volume of 25-30 ml with milli-Q. In order to determine heavy metals in water samples, 100 ml of seawater digested at 100 °C in 5ml nitric acid (HNO3) for 8 hours. The digested seawater samples were kept at room temperature for 12 hours.

2.3. Analysis of metal concentration

The concentrations of Cd, Co, Cr, Cu, Fe, Mn, Ni, and Pb in the digested samples were determined three times by the fast sequential atomic absorption spectrometer. Replicate blanks, Tissue Standard Reference Material (SRM 2976) and certified international marine sediments (BCSS1) were used as standard reference materials for analytical quality control and assurance. Metal concentrations as standard reference materials were between 73% and 85% confidence interval of certified values.

2.4. Statistical analysis

One-Way analysis of variance (ANOVA) was used to indicate significant differences in metal levels between heavy metals concentration in the samples of various stations and between seasons (Hédouin *et al.*, 2009). A Pearson correlation was used to identify differences between metal concentrations in water, sediment, and oyster tissue (Chouvelon *et al.*, 2009).

3. Results

3.1. Metal concentrations in water

The mean concentrations of the investigated metals in seawater, sediment, and oyster tissues are given in Table 1. Accumulation of the mean values of heavy metals in seawater was higher than permissible limits at sites I and II (Beris and Tis), while heavy metal levels were found to be lower or equal to permissible limits established by FAO at site III (Lippar). Average heavy metal concentrations of examined seawater in the three study sites were significantly higher in summer than winter (0< 0.05). The study found that the maximum mean concentration of Cu, Zn, Pb, Cd (station II, Tis), Ni (station I, Beris) and Fe (station III, Lippar) was observed during winter, while the highest concentration of Cr and Mn were found at station I during summer. On

Table 1. Element concentrations in seawater (μg L^{-1}), Sediment (μg g^{-1}) and Oyster tissue (μg g^{-1}) from Chabahar coast during winter and summer 2014

	Sediment	Tissue	Water			Sediment	Tissue	Water
Mn	40.89	58.46	2.99		Mn	42.33	46.15	3.30
Fe	12.83	0.96	10.39		Fe	11.52	1.42	12.47
Ni	12.29	17.57	5.08		Ni	7.50	15.73	2.89
Cr	17.30	14.33	8.69		Cr	23.42	13.32	14.42
Cd	0.32	0.09	0.11		Cd	0.19	0.12	0.09
Pb	12.52	3.94	1.05		Pb	9.67	2.13	1.02
Zn	14.62	78.98	6.06		Zn	16.93	87.73	4.80
Cu	27.44	78.88	1.05		Cu	9.89	53.44	0.96
		Winter		Station III			Summer	
	Sediment	Tissue	Water			Sediment	Tissue	Water
Mn	38.93	41.97	1.91		Mn	49.39	26.83	2.21
Fe	33.39	2.31	15.97		Fe	36.21	2.72	14.86
Ni	23.86	34.31	9.09		Ni	23.12	25.79	13.43
Cr	32.08	13.13	14.06		Cr	0.48	11.42	15.94
Cd	0.57	0.36	0.15		Cd	0.41	0.41	0.14
Pb	25.46	15.77	4.08		Pb	25.87	13.48	3.82
Zn	38.89	172.51	20.40		Zn	37.84	140.87	16.25
Cu	42.20	120.50	5.18		Cu	47.96	130.85	3.04
		Winter		Station II			Summer	
	Sediment	Tissue	Water			Sediment	Tissue	Water
Mn	76.15	44.48	6.37		Mn	80.40	35.68	7.82
Fe	48.22	5.64	13.92		Fe	47.02	5.24	22.33
Ni	10.60	13.91	15.46		Ni	13.89	14.73	14.81
Cr	42.55	20.85	19.36		Cr	46.39	25.21	18.18
Cd	0.36	0.10	0.09		Cd	0.20	0.07	0.09
Pb	18.59	9.77	1.72		Pb	13.41	8.83	0.88
Zn	26.20	87.75	7.60		Zn	20.44	115.03	12.05
Cu	30.46	91.08	2.42		Cu	20.83	86.33	2.18
		Winter		Station I			Summer	

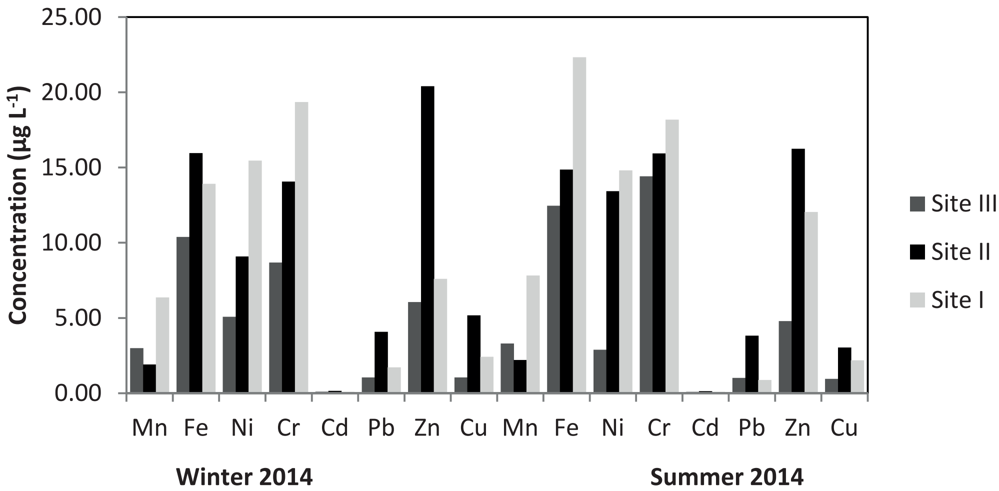

Figure2. Heavy metal concentration in seawater (µg L^{-1}) from Chabahar coast during winter and summer 2014

the other hand, the lowest concentration of Cu, Zn, Pb, Ni, and Cd (station III) were obtained during summer, while Fe and Mn showed minimum values at stations III and II during winter (Figure 2). Considering all heavy metals and sampling sites, the calculated mean values decreased in the following order: Cr > Fe > Ni > Zn> Mn > Cu > Pb > Cd (Station I- sewage drain and spilled fuel and oil from boats in harbour), Zn > Fe > Cr > Ni > Cu > Pb > Mn > Cd (Station II, Industrial activity and boat sewage and cleaning products), Cr> Fe >Zn > Ni > Mn > Pb> Cu > Cd (Station III, open area).

3.2. Metal concentrations in sediments

The results of heavy metals concentration in sediment samples in the selected study locations during two seasons are given in Figure 3. Although the observed values for concentration of heavy metals in sediment samples revealed a similar pattern with that was observed in seawater, accumulation of heavy metals were in significantly higher range in compare to seawater ($P<0.05$). The highest concentration of Cu, Zn, Pb, Ni and Cd were detected in station II and Cr, Fe, and Mn showed a maximum value at station

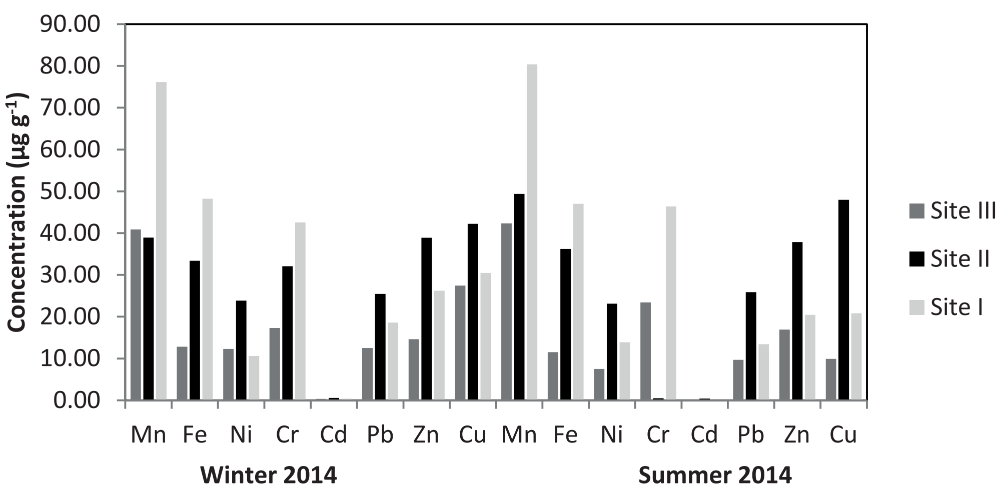

Figure 3. Heavy metal concentration in sediment (µg g^{-1}) from Chabahar coast during winter and summer 2014

I, while the lowest values were recorded in sediment samples at site III. This is not unexpected because of discharge domestic or industrial sewage to the water body in sites I and II. The metal concentrations in sediment samples were as follows: Mn > Fe > Cu > Zn > Cr > Pb > Ni > Cd (Stations I and II), Mn > Cr > Cu > Zn > Fe > Pb > Ni > Cd (Station III).

3.3. Metal concentrations in oyster soft tissues

The concentrations of the eight heavy metals under investigation Mn, Cr, Cu, Zn, Fe, Pb, Ni, and Cd for the studied oyster tissue are given in Figure 4. Concentration of the heavy metals significantly varied in the different samples collected from different locations. The results showed that heavy metal concentrations reached the highest levels among the samples from site II. Zn represented the first highest metals in the samples of all metals measured in the present study; however, Cd was the lowest one of accumulation. The concentration of Cu showed a wide range during winter and summer (78.88 to 120.50 and 53.44 to 130.85, respectively). During the winter, higher concentrations of Cu, Zn, Pb and Cd were observed in the samples from site II, and the highest Cr and Ni concentrations were in samples from site I. In the case of Mn and Fe concentration, the highest values were in samples from site I during the summer. Concentrations of Fe, Ni, Cr, Cd, Pb, Zn, and Cu in the samples at site III were considerably less than the sites I and II during the study period. Whereas, Mn concentration in samples at site III showed high levels compared to those at sites I and II. The heavy metals accumulation in oyster tissue was higher than the permissible limits in two sites (I and II) under investigation. The heavy metals concentration in sediments at sites I and II occurs in descending order of Zn > Cu > Mn > Ni > Pb > Cr > Fe > Cd, except for station I where Ni concentration was higher than Pb. The concentrations ranges of heavy metal in site III were found as follows: Zn > Cu > Mn > Ni > Cr > Pb > Fe > Cd. The correlation between heavy metal pairs in seawater, sediment, and oyster tissue from the Chabahar coast are shown in Table 2. The Pearson correlation shows a positive correlation between the heavy metals, except that between Cd and Cr, Cr and Ni that showed some negative correlations. This is showing that some heavy metals have the same origin. The correlation between Zn and Pb, Cu, and Pb

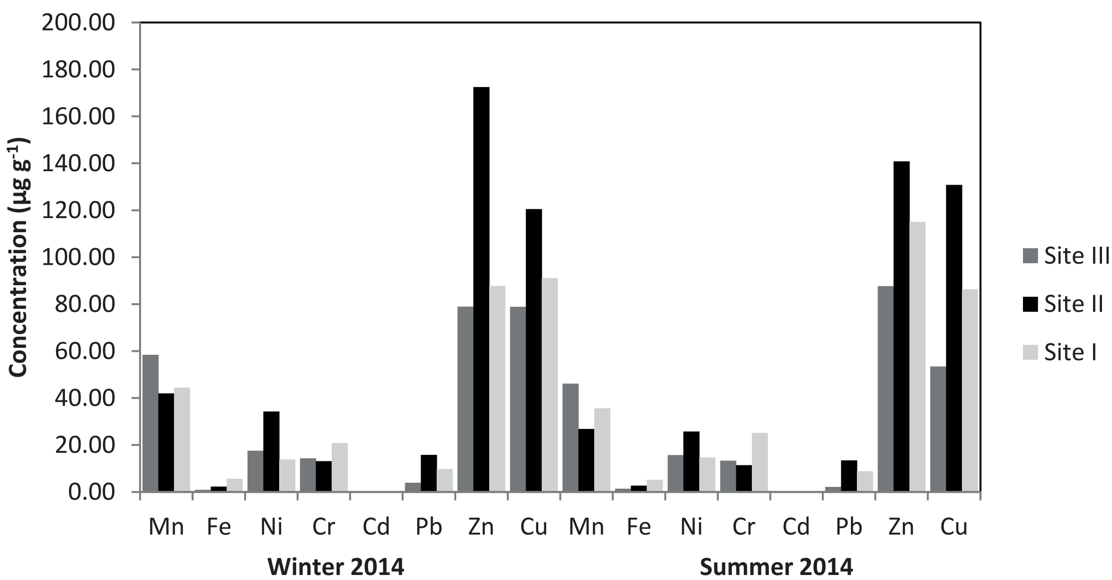

Figure 4. Heavy metal concentration in oyster tissue (µg g^{-1}) from Chabahar coast during winter and summer 2014

Table 2. Correlation coefficients between metal concentrations in seawater, sediment and oyster tissue

	Mn	Fe	Ni	Cr	Cd	Pb	Zn	Cu
Seawater								
Mn	1							
Fe	0.61*	1.00						
Ni	0.58*	0.63	1.00					
Cr	0.65*	0.63	-0.78	1.00				
Cd	0.74**	-0.05	0.01	-0.28	1.00			
Pb	0.62*	0.03	0.23	0.06	0.92**	1.00		
Zn	0.32	0.46	0.39	0.17	0.85**	0.87**	1.00	
Cu	0.29	0.34	0.38	0.23	0.81**	0.86**	0.93**	1
Sediment								
Mn	1							
Fe	0.80**	1.00						
Ni	0.28	0.30	1.00					
Cr	0.67*	0.51	-0.36	1.00				
Cd	0.14	0.14	0.58	-0.76**	1.00			
Pb	0.13	0.47	0.90**	0.29	0.60*	1.00		
Zn	0.14	0.46	0.89**	0.24	0.57*	0.98	1.00	
Cu	0.18	0.38	0.89**	0.41	0.65*	0.95**	0.87**	1
Oyster Tissue								
Mn	1							
Fe	0.42	1.00						
Ni	0.30	0.35	1.00					
Cr	0.05	0.83**	-0.60*	1.00				
Cd	0.59*	0.25	0.89**	-0.65	1.00			
Pb	0.63*	0.33	0.75**	-0.08	0.78**	1.00		
Zn	0.60*	0.00	0.91**	0.28	0.85**	0.88**	1.00	
Cu	0.62*	0.15	0.75**	0.25	0.85**	0.94**	0.81**	1

* = Correlation is significant at 0.05 level (2 -tailed)

** = Correlation is significant at 0.01 level (2-tailed)

were in high significant level in sediment and oyster tissue. The geochemical behaviours of Zn and Pb are similar in most natural processes. This could explain the high relationship and indicate minimum anthropogenic inputs.

4. Discussion

Heavy metals are elements that are characterized by their high atomic weight. Utilization of heavy metals in industry, marine transport, and agriculture leads to wide distribution of these components in marine environment. Their toxicity, potential for human exposure and environmental occurrence is depended on a number of factors such as their source and concentration (Wu and Zhang, 2010). This study focuses on seasonal fluctuations of heavy metals in the three different sampling sites to provide a useful data set for monitoring Chabahar coast. The results of this study showed that values of heavy metals in seawater, sediment and tissue of organism were higher than permissible limits at Tiss and Beris Port (Sites I and II). A possible explanation for this might be discharging sewage directly into a nearby body of water, wastes discharged from factories and industrial zone, chemicals of industrial effluent and products of ship and boats.

The results also showed relatively high concentrations of heavy metals during winter coincide principally with decreasing rate of organic matter decomposition, due to low water temperature. The most interesting finding was that the concentration of Cu was significantly highest in seawater and sediment at the polluted sites (I, and II) during the seasons ($P< 0.05$). These results are in agreement with those obtained by Bazzi (2014) who reported the same patterns in Gulf of Chabahar. These results are likely related to corrosion of ship hulls docked at ports, system failures and organotin compounds used in anti-fouling to coat the bottoms of ships. The current study found that the maximum and minimum concentration of Zn in seawater and sediment from Chabahar coast was during winter and summer, respectively. The results corroborate the ideas of Coale *et al.* (2003) who stated Zinc decreases as a result of phytoplankton consumption during their bloom in summer. The values of metal concentration in site III are less than permissible limits and it is due to geological characteristics of the region and is not related to environmental pollution. This fact further supports the idea of Borrego *et al.* (2002) who evaluated geochemical characteristics of heavy metal pollution in surface sediments.

Sediments known as a valuable indicator for marine environmental pollution; they act as permanent and temporary reservoir of heavy metals in environment (Loska and Wiechuła, 2003). Results of this study showed same pattern in concentration of heavy metal in seawater and the corresponding sediments at the same location. In the present study, concentration of heavy metals in sediment was more than seawater. These results seem to be consistent with Bazzi (2014) findings, which showed Mn, Cr and Zn presented the highest heavy metal concentration in water and sediments on Chabahar Coast. The explanation for maximum concentration of heavy metals in sediment is due to quickly removal of heavy metals from water bodies and depositing in sediment (Asiima *et al.*, 2015). The high Pb content in sediment in site II compare with other regions may be due to the decomposition organic matter. Furthermore, the highest concentration of Zn was at site II during winter and summer.

Oysters are suitable bio-indicator for monitoring and tracking of metal pollution because of their potential to deposit pollution components in their tissues. Concentration of the heavy metals significantly varied in oysters collected from different locations. Results of this study showed that heavy metal concentrations reached the highest levels in oysters from site II. Moreover, heavy metal concentration in tissue of *Saccostreacucullata* followed a same pattern observed in corresponding seawater. These results are consistent with those of Ashraf (2005) who stated similar factors affect on heavy metal levels in seawater and organisms in the same region. On the other hand, the level of metal deposition in oyster tissues shows the same trend with the sediments. As a filter feeding behaviour, some metals from sediments and water, enter to their bodies and they accumulate metals from both the sediment and water. Thereby, it indicated a strong relationship between marine environment and biological concentration.

Another important finding was that the heavy metal concentrations in oyster tissue from site III were the lowest mean values during the study period. This may be due to the fact that site III is away from industrial activity and human impacts. Zn represented the first highest metals in oyster tissue and it may be due to the location of sites where were close to boating activities, fish landing, anthropogenic activities and sightseeing view place. Regarding to high concentration of Cu and Zn in oyster tissue, *Saccostreacucullata* can be introduced as a bio-indicator for mentioned metals. The high concentration of these metals is most probably linked to their physiological characteristics, metabolic activities, and different species of phytoplankton consumption.

In fact, bivalves are useful in bio-monitoring metal contamination in aquatic ecosystems. Compared to the non-polluted area, the higher amount of lead (Pb), cadmium (Cd) and nickel (Ni) were detected in oysters collected around industrial areas of sites I and II. Alarmingly, all the spices contained Pb, Cd, and Ni was higher than the tolerance limits proposed by Food and Agricultural Organization (FAO). The dominant sources of heavy metal pollution are industrial activities; other major sources include Boat sewage, cleaning products, spilled fuel and oil, trash and aquatic invasive species, which are all potential sources of pollution. Sediment is considered as a suitable indicator of heavy metal pollution due to adsorptive sink in coastal and marine environments. Moreover, marine organisms can consider as a suitable bio-indicator to determine the level of contamination.

Conclusion

This study highlights the accumulated metal concentrations measured in seawater, sediment, and oyster tissue that vary according to the seasonal changes and with the influence of seasonal fluctuations of biotic and abiotic factors. Seawater temperature has an active role in organic matter decomposition within the sediments and water that greatly influence the mobility of heavy metals. Furthermore, the occurrence of elevated levels of heavy metals especially in the sediments and marine organisms can be a good indication of man-induced pollution due to their potential to deposit pollution components.

In fact, high levels of heavy metals are often attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering. A possible explanation for high level of Mn, Cr, and Zn concentration in water and sediments might be discharging sewage directly into a nearby body of water, wastes discharged from factories and industrial zone, chemicals of industrial effluent and products of ship and boats along the some part of Chabahar coast. Toxic heavy metals, such as copper, zinc, and lead from bottom paints, can get into the food chain through bottom-dwelling creatures or may settle into the sediments, potentially increasing the cost of dredge spoil disposal. Since differences in the sampling times, size of oysters, genetic differences, individual maturation of organisms, can also influence the result. As a suggestion the association of these factors should be investigated in future studies.

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