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Assessment of heavy metals pollution in muscle of sole (*Cynoglossus arel*), spiny lobster (*Panulirus homarus*) and sediments in the northern coasts of the Oman Sea during pre and post monsoon

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Abstract

Concentrations of lead, copper, cadmium, and mercury were determined in sediments and two selected marine organisms (*Cynoglossus arel* and *Panulirus homarus*) collected from Jask Port, Darak Port, Pozm Port, Gulf of Chabahar and Gowatr Port in the north coasts of the Oman Sea during pre (spring) and post (autumn) monsoon in 2017. Heavy metals concentrations were determined by atomic absorption spectrophotometry. Results showed that there are statistically significant differences between studies concentrations in sediments during pre and post-monsoon at sample locations ($p<0.05$). Analysis of variance revealed a significant difference in the muscle of *C. arel* and *P. homarus* in pre and post-monsoon between the different sites ($p<0.05$), except for copper in the muscle of *P. homarus* in the post-monsoon ($p>0.05$). In sediments and muscle of *C. arel* and *P. homarus* the trend in the mean metal concentrations was: Cu> Pb> Cd> Hg; Cu> Pb> Cd> Hg and Cu> Hg=Pb> Cd, respectively. Also, the trend in the mean metal concentrations in the samples was sediment > *P. homarus*> *C. arel*. However, paired t-test analysis for comparing heavy metal concentration in pre and post-monsoon showed only a significant difference for cadmium in the muscle of *P. homarus* ($p<0.01$). Totally, by comparing the results obtained from this study indicated that the accumulation of lead, copper, cadmium, and mercury in sediments and muscle *C. arel* and *P. homarus* at the Gulf of Chabahar and Jask Port is higher than other regions of the Iranian coasts along the Oman Sea which studied in current research.

Keywords: Oman Sea, Heavy metals, Sediment, *Cynoglossus arel, Panulirus homarus*

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Introduction

The word heavy metal refers to a group of metals and semi-metals whose density is more than 4 times in the water density (1 $cm^{-3}g^{-1}$) (Duruibe *et al*., 2007). These Materials are an integral component of the environment and living matter (Nicholas *et at*., 1998; Carolyn *et al*., 2004). These compounds in the water can originate both from natural sources, industrial, agricultural and domestic activities in the drainage basin of a water system. As the metal levels in many aquatic ecosystems increase due to anthropogenic activities, they raise the concern on metal bioaccumulation through the food chain and related human health hazards (Nzeve, 2015).

Once the heavy metals come into the aquatic environment, they scatter in various components such as water, suspended solids, sediments, and biota. Contamination of sediments by heavy metals and other pollutants is considered by many regulatory agencies to be one of the major threats to aquatic ecosystems. The importance of sediments as a sink for a range of substances, including nutrients, hydrocarbons, pesticides and heavy metals has been highlighted in many past studies (Baldwin and Howitt, 2007). Sediments are one of the possible media in monitoring the health of aquatic ecosystems. They are ecologically important components of the aquatic habitat and play a significant role in maintaining the trophic status of any water body (Sigh *et al*., 1997). In this respect, Abraha *et al*. (2012) reported that sediments have a significant role in the demobilization of contaminants in aquatic systems under favorable conditions and interactions between water and sediments. Aquatic animals are also used as bio-indicator because it is easy to be obtained in large quantity, the potential to accumulate metals, long lifespan, easy to be a sample and optimum size for analysis (Anim-Gyampo *et al*., 2013). According to importance and role of endemic or abundant species as bioindicator on environment health, sole (*Cynoglossus arel*), spiny lobster (*Panulirus homarus*) selected due to: geographical distribution and economical and fisheries value in northern coasts of the Oman Sea.

Multiple parameters, such as age, length, weight, gender, nutritional habits, ecological needs, Concentration of heavy metals in water and sediment, seasons, and physical and chemical properties of water (salinity, water hardness and temperature) can play an important role in the heavy metal assimilated in sediments and the muscle of the marine organisms (Demirak *et al*., 2006).

Above-mentioned topics, with combination of monitoring and measurements of water and sediment quality, can provide a good indication of conditions and potential risks to a water body (Alaa and Osmanl, 2010). For this purpose, this study was undertaken to investigates: 1) to assess, the concentration of four heavy metals including lead, copper, mercury and cadmium in edible tissues of sole (*C. arel*), spiny lobster (*P. homarus*) and surface sediment from five different stations in northern parts of the Oman sea 2) to compare the concentration of these metals with global standards.

Materials and methods

Description of the study area

The study areas Jask Port (Longitude : 57˚ 77' 95″; Latitude: 25˚ 65' 77"), Darak Port (Longitude: 59˚ 47' 18"; Latitude: 25˚ 47' 55"), Pozm Port (Longitude :60˚ 31' 12"; Latitude: 25˚ 35' 42"), Chabahar Gulf (Longitude : 60˚ 49' 59"; Latitude: 25˚ 30' 19"), and Gowatr Port (Longitude :61˚ 52' 12"; Latitude: 25˚ 18' 35"), are located in the northern coasts of the Oman Sea (Fig. 1). The north-western part of the Arabian Sea is connected to the Persian Gulf (Sheppard *et al*., 2010), by the Sea of Oman is situated in the subtropical zone and has the total area of 94,000 km^2. (Piontkovski *et al*., 2012). Being situated between the shallow high salinity waters of the Persian Gulf and the deeper Arabian Sea, the Sea of Oman possesses a unique hydrological regime. In winter, during the Northeast Monsoon, the current, transporting, the Arabian Sea water mass from the oceanic regions in the gulf heads towards its inner path along the northern (Iranian) coast. In summer, during the southwest monsoon, the sea is influenced by the outflow of high saline Gulf water mass. The current exits from the Gulf in the Strait of Hormuz at a depth of about 100 m, cascades down to the bottom and propagates along the Omani coast towards the open Arabian Sea. A well-pronounced density front separates high saline deep water in the Gulf from fresher surface waters in the Sea of Oman (Swift *et al*., 2003). As far as seasonal variations are concerned, the most prominent feature of the Sea of Oman is the seasonal upwelling along the Iranian coast, with a peak in February-March, although the onset of the Northeast monsoonal winds could occur in December. During the Northeast monsoon, the Oman Coastal Current reverses to southeastward flow (Piontkovski *et al*., 2012).

Sampling and analytical techniques

During a field reconnaissance (February, 2016) the five stations were selected to represent different sub basins that influence of industrial and human activities on the Iranian coasts along the Oman Sea for the first time (Fig. 1). For these purposes, sampling of heavy metals in sediments and edible tissue of *P. homarus* and *C. arel* was carried out once a month for two seasons during pre-monsoon (spring) and post monsoon (autumn) in 2017 at all the sampling stations.

Sediment samples were taken using an Eckman grab. For each sample, three sediment grabs were randomly taken, homogenized and kept in clean polyethylene bags. The polythene bags were then labeled to indicate the sampling station and date of sampling. The samples were then stored in ice box for transportation to the laboratory. In the laboratory, the samples were kept in a freezer at –20˚C until they were processed for heavy metal analysis. Gill net for sole and trap / diving for spiny lobster specimens were used in five sampling sites. A total of 100 samples of sole and 100 samples of spiny lobster samples were caught during the study period. After obtaining samples they were immediately kept in pre-cleaned polythene bags, sealed, labeled and kept in ice boxes for transportation to the laboratory at Islamic Azad University - Bandar Abbas Branch. In the laboratory, the samples were then kept in a deeper freezer until muscle tissues were extracted for analysis.

Processing and digestion of sediments for metal analysis

Each sediment sample was thawed at room temperature (25˚C – 28˚C) and put into pre-acid cleaned evaporating beakers. The sediments were then dried at 70˚C for 48 hours in an oven until a constant weight was obtained. The dried sediment samples were ground using a porcelain mortar and pestle and sieved through a 2 mm mesh plastic sieve.

For each sediment sample, 2 g was weighed using electronic weighing balance into 100 mL acid cleaned beakers. Digestion was done using concentrated nitric acid (analytical grade) and hydrogen peroxide. All the digested samples were filtered using Whatman 0.42µm filter paper into a 50 mL volumetric flask and topped up to the mark with distilled water. The filtrate was analyzed for heavy metal analysis using AAS (Model Fs 240 vari). A blank solution was similarly prepared.

Processing and digestion of tissues for metal analysis

The deep frozen samples for the two species (*C. arel* and *P. homarus*) were thawed at ambient laboratory temperature overnight. The skin of each sample has removed using plastic knives to avoid metal contamination and this was followed by extraction of muscles. Muscles were put in a pre-acid washed and oven-dried crucibles. The samples were then dried to a constant weight in an oven at 50˚C. The dried samples were allowed to cool in a desiccator at room temperature. After cooling, 2 g of muscles tissues were accurately weighed using an electronic weighing balance and transferred into a clean beaker. Dried samples were digested in triplicates according to the method described in APHA (2005). To each weighed muscle, 18 mL of concentrated nitric acid was added and heated at 100 ˚C on a hot plate in a fume hood chamber. A few drops of hydrogen peroxide (analytical grade) were added until there were no brown fumes.

The digested tissue sample solution has been filtered using Whatman 0.42µm filter paper in a 25 mL volumetric flask and topped to the mark with distilled water. The filtrate solutions were put into 60 mL pre – acid cleaned plastic bottles (plate 3.4) and metal analysis done using a computerized Varian Atomic Absorption Spectrophotometer (Model Fs 240 vari) at Mines and Geological department under working conditions as shown in Table 3.2. Blank solutions were prepared in a similar manner as for the samples.

Figure 1: Map showing the location of the study area and the position of the stations in the north costs of the Oman Sea.

Statistical data analysis

Data analysis was done using a computerized statistical program (SPSS, 17). The data were subjected to one way analysis of variance (ANOVA) and significant differences accepted at $p \leq 0.05$. Where significant differences were found, the mean values were separated using post-hoc Duncan test. Paired-Samples T-test analysis was done to determine associations among heavy metals in per and post-monsoon season.

Results

Heavy metals in the sediment

The temporal variations of the mean heavy metal (Pb, Cu, Cd and Hg) concentrations (ppm) at the five sampling sites in the pre and post monsoon during the study period showed in Table 1. One way ANOVA revealed significant differences in sediments between the different sites (Table 1). Based on the results, the highest concentration of Pb was recorded in the pre monsoon (1.94±0.20 ppm) and post monsoon (2.02±0.22 ppm) at the Gulf of Chabahar ($p<0.05$). In the case of Cu, the highest concentrations were observed pre and post –monsoon, at the Gulf of Chabahar (respectively 24.54±5.76 and 29.26±4.97 ppm) and Jask Port (respectively 24.16±3.86 and 26.15±3.86 ppm) ($p<0.05$). Also, the highest concentrations of cadmium and mercury in the pre monsoon were 0.90±0.28 and 0.42±0.25 at the Gulf of Chabahar and the highest concentrations of these metals in the post-monsoon, were obtained 0.74±0.16 and 0.87±0.16 ppm for cadmium and 0.33±0.19 and 0.35±0.19 ppm for mercury at two stations on Jask Port and Chabahar Gulf, respectively ($p<0.05$). The lowest amount of all metals studied was observed at the stations of the Darak and Gowatr Ports (Table 1).

Table 1: Heavy metals concentration (ppm dw) in the sediments sample collected from the north coasts of Oman Sea on the pre and post monsoon.

Station	Metals			
	Pb	**Cu**	**Cd**	**Hg**
Pre -Monsoon				
Jask Port	1.51±0.30[c]	24.16±3.86[a]	0.63±0.29[b]	0.33±0.15[ab]
Darak Port	0.64±0.20[d]	17.46±2.02[b]	0.16±0.07[d]	0.15±0.05[c]
Pozm Port	1.73±0.20[b]	18.67±1.33[b]	0.57±0.19[b]	0.23±0.10[bc]
Chabahar Gulf	1.94±0.20[a]	24.54±5.76[a]	0.90±0.28[a]	0.42±0.25[a]
Gowatr Port	0.71±0.14[d]	20.12±4.02[b]	0.36±0.08[c]	0.18±0.09[c]
Post -Monsoon				
Jask Port	1.65±0.17[b]	26.15±3.86[a]	0.74±0.11[a]	0.33±0.11[a]
Darak Port	0.70±0.19[c]	17.11±2.41[b]	0.13±0.04[d]	0.16±0.07[c]
Pozm Port	1.61±0.24[b]	19.42±3.76[b]	0.61±0.20[b]	0.26±0.13[ab]
Chabahar Gulf	2.02±0.22[a]	29.26±4.97[a]	0.87±0.16[a]	0.35±0.19[a]
Gowatr Port	0.72±0.18[c]	17.46±2.21[b]	0.30±0.08[c]	0.19±0.07[c]

Paired t-test analysis showed that there was no significant difference (p>0.05) in mean heavy metals concentration levels in sediments in the different sites in pre and post monsoon (Table 3). Also, Based on the results, the mean negative differences of Pb (-0.72), Cu (-1.09) and Cd (-0.15) indicate that the mean concentration of these metals in the post monsoon (autumn) has been higher compared with the pre monsoon (spring). Contrary to these results, the concentration of Hg (0.15) in pre monsoon was higher compared with the post monsoon (Table 2).

Table 2: Comparison of heavy metals concentration (ppm dw) in sediments at pre and post –Monoson

Group		No.	Mean±SD	df	*p*	t
Pb	Pre -Monsoon	50	1.31±0.57	49	0.47	-0.72
	Post -Monsoon	50	1.34±0.57			
	Mean difference	-	-0.03±0.29			
Cu	Pre -Monsoon	50	20.99±4.61	49	0.27	-1.09
	Post -Monsoon	50	21.88±6.04			
	Mean difference	-	-0.88±5.73			
Cd	Pre -Monsoon	50	0.52±0.32	49	0.87	-0.15
	Post -Monsoon	50	0.53±0.30			
	Mean difference	-	-0.005±0.26			
Hg	Pre -Monsoon	50	0.26±0.17	49	0.88	0.15
	Post -Monsoon	50	0.26±0.14			
	Mean difference	-	0.003±0.17			

Heavy metals in sole (C. arel)

The mean metals levels in muscle of sole (*C. arel*) observed in this study showed modest variation and are presented in Table 4. One way ANOVA revealed that there was a significant variations in mean metals concentration levels observed in sole muscles tissues in different sampling sites (p<0.05). Based on the results, the highest concentrations of Pb and Cu in the pre monsoon were found 0.28±0.09, 0.35±0.08 (ppm) and 3.86±1.06, 3.79±1.16 (ppm) at Jask Port and the Gulf of Chabahar (p<0.05), respectively. The highest mean Pb concentration levels in the post monsoon was 0.43±0.18 (ppm) at Jask Port and the highest mean Cu concentration levels was observed 4.62±1.25 (ppm) at Chabahar Gulf (p<0.05). The highest concentration of Cd in the pre (0.23±0.07 ppm) and post monsoon (0.21±0.07 ppm) were recorded in the Jask Port. The highest concentrations of Hg observed in both cases of pre (0.43±0.08 ppm) and post (0.42±0.11 ppm) monsoon were observed at the Chabahar Gulf (p<0.05). The lowest amount of the mean heavy metals concentration studied like the results of the sediments obtained from Darak and Gowatr Ports (Table 3).

Table 3: Heavy metals concentration (ppm dw) in the muscle of sole *Cynoglossus arel* at the pre and post monsoon.

Station	Metals			
	Pb	**Cu**	**Cd**	**Hg**
Pre -Monsoon				
Jask Port	0.35±0.08^{a}	3.86±1.06^{a}	0.23±0.07^{a}	0.20±0.07^{c}
Darak Port	0.13±0.04^{c}	2.18±0.92^{bc}	0.07±0.01^{c}	0.09±0.02^{d}
Pozm Port	0.19±0.04^{b}	2.55±0.81^{b}	0.16±0.04^{b}	0.32±0.09^{b}
Chabahar Gulf	0.28±0.09^{a}	3.79±1.16^{a}	0.17±0.05^{b}	0.43±0.08^{a}
Gowatr Port	0.20±0.7^{b}	1.47±0.66^{c}	0.09±0.01^{c}	0.12±0.04^{d}
Post -Monsoon				
Jask Port	0.43±0.18^{a}	3.39±1.54^{b}	0.21±0.07^{a}	0.25±0.08^{b}
Darak Port	0.12±0.04^{c}	2.85±0.78^{b}	0.07±0.01^{c}	0.10±0.02^{c}
Pozm Port	0.21±0.06^{bc}	3.17±1.17^{b}	0.15±0.03^{b}	0.32±0.10^{b}
Chabahar Gulf	0.22±0.07^{b}	4.62±1.25^{a}	0.19±0.08^{ab}	0.42±0.11^{a}
Gowatr Port	0.19±0.05^{bc}	1.60±0.59^{c}	0.09±0.01^{c}	0.12±0.02^{c}

Also, paired t-test analysis indicated that there were no significant differences of heavy metals levels in the pre and post monsoon between the sites (Table 4). The negative mean differences of lead, copper and mercury in Table 5 indicate that the mean concentration of these three metals in the post monsoon (autumn) was higher than compared to the pre monsoon (spring). However, the amount of Cd was higher in the pre monsoon (spring).

Table 4: Comparison of heavy metals concentration (ppm dw) in the muscle of sole *Cynoglossus arel* at the pre and post –monsoon.

Group		No.	Mean±SD	df	*p*	t
Pb	Pre -Monsoon	50	0.23±0.10	49	0.77	-0.29
	Post - Monsoon	50	0.24±0.14			
	Mean difference	-	-0.004±0.11			
Cu	Pre -Monsoon	50	2.77±1.30	49	0.09	-1.69
	Post - Monsoon	50	3.12±1.14			
	Mean difference	-	-0.35±1.49			
Cd	Pre -Monsoon	50	0.14±0.07	49	0.81	0.23
	Post - Monsoon	50	0.14±0.07			
	Mean difference	-	0.001±0.04			
Hg	Pre -Monsoon	50	0.23±0.14	49	0.58	-0.55
	Post - Monsoon	50	0.24±0.14			
	Mean difference	-	-0.008±0.11			

Heavy metals in Lobster (Panulirus homarus)

The mean concentration levels for heavy metals recorded during the study in different sites in spiny lobster muscle tissues showed variations (Table 5). By using one way ANOVA, it was found there was a significant difference in the levels of the mean heavy metals except copper in per-monsoon condition in the sampling sites. Based on the results, the highest mean Pb levels were recorded 0.71±0.12 and 12.12, in both cases of pre and post monsoon, at the Jask Port (p<0.05). The highest mean Cu levels in the pre- monsoon was observed at Jask station 14.10±2.30 (ppm) (p<0.05). The highest mean Cd and Hg concentration levels in the pre–monsoon were recorded 0.24±0.19 and 0.24±0.09 ppm and 0.20±0.04 and 19.19±0.05 ppm in Jask Port and the Gulf of Chabahar, respectively. While in the post monsoon, the highest mean Cd concentration levels were recorded 0.41-0.0 0 and 0.39±0.08 (ppm) at the Chabahar Gulf and Pozm Port (p<0.05). However, the highest mean Hg concentration was observed at the Jask Port 0.20 ± 0.04 ppm (p<0.05) (Table 5).

Table 5: Heavy metals concentration (ppm dw) in the muscle of *Panulirus homarus* at the pre and post monsoon.

Station	Metals			
	Pb	Cu	Cd	Hg
Pre -Monsoon				
Jask Port	0.71±0.12[a]	14.10±2.30[a]	0.24±0.11[a]	0.20±0.04[a]
Darak Port	0.16±0.06[c]	12.03±2.13[b]	0.11±0.03[b]	0.08±0.008[b]
Pozm Port	0.44±0.18[b]	12.86±1.32[ab]	0.21±0.08[a]	0.17±0.07[a]
Chabahar Gulf	0.55±0.26[b]	13.12±1.64[ab]	0.24±0.09[a]	0.19±0.05[a]
Gowatr Port	0.23±0.09[c]	13.43±1.79[ab]	0.11±0.08[b]	0.08±0.01[b]

Table 5 continued:

Post -Monsoon				
Jask Port	0.74±0.12[a]	14.33±1.88[a]	0.21±0.08[b]	0.20±0.04[a]
Darak Port	0.18±0.05[d]	12.72±2.50[a]	0.12±0.05[c]	0.09±0.01[c]
Pozm Port	0.36±0.10[c]	13.25±1.47[a]	0.39±0.08[a]	0.16±0.04[b]
Chabahar Gulf	0.46±0.09[b]	14.33±1.88[a]	0.41±0.09[a]	0.16±0.06[b]
Gowatr Port	0.20±0.12[d]	13.48±1.17[a]	0.20±0.08[b]	0.08±0.01[c]

Paired t-test results indicated no significant variation in mean Pb, Cu and Hg concentration levels in the muscle issues of spiny lobster among the pre and post monsoon. While the mean Cd concentration levels have a significant difference (p<0.01). The negative mean differences of Cu and Cd in Table 6 indicates the mean concentration of these metals in the post monsoon (autumn) were higher than compared to the pre monsoon (spring). However, the amount of Pb and Hg levels were higher in the post monsoon (spring) (Table 6).

Table 6: Comparison of heavy metals concentration (ppm dw) in the muscle of *Panulirus homarus* at the pre and post –monsoon.

Group		No.	Mean±SD	df	*p*	t
Pb	Pre -Monsoon	50	0.42±0.26	49	0.25	1.14
	Post -Monsoon	50	0.39±0.22			
	Mean difference	-	0.02±0.17			
Cu	Pre -Monsoon	50	13.11±1.92	49	0.29	-1.05
	Post -Monsoon	50	13.44±1.79			
	Mean difference	-	-0.33±2.20			
Cd	Pre -Monsoon	50	0.18±0.10	49	0.00	-4.09
	Post -Monsoon	50	0.27±0.13			
	Mean difference	-	-0.08±0.14			
Hg	Pre -Monsoon	50	0.14±0.07	49	0.44	0.68
	Post -Monsoon	50	0.14±0.06			
	Mean difference	-	0.005±0.06			

Paired t-test analysis calculated to obtain associations between heavy metals variables in the pre (spring) and post (autumn) monsoon seasons in sediments and edible muscle of two marine organisms *C. arel* and *P. homarus* samples just showed significant differences in Cd levels in muscle of *P. homarus* (p=0.000; t=-4.09). Based on the results, the mean Cd concentration levels has increased significantly from 0.18 ppm in the pre monsoon and reached to 0.27 ppm in the post monsoon season.

Discussion

Our study reported for the first time, the accumulation pattern of lead, copper, cadmium and mercury at the five stations from West to East of Iranian costs along the Oman Sea including: Jask Port, Darak Port, Pozm Port, Gulf of Chabahar and Gowatr Port during pre (spring) and post (autumn) monsoon season in sediments and muscle tissues of *P. homarus* and *C.*

arel. Numerous studies described the comparison of heavy metals residues in sediments and marine organisms (Tchounwou *et al*., 2012). In summery the order of Pb, Cu, Cd and Hg concentrations in sediments, *P. homarus* and *C. arel* muscle tissues from the Oman Sea was Cu> Pb> Cd> Hg; Cu> Pb> Cd> Hg and Cu> Hg=Pb> Cd, respectively.

Also, Based on the results obtained from the present study were observed significant increase only of Cd levels in the muscles of *P. homarus* during post-monsoon.

This results, attributed to industrial activities in the catchment especially the shipping of vessels moving, mining, smelting, and manufacturing of batteries, and alloys. Therefore, in the post monsoon with increasing rainfall Cd compounds get into sediments through surface runoff, which due to the water circulation, cause the entry of heavy metals into the water columns and, consequently, their absorption by the aquatic organisms. The correlation between heavy metals is influenced by physical and chemical processes occurring in an aquatic environment (Baeyens *et al.*, 1998). copper can get into aquatic ecosystems from diverse sources, for example from Cu compounds used in fungicides, algaecides, insecticides, wood preservatives, electroplating and azo dye manufacture (Akan *et al*., 2010). Also, from Cu compounds added in fertilizers and animal feeds as a nutrient to support plant and animal growth. Copper compounds are also used in food additives and copper salts in water supply systems to control biological growths in reservoirs and distribution pipes (WHO, 2004; Eaton, 2005). Copper is an essential substance to human life, however, in high concentrations, it can cause anemia, liver and kidney damage, stomach and intestinal irritation (Turnlund, 1998). Copper is a natural element which is widely distributed in soils, rocks and in rivers. It is released into the water as a result of natural weathering of soil and discharges from industries and sewage treatment plants (Romo-Kroger *et al*., 1994; Hutchinson, 2002). Copper in surface water from extensive use of pesticide sprays which contain Cu compounds for agricultural purposes (Al-Weher, 2008). In the dissolved form, Cu is potentially very toxic to aquatic animals and plants, especially for young life-stages such as fish larvae. However, the toxicity is greatly reduced when Cu is bound to particulate matter in the river water and when the water is hard (Nzeve *et al*., 2015). The observed of high mean of Cu concentration levels in the present study could be attributed to human activity in the catchment to increasing of heavy metals in sediments. Because often industrial and fishing activities in Sistan and Balochestan province are concentrated in Jask Port and the Gulf of Chabahar and the other parts of this province, these activities have not been expanded. Also, by comparing the results among the different sites, the highest values for metals in the north costs of the Oman Sea were recorded in Jask Port in the western part and the Gulf of Chabahar in the eastern part, while the lowest once has observed on Darak Port and Pozm Port at center part of the north costs of the Oman Sea. These results indicated that Jask Port and the Gulf of Chabahar were more polluted than Darak port, Pozm Port and Gowatr Port, because all of the wastes and industrial effluents have been entered through Jask Port and the Gulf of Chabahar. However, observed the high concentrations of heavy metals in these areas may be attributed to sewage and wastes discharged from industrial activities related to Chabahar Free Trade-Industrial Zone, shipping activity (such as repairing, fueling, greasing and painting of fishing ships) and boats. Also, this high concentration of heavy metals at Jask Port and the Golf of Chabahar may be due to corrosion of ships hulls coatings and anti-fouling paints at these areas (Bazzi, 2014). In addition, the reason for the high adsorption of heavy metals in the surface sediments of the northern coasts of the Oman Sea can be explained by this fact that sediments are the main receiving areas and in fact the storage of various pollutants such as heavy metals (Davari *et al*., 2010; Abdolahpur *et al*., 2013). Therefore, the highest rate of adsorption and accumulation of heavy metals has been observed is expected in the sediment samples, which in accordance with the results of this study. The low concentration of heavy metals in the marine organism compared with sediments may be due to low levels of proteins binding with heavy metals (Allen-Gill and Martynov, 1995). After sediments, *P. homarus* had higher heavy metals levels in the muscles tissues followed by *C. arel*. The variations in the Cu levels could be attributed to differences in feeding habits of the fish species (Yousafzai *et al*., 2012). *P. homarus* is reef dwelling species, most abundant on coral and coastal fringing rocky reefs and the areas surrounding them. Prefers to scavenge the bottom for of fish species, crustaceans and mollusks that could have led to high Cu levels as compared to *C. arel* which is one of the most important predators in benthic communities (Pereira and Josupeit, 2017). Also, According to Viarengo (1989), the ability of organisms to absorb, accumulate and remove or detoxification of heavy metals is fundamentally different. Species that have varying amounts of metallothionein and lysozyme can eliminate the toxicity of heavy metals. The mean Pb and Cu concentration levels in the muscle tissues of sole and spiny lobster had caught in the north costs of the Oman Sea were higher and mean Cd concentration levels were was lower than heavy metals levels recorded in the muscle tissues of same species from Coastal Waters of Ondo State, Nigeria (Olusola and Festus, 2015). Studies carried out by Safahieh *et al*. (2011) at Musa estuary (Persian Gulf) showed high mean Pb (0.88 μg g^{-1}) and low mean Cu (2.44 μg g^{-1}) and Cd (0.07 μg g^{-1}) levels in sole (*Euryglossa orientalis*). Also, mean Cu levels of 1.20 μg g^{-1} have been observed in sole (*Psettodes erumei)* in Bushehr province (Persian Gulf) (Hosseini *et al*., 2015), which was higher than Cu levels observed in *P. homarus* and *C. arel*.

However, they were lower than mean Pb (1.32 μg g^{-1}) and Cd (1.60 μg g^{-1}) values recorded in muscle tissues of *P. erumei*. Also, studies carried out by Sadough Niri *et al*. (2012) in two fishing areas, from the northern Persian Gulf that names Hendijan Port (Bahrekan) in Khuzestan province and Deylam Port in Bushehr province has shown very high mean Pb (respectively 5.20 and 4.69 ppm) and Cd (respectively 0.70 and 0.49 ppm) levels in *E. orientalis* sole fish compared with *C. arel* and *P. homarus*.

The local distribution of metals in sediments and *P. homarus* muscle tissues gave the similar pattern to that found in sediments. The order of heavy metal concentration in sediments and the marine organism (*P. homarus* and *C. arel*) from the north coasts of the Oman Sea was sediments> *P. homarus*> *C. arel*. These results indicated that the accumulation of the mean heavy metals is predominant in sediments rather than of *P. homarus* and *C. arel* muscle tissues. This can be interpreted as sediments act as the reservoir for all the contaminants and dead organic matter descending from the ecosystem above (Bazzi, 2014).

The level of Pb, Cu, Cd and Hg in sediment samples of the north costs of the Oman Sea was higher than LAL-USEPA levels. However, their amounts were very lower than the HAL-USEPA levels. Also, the levels of Pb, Cu, Cd and Hg in sediments were low and below both levels of the international limits CCME (PEL and ISQGs) and NOAA (ERM and ERL) (Table 7). However, sediments have the capacity to accumulate more heavy metals with time and demobilize them back to the water and the food chain (WHO, 2008). Compared to other studies, Lakshmanasenthi *et al*. (2013) obtained higher heavy metals levels in sediments compared with fishes of estuaries of Bay of Bengal. Bazzi (2014) were determined heavy metals levels in sediments (Pb 25.63 to 28.23; Cu 46.79 to 54.76 and Cd 0.53 to 0.63 μg g^{-1}) from the intertidal zone at the Gulf of Chabahar, which was higher compared with our results. Anand and Kala (2015) by examining the concentration of heavy metals in the Coastal Environments along the Foremost Places of South-East Coast of India reported concentrations of Cadmium in the coastal seawater from 0.9 to 3.42(ppb), Lead from 5.32 to 21.60 (ppb) and Copper from 0.78 to 4.36 (ppb). Studies by Coulibaly *et al*. (2012) by examined of the seasonal variations of some heavy metals in sediments of Biétri Bay, in Ebrié Lagoon, Ivory Coast showed a significant difference in the levels of mercury, cadmium and lead in different seasons. The different forms and concentrations of heavy metals in the sediment of aquatic medium determine their bioavailability and toxicity. Thus, the study of the different fractions of the elements in the sediment was vital because the total concentrations are not representing the real degree of the potential contamination. Heavy metals can be bound to or occluded in amorphous materials, adsorbed on clay surfaces or iron/manganese oxyhydroxides, co-precipitated in secondary minerals such as carbonates, sulfates, or oxides, complexes with organic materials, or included in the lattice of primary minerals such as silicates (Peng *et al*., 2009). Ayling (1974) suggested that different mechanisms exist for the uptake of Cu, Cr, Zn and Cd within the Oyster *Crassostrea gigs*. These mechanisms may vary within 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. Also, Olusola and Festus (2015) with investigation assessed and monitored accumulation levels of Pb, Cu and Cd found lower mean levels in sediments ranging from Pb 0.9-1.00; Cu 0.24-1.44 and Cd 0.35-0.38 mg kg^{-1} collected from the ocean shoreline in the coastal waters of Ondo State, Nigeria. Other studies carried out have revealed lower mean Cu (17.24 μg g^{-1}) and Cd (<0.5 μg g^{-1}) concentration levels in the northwest sediments of the Persian Gulf, Iran (Azimi *et al*., 2012), the mean Cu (19.89 μg g^{-1}) concentration levels in Aden Port, Yemen (Nasr *et al*., 2006) and the mean Cd (35.81 ppm) and Hg (0.28 ppm) concentration levels in the Red Sea–Jeddah Coast, KSA (Ali *et al*., 2011). Azimi *et al*. (2012) obtained higher Hg (1.93ppm) and Pb (5.63 ppm) levels in sediments in the northwest of the Persian Gulf, Iran.

Also, Nasr *et al.,* (2006) were observed higher Pb (77.28 ppm) levels in Aden Port, Ali *et al*. (2011) reported higher Cu (24.98 ppm) and Pb (42.51 ppm) levels in the Red Sea–Jeddah Coast, KSA.

The Pb levels recorded in the muscles of the two selected the marine organisms were low and below the international limits (WHO, range from FDA and NHMRC) and Iranian National Standards (Table 8). The mean Cu levels recorded during this study in *C. arel* sole were lower than the recommended limit for Cu by WHO, NHMRC and the Institute of the national standard center of Iran in fish and fish products. While the levels of these metals in the edible muscle of *P. homarus* only were lower than the Iranian National Standards. The Cd concentration levels recorded in muscles of *C. arel* sole only were lower the recommended limit of WHO and FDA. Also, the mean Cd concentration levels in muscles of *P. homarus* on the pre-monsoon season lowered the recommended limit of WHO and FDA. But, the mean Cd levels recorded during this study in the post monsoon season have been only lower than the acceptable limit suggested by the standard value of FDA. The Hg concentration levels obtained in the two marine organisms during this study in the pre and post monsoon did not exceed the range from FDA, NHMRC and acceptable limit suggested by the Institute of the national standard center of Iran (Table-8). Because a metal concentration in the aquatic environment is low and considered to be naturally occurring or background does not mean that the concentration could not cause adverse ecological effects (USEPA, 1976). The presence of one metal can significantly affect the impact that another may have on an organism. The effect may be synergistic, additive or antagonistic (Olusola and Festus, 2015).

Table 7: Comparison of heavy metals in sediments with global standards.

Metal	USA Deposition Guide (NOAA)		Canadian Deposition Guide (CCME, 1999)		United States Environmental Protection Agency (USEPA, 1990)		Present study
	ERM	ERL	PEL	ISQGs	LAL	HAL	
Pb	218	46.7	112	30.2	2	218	(1.31) Pre -Monsoon (1.34) Post -Monsoon
Cu	270	34	108	18.7	2	270	(20.99) Pre -Monsoon (21.88) Post -Monsoon
Cd	9.6	1.20	4.20	0.70	0.04	9.6	(0.52) Pre -Monsoon (0.53) Post- Monsoon
Hg	0.71	0.15	0.13	0.01	0.01	0.71	(0.26) Pre -Monsoon (0.26) Post -Monsoon

Table 8: Comparison of heavy metals in the muscle of marine organisms with global standards (μg kg^{-1} Dry Weight)

Standards/ Species	Heavy metals				References
	Pb	Cu	Cd	Hg	
WHO	0.5	10	0.20	0.10	WHO (1995)
FDA	5	-	1	0.10-0.50	FDA (2011)
NHMRC	5/1	10	0.05	1	Darmono and Denton (1990)
Iranian National Standards	1	20	0.10	500*	Movahed *et al.* (2013)
Cynoglossus arel	0.23	2.77	0.14	0.23	Pre -Monsoon (present study)
	0.24	3.12	0.14	0.24	Post -Monsoon (present study)
Panulirus homarus	0.40	13.27	0.18	0.14	Pre -Monsoon (present study)
	0.39	13.44	0.27	0.14	Post -Monsoon (present study)

In conclusion, the findings of the present study demonstrated that seasonal variations in the levels of lead, copper, cadmium and mercury in sediments and *P. homarus* and *C. arel* edible muscles during pre and post monsoon, except copper in the muscle of *P. homarus* at post monsoon. Also, by comparing the results obtained in pre and post-monsoon in different samples was observed concentration of Cd in the muscle of *P. homarus* being significantly higher in post monsoon than pre-monsoon. The highest levels of heavy metal concentrations were recorded in both Jask Port and the Gulf of Chabahar area. The main routes of exposure to heavy metals in the studied area are the human activity such as repairing, fueling, greasing and painting of fishing vessels and boats. The present study showed the impact of anthropogenic activity as a source of pollution for heavy metals studied in the north coasts of the Iranian Sea of Oman. The Pb, Cu, Cd and Hg concentrations in sediments were below USEPA, CCME and NOAA set limits for the survival of aquatic organisms. While the concentrations of heavy metals studied in the muscle tissues of *P. homarus* and *C. arel* during pre and post-monsoon showed that the exposes the consumers of fish and fish products from the north coasts of the Iranian Sea of Oman is a health risk.

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