

Original Article

Risk Assessment and Heavy Metal Contamination in Fish (*Otolithes ruber*) and Sediments in Persian Gulf

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Abstract

Introduction: Heavy metal pollution is one of the most serious environmental issues globally. This research investigated the heavy metal concentrations in sediments and fish in Persian gulf.

Materials & Methods: For determination of heavy metal concentrations in sediments eight sampling stations were selected to measure Cd, Zn, Ni, Pb, and Fe in the sediments and in the muscle tissue of *Otolithes ruber* from the northern part of the Hormuz strait (Persian gulf). Samples were then prepared according to MOOPAM for metal analysis. Heavy metal concentrations were analyzed using inductively coupled plasma-optical emission spectrometry (ICP-OES). Analysis of variance (ANOVA) was done to evaluate significant differences in elements' concentrations during sampling periods.

Results: Analysis of the potential ecological risk of sediment heavy metal concentrations showed that most sample sites in the northern part of the Hormuz strait (Persian Gulf) presented a low ecological risk. Regarding enrichment of Cd, Zn, Ni, Pb, and Fe the highest EF belonged to Pb. The obtained mean enrichment factor (EF) values for various metals were between *no enrichment* and *moderate enrichment*.

Conclusion: The concentrations of heavy metal in the edible part of *O. ruber* did not exceed the permissible limits proposed by NOAA, FAO, and WHO standards and thus are suitable for human consumption, except for Pb and Cd.

Keywords: Heavy metal, Sediment, Ecological risk, Persian Gulf, Fish

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Introduction

Heavy metals in marine sediments have natural and anthropogenic origin: distribution and accumulation are influenced by sediment texture, mineralogical composition, reduction/oxidation state, desorption processes, and physical transport. Moreover, metals can be absorbed from the water column onto fine particles' surfaces and move thereafter toward sediments. Moreover, metals participate in various biogeochemical mechanisms, have significant mobility, can affect the ecosystems through bio-accumulation process, and are potentially toxic for environment and for human life ^[1]. Rapid industrialization and urbanization have led to the high accumulation of heavy metals and organic pollutants in soil, water, sediment, street dust, as well as organisms in urban areas ^[2]. Due to their toxicity, bioaccumulation, persistence, and bio magnifications through food chains, heavy metals posed a potential threat to ecological system as well as human health, and gradually drew a wide concern ^[3]. Marine sediments can be sensitive indicators for monitoring contaminants in aquatic environments ^[3]. Sediments have the capacity for accumulating heavy metals from overlying waters; therefore, the enrichment of heavy metals in sediments is often a preferred indicator of the contamination status ^[4]. Sediments also provide habitat and a food source for benthic fauna ^[5]. They have been used to assess the pollution of water bodies and reflect the pollution source extensively which can

provide the information of historical deposition of pollutants ^[6]. Furthermore, sediments could also be a secondary contamination source because pollutants may be directly and indirectly toxic to the aquatic biota and even other organisms throughout the marine food web ^[3]. Various studies have demonstrated that marine sediments from industrialized coastal areas are greatly contaminated by heavy metals; therefore, the evaluation of metal distribution in surface sediments is useful to assess pollution in the marine environment ^[7]. Fishes are widely consumed by humans in the world due to their high protein supply and omega-3 fatty acids that help to reduce the risk of certain types of cancer and cardiovascular diseases ^[8]. However, fishes can accumulate high concentrations of metals absorbed from the water and their food ^[9]. Approximately 90% of human health risk related to fish consumption is associated to metal-contaminated fish ^[10]. The Persian Gulf is a strategic region in the Middle East. Additionally, it is well known as the most active oil production region in the world ^[11]. This Gulf during the last three decades has been affected by two major oil spills. The first occurred in the Iran-Iraq War in 1983, and the second happened during the 1991 Gulf War ^[11]. Also, this region has a complex and interesting ecosystem and is influenced by anthropogenic activities including shipping and transportation, the oil and petrochemical industry, fishing, agriculture, harbor, mining, residential, and commercial

wastewater^[12]. Also, the Persian Gulf is the main source of fishery in the south of Iran^[13]. The objectives of the present study were to assess the extent and ecological risk assessment of heavy metals (Cd, Zn, Ni, Pb, and Fe) in the surface sediments and *Otolithes ruber* from the northern part of Persian Gulf.

Materials and Methods

Surface sediments and fish samples were collected from eight sites in northern part of the Hormuz strait (Persian Gulf), including Eskele 20 (St1), Banagostar(St2), Shour-e-aval (St3), Bustanoo (St4), Kanaf (St5), Souro (St6), estuary(St7), and Sabzbandar(St8)(Fig. 1). Surficial sediments were collected by Peterson grab in the summer of 2015 (Fig. 1). The samples selection according to USGS standard^[14]. Stations were selected first, to cover all Hormuz strait Berths and second to assess all processes of shipping and industry in this area. Samples for metal analysis are prepared according to MOOPAM. They were collected using a zinc-plated Peterson grab. A Teflon spatula was used to extract the sediment samples from the center of grab. After collecting samples, the surface sediment samples were immediately packed in airtight pre-labeled polyethylene bags and preserved at 4° C till the metal analysis. Grain size fractions less than 63 µm were separated for geochemical analysis^[15]. The concentration of elements (Cd, Zn, Ni, Pb, and Fe) in sediment samples was determined by atomic absorption spectroscopy (Varian AA-30 model). All the sediment samples were gently

air-dried at 50° C and then sieved. The sediments were weighted and placed into a Teflon beaker and were digested using 7 mL of aqua regia (1:3HCl:HNO₃). The mixture was heated at 95° C for 1 h and refluxed for 5–10 min until the brown fumes were no longer visible, then after cooling, 5 mL of hydrogen fluoride (HF) was added. Then, samples were refluxed to room temperature. Sediment samples were filtered by Whatman 0.45 µm membrane and brought to 50 mL volume using 1 N HCl^[16]. Finally, heavy metal concentrations were analyzed using inductively coupled plasma-optical emission spectrometry (ICP-OES).

A total of 80 individuals of *Otolithes ruber* were collected from 8 sampling sites in the northern part of Persian Gulf in summer of 2015. In the laboratory, the samples were cleaned up with tap water and deionized water, so that the dorsal muscle samples of each fish were removed for metal analysis. The muscle was preferred because it is a major target tissue for metal storage^[17] and is the main edible part of fish. So, metal assessment in muscle would determine status of public health risk^[18]. The fish samples were collected in sterile polythene bags and kept in the laboratory deep freezer (-20° C) to prevent deterioration until further analysis. According to AOAC (1995), then an acid mixture (10 mL, 70% high purity HNO₃ and 65% HClO₄, 4:1 v/v) was added to the beaker containing 1 g of the dry sample. Later, the mixture was digested at 80° C until the transparent solution was achieved. After cooling, the digested samples

were filtered using Whatman no. 42 filter paper and the filtrate was diluted to 50 mL with

distilled water.

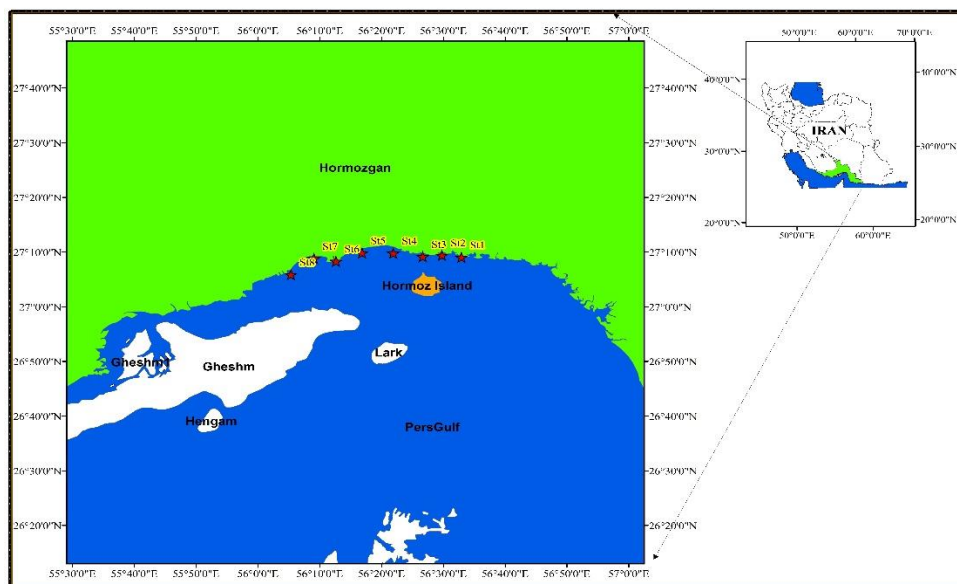


Fig. 1. Sediments' sampling stations in north part of Persian gulf (south Iran)

Assessment of sediment pollution: Enrichment factor (EF) technique was applied to assess the level of contamination in the sediments of Persian gulf North part. According to this technique, metal concentrations were normalized to metal concentrations of average shale [19]. Widely used elements for normalization are Fe [19] and Al [20]. In this study, iron has also been used as a conservative tracer to differentiate the metal contamination with respect to the average shale to quantify the extent and degree of metal pollution. To assess the level of metal enrichment in sediment samples of study area enrichment factor (EF) was computed using the following equation:

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

Where:

M_{sample} concentration of the examined metal in the examined sediment

Fe_{sample} concentration of the reference metal in the examined sediment

$M_{\text{average shale}}$ concentration of the examined metal in the average shale

$Fe_{\text{average shale}}$ concentration of the reference metal in the average shale

According to Chen et al. [20], $EF < 1$ indicates no enrichment, $EF < 3$ is minor enrichment, $EF = 3-5$ is moderate enrichment, $EF = 5-10$ is moderately severe enrichment, $EF = 10-25$ is severe enrichment, $EF = 25-50$ is very severe enrichment, and $EF > 50$ is extremely severe enrichment. The values of the average shale used in this work are from central Persian Gulf sediments [21].

Potential ecological risk index (RI) is introduced to assess the degree of heavy metal pollution in soil or sediments which was originally proposed by Hakanson and widely used [22]. The value of RI can be calculated by the following formulas [22].

$$C_f^i = C_{\text{surface}}^i / C_{\text{reference}}^i$$

$$E_r^i = T_r^i \times C_f^i$$

$$RI = \sum E_r^i$$

Where RI is the sum of potential risk of individual heavy metal, E_r^i is the potential risk of individual heavy Metal, and T_r^i is the toxic-response factor for a given heavy metal. Further, C_f^i is the contamination coefficient, C_{surface}^i is the present concentration of heavy metals in sediments, and $C_{\text{reference}}^i$ is the average of heavy metal concentrations in shale.

Table 1. Background reference values (mg/kg, C_R^i) and toxicity coefficient (T) of heavy metals [21, 22].

| Elements | Cd | Zn | Ni | Pb |
|----------|-----|----|----|-----|
| C_R^i | 2.7 | 69 | 86 | 4.5 |
| T | 30 | 1 | 5 | 5 |

Table 2. Relationship among RI, E_r^i and pollution levels

| Ei | Grade of ecological risk of single metal | RI value | Grade of potential ecological risk of environment |
|----------------|--|----------------|---|
| Ei<40 | Low risk | RI<150 | Low risk |
| 40 < Ei < 80 | Moderate risk | 150 < RI < 300 | Moderate risk |
| 80 < Ei < 160 | Considerable risk | 300 < RI < 600 | Considerable risk |
| 160 < Ei < 320 | High risk | RI > 600 | Very high risk |
| Ei > 320 | Very high risk | | |

Results

Heavy metals concentrations found in sediment: The concentrations of heavy metals in sediments from northern Persian Gulf are tabulated in Table 3. These sediment samples at St4 represented the highest concentrations (in mg/kg) of Cd (0.43), Ni (42.38), and Pb (10.12). Exports of goods as well as loading and unloading of organic and inorganic have increased the

pollution in this site. The highest concentrations of Fe (22400) and Zn (112.31) mg/kg were found in St7. However, the lowest concentration of Ni (24.63) and Pb (5.32) mg/kg were found in St6. The lower Fe (10800) and Zn (25.83) mg/kg were detected in St2 and St3, respectively. Also, the lowest concentration of Cd (0.12) was found in Station 1.

Table 3. Elemental concentration of surficial sediments of North Persian Gulf (mg/kg)

| Station | Cd | Zn | Ni | Pb | Fe |
|--|------|--------|-------|-------|-------|
| 1 | 0.12 | 61.45 | 37.28 | 6.71 | 22100 |
| 2 | 0.16 | 37.76 | 32.71 | 9.62 | 10800 |
| 3 | 0.23 | 25.83 | 30.85 | 8.86 | 17300 |
| 4 | 0.43 | 57.22 | 42.38 | 10.12 | 19800 |
| 5 | 0.24 | 92.60 | 34.29 | 7.40 | 16200 |
| 6 | 0.21 | 33.62 | 24.63 | 5.32 | 18000 |
| 7 | 0.19 | 112.31 | 38.37 | 7.03 | 22400 |
| 8 | 0.28 | 98.69 | 31.90 | 6.83 | 20500 |
| Min | 0.12 | 25.83 | 24.63 | 5.32 | 10800 |
| Max | 0.43 | 112.31 | 42.38 | 10.12 | 22400 |
| Average | 0.23 | 64.93 | 34.05 | 7.73 | 18388 |
| Persian gulf standard ^[21] | 2.7 | 69 | 86 | 4.5 | 20000 |
| Mean crust ^[23] | 0.3 | 75 | 75 | 14 | 46000 |

Heavy metal concentrations in fish tissue are presented in Table 7. According to the results of Table 4, the highest mean values of Ni (15.40 mg/kg) and Fe (34.78 mg/kg) were recorded in the station 5. For Cd, station 3 showed higher mean values with 0.12 ppm, and the higher Zn concentration was observed in station 6 with

32.82 ppm. The highest mean value of Pb (5.02) ppm was recorded in station 2. The lowest concentration of Cd (0.057) and Ni (9.21) mg/kg were found in station 1. For Zn and Pb, station 5 showed the lowest concentrations with (14.95) and (0.86) mg/kg, respectively, and the lower Fe concentration was observed in station 4 with 11.21 mg/kg.

Table 4. Heavy metal concentrations (mg/kg) of tissue from *O. ruber* sampled in the northern part of the Persian Gulf (mg/kg)

| Station | Cd | Zn | Ni | Pb | Fe |
|---------|-------|-------|-------|------|-------|
| 1 | 0.057 | 19.46 | 9.21 | 2.34 | 17.74 |
| 2 | 0.085 | 20.32 | 10.86 | 5.02 | 16.87 |
| 3 | 0.12 | 21.74 | 12.57 | 3.85 | 19.60 |
| 4 | 0.11 | 26.67 | 14.63 | 3.40 | 11.21 |
| 5 | 0.091 | 14.95 | 15.40 | 0.86 | 34.78 |
| 6 | 0.10 | 32.82 | 13.76 | 1.63 | 31.41 |
| 7 | 0.081 | 24.53 | 13.85 | 4.57 | 20.82 |
| 8 | 0.077 | 25.62 | 11.71 | 3.63 | 24.63 |
| Min | 0.057 | 14.95 | 9.21 | 0.86 | 11.21 |
| Max | 0.12 | 32.82 | 15.40 | 5.02 | 34.78 |
| Average | 0.090 | 23.26 | 12.74 | 3.16 | 22.13 |

Discussion

Metal concentrations in the sediments of north part of Hormuz strait (Persian Gulf) were compared to those of other studies performed in other areas of the world (Table 5). Several studies have been conducted on heavy metal contamination throughout the world, including Iran [25-36]. According to the reported results, the mean concentrations of heavy metals in present study were lower than mean crust [23] concentrations. The heavy metal contents in the current study were more than those found in Adriatic sea [30] and Mediterranean sea [24], while they were lower than those in Pichavaram mangrove [32], Izmit Bayn [28], Gulf of Tunis [25],

and Astakos bay [32]. The concentration of Zn in this study was higher in comparison to the ones observed in Mahshahr creeks [34]. Furthermore, the comparison of the present results with previous studies in this area showed that the levels of Zn, Ni, and Pb were lower than those reported by Farsad et al. (2011) (Table 5). Two sets of SQGs developed for marine and estuarine ecosystems [35, 36] were applied in this study to assess the ecotoxicological risk assessment of heavy metals in sediments: (a) the effect range low (ERL) effect range median (ERM), and (b) the threshold effect level (TEL) probable effect level (PEL) values. Low-range values (i.e.,

ERLs or TELs) are concentrations below which adverse effects upon the sediment dwelling fauna would be infrequent. In contrast, the ERLs and PELs represent chemical concentrations above which adverse effects are likely to occur^[35]. The concentrations of Cd, Zn,

and Pb in present study were lower than the corresponding values of the ERL, PEL, and TEL (Table 5). The mean concentration of Ni in this study was more than TEL and ERL but was lower than PEL.

Table 5. Comparison of concentration of heavy metals found in sediment of North Persian Gulf in Iran and other countries(mg/kg)

| Location | Cd | Zn | Ni | Pb | Fe |
|--|-----------|-----------|--------|-----------|---------------|
| Adriatic sea, Italy ^[30] | 0.20 | 95.8 | - | 4.43 | 8800 |
| Pichavaram mangrove ^[31] | 6.96 | 89 | 62 | 11.2 | 32482 |
| Izmit Bay(Turkey) ^[28] | 2.5–9.5 | 440–1,900 | - | 55.2–172 | - |
| Gulf of Tunis(Tunisia) ^[25] | 0.07–0.67 | 75–249 | - | 18.7–98.8 | 25,731–47,922 |
| Astakos bay, Greece ^[32] | 3.25 | 89 | - | 28 | - |
| Mahshahr creeks (Persian Gulf) ^[33] | - | 43 | 70 | 25 | - |
| Mediterranean sea ^[24] | 0.0011 | 0.02 | 0.13 | 0.0057 | 1.29 |
| North part of Persian gulf ^[26] | – | 139.88 | 138.47 | 18.20 | – |
| ERL ^[34] | 5 | 120 | 30 | 35 | – |
| PEL ^[35] | 3.5 | 315 | 36 | 91.3 | – |
| TEL ^[36] | 0.6 | 123 | 18 | 35 | – |
| Persian gulf | 0.23 | 64.93 | 34.05 | 7.73 | 18388 |

*References

Enrichment factor: The obtained mean enrichment factor (EF) values for various metals were between no enrichment and moderate enrichment. The maximum mean EF value

belonged to Pb (Pb =4.04) indicating moderate enrichment and also the minimum mean EF value was seen for Cd (Cd =0.003) showing no enrichment (Table 6).

Table 6. Enrichment factors (EF) of metals for sediment samples

| Station number | Cd | Zn | Ni | Pb |
|----------------|-------|------|------|------|
| 1 | 0.003 | 0.79 | 0.37 | 1.36 |
| 2 | 0.010 | 1 | 0.69 | 4.04 |
| 3 | 0.009 | 0.41 | 0.39 | 2.31 |
| 4 | 0.015 | 0.82 | 0.48 | 2.31 |
| 5 | 0.010 | 1.67 | 0.48 | 2.04 |
| 6 | 0.008 | 0.52 | 0.30 | 1.31 |
| 7 | 0.062 | 1.47 | 0.39 | 1.4 |
| 8 | 0.009 | 1.41 | 0.34 | 1.5 |
| Mean | 0.015 | 1.01 | 0.43 | 2.03 |

Potential ecological risk: Potential Ecological Risk values are shown both individually and totally in Table 7. The order of potential ecological risk factor of heavy metal in sediments of North Persian Gulf was Pb>Cd>Ni>Zn (Table 7). Based on the calculations of the respective index for the selected metals (Cd, Zn, Ni and Pb), it was observed that all of the samples were within the grade of "low ecological risk" as their individual (E_r^i) values were all below 40. Values of E_r^i for Cd ranged from 1.32 (St. 1) to 4.77 (St. 4). For Zn, E_r^i values ranged from 0.41 (St. 3) to 1.64

(St. 8). For Ni, E_r^i values ranged from 1.43 (St. 6) to 2.46 (St. 4). For Pb, it ranged from 5.9 (St. 6) to 11.2 (St. 4). Based on the calculated RI values which is the summation of the calculated E_r^i values, for different elements across the site, it was similarly observed that all the site samples were within the lowest grade of potential ecological risk values (RI<150). RI varied between 10.12 and 17.19 for all metals and the general average was calculated as 13.79. Station 4 (17.19) had higher values of RI while the lowest values was detected at station 6 (10.12).

Table 7. Ecological risk factor (E_r^i) and the potential ecological risk index (RI) of heavy metals in surface sediments of North Persian Gulf

| Station number | Cd | Zn | Ni | Pb | Potential toxicity response indices for heavy metals (RI) | Risk grade |
|----------------|------|------|------|-------|---|------------|
| 1 | 1.32 | 0.89 | 2.16 | 7.45 | 11.82 | Low |
| 2 | 1.77 | 0.54 | 1.9 | 10.65 | 14.86 | Low |
| 3 | 2.55 | 0.41 | 1.79 | 9.8 | 14.55 | Low |
| 4 | 4.77 | 0.82 | 2.46 | 11.2 | 17.19 | Low |
| 5 | 2.64 | 1.34 | 1.99 | 8.2 | 14.17 | Low |
| 6 | 2.31 | 0.48 | 1.43 | 5.9 | 10.12 | Low |
| 7 | 2.1 | 1.62 | 2.23 | 7.8 | 13.75 | Low |
| 8 | 3.09 | 1.43 | 1.85 | 7.55 | 13.92 | Low |
| Average | 2.56 | 0.94 | 1.97 | 8.56 | 13.79 | Low |

The reported results in the literature showed that the metal contents in the fish muscles varied depending on the location and the species that were caught (Table 8). The concentration of Cd, Ni, and Fe in this study was lower than Shat al Arab ^[37] and northwest Persian Gulf ^[38]. The concentration of Pb in present study was more than northwest Persian Gulf ^[38]. The concentrations of Zn, Ni, and Fe in present study were lower than Agriculture Organization (FAO) (1983) and WHO (1996) standards. But, the Pb concentration was more than Agriculture Organization (FAO) (1983) and WHO (1996) standards. Also, the concentration of Cd in the current study was higher than WHO (1996) standards. Therefore, the concentrations of heavy metals in the edible part of *O. ruber* did not exceed the permissible limits proposed by NOAA (2009), FAO (1983), and WHO (1996) (Table 8) and are suitable for human

consumption, except for Pb and Cd. Alahverdi and Savabieasfahani also indicated that the mean concentrations of metals in the sediment were: Pb (42.4 ± 2.7), Cd (7.4 ± 1), Ni (38.1 ± 3.7), and Cu (8.3 ± 1.2) $\mu\text{g g}^{-1}$ dry weight in the Bushehr Province on the Coast of the Persian Gulf ^[39]. Janadeleh et al. reported that the mean concentrations of iron, nickel, lead, and zinc were as: 40991 mg/kg, 65 mg/kg, 31 mg/kg, and 60 mg/kg, respectively in surface sediment of the study area. All heavy metal concentrations in that study were more than heavy metal concentrations of the present study ^[40]. Mortazavi and Sharifian were reported that Mercury concentration was 0.373 $\mu\text{g/g}$ for *Liza abu*, 1.172 $\mu\text{g/g}$ *Sparidentex hasta*, 0.445 $\mu\text{g/g}$ for *Acanthopagrus latus*, 0.390 $\mu\text{g/g}$ for *Thunnus tonggol*, and 0.360 $\mu\text{g/g}$ for *Fenneropenaeus indicus* in Mosa Bay, Persian Gulf^[41].

Table 8. Comparison of heavy metal accumulation in fish (*Otolithes ruber*) muscles with the reported values in other lakes (mg/kg)

| Locations and standards | Cd | Zn | Ni | Pb | Fe | |
|-------------------------|-------|-------|-------|------|-------|-----------------|
| shat al arab | 11.9 | - | 12 | - | 1.7 | ^[37] |
| Northwest Persian gulf | 0.28 | - | 42.01 | 0.45 | 87.02 | ^[38] |
| FAO | 0.5 | 300 | 55 | 2 | 180 | ^[42] |
| WHO | 0.020 | 50 | 30 | 0.5 | 109 | ^[43] |
| NOAA | 4 | 150 | 52 | 128 | 250 | ^[44] |
| Persian gulf | 0.090 | 23.26 | 12.47 | 3.16 | 22.13 | Present study |

Conclusion

This study provided important information on heavy metal concentrations in surface sediments and *O. ruber* from the area of study. The major findings of this study confirmed that heavy metal concentrate in the muscle tissue of *O. ruber* from the area of study. The concentrations of heavy metal in the edible part of *O. ruber* did not exceed the permissible limits proposed by

NOAA (2009), FAO (1983), and WHO (1996). Further, they were reported to be suitable for human consumption, except for Pb and Cd. Analysis of the potential ecological risk of sediment heavy metal concentrations showed that most sample sites in the northern part of the Hormuz strait (Persian Gulf) presented a low ecological risk.

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