

# Assessment of metal contamination in water, sediment, and tissues of *Arius thalassinus* fish from the Red Sea coast of Yemen and the potential human risk assessment

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**Abstract** Heavy metal pollution is one of the most serious environmental issues globally. To evaluate the metal pollution in the Red Sea coast of Hodeida, Yemen Republic, the concentrations of Fe, Cu, Ni, Pb, and Cd in water, sediment, and some vital organs of sea catfish, *Arius thalassinus* collected from polluted and unpolluted sites, were determined. The risk of these metals to humans through fish consumption was then assessed. The results showed that the concentration order of metals in water, sediment, and fish tissues were Fe>Cu>Ni>Pb>Cd. The levels of studied metals in water, sediment, and fish tissues were significantly higher in the polluted site than those of the unpolluted site, with few exceptions. Linear correlation incorporating paired variables (water-sediment, water-fish, and fish-fish) exhibited several significant correlations indicating a common metal pollution. The risk assessment performed revealed that fish consumption was safe for consumers. This field investigation provides a baseline data on metal pollution in this region.

**Keywords** Heavy metal · *Arius thalassinus* · Red Sea · Risk assessment · Pollution

## Introduction

Marine ecosystems are affected by many types of pollutants because of their proximity to sources of contamination and

decrease their natural ability and capacity (Cohen et al. 2001). Heavy metals are one of the main groups of those pollutants which could enter aquatic systems via anthropogenic activities and/or atmospheric deposition. Significant quantities of heavy metals are discharged into aquatic systems which can be strongly accumulated and biomagnified along water, sediment, and aquatic food chain, resulting in devastating effects on the ecological balance of the recipient environment and on a variety of aquatic organisms (Ben Ameer et al. 2012).

Among aquatic species, fish are particularly vulnerable because they cannot escape from the detrimental effects of pollutants (Mahboob et al. 2014). The accumulation of heavy metals in fish due to anthropogenic activities has become internationally an important issue, not only because of the threat to fish but also due to the health risks associated with fish consumption (Rahman et al. 2012).

The Red Sea is recognized as one of the world's most unique coastal and marine environments due to its role as an important repository of marine biodiversity, its complex systems of coral reefs, sea grass beds, and other diverse coastal habitats (Dicks 1987). The Hodeida governorate is located on the Red Sea coast. It is Yemen's seventh largest governorate by area and the second largest by population. In a developing country like Yemen, little attention has been paid to problems of aquatic pollution by anthropogenic sources as economic and social problems have been afforded greater priority than environmental contamination. The coastline of Hodeida has been extensively modified. Dredging and reclamation, industrial and sewage effluents, hyper saline water discharges from desalination plants, and oil pollution are examples of anthropogenic stresses that contribute to environmental degradation in the Red Sea. There has been no strict control on these releases and these contaminants are discharged without any treatment along the coastline. Therefore, there is a situation of mixture of different types of environmental contaminations that makes it difficult to assess the real effects of pollution in

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the components of this aquatic system (Al-Shiwafi et al. 2005).

The present study was carried out in view of the scarcity of information about aquatic pollution and heavy metal concentrations in water, sediment, and marine organism from the coastal area of Hodeida as well as human health risk associated with fish consumption. Therefore, the aim of this work was to provide baseline information on pollution situation and heavy metals distribution in different compartments of the aquatic environment of the Red Sea coast of Hodeida, Yemen Republic. Moreover, to evaluate the health risks of Fe, Cu, Ni, Pb, and Cd via consumption of the *A. thalassinus* by applying the non-carcinogenic target hazard quotient (THQ).

## Materials and methods

### The study area

Hodeida, Yemen Republic is one of the main cities on the Red Sea. From the last few decades and up till now, this city has suffered increasing population, urbanization, and industrial activities. The present study was conducted on two sites located on the Red Sea coast of Hodeida. The first site (unpolluted) is located near Al-Nukhailah beach about 20 km south of Hodeida city with global positioning system

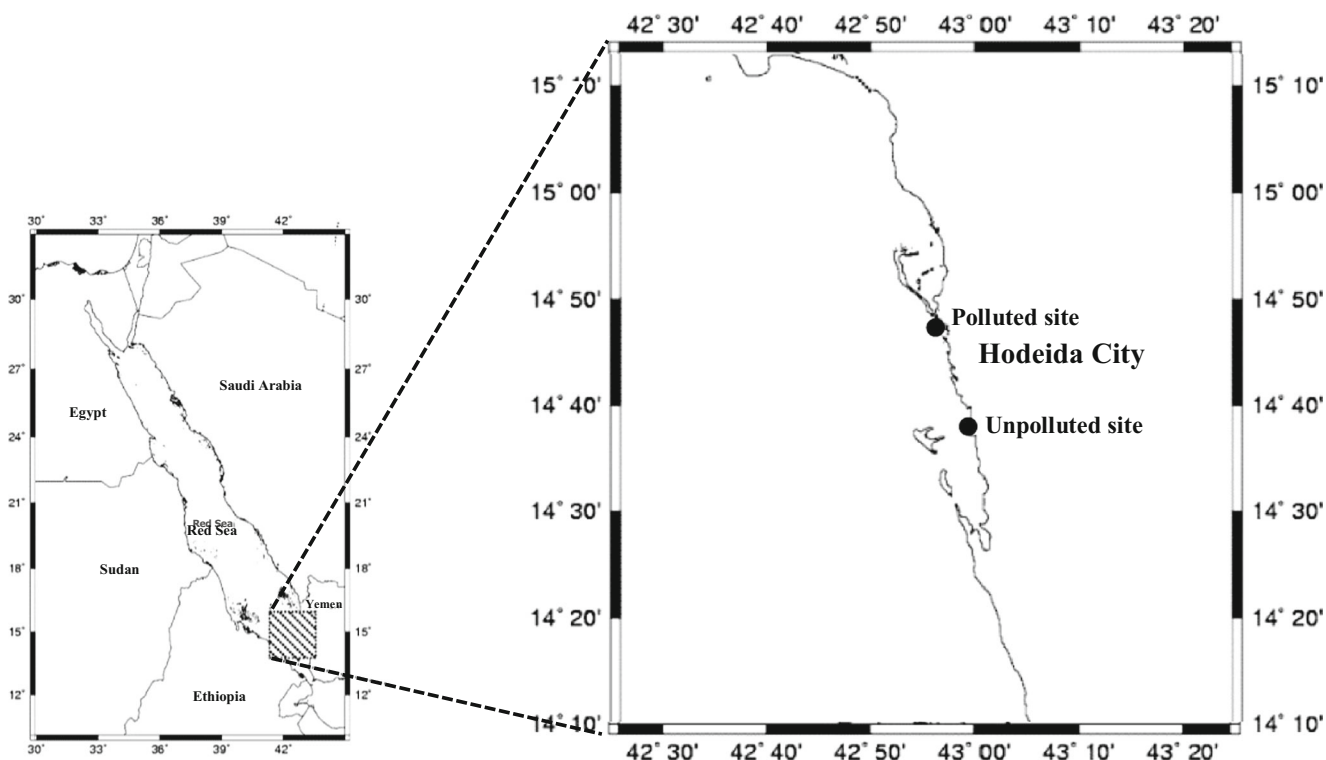
(GPS) reading of  $14^{\circ} 38' 29.87''$  N and  $42^{\circ} 58' 37.97''$  E (Fig. 1). It is considered a reference site as it is situated far away from urban activities where there are no nearby inhabitants, and is thus considered to be very close to an undisturbed natural environment. The second site (polluted) is located along the coast of Al-Shaykh Younes facing Hodeida city at GPS reading of  $14^{\circ} 47' 40.30''$  N and  $42^{\circ} 55' 49.05''$  E (Fig. 1). It is mainly contaminated with continuous untreated industrial and domestic wastewater due to a variety of anthropogenic activities from the nearby city and villages.

### Samples collection

The studied samples of water, sediment, and fish were collected during the summer season, 2012.

Eight water samples were taken with a water sampler from each sampling site (eight samples/site) at water depth of about 30 cm. Water samples were collected using cleaned polypropylene bottles, which were acid-washed and rinsed with distilled water, taking care not to incorporate sediment into the samples. The samples were filtered through Whatman 0.45- $\mu\text{m}$  glass-fiber for obtaining filtered water, acidified with 1 M ultrapure nitric acid (to  $\text{pH} < 2.0$ ) to minimize precipitation, and stored at  $4^{\circ}\text{C}$  for metal analysis.

Eight samples of core sediment up to 20 cm in depth were taken from each sampling site (eight samples/site). Sediment



**Fig. 1** Map of the studied sites

samples were taken using polyvinyl chloride (PVC) corers and well mixed after being freed from coarse shell fragments and visible sea grass leaves when present. After sampling, each sediment sample was sealed in clean polypropylene bags, kept at 4 °C to decrease chemical and biological reactions, and transported to the laboratory immediately for further analysis. The samples were oven dried at 80 °C for 8 h to gain constant weight. The dried samples were then ground using mortar and pestle and sieved through a 63-µm plastic sieve. The samples were packed in polypropylene bags and stored at 4 °C for metal analysis.

Sea catfish, *Arius thalassinus* is a marine bony fish that have high commercial and economic value for the Yemeni people. It is caught throughout the year, mostly near the coast line, and it is a wide-spread food fish species in Yemen (Walczak 1976).

A total number of 20 adult fish (10 fish/site) with average body weight of 160±4.3 and average body length of 24.4±0.35 were collected with the help of local fishermen in a range of 1 km<sup>2</sup> around the reported GPS of each site.

Fish samples were dissected to isolate the desired organs of liver, kidney, gill, muscle, and intestine. Care was taken during dissection of the internal organs to prevent any injuries or metal contamination of the samples by using stainless steel dissecting kits. Samples were packed in polyethylene bags and frozen at -20 °C until metals analysis.

#### Determination of heavy metal

Prior to analysis, all glassware were acid-washed, rinsed in deionized water, and air-dried for 12 h prior to usage.

Dry sediment samples (0.5 g) were digested with 2 ml of HNO<sub>3</sub> (65 %) and 6 ml HCL (37 %) in microwave digestion system for 30 min.

The stored fish tissues were oven dried at 80 °C for 8 h until completely dried, and the dry tissues (1.0 g) were digested with 6 ml of HNO<sub>3</sub> (65 %) and 2 ml of H<sub>2</sub>O<sub>2</sub> (30 %) in microwave digestion system for 30 min.

After digestion, all samples were cooled and diluted to 10 ml with deionized water as described by Mendil and Uluözlü (2007). Digestion program for microwave system was as 2 min for 250 W, 2 min for 0 W, 6 min for 250 W, 5 min for 400 W, 8 min for 550 W, vent 8 min, respectively. Heavy metals (Fe, Cu, Ni, Pb, and Cd) concentrations in water, sediment, and fish tissues samples were determined using atomic absorption spectrophotometer (Nova A 300 Analytikjena AG, Germany) using two replicates for each sample with an error of 10 %. Metal concentrations were expressed as milligram per liter (mg/l) for water samples and as milligram per kilogram dry weight (mg/kg dry wt.) for sediment and fish tissues samples.

#### Quality assurance and quality control (QA/QC) procedures

Blank samples were prepared and treated exactly in the same way as the samples. The concentrations were determined using standard solutions prepared in the same acid matrix. Standards for instrument calibration were prepared on the basis of mono-element certified reference solution (Merck). Standard reference materials of water (SRM 1643e), sediment (SRM 2702), and fish (Lake Superior fish 1946), purchased from the National Institute of Standards and Technology (NIST; USA), were used to validate analysis, and the metal average recovery percentages in all measured samples ranged from 90 to 109 %.

#### Health risk assessment

To estimate the human health risk associated with consumption of fish edible tissue, the target hazard quotient (THQ) was calculated according to the method provided in the US EPA Region III Risk-Based Concentration Table (USEPA 2010). Although the methodology for estimation of THQ (non-carcinogenic) does not provide a quantitative estimate on the probability of an exposed population experiencing a reverse health effect, it offers an indication of the risk level due to pollutant exposure. This method is described in the following equation:

$$ADD(\text{mg/kg/day}) = (C \times IR \times EF \times ED) / (BW \times AT)$$

$$THQ = ADD / \text{Oral RfD}$$

where, ADD is average daily dose of a specific metal over a lifetime, C is the mean metal concentration in muscle tissue (mg/kg dry wt.), IR is the ingestion rate (0.018 kg/day for normal adult Yemeni consumers according to FAO (2002) or 0.1424 kg/day for subsistent fishermen or other habitual fish consumers according to USEPA (2000)), EF is the exposure frequency (365 days/year), ED is the exposure duration over a lifetime (70 years), BW is the body weight (60 kg for normal Yemeni adults (Walpole et al. 2012)), AT is the average life time (70 years×365 days/year), and oral RfD is the oral reference dose (Fe=0.7, Cu=0.04, Ni=0.02, Pb=0.004, Cd=0.001 mg/kg/day) according to USEPA (2010). THQ values less than 1.0 indicate that adverse health effects are not likely to occur but if the ADD of certain metal exceeds its oral RfD and thus the THQ is greater than or equal to 1.0, it is probably that adverse health effects are expected to occur.

The hazard index (HI) from THQ can be expressed as the sum of the target hazard quotients (USEPA 2011) as follows:

$$HI = THQ_{Fe} + THQ_{Cu} + THQ_{Ni} + THQ_{Pb} + THQ_{Cd}$$

## Statistical analysis

Statistically, the analysis of the present results was carried out using the Statistical Package for the Social Sciences (SPSS) version 18 released in 2009 (SPSS, Inc., Chicago, IL, USA) software for windows. Student's *t* test was used to estimate the significant differences between the levels of metals in water or sediment or tissues of sea catfish collected from the unpolluted and polluted sites. Duncan's test was applied in order to estimate the homogeneity (similarities) between the concentrations of the metal in various tissues collected from the same area. The regression analysis and Pearson's correlation coefficient (*r*) were used to fit the relationship between the levels of metals in water, sediment, and fish tissues. In all cases, statistical significance was estimated at  $P < 0.05$ .

## Results

### Heavy metals in water and sediment

The levels of Fe, Cu, Ni, Pb, and Cd in water and sediment samples collected from the unpolluted and polluted sites are recorded in Table 1. The relationship between metals level in water and sediment are represented by correlation coefficient (*r*) in Table 2. All metals concentrations in water and sediment samples of the polluted site were significantly higher than

**Table 1** Heavy metals concentration in water (mg/l) and sediment (mg/kg dry wt.) samples (mean±SE, *n*=8)

|          | Site        |             |
|----------|-------------|-------------|
|          | Polluted    | Unpolluted  |
| Fe       |             |             |
| Water    | 0.09±0.005* | 0.05±0.006  |
| Sediment | 199.8±0.2*  | 100.5±0.7   |
| Cu       |             |             |
| Water    | 0.08±0.008* | 0.02±0.005  |
| Sediment | 76.0±3.1*   | 39.1±1.6    |
| Ni       |             |             |
| Water    | 0.05±0.006* | 0.01±0.001  |
| Sediment | 42.2±2.5*   | 8.63±0.8    |
| Pb       |             |             |
| Water    | 0.03±0.004* | 0.003±0.001 |
| Sediment | 6.0±0.15*   | 4.4±0.12    |
| Cd       |             |             |
| Water    | 0.02±0.004* | 0.003±0.001 |
| Sediment | 5.5±0.17*   | 2.8±0.22    |

\* Significant difference ( $P < 0.01$ ) in comparison with the unpolluted site

**Table 2** Pearson's correlation coefficient (*r*) between heavy metals level in water (mg/l) and sediment (mg/kg dry wt.) samples collected from the polluted site

|       | Sediment |         |         |         |         |
|-------|----------|---------|---------|---------|---------|
|       | Fe       | Cu      | Ni      | Pb      | Cd      |
| Water |          |         |         |         |         |
| Fe    | +0.49    | +0.36   | -0.94** | -0.64*  | +0.87** |
| Cu    | -0.05    | +0.05   | +0.69*  | +0.19   | -0.20   |
| Ni    | -0.49    | -0.42   | +0.71*  | +0.55*  | +0.04   |
| Pb    | -0.89**  | -0.85** | +0.71*  | +0.90** | -0.03   |
| Cd    | -0.06    | 0.02    | +0.47   | +0.13   | +0.20   |

\* Significant correlation ( $P < 0.05$ ); \*\* Significant correlation ( $P < 0.01$ )

those of the reference site (Table 1). The Fe level in the polluted water correlated positively with Cd (+0.87) and negatively with Ni and Pb (-0.94 and -0.64, respectively) concentrations in sediment of the same site. Direct relationship was recorded between Cu level in water and Ni (+0.69) content in sediment of the polluted site. Nickel content of the polluted water showed significant positive relationships with Ni and Pb contents in the polluted sediment with correlation coefficient +0.71 and +0.55, respectively. Furthermore, the level of Pb in the polluted water correlated positively with Ni and Pb (+0.71 and +0.90, respectively) and negatively with Fe and Cu (-0.89 and -0.85, respectively) contents in the polluted sediment (Table 2).

### Metals bioaccumulation in fish tissues

The concentrations of Fe, Cu, Ni, Pb, and Cd in the muscle, liver, kidney, intestine, and gill of the sea catfish, *A. thalassinus*, are recorded in Table 3.

The concentrations of the studied metals in all selected tissues of the polluted fish were significantly higher than those of the unpolluted fish except the intestinal Cu content that revealed the opposite behavior as well as the intestinal Fe and gill Cu contents that showed insignificant differences. Among the tissues of the polluted fish, the intestine tissue showed the lowest concentration of Fe (3.1±0.5), whereas the highest concentration was detected in the hepatic tissue (9.2±1.1). The accumulation pattern of Fe in tissues of fish sampled from the polluted site was liver>kidney>gill>muscle>intestine. The recorded lowest value of Cu concentration was detected in the intestine (0.25±0.03) and the highest value was in the liver and kidney tissues (1.3±0.18 and 1.4±0.17, respectively) of fish collected from the polluted site (Table 3).

The gill accumulated the highest content of Ni (1.8±0.17) whereas the muscle accumulated the lowest content (0.42±0.08) in tissues of contaminated fish. The arrangement order of Pb content in tissues of the polluted fish was intestine>

**Table 3** Heavy metals concentration (mg/kg dry wt.) in the studied tissues of sea catfish, *A. thalassinus* (mean±SE, n=10), the minimum and maximum values were represented in parentheses

|                 | Muscle                                   | Liver                                  | Kidney                                   | Intestine                                | Gill                                    |
|-----------------|--|--|--|--|---|
| <b>Fe</b>       |  |  |  |  |   |
| Unpolluted site | 0.7±0.2 <sup>a</sup><br>(0.3–1.9)        | 3.6±0.5 <sup>c</sup><br>(2.4–4.6)      | 3.9±0.22 <sup>c</sup><br>(2.7–4.6)       | 2.5±0.3 <sup>b</sup><br>(2.0–4.0)        | 3.2±0.4 <sup>bc</sup><br>(2.1–4.5)      |
| Polluted site   | 3.9±0.6 <sup>ab</sup> **<br>(3.1–6.4)    | 9.2±1.1 <sup>d</sup> **<br>(6.2–13.8)  | 6.2±0.6 <sup>c</sup> **<br>(4.4–6.9)     | 3.1±0.5 <sup>a</sup><br>(1.7–4.1)        | 5.4±0.4 <sup>bc</sup> **<br>(4.6–6.4)   |
| <b>Cu</b>       |  |  |  |  |   |
| Unpolluted site | 0.20±0.02 <sup>a</sup><br>(0.18–0.33)    | 0.38±0.05 <sup>ab</sup><br>(0.28–0.55) | 0.21±0.03 <sup>a</sup><br>(0.12–0.36)    | 0.48±0.09 <sup>b</sup><br>(0.4–0.5)      | 0.87±0.15 <sup>c</sup><br>(0.5–1.5)     |
| Polluted site   | 0.34±0.02 <sup>a</sup> **<br>(0.24–0.42) | 1.3±0.18 <sup>b</sup> **<br>(0.7–2.0)  | 1.4±0.17 <sup>b</sup> **<br>(0.5–1.7)    | 0.25±0.03 <sup>a</sup> *<br>(0.2–0.4)    | 1.1±0.17 <sup>b</sup><br>(0.6–1.8)      |
| <b>Ni</b>       |  |  |  |  |   |
| Unpolluted site | 0.18±0.03 <sup>ab</sup><br>(0.1–0.3)     | 0.11±0.005 <sup>a</sup><br>(0.1–0.14)  | 0.13±0.007 <sup>a</sup><br>(0.10–0.16)   | 0.29±0.04 <sup>c</sup><br>(0.2–0.4)      | 0.24±0.04 <sup>bc</sup><br>(0.2–0.4)    |
| Polluted site   | 0.42±0.08 <sup>a</sup> *<br>(0.2–0.7)    | 1.21±0.23 <sup>b</sup> **<br>(0.3–2.2) | 1.27±0.13 <sup>b</sup> **<br>(0.7–1.8)   | 0.47±0.05 <sup>a</sup> *<br>(0.3–0.6)    | 1.8±0.17 <sup>c</sup> **<br>(1.1–2.5)   |
| <b>Pb</b>       |  |  |  |  |   |
| Unpolluted site | 0.07±0.005 <sup>b</sup><br>(0.05–0.09)   | 0.04±0.01 <sup>a</sup><br>(0.02–0.06)  | 0.07±0.01 <sup>b</sup><br>(0.04–0.09)    | 0.04±0.004 <sup>a</sup><br>(0.03–0.05)   | 0.14±0.02 <sup>c</sup><br>(0.1–0.2)     |
| Polluted site   | 0.20±0.03 <sup>a</sup> **<br>(0.1–0.3)   | 0.21±0.03 <sup>a</sup> **<br>(0.1–0.3) | 0.21±0.03 <sup>a</sup> **<br>(0.1–0.3)   | 0.72±0.05 <sup>c</sup> **<br>(0.5–0.9)   | 0.34±0.02 <sup>b</sup> **<br>(0.2–0.4)  |
| <b>Cd</b>       |  |  |  |  |   |
| Unpolluted site | 0.013±0.003 <sup>a</sup><br>(0.01–0.03)  | 0.04±0.01 <sup>b</sup><br>(0.02–0.09)  | 0.027±0.005 <sup>ab</sup><br>(0.02–0.04) | 0.03±0.004 <sup>b</sup><br>(0.02–0.05)   | 0.056±0.004 <sup>c</sup><br>(0.04–0.07) |
| Polluted site   | 0.1±0.02 <sup>ab</sup> **<br>(0.06–0.15) | 0.15±0.02 <sup>b</sup> **<br>(0.1–0.2) | 0.09±0.02 <sup>ab</sup> **<br>(0.06–0.1) | 0.05±0.006 <sup>a</sup> *<br>(0.04–0.07) | 0.27±0.04 <sup>c</sup> **<br>(0.2–0.4)  |

Means marked with the same superscript lowercase letter in the same row are not significantly different. \* and \*\* pointed to significant differences in comparison with the unpolluted site at confidence level 0.95 (P<0.05) and 0.99 (P<0.01), respectively

gill>kidney~liver>muscle, whereas in case of Cd the order was gill>liver>kidney~muscle>intestine (Table 3).

**Relationship between metal concentrations in water and fish tissues**

The correlation between metals content in water and fish tissues are represented by correlation coefficient (r) in Table 4.

The level of Fe in the polluted water showed positive correlations with the hepatic Fe and Pb contents (+0.76 and +0.77, respectively) as well as with the kidney contents of Ni, Pb, and Cd (+0.72, +0.95, and +0.78, respectively) in fish of the polluted site. On the other hand, significant negative relationships were recorded between aquatic Fe and Cu contents in all tissues except the liver. The relationship between aquatic Cu and the bioaccumulated Cd was negative in all tissues except in the liver that showed positive correlation (+0.76). Another negative correlation existed between aquatic Cu and the intestinal and hepatic Fe contents (–0.66 and –0.73, respectively). On the other side, the aquatic Cu related directly with Pb contents in the gill and intestine tissue (+0.57).

Nickel level in the polluted sea water revealed strong negative relationships with Cd contents in the kidney, gill, and intestine (–0.89, –0.73 and –0.99, respectively) as well as with the hepatic Cu and Pb contents (–0.99 and –0.59, respectively). Furthermore, significant direct correlations were detected between aquatic Ni and Pb contents in the gill and intestine tissue (+0.75 and +0.61, respectively).

The level of Pb in the polluted water showed significant negative relationships with the kidney, gill, and muscle contents of Ni as well as with the intestinal Cd content. Inversely, positive correlations were found between the aquatic Pb with the gill and intestine Pb contents (+0.99 and +0.70, respectively).

The aquatic Cd content correlated reversely with Fe contents in the gill and intestine (–0.65 and –0.68, respectively) as well as with Cu contents in the liver, kidney, and gill tissues (–0.95, –0.65, and –0.79, respectively). Moreover, there were significant positive correlations between aquatic Cd with Pb contents in the gill and intestine tissues (+0.59 and +0.63, respectively) (Table 4).



**Table 4** Pearson's correlation coefficient ( $r$ ) between heavy metals level in the polluted water (mg/l) and tissues (mg/kg dry wt.) of sea catfish, *A. thalassinus* collected from the polluted site

|           | Fe (water) | Cu (water) | Ni (water) | Pb (water) | Cd (water) |
|-----------|------------|------------|------------|------------|------------|
| <b>Fe</b> |            |            |            |            |            |
| Liver     | +0.76*     | -0.73*     | -0.35      | -0.05      | -0.42      |
| Kidney    | -0.32      | +0.70*     | +0.22      | -0.28      | +0.50      |
| Gill      | +0.06      | -0.24      | -0.71*     | -0.89**    | -0.65*     |
| Muscle    | -0.75*     | +0.74*     | +0.37      | +0.05      | +0.44      |
| Intestine | +0.55*     | -0.66*     | -0.19      | +0.21      | -0.68*     |
| <b>Cu</b> |            |            |            |            |            |
| Liver     | +0.37      | -0.90**    | -0.99**    | -0.75*     | -0.95**    |
| Kidney    | -0.56*     | -0.54*     | -0.24      | +0.31      | -0.65*     |
| Gill      | -0.53*     | -0.16      | -0.59*     | -0.65*     | -0.79*     |
| Muscle    | -0.59*     | +0.99**    | +0.84**    | +0.47      | +0.88**    |
| Intestine | -0.83**    | +0.42      | +0.07      | -0.05      | +0.04      |
| <b>Ni</b> |            |            |            |            |            |
| Liver     | -0.26      | -0.59*     | -0.25      | +0.32      | -0.65*     |
| Kidney    | +0.72*     | +0.09      | -0.16      | -0.60*     | +0.28      |
| Gill      | +0.35      | -0.20      | -0.66*     | -0.96**    | -0.28      |
| Muscle    | -0.03      | +0.31      | -0.23      | -0.68*     | +0.14      |
| Intestine | -0.39      | -0.28      | +0.14      | +0.66*     | -0.61*     |
| <b>Pb</b> |            |            |            |            |            |
| Liver     | +0.77*     | -0.31      | -0.59*     | -0.89**    | -0.19      |
| Kidney    | +0.95**    | -0.70*     | -0.50      | -0.37      | -0.39      |
| Gill      | -0.54*     | +0.57*     | +0.75*     | +0.99**    | +0.59*     |
| Muscle    | 0.003      | +0.01      | -0.51      | -0.83**    | -0.20      |
| Intestine | -0.97**    | +0.57*     | +0.61*     | +0.70*     | +0.63*     |
| <b>Cd</b> |            |            |            |            |            |
| Liver     | +0.15      | +0.76*     | +0.53*     | -0.01      | +0.85**    |
| Kidney    | +0.78*     | -0.90**    | -0.89**    | -0.72*     | -0.76*     |
| Gill      | +0.45      | -0.98**    | -0.73*     | -0.28      | -0.88**    |
| Muscle    | +0.16      | -0.51      | +0.01      | +0.50      | -0.33      |
| Intestine | +0.39      | -0.81**    | -0.99**    | -0.85**    | -0.88**    |

\* Significant correlation ( $P < 0.05$ ); \*\* Significant correlation ( $P < 0.01$ )

#### Relationships between metals concentration in fish tissues

The interrelationship between the concentrations of the investigated metals with each other in the selected tissues of the polluted fish are represented by correlation coefficient ( $r$ ) in Table 5.

In the liver tissue, there are direct relationships between Fe and Pb (+0.58) as well as between Cu and Cd contents (+0.56) while Ni content exhibited inverse correlations with Cu and Cd contents (-0.85 and -0.61, respectively). In the kidney tissue, significant positive correlation existed between Cu and Pb contents with correlation coefficient of +0.61. The gill tissue showed inverse correlations between Cd with Ni and Pb contents (-0.64 and -0.72, respectively). In addition, Cd exhibited positive relationship with Cu contents in the gill

**Table 5** Interrelationships between heavy metals concentration with each other in the tissues of sea catfish, *A. thalassinus* collected from the polluted site

|   | Liver   | Kidney | Gill   | Muscle  | Intestine |
|---|---------|--------|--------|---------|-----------|
| <b>Correlation coefficient (<math>r</math>)</b> |         |        |        |         |           |
| <b>Iron</b>                                     |         |        |        |         |           |
| Copper  | -0.16   | +0.38  | -0.28  | +0.25   | -0.14     |
| Nickel  | +0.15   | +0.18  | -0.25  | +0.93** | +0.04     |
| Lead  | +0.58*  | +0.47  | +0.03  | -0.01   | +0.28     |
| Cadmium   | +0.17   | -0.16  | +0.20  | -0.64*  | -0.15     |
| <b>Copper</b>                                   |         |        |        |         |           |
| Nickel  | -0.85** | +0.43  | -0.51  | +0.45   | -0.58*    |
| Lead  | -0.03   | +0.61* | -0.72* | -0.40   | -0.60*    |
| Cadmium   | +0.56*  | +0.16  | +0.70* | -0.69*  | -0.67*    |
| <b>Nickel</b>                                   |         |        |        |         |           |
| Lead  | -0.24   | +0.28  | +0.25  | -0.03   | +0.17     |
| Cadmium   | -0.61*  | +0.11  | -0.64* | -0.64*  | +0.40     |
| <b>Lead</b>                                     |         |        |        |         |           |
| Cadmium   | -0.27   | -0.01  | -0.72* | +0.28   | +0.04     |

\* Significant correlation ( $P < 0.05$ ); \*\* Significant correlation ( $P < 0.01$ )

tissue with correlation coefficient of +0.70. In the muscle tissue, positive correlations were found between Ni and Fe content with correlation coefficient of +0.93. On the other hand, negative correlations existed between Cd with Fe, Cu, and Ni. The intestinal tissue exhibited negative relationships between Cu with Ni, Pb, and Cd contents (-0.58, -0.60, and -0.67, respectively) (Table 5).

#### Human health risk assessment

The values of ADD, THQ, and HI are calculated and recorded in Table 6. The mean metals concentrations in fish muscle were used to conduct THQ for human fish consumption. It is clear that all THQ values for all estimated metals were lower than 1.0 at both ingestion rates. In addition, the HI values of the five heavy metals in the muscle tissue were lower than one for all consumers at both studied sites.

## Discussion

#### Heavy metals in water and sediment

The significant high levels of the studied metals in the water of the polluted site may be attributed to the discharge of untreated sewage, agricultural and industrial effluents into this site. Varol (2011) attributed the contamination of coastal areas with heavy metals to the higher population densities of urbanization and industrialization. Due to the presence of some

**Table 6** Average daily dose (ADD; mg/kg/day), target hazard quotient (THQ), and hazard index (HI) of the estimated metals caused by consumption of muscle tissue at normal (N) and subsistence (S) ingestion rate

| Site       | Ingestion rate | Average daily dose |        |        |         |         | Target hazard quotient |       |       |       |       | HI   |
|------------|----------------|--------------------|--------|--------|---------|---------|------------------------|-------|-------|-------|-------|------|
|            |                | Fe                 | Cu     | Ni     | Pb      | Cd      | Fe                     | Cu    | Ni    | Pb    | Cd    |      |
| Unpolluted | N              | 0.0002             | 0.0001 | 0.0001 | 0.00002 | 0.00001 | 0.0003                 | 0.002 | 0.003 | 0.006 | 0.004 | 0.01 |
|            | S              | 0.0010             | 0.0004 | 0.0004 | 0.00010 | 0.00003 | 0.0020                 | 0.010 | 0.018 | 0.040 | 0.026 | 0.10 |
| Polluted   | N              | 0.0010             | 0.0001 | 0.0001 | 0.00006 | 0.00006 | 0.0020                 | 0.003 | 0.006 | 0.020 | 0.033 | 0.06 |
|            | S              | 0.0080             | 0.0007 | 0.0008 | 0.00040 | 0.00040 | 0.0110                 | 0.017 | 0.042 | 0.114 | 0.220 | 0.40 |

industrial resources such as electric power station, ship maintenance factories, metal plating, coating operations, transportation equipment, machinery, and baking enamels in Hodeida, the body surface of the sea at this area may directly and/or indirectly be contaminated with heavy metals. The concentration order of metals in water and sediment samples were Fe < Cu < Ni < Pb < Cd.

Analysis of heavy metals in sediments offers more convenient and more accurate means of detecting and assessing the degree of water pollution (Tam and Wong 2000). The higher heavy metals contents in the sediments of the polluted site may refer to several reasons including sewage discharge of Hodeida city, the industrial pollution, ship wrecks, and oil enrichment in the vicinity areas. Vicente-Martorell et al. (2009) attributed the abnormal high level of heavy metals in the sediments to the industrial and urban discharges. The concentrations of heavy metals in the sediments were hundreds-fold higher than those of the overlying water. This was mainly due to the presence of large quantities of organic and inorganic pollutants, disposal of domestic and industrial effluent in the water, and the deposition of dead organisms and other particulate matter with high metal contents. In addition, some metals may interact with organic matter in the water and settle down causing the precipitation of metals in the sediments (Gupta et al. 2009). When contaminated particulate or sedimentary material is dispersed through an ecosystem, it equilibrates with water, detritus, and living food materials, resulting in ongoing contamination of all environmental compartments (Pyle et al. 2005). The detected positive correlation between the concentration of each metal in the water and sediment, in the present study, supports this argument.

Taking into consideration that the Red Sea is an enclosed sea and has a slow turnover rate of 6 years for the surface water layer and 200 years for the whole water body as well as it is the most saline water body of the world seas (Morcos and Varelly 1970), the accumulation of chemical pollutants is expected to increase annually in all its components, changing its quality and affecting its aquatic life.

#### Metals bioaccumulation in fish tissues

Fish accumulate metals mainly in the liver, kidney, gill, and other organs in much smaller amounts. In addition, differences in metal concentrations in various tissues may result from their different capability to induce metal binding proteins such as metallothioneins (MTs) (Tuzen and Soylak 2007). The main reason for the elevated concentrations of metals in tissues of the polluted fish in comparison to the corresponding reference concentrations may be due to high levels and continuous discharges of metals into this site that increases uptake of these metals from water leading to their bioaccumulation in different fish tissues.

Iron and copper showed the same behavior regarding their contents arrangement in tissues of contaminated fish as their highest contents were recorded in the hepatic tissue whereas the lowest were in the muscular and intestinal tissues. A similar trend was observed in the findings of Kojadinovic et al. (2007) in the liver tissue of four pelagic fish collected from the Indian Ocean. According to the fact that the liver is the main metabolic organ, the amount of pollutants in fish liver is directly proportional to the degree of pollution in the aquatic environment (Tapia et al. 2012). Many experimental and field studies showed that the liver is the target organ for bioaccumulation of many metals and highly active in the uptake and storage of metals because of its role in the storage, redistribution, detoxification, and transformation of contaminants (Dural et al. 2007; Yilmaz et al. 2010). The high bioaccumulation of Cu in the liver could be attributed to the specific metabolic processes and enzyme-catalyzed reaction involving Cu taking place in the liver ((El-Shahawi and Yousuf 1998). The positive relationship that was noticed between the hepatic contents of Cu and Cd, in the present work, was in agreement with the previous study of Kamunde and MacPhail (2011). They imputed the enhanced hepatic copper accumulation in the presence of Cd to the induction of metal binding proteins (MTs) and its stability state upon binding of various metal ions.

The lowest Fe and Cu concentrations were detected in the muscle tissue which may be related to the increase of their deposition in the hepatic tissue (Pyle et al. 2005). In addition,

Yilmaz et al. (2007) showed that the low metals concentration in the muscle may reflect the low MTs content. Moreover, the presence of mucous layer coating the fish skin surface serves as a barrier which protects the integrity of fish flesh from the invasion of metals from the media.

Fish intestine compared to other organs acts as a transient site for most metal accumulation in fish (Marijic and Raspor 2007). Jezierska and Witeska (2006) concluded that dietary uptake of Fe elevates its content in the fish intestinal tissue followed by gradual depletion as a result of excretion and/or absorption. The low content of Fe in fish intestine may be explained by the inverse correlation detected between the intestinal Fe and aquatic Cd content. This finding as well as the negative relationship recorded between Fe content in the gill and waterborne Cd may indicate that both of them have the same uptake pathway. Bury and Grosell (2003) concluded that in vivo branchial absorption of Fe was inhibited by waterborne Cd. Moreover, Cooper et al. (2006) also provided indirect evidence that Fe and Cd may share a common transport pathway in piscine intestine. The negative correlation found between intestinal Fe and waterborne Cu may be attributed to the common absorption pathway of both metals. Nadella et al. (2007) reported that the intestinal Cu absorption in rainbow trout, *Oncorhynchus mykiss*, was dependent on the luminal proton gradient, thereby suggesting the involvement of divalent metal transporter 1 (DMT1) in Cu absorption. They deduced that the uptake of dietary Fe and Cu likely occurs through the same pathway, probably DMT1. They also observed that Cu absorption and transportation through the blood was significantly inhibited by high Fe level which was in agreement with inverse correlations recorded between Cu contents in, approximately, all studied tissues with aquatic Fe. The negative correlations presented between Cu and Cd in the muscle and intestine as well as between the liver, kidney, and gill contents of Cu with waterborne Cd were highly parallel to the findings of Komjarova and Blust (2009) and Kamunde and MacPhail (2011) who revealed that there is an antagonism between Cu and Cd. In addition, the negative correlation recorded between Cu and Pb content in the intestine tissue demonstrated and explained the low level of Cu in the intestine of the polluted fish in comparison with that of the unpolluted fish as Pb exhibited its highest concentration in the intestinal tissue of the polluted fish.

The highest amounts of Ni and Cd were detected in the gill tissue of the polluted fish which may be a result of MTs that trap heavy metals. Many authors showed marked high accumulation of Ni and Cd in fish gills (Kalay et al. 1999; Abu Hilal and Ismail 2008; Vinodhini and Narayanan 2008). Gill epithelia are usually covered with a layer of protecting mucus functioning as an ion exchange system. In addition, the filamentous protecting mucus may act as an ion exchanger system with different affinities associated with different metals

(Playle and Wood 1990). It seems likely that mucus can compete with other ligands in forming mucus–metal complexes within the fish gill microenvironment (Tao et al. 2001). Thus, the high metals concentration in the fish gill may be ascribed to the element complexation with the mucus. The low levels of Cd found in other tissues in comparison to the gills confirmed that the gills are the main uptake route of Cd from water (Yilmaz et al. 2010).

The high bioaccumulation of Cd in the liver of fish may be due to its strong binding with cystine residues of MT (Kaoud and El-Dahshan 2010). Positive correlations were detected between Cd and Cu contents in the liver and gill of the polluted fish. It is thus possible that hepatic and gill MTs could act as a sink for toxic Cd or for the excess Cu in the polluted fish. Similarly, Pelgrom et al. (1995) noticed high accumulation of Cd in various tissues of *Oreochromis mossambicus* in the presence of copper. In addition, the existence of high waterborne Cu increases the Cd content in the liver which could be attributed, according to Kamunde and MacPhail (2011), to the induction of MTs that bind Cd. This is compatible with our findings which revealed direct correlation between waterborne Cu with the hepatic Cd content. The recorded strong negative relationships between, almost, all tissues Cd contents with aquatic Cu were in agreement with many prior studies reported that the availability of Cu in water caused reduction in tissues Cd concentrations such as that in Juvenile tilapia, *O. mossambicus*, (Pelgrom et al. 1994) and zebrafish, *Danio rerio* (Komjarova and Blust 2009). The inverse relationship found between aquatic Pb and intestinal Cd content explained the low content of Cd in fish intestine. The calcium transport pathways participate in the absorption of dietary Cd and Pb (Alves and Wood 2006). Therefore, the competition between Cd and Pb for the intestinal uptake sites causes inhibition of Cd content in the entity of waterborne Pb. Cadmium is antagonistic to the essential trace elements and competes with nutrient elements for binding sites (Kojadinovic et al. 2007). Thus, all negative correlations found between Cd content in different tissues with Fe, Cu, and Ni contents either in water or in tissues could be explained according to previous explanation.

The higher Pb content in intestine tissue in comparison with gill indicated that uptake of this metal from food is more important than that from water. Many authors also observed high concentrations of Pb in the intestinal tissue of fish from natural water bodies (Kock et al. 1998; Soltan et al. 2005). The positive correlations recorded between the levels of Cu, Ni, Pb, and Cd in the polluted water with Pb contents in intestine and gill tissues of the polluted fish may be explained by the high concentration of lead detected in these tissues. Similarly, Komjarova and Blust (2009) showed that existence of Cu, Ni, and Cd in water increased the intestinal uptake of Pb by fish. Marijic and Raspor (2007) revealed that Pb form complexation with the intestinal amino acids and small peptides with



respect to high affinity for protein thiol-group which was present within the fish intestine.

Masoud et al. (2007) found that the gill exhibited high accumulation of Pb and attributed this to the similarity of lead and calcium in their deposition and mobilization from the gill. Rogers and Wood (2004) showed that fish may uptake Pb and Cd via the branchial  $\text{Ca}^{2+}$  uptake pathways and thus compete with each other for this uptake sites leading to inverse relationship between them at gill uptake sites which agreed with the negative correlation, presented in our data, between Pb and Cd in gill tissues.

#### Health risk assessment

A health risk assessment approach was conducted to assess the current risk status associated with the consumption of fish-edible tissues from the studied sites at two different consumption rates, normal and subsistence ingestion rates. The calculations showed that for an average Yemeni person, the values of ADD of fish muscle lead to metal consumption very much lower than the oral RfD guidelines for all studied metals, thus strongly indicating no health risk due to consumption of metal-contaminated muscle in *A. thalassinus*. The target hazard quotient (THQ) values of Fe, Cu, Ni, Pb, and Cd for the muscle tissue did not exceed one, indicating that there is no health risk from consuming the investigated fish. Analogously, the HI values ranged from 0.01 to 0.06 and from 0.10 to 0.40 for both normal and subsistence ingestion rates, respectively, being less than 1.0 suggested that the health risk was insignificant. Thus, the consumption of muscle tissue of *A. thalassinus* at current ingestion rate poses no health problems for consumers.

#### Conclusion

This field investigation revealed higher degree of pollution in the coastal area of Hodeida city in comparison to the other clean environment. The THQ of individual metals and the HI of all five metals were lower than one, indicating that exposure to these metals in fish posed no chronic-toxic risk to the local community under the current fish consumption rate. The present results provide baseline information on the metal pollution in this coastal area and could be used as a contribution to the knowledge and rational management of this region in the future and would serve as a baseline against which future anthropogenic effects can be assessed. In addition, metals profiling from the present study will be useful for future comparative metals pollution studies and monitoring works on the assessment of land bases and marine pollution inputs. Efforts are needed to protect the coastal area of Hodeida from pollution and also to reduce environmental

risks. This study and the valuable data will pave the way for future research on this area.

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