INVESTIGATION OF TOXIC METALS POLLUTION IN WATER, SEDIMENT AND FISH AT ADEN COAST, GULF OF ADEN, YEMEN

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ABSTRACT

Objectives: This study investigates concentrations of toxic metals, Lead and Cadmium in water, sediments and fish organs (muscle, liver and gill). Collected from Aden coast, gulf of Aden in Yemen. Water samples and sediment were taken from nine locations.

Methods: Four fish species were collected (Lethrinus mahsena; Epinephelus areolatus; Thunnus tonggol and Sphyraena jello) were collected from the local commercial fishermen of Aden city during winter 2011. Summer 2012 and winter 2013.

Results: Lead concentration in sea water is 0.045-0.055 mg/l and Lead concentration in Sediment 33.512-35.726 µg/g dry wt., Cadmium concentration in sea water 0.006-0.010 mg/l, Cadmium concentration in sediment 1.944-2.004 µg/g dry wt., lead concentration is the highest in most fish gill samples 0.047-0.727 µg/g dry wt., where as in muscles is the lowest 0.020-0.116 µg/g dry wt., and in liver was 0.038-0.267 µg/g dry wt. Cadmium concentration is the highest in most fish gill samples 0.033-0.609 µg/g dry wt., where as in muscles is the lowest 0.018-0.073 µg/g dry wt., and in liver was 0.028-0.209 µg/g dry wt.

Conclusion: By comparing the results obtained with other data obtained from the local and international studies, in addition to, comparing the results standard levels of these metals contaminated and adopted internationally and domestically and the pollution levels in Yemen is currently within the lower limits of pollution.

Keywords: Aden, Cadmium, fish, Lead, sediments, water, Yemen.

INTRODUCTION

The pollution of aquatic systems has become a major concern worldwide. There are a variety of sources that will pollute aquatic systems with heavy metals. These include animal matter, wet and dry fallouts of atmospheric particulate matter and human activities. The concentration, bioavailability and toxicity of heavy metals in aquatic systems can be affected by various factors, including pH and temperature. Poor quality of surface water is caused in two ways. The pollution of surface water can either be due to point source (PS) or nonpoint source pollution (NPS). Point source pollution is mainly municipal sewage discharge and industrial wastewater loads. Municipal sewage discharge is from urban or highly residential areas, while industrial wastewater is from a variety of manufacturers. When rainfall or irrigation water runs over land it will carry and deposit pollutants into rivers, lakes and coastal waters. This is seen as nonpoint source pollution. Heavy metals will be distributed between the aqueous phase and bed sediments in aquatic systems. Only a small percentage of the free metal ions stay dissolved in water. The majority of the ions get deposited in the sediment due to adsorption, hydrolysis and co-precipitation of the free ions. As an important component of water environment, sediment is not only the place where pollutants accumulate from the water body, but also it is a secondary pollution source which has a potential impact on water quality. Sediment represents one of ultimate sinks for heavy metals discharged into the aquatic environment. Therefore, sediment quality is a good indicator of pollution in the water column, where it tends to concentrate the heavy metals. Heavy metals are distributed in sediments in four fractions, as exchangeable bound, iron–manganese oxide, organic.
matter and residual species\(^5\). In order to protect the aquatic life community comprehensive methods for identifying and assessing the severity of sediment contamination. Due to the ecological importance and the persistence of pollutants in the aquatic ecosystem, sediments are more appropriate to be monitored in environmental evaluations and understand their potential toxic impacts\(^6\). Sediment pollution, especially from heavy metals, has an important impact on the water environment and a direct potential threat on human and aquatic\(^5\). Fishes represent the peak of consumers in the water system. Fishes have ability to collect these metals in concentrations higher than water and sediments because of feed on organic materials in aquatic environments\(^9\). Fishes have been found to be good indicators of the heavy metal contamination levels in the aquatic systems because they occupy different trophic levels\(^10\). According to there are two main routes of heavy metals exposure: The primary route of intake of these chemicals in fish species is via gill or transport of dissolved contaminants in water across biological membranes and ionic exchange. The secondary route is through the intestine by food or sediment particles with subsequent transport across the gut. The food may also be important source for heavy metal accumulation in fish\(^11\). In aquatic ecosystem, metals are transferred to the fish through food chain that could ultimately affect the health of people consuming this fish. Accumulation of these metals in the bodies of fish affected by different factors such as pH, water hardness and level of pollution in the surrounding water added to the age and physiological situation of fish\(^12\). Industrial and domestic waste containing heavy metals and hydrocarbon accumulate in aquatic food chains as possible to cause acute and chronic damages in fish communities and lead to reducibility to growth and reproduce\(^13\).

Lead, in water, accumulates in the body of fish and other marine organisms and it is eventually ingested by humans who consume these fish and seafood products\(^14\). The presence of Pb in the human body causes damage to the nervous system through several mechanisms. Neuropsychological research over the years has revealed that Pb exposure can result in declines in intelligence, memory, processing speed, comprehension and reading, visuospatial, motor and executive skills. Among the cognitive deficits induced by Pb toxicity, visuospatial deficits appear to be major. Anxiety, depression and phobia can also occur, while outcome, intervention, and rehabilitation results are largely dependent on the level of toxic exposure. There is also a growing evidence of antisoical behaviour linked to early Pb exposure\(^15\). Cadmium in water can be accumulated in the body of marine organisms and can eventually enter the body of humans who consume these seafood products. The concentrations of Cd call for caution as cumulative effects might constitute health hazards to aquatic life including man who feeds on fish\(^16\). In the late 1960s environmental cadmium contamination was established as the cause of an epidemic of bone disease (itai-itai disease) in Japan. Since that time, increasing scientific interest has been devoted to cadmium as an environmental contaminant. Awareness is now been disseminated in some countries concerning the small margin of safety between existing intake levels and levels that may cause adverse health effect to the population\(^17\). Determining the levels of such heavy metals and comparing the levels with guidelines will establish the potential health risk from the consumption of such fish species. Therefore, it is important to determine the concentrations of non-essential metals in fish in order to evaluate the possible risks of fish consumption. This can serve as an indicator for the extent of pollution in Yemen coastal waters.

**MATERIALS AND METHODS**

Water samples and Sediment samples were collected from Winter 2011, Summer 2012 and Winter 2013, from 9 sampling sites (Fig. 1) during which a total of 27 Sample of Surface Seawater, 27 Sample of Sediments, 36 Sample of Muscles Fish, 36 Sample of Liver Fish and 36 Sample of Gills Fish were collected and analyzed.

![Figure 1: Sampling locations (1–9) along the Coast of Aden, Yemen.](image)

Sea water samples were collected using cleaned plastic water sampler. Each sample was taken in 1 liter polyethylene bottles. All water samples were immediately brought to the laboratory where filtered through whatman No.41 (0.45 μm pore size) filter paper. The samples were acidified with 2ml nitric acid to prevent precipitation of metals, reduce adsorption of the analyses onto the walls of containers and to avoid microbial activity, and then stored at 4°C until the chemical analyses. Surface Seawater Digestion for Pb and Cd Analysis by GFAAS, Five milliliter of concentrated HCl was added to 250 ml of each surface seawater sample placed in 600 ml beaker and evaporated to 25 ml volume. The concentrate was transferred to a 50 ml volumetric flask and diluted to mark with deionized water. Prior analysis, the solutions were filtered through Whatman No.41 (0.45 μm pore size) filter paper. Analyzed for Lead (Pb) and Cadmium (Cd) using Buck Model 210 VGP, USA Made -Graphite furnace Atomic Absorption Spectrophotometer (GF AAS) in Seawater samples, before proceeding Method 200.13\(^18\). Sediments samples were collected by a polyethylene corer in accord with standard methods.
Table 1: Operational parameters for Pb and Cd analysis by GFAAS in surface seawater and Fish

| Element       | Pb          | Cd          |
|---------------|-------------|-------------|
| Sample        | Sw+F        | Sw+F        |
| Method        | GFAAS       | GFAAS       |
| Wavelength    | 283.3       | 228.8       |
| T Drying, °C  | 110-130     | 110-130     |
| T Pyrolysis, °C | 1000       | 900         |
| T Atomization, °C | 1900       | 1200        |
| T Clean out, °C | 2500       | 2500        |
| T Ramp, °C    | 1400        | 1300        |
| Matrix modifier | NH₄H₂PO₄+Mg (NO₂) | NH₄H₂PO₄+Mg (NO₂) |
| Slit width (nm) | 0.8         | 1.2         |
| Argon flow (ml min⁻¹) | 250        | 250         |
| Injection volume (µL) | 20         | 20          |
| Instrumental detection limit (µg/l) | 0.02        | 0.005       |

Sw Sea water, F Fish, GFAAS graphite furnace atomic absorption spectrometry.

The procedure for the extraction heavy metals was based on Standard Method 3051A (Microwave – assisted acid digestion of sediments)⁵⁹. About 0.25 g of dry sediment sample was accurately weighed and digested with 6 ml of concentrated nitric acid (HNO₃ 65%) . 1 ml of Perchloric acid (HClO₄ 65%) and 1 ml of hydrogen peroxide (H₂O₂ 30%). Milestone Stard D Microwave Digestion Lab station with internal Temperature sensor and 260 terminal teach screen With HPR1000/105 High Pressure Segmented Rotar (Application Note HPR-EN-33). Microwave Program 2 Steps (1) 15.00 Min (temperature 200°C) (2) 15.00 Min, (temperature 200°C). After Finish left vessels 20 min until reach the room temperature, then the digested portion was diluted to a final volume of 50 ml using deionized water. Pb and Cd Analyzed by GFAAS without further treatment.

RESULTS

**Heavy metals in filtered water surface** - The highest concentration of Pb in filtered water surface of Aden was 0.055 mg/l on winter 2011 and the lowest concentration was 0.045 mg/l on summer 2012. The highest concentration of Cd in filtered water surface of Aden was 0.010 mg/l on summer 2012 and the lowest concentration was 0.006 mg/l on winter 2011 (Table 5).

**Heavy metal in surface sediment** - The highest concentration of Pb in sediments of Aden was 35.104 µg/g (dry wt.) on summer 2012 and the lowest concentration was 33.507 µg/g (dry wt.) on winter 2011. The highest concentration of Cd in sediments of Aden was 2.111 µg/g (dry wt.) on summer 2012 and the lowest concentration was 1.775 µg/g (dry wt.) on winter 2011 (Table 5).

**Heavy metals in muscles fish** - The highest concentration of Pb in the muscles of fish was 0.075 µg/g (dry wt.) on winter 2013 and the lowest concentration was 0.059 µg/g (dry wt.) on winter 2011. Also, the highest concentration of Cd in the muscles of fish was 0.052 µg/g (dry wt.) on Summer 2012, whereas the lowest concentration was 0.024 µg/g (dry wt.) on winter 2011 (Table 5). However, the highest concentration of Pb in the muscles of fish was 0.116 µg/g (dry wt.) on large *E. areolatus* and the lowest concentration was 0.020 µg/g (dry wt.) on small *S. jello*. Also, the highest concentration of Cd in the muscles of fish was 0.073 µg/g (dry wt.) on large...
Heavy metals in liver- the highest concentration of Pb in the Liver of fish was 0.150 µg/g (dry wt.) on summer and the lowest concentration was 0.110 µg/g (dry wt.) on winter 2011 ; on the other hand, the highest concentration of Cd in the Liver of fish was 0.130 µg/g (dry wt.) on winter 2013, whereas the lowest concentration was 0.057 µg/g (dry wt.) on winter 2011 (Table 5). The highest concentration of Pb in the Liver of fish was 0.267µg/g (dry wt.) on large L. mahsena and the lowest concentration was 0.038 µg/g (dry wt.) on small S. jello. Also, the highest concentration of Cd in the Liver of fish was 0.172 µg/g (dry wt.) on Large T. tonggol, whereas the lowest concentration was 0.028µg/g (dry wt.) on small S. jello (Table 6).

**Table 3: Operational parameters for Pb and Cd Analysis by FAAS in Surface Sediment**

| Element | Pb | Cd |
|---------|----|----|
| Sample  | Sediment | Sediment |
| Method  | Flam | Flam |
| Wavelength (nm) | 283.3 | 228.8 |
| Flam mode | C2H2/Air | C2H2/Air |
| Slit width (nm) | 0.7 | 0.7 |
| Lamp current (mA) | 2 | 2 |
| Energy (eV) | 2.874 | 3.214 |
| Instrumental detection limit (µg/l) | 0.08 | 0.01 |

Heavy metals in gill- the highest concentration of Pb in the Gill of fish was 0.290 µg/g (dry wt.) on Summer and the lowest concentration was 0.212 µg/g (dry wt.) on winter 2013. Also, the highest concentration of Cd in the Gill of fish was 0.348 µg/g (dry wt.) on summer, whereas the lowest concentration was 0.092 µg/g (dry wt.) on winter 2011 (Table: 5). The highest concentration of Pb in the Gill of fish was 0.727 µg/g (dry wt.) on small L. mahsena and the lowest concentration was 0.047 µg/g (dry wt.) on small S. jello ; but , the highest concentration of Cd in the Gill of fish was 0.609 µg/g (dry wt.) on small E. areolatus, whereas the lowest concentration was 0.033 µg/g (dry wt.) on small S. jello (Table 6).

**Table 4: Standard Concentration for Metals Analyzed in Surface Sediment**

| Element | Pb | Cd |
|---------|----|----|
| Sample  | Sediment | Sediment |
| Method  | flam | flam |
| Standard | 1.0, 2.0, 4.0, | 0.05, 0.1, |
| Solutions (mg/l) | 8.0 | 0.15, 0.20, |
|           |       | 0.25 |

DISCUSSION

The heavy metals Pb and Cd were noticeably abundant in samples collected in the summer. The abundance of metals in filtered surface water samples is interpreted to be due to amount of draining sewage on summer were higher compared with winter and also due to high water temperature on summer season. The heavy metals Pb and Cd were noticeably abundant in samples collected from sites located near the traffic ways and from areas near sewage discharge. However, our results are in a good agreement with those found by showed that the concentration of Pb was 0.034 ± 0.002 mg/l and Cd was 0.012±0.001 mg/l in water from the Kolleru Lake, India, on summer. Besides, pointed out that the concentration of Pb was 0.03-0.07 mg/l, which is below the permissible limit of 0.1 mg/l set for inland surface water, in the water samples collected from sea water in Międzyzdroje, Baltic coast, Poland.

Heavy Metals in Surface Sediments, The highest concentration Pb and Cd were in the surface sediments of Aden coast was obtained on summer. This result may be explained by the fact that during summer that region faced with decreasing internal currents and water supplies that might have caused increase of heavy metals comparison with winter season, wastewater discharge from the city. The obtained results for Cadmium and their interpretation are comparable to the concentration of Cd was 0.20- 5.80 µg/g (dry wt.) in summer from Red Sea coast, Al Hodeidah, Yemen, and a good agreement with those found by pointed out that the concentration of Pb was 61.620 µg/g (dry wt.) in northern Delta Lakes, Egypt. Heavy metals in Muscles Fish, The highest concentration of Cd in muscles of fish species studied was on summer. The present high concentration of Cd in summer may be attributed to various factors as sewage, increase in the water temperature during summer increases heavy metals uptake in the fishes as compared to the winter season, higher metallic rate in the fishes. The highest concentration of Pb and Cd in muscles of fish of Epinephelus areolatus and Lethrinus mahsena. The present high concentration of Pb and Cd in E. areolatus and L. mahsena have been usually attributed to their habitat and feeding behavior. E. areolatus and L. mahsena tend to be near the sediment region. The present high concentration of Pb and Cd in Aden may be due to the anthropogenic activities, feeding behavior of the fish species, fat content, the in their diet uptake. Some of these values are very small compared with the obtained readings in the current study, however, not be surprising because the marine environment of the ocean is generally less polluting for the sea that are surrounded by industrialized countries, or that are over a lot of ships, such as the Gulf of Aden line. The mean concentrations of Pb and Cd were considered low and under the recommended safe levels set by [FAO/WHO and Yemen standard. However, current results are in a good agreement with those found, showed that the concentration of Pb was 0.10±0.09 µg/g (dry wt.) in muscles of Epinephelus areolatus from Tuticorin, India. Besides, pointed out that the concentration of Pb was 0.20±0.02 µg/g (dry wt.), in L. mahsena from Red Sea, Egypt, pointed out that the concentration of Pb was 0.118- 0.193 µg/g (wt. wt.) and Cd was 0.013- 0.023 µg/g (wt. wt.), in tuna fish Hadramout Coast Yemen pointed out that...
the concentration of Pb was 0.12±0.21 µg/g (dry wt.), in L. mahsena from Red Sea of Al-Cornish Hodeidah Yemen; pointed out that the concentration of Cd was 0.09± 0.010 µg/g (wet wt.), in T. albacares from Aden Yemen;\(^\text{34}\) pointed out that the concentration of Pb was 0.10 - 2.10 µg/g (dry wt.), Cd was 0.06-1.06 µg/g in L. mahsena from Jeddah Coast, Saudi Arabia. Heavy metals in Liver Fish, The highest concentration of Pb and Cd in Liver of fish, were observed to be more concentrated in the larger sizes of fish. The present high concentration of Pb and Cd in the larger sizes of fish may be attributed to various factors as large fish that prey upon smaller fish can accumulate more of the chemical in their bodies. It is better to eat the smaller fish within the same species, the strong affinity of metallothionine protein with these elements. This is usually more pronounced in bigger fishes. The highest concentration of Cd in Liver of fish species studied was on summer.

**Table 5: The mean of concentration (mg/l) for lead and cadmium during the seasons in water, sediments and fish organs (muscle, liver and gill) of Aden station, Yemen coast.**

| Samples          | Metal ion | Seasons       | Total mean±SD |
|------------------|-----------|---------------|---------------|
|                  |           | Winter 2011   | Summer 2012   | Winter 2013 |
| Filtered surface water | Pb        | 0.055±0.004   | 0.045±0.007   | 0.051±0.005 |
|                   | Cd        | 0.006±0.002   | 0.010±0.003   | 0.009±0.001 |
| Sediments        | Pb        | 33.507±1.132  | 35.104±0.416  | 34.688±1.285 |
|                   | Cd        | 1.775±0.201   | 2.111±0.839   | 2.045±0.310 |
| Muscles          | Pb        | 0.059±0.028   | 0.071±0.032   | 0.075±0.035 |
|                   | Cd        | 0.024±0.010   | 0.052±0.021   | 0.041±0.019 |
| Liver            | Pb        | 0.110±0.042   | 0.150±0.096   | 0.144±0.086 |
|                   | Cd        | 0.057±0.026   | 0.126±0.093   | 0.130±0.070 |
| Gill             | Pb        | 0.243±0.225   | 0.290±0.366   | 0.212±0.219 |
|                   | Cd        | 0.092±0.075   | 0.348±0.354   | 0.310±0.296 |

The present high concentration of Cd in summer may be attributed to various factors as differences in local pollution, industrial wastes, bioavailability of metals (variations among physiochemical factors) and fish metabolism (growth cycle, reproduction and feeding). physiological changes, minor role of annual cycles of pH and metal concentration in the water and metal level of the diet in the seasonal pattern of metal concentration in liver. The highest concentration of Pb in Liver of L. mahsena, but, the highest concentration of Cd in the Liver of E. areolatus. The present high concentration of Pb and Cd in L. mahsena and E. areolatus may be attributed to various factors, demersal or bottom-dwelling species, food preferences, organism mobility or other attributes of behavior with respect to the environment, strong binding with cystine residues of MT, lipid content in the tissue and excretion percentage of these toxic metals from their body, increased metabolic rate, water temperature. However, our results are in a good agreement with those found in a previous study\(^\text{8}\) pointed out that the concentration of Cd was 0.6 µg/g (dry wt.) in liver of *Epinephelus fasciatus* from Jordan; and\(^\text{35}\) pointed out that the concentration of Cd was 0.25±0.04 µg/g (dry wt.) in liver of *E. areolatus* from Hong Kong, China.

The highest concentration of Cd was found in Gill of fish on summer. These observations probably as indicated in the case of liver tissue which mentioned above, beside that in water, gills are the main surface during exchange of ions metals. The highest concentration of Pb in Gill of fish of *L. mahsena*, but, the highest concentration of Cd and As in the Gill of fish of *E. areolatus*; on the other hand, the highest concentration of Hg in the Gill of fish of *T. tonggol*. The present high concentration of Pb in *L. mahsena* may be attributed to various factors, the metal complexion with the mucus that is impossible to remove completely from the lamellae before analysis, the similarity of lead and calcium in their deposition and mobilization from the gill, the result of a water contamination caused by environmental pollution. However, our results are in a good agreement with those found in a previous study\(^\text{38}\) showed that the concentration of Cd was 0.12-0.35 µg/g (dry wt.) in Gill of *Mystusseenghala* and *Wallago attu* from Pakistan;\(^\text{39}\) in summer; and showed that the concentration of Cd was 0.30±0.01 µg/g (dry wt.) in Gill of *Scardinius erythrophthalmus* from the Topolitsa reservoir (Bulgaria), on summer; and another study\(^\text{40}\) showed that the concentration of Cd...
was 0.34 ± 0.20 µg/g (wt. wt.) in Gill of of *Cyprinus carpio* from Sikkak dam at Ainoyucef (Wilaya of Tlemcen) Algeria, in summer. Similar results are found in high Pb concentrations in gills were recorded by 36, 41, 42, 43. However, current results are in a good agreement with those found in a previous study, 33 showed that the concentration of Pb was 1.20±0.95-1.70±0.90 µg/g (wt. wt.) in Gill of *Lethrinus lentjan* and Cd was 0.25±0.07 - 0.54±0.10 µg/g (wt. wt.) in Gill of *Epinephelus sexfasciatus* during winter from Red Sea, AL-Hudaydah, Yemen. and 31 showed that the concentration of Pb was 0.23±0.02 µg/g (dry wt.) in Gill of *L. mahsena* and Cd was 0.54±0.07 µg/g (dry wt.) in Gill of *Epinephelus spp* from Red Sea, Egypt. and 44 showed that the concentration of Cd was 0.117±0.087 µg/g (dry wt.) in Gill of *Epinephelus coioides* from northern of Persian Gulf. The present high concentration of Cd in *E. areolatus*; suggesting that their gills could accumulate trace metals from the environment. *E. areolatus* are both omnivorous feeders (the former being a bottom feeder), metabolism, biological and ecological factors such as feeding habits and habitat, have a close relationship with sediment. A possible explanation for this might be to electricity generating stations cooling waters and effluents, sewage disposals and storm waters. Another possible explanation for this is that Scrap-iron store at Labour Island is the most likely source of Pb and Cd in the Seawater, Oil Harbour and municipal sewages are expected sources to be responsible for elevated tissue Cd concentrations in Seawater from Sahel Abyen and Sira Island. This result may be explained by the fact that Aden area was the highest among the other stations. It is assumed that the presence of the area near the strait of Bab Al-Mandab, where the water coming from both the Red Sea and the Indian Ocean mixes and consequently changes its nature, especially during the seasonal monsoons, is responsible for elevated Gill tissue Cd concentrations in the fish inhabiting the Aden area. However, some sewage outflow can be additional anthropogenic sources of Cd in the area. Apparently, Cd contamination gradients exist in the Gulf of Aden waters.

**CONCLUSION**

In conclusion, heavy metals accumulate in various tissues of fish with different amount. Overall, accumulation of metals in muscle was lower than liver and gills. The results present new information on the distribution of these metals in liver, gills and muscle of *Lethrinus mahsena, Thunnus tonggol, Sphyraena jello* and *Epinephelus areolatus*. Generally, this research showed that concentrations of heavy metal in the Aden Coast, Gulf of Aden, Yemen were so far significantly lower than effects range low (ERL) and lower than the maximum permissible concentration for various countries. According to the fish samples analyses, concentrations of heavy metal in fish species tissues were well within the limits set by the (FAO/WHO (2004) and Standard Specification for Yemen, 2006). Recommendations and showed that the fish from investigated region are safety for consumers.

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**AUTHOR’S CONTRIBUTION**

The manuscript was carried out, written, and approved in collaboration with all authors.

**CONFLICT OF INTEREST**

No conflict of interest associated with this work.
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