



Bioaccumulation of Some Hazardous Heavy Metals in Three Edible Fishes Caught from Gorgan Bay, Caspian Sea

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Abstract

A survey was conducted to evaluate heavy metal accumulation in edible fishes caught from Gorgan Bay, as a part of the Caspian Sea ecosystem. Fish samples including mullet, roach, and common carp were just randomly prepared from fishing locations in three different areas of Gorgan Bay including estuary, channel, and Miankale wetland twice the fishing seasons. A total of 135 samples were measured using an atomic absorption spectrometer to 4 heavy metals of arsenic (As), lead (Pb), cadmium (Cd), and copper (Cu) (a total of 540 metal samples). The highest accumulation of As was related to the mullet of estuary, and the lowest contamination was related to the mouth of the channel. In relation to Cd, the highest pollution in all three species related to the fishes caught from the estuary and the lowest accumulation related to the Miankaleh wetland. In the case of metal Pb, results showed the most contamination related to the Mullet caught from Miankaleh wetland and the lowest accumulation related to the roach of the mouth of the channel. In all three fish species, the highest contamination of Cu was in the estuary and the lowest was in the fish caught from Miankaleh wetland. In general, the highest accumulation belonged to all three species located in estuaries. The probable reason can be pouring river pollution to the bay more than wetlands and channel. Due to the enclosure of the Caspian Sea and consequently the Gulf of Gorgan, limiting the fishing times and points to the periods of least incoming pollutants is very helpful for public health, especially food hygiene and fishing from this ecologically sensitive and vulnerable basin.

Keywords Pollution · Heavy metals · Edible fish · Fishing · Caspian Sea

Introduction

Heavy metals, which have relatively higher density than water, have been growing global concerns about environmental pollution in recent years [1]. Human exposure has also increased significantly as a result of the increasing use of these metals in industry, agriculture, and domestic applications [1]. Some of these heavy metals such as cobalt, iron, nickel, magnesium, Cu, manganese, zinc, chromium, and selenium have functional roles in the body and are essential

for various physiological and biochemical functions of the body [2]. However, high amounts of these metals can be harmful to the body, while some other heavy metals such as Cd, As, Pb, and silver, even in small amounts, have negative effects on the body [2].

Numerous physical, chemical, and biological factors are effective in the accumulation of heavy metals in tissues, including environmental parameters (temperature, salinity, pH, dissolved oxygen, concentration of other chemicals), type of pollutant (organic or inorganic), pollutant properties (hydrophilic or lipophilic and degradation resistance), biological parameters (such as evolution, age and sex, reproductive status and available nutrition), and bioavailability (content of exposure to pollutants, chemical inputs, transport mechanisms, and level of contamination) [1]. Due to their ability to accumulate in different tissues and non-degradability, as well as resistance to biological changes, even at low concentrations, heavy metals can be present throughout the food chain upon entering the environment. They are able to be stored gradually in the tissues of consumers,

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thereby causing many risks, toxic, acute, and chronic and even adverse genetic effects [3].

The closed ecosystem of the Caspian Sea made it residence of unique animals and plants, but at the same time made it very vulnerable to agricultural, industrial, and oil pollution [3]. Adjacent to the cities, various industries, agricultural fields, and the rivers reach the Caspian Sea, which contain suspended solids and sediments, which cause many pollutants such as toxins and chemical fertilizers, municipal and industrial effluents, and metal accumulation.

Gorgan Bay (approximate length 60 km and maximum width 12 km) is a triangular and semi-enclosed bay at the southeastern end of the Caspian Sea on the coastline of Golestan province in Iran. The bay was formed during the Holocene/New Caspian geological period by a sandy promontory called the Miankaleh Coastal Fence System [4]. The length of the bay is about 70 km and in the west–east direction and its width is between 13 and 14 km, its watershed area is more than 3000 square kilometers [5].

Fishing activities are performing throughout this vulnerable ecosystem, and there are some valuable species consumed by humans. Therefore, this study was conducted on evaluation of some heavy metal accumulation in edible fish caught from Gorgan Bay as a part of the Caspian Sea ecosystem.

Materials and Methods

Preparing Fish Sample and Fishing Location

Three points of Gorgan bay were selected as experimental fishing areas, including estuary, channel, and Miankale. According to the literatures, we chose three different species which have been frequently consumed by humans. These three edible fishes consist of mullet, roach, and common carp. Sampling was done twice a period of fishing season by random. Fifteen samples of each selected species were prepared from every fishing location through putting them inside an ice box and transferred to the laboratory for heavy metal measurements.

Laboratory Activities and Heavy Metal Measurement

For preparing samples, they were washed with double-distilled water in the laboratory, and after recording total length and weight, their muscle was cut with a ceramic knife. The samples were placed in a freezer to be dried. After drying, they were stored in polyethylene bags until acidic digestion [6]. The method proposed in MOOPAM was used to digest the samples [7].

According to this method, for measuring heavy metals, 0.3 g of each prepared sample was poured into polypropylene tubes and 4 ml of nitric acid was added to tubes for 1 h at room temperature; then, in a hot plate at 90 °C, digestion was performed for 3 h. Then, 5 ml of nitric acid was added and put at room temperature overnight and then put in a hot plate for 3 h for digestion at 90 °C. Finally, the samples were added to distilled water and 1 ml of 2% potassium dichromate to a final volume of 50 ml [8]. All metals were measured by atomic absorption spectrometry (AAS).

Data Statistical Analysis

SPSS 24 software was used for data analysis. The normality of the data was determined by Shapiro–Wilk and Kolmogorov–Smirnov tests. One-way analysis of variance (ANOVA) was used to examine the presence or absence of significant differences in the concentration of heavy metals among different fishes.

Results

Arsenic (As)

The average amount of As in the muscle of the three fishes is shown in Fig. 1 in terms of micrograms per kilogram of fresh weight (ppb). Statistical analysis showed that the amount of As in the three species and the different sampling areas were significantly different ($P < 0.05$); among the fishes, mullet (9.40) showed the lowest accumulation, but the highest accumulation appeared in mullet (112.6). In general, the highest amount of As was in mullet and the lowest in roach. The results of As measurements in fish samples belonging to the fishing areas revealed that the most As concentration was within samples from estuary and the lowest in the mouth of the channel (Fig. 1).

Cadmium (Cd)

The average amount of cadmium is shown in Fig. 2 in terms of micrograms per kilogram of fresh weight (ppb). Statistical analysis of the results shows that the amount of Cd in the three studied species and different sampling areas is significantly different ($P < 0.05$). The mullet and carp (205 ppb) captured from the estuary showed the highest and the roach of Miankaleh (50 ppb) showed the lowest Cd. In general, the amount of Cd was the highest in carp and the lowest in roach. Also, in the studied areas, the highest Cd concentration was observed in fishes sampled from estuary and the lowest from the Miankaleh area (Fig. 2).

Fig. 1 Mean As ($\mu\text{g}/\text{kg}$) in muscle tissue of common carp, mullet, and roach in different areas of Gorgan Bay

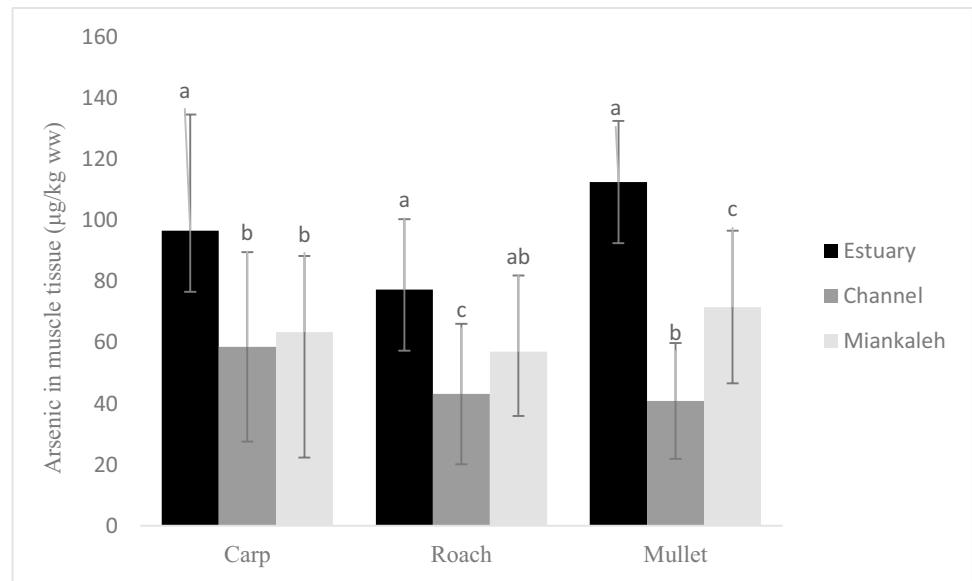
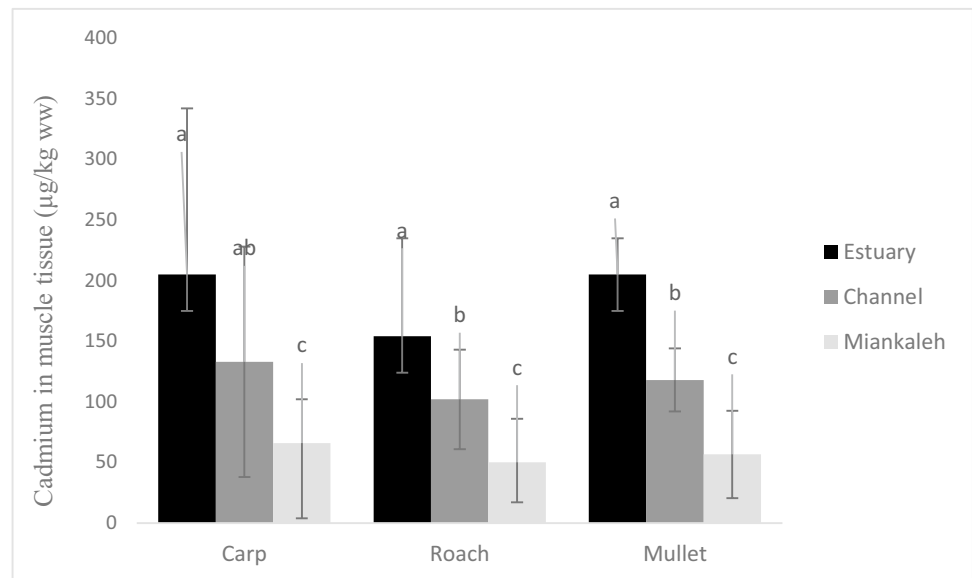


Fig. 2 Mean Cd ($\mu\text{g}/\text{kg}$) in muscle tissue of common carp, mullet, and roach in different areas of Gorgan Bay



Copper (Cu)

The average amount of Cu in terms of ppb of fresh weight in the muscle of three species from capture regions of Gorgan Bay is presented in Fig. 3. Statistical analysis of the results shows that the amount of Cu in the three species and different sampling areas is significantly different ($P < 0.05$); the highest Cu contamination belonged to the mullet of the estuary (2106 ppb) and the lowest concentration evaluated in mullets captured from Miankaleh (509 ppb). Throughout the studied areas, the highest Cu concentration was measured in the samples from estuary and the lowest from Miankaleh area (Fig. 3).

Lead (Pb)

Pb concentration within the muscle of fishes caught from the three different regions of Gorgan Bay is shown in Fig. 4. Statistical analysis showed that the amount of Pb in the three species and different sampling areas was significantly different ($P < 0.05$); the highest and lowest Pb pollution was related to the mullet captured from Miankaleh (768 ppb) and roach captured at the mouth of the channel (406 ppb), respectively. The amount of Pb was the highest in mullet and the lowest in carp samples. The most Pb concentration was measured within samples from the Miankaleh area and the lowest from the channel (Fig. 4).

Fig. 3 Mean Cu ($\mu\text{g}/\text{kg}$) in muscle tissue of common carp, mullet, and roach in different areas of Gorgan Bay

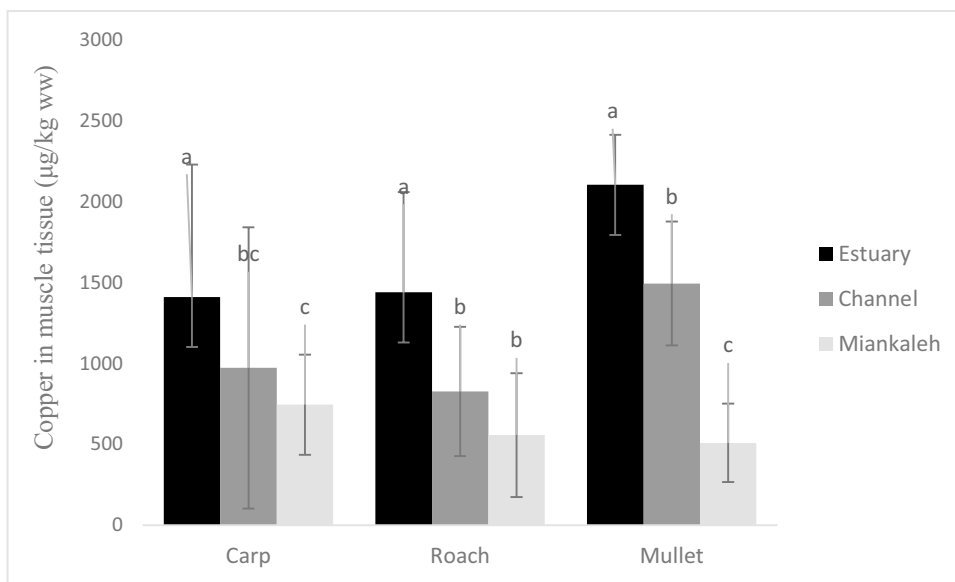
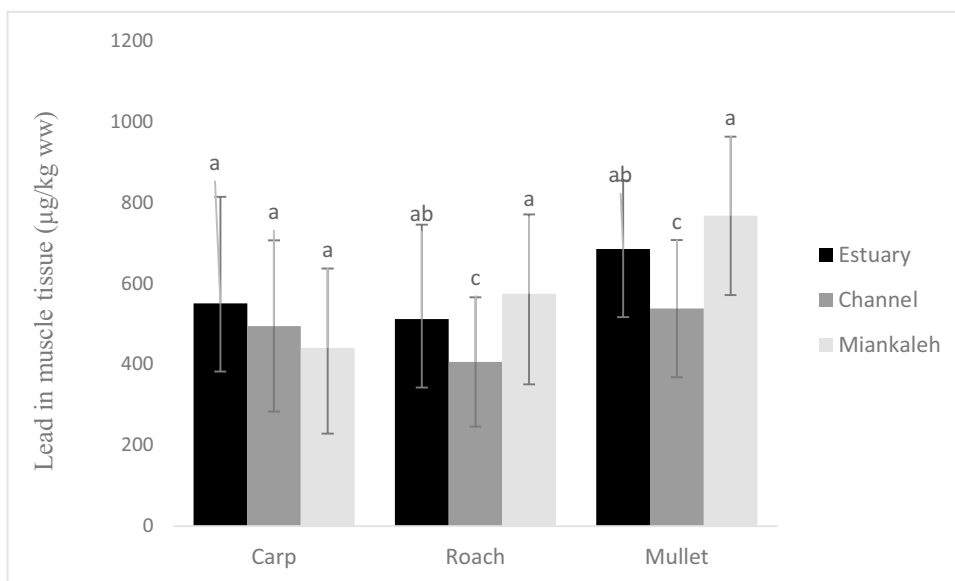


Fig. 4 Mean Pb ($\mu\text{g}/\text{kg}$) in muscle tissue of common carp, mullet, and roach in different areas of Gorgan Bay



Discussion and Conclusion

One of the fishing points in this study was Miankaleh wetland, which is located in the southern part of the Caspian Sea and is a closure ecosystem. As a result of geographic situation, the residence time of various pollutants is high at such ecosystem. What is more, Pb is found in petroleum compounds and municipal and agricultural wastewaters, which are abundant in adjacent areas of Miankaleh, and their final destination is wetland. On the other hand, in this study, the most bioaccumulation of Pb was recorded from this area. Miankaleh is the attractive location for hunters, so bullet casings could be the reason of some part of Pb pollution. Also, motor boat traffic in Miankaleh caused oils and

fuel releasing Pb. Salehi Borban et al. [9] evaluated the accumulation of six heavy metals (Cd, As, Pb, Cu, iron, and mercury) in three edible species, including Kutum, Kilka, and mullet, caught from Mahmoud Abad region. Pb amount of fish muscle consisted of mullet (0.3593) > Kilka (0.2707) > Kutum (0.02497) ppm, respectively. Comparison of Pb levels in the muscles of the studied species with the global standards WHO, FAO, NHMRC, UKMAFF, and EC shows that the level of Pb accumulation in fish muscles was more than the EC and lower than other standards [9]. In the present study, the evaluation of heavy metals among species showed that the most contaminated species was mullet. This fish is a benthic and omnivorous one which feeds on plant matter and algae and sediment ingredients. The life style of mullet makes it to be exposed to heavy

metals accumulated in the sediments of the aquatic environment more than two other studied species. Pb accumulation in all caught fish is less than WHO, MAFF, and NHMRC standards, and all are higher than EC and EU standards. Except carp and roach caught from the mouth of the channel and carp caught from Miankaleh wetland, Pb accumulation in other fishes was higher than FAO and FAO/WHO standards.

Regarding the accumulation of Cd metal in the studied fishes, the amount of metal accumulation is less than the standards provided by WHO and FAO/WHO. With the exception of fish caught from Miankaleh wetland, Cd accumulation in other fish is higher than EU standard. In carp and mullet caught from the estuary, Cd accumulation is higher than MAFF and NHMRC standards, but in other samples, it is less than these two standards. In general, Cd contamination in fish caught from estuaries is more than others.

Cd has been known as one of the most toxic pollutants, because of its ability of interference in a variety of activities of organs and tissues with short- or long-term exposure [10]. This element has especial properties which causes its wide use in several industrial processes, such as zinc and Pb mining, zinc refining process, and smelting. Furthermore, Cd is frequently used in lamp working, coloring borosilicate glass, in manufacturing of nickel-Cd batteries, as a stabilizer in PVC products, as an anti-corrosive agent in electroplating (Cd oxide), as electrodes in batteries, as a coloring agent, as catalysts, in semiconducting solar panels (as Cd telluride), in infrared optical windows, in ceramic glazes, in phosphate fertilizer production, etc. [6]. Sattari et al. [11] showed the highest accumulation of Cd in Kutum (0.08), Kilka (0.042), and mullet (0.06), respectively. Comparison of Cd in the muscle of the three species with the mentioned standards showed that the amount of Cd in all three species was higher than the EC standard and in this respect is similar to the As in the present study because in the present study, As accumulation has been higher than the EC standard.

The amount of Cu in study by Burban et al. [12] was as Kutum (1.2156) > mullet (0.8011) > Kilka (0.64) and was less than the international standards; like in the present study, Cu was less than the existing standards. In the case of Cu, the accumulation rate in all fish was lower than the international standards, and fish caught from the estuary were more contaminated than others.

In this investigation, the most level of As was detected in fishes caught from estuary points. Previous literatures revealed that accumulation and proportion of As in fish muscles are affected by its concentration throughout the aquatic ecosystem, trophic level, and feeding habitat of fish (y.[13]. Therefore, the estuary accumulated As more than other stations, and mullet is a more susceptible species to environmental As accumulation. Because the

International Agency for Research on Cancer identified As as a first class of carcinogen for humans [14], monitoring of this heavy metal in what humans consume, whether beverages or food productions, could be helpful to public health and especially food hygiene. Ashja Ardalan et al. [15] monitored Pb and Cu in the muscle of Haji Tarkhan perch (*Perca fluviatilis*) of Anzali wetland comparing the results of the present study cleared that all are in almost the same numerical range, and there is not much difference in the results obtained from the accumulation of these metals. In both studies, Cu was higher than Pb, all of which were lower than international standards (Ashja [15]. The difference in results might be related to the difference in location and species studied.

Accumulation of Cu and Pb was monitored in the muscle tissue of Persian sturgeon and stellate from the southern basin of the Caspian Sea; the maximum levels of Cu and Pb in both species were slightly higher than the maximum accumulation of these two heavy metals in the present study, but all of them are less than the limits of international standards [16]. Cu is an essential element, but it can also pose a potential risk. Differences in Cu accumulation in different species may be due to differences in the type of species, water temperature, sampling site, and aquatic feeding habits [17].

The ecology of Gorgan Bay is influenced by the Caspian Sea, rivers, and Miankaleh wetland [18]. As mentioned, the accumulation of heavy metals As, Cd, and Cu in two species of mullet and roach and also the accumulation of Pb in carp in the estuary sampling area were the highest. The estuary is the final part of the river that develops and enters the seas or bays and is the confluence of salt and fresh water. The highest accumulation of metals in this area (estuary) indicates that this sampling area is more polluted than the other two points. According to the studies, there are a relatively large number of agricultural and rice fields, fish, and poultry centers and various industries such as dyeing industries around the Gulf of Gorgan [19].

The vast majority of industrial and production units contain industrial wastewater such as As and Pb, which do not have a treatment system, and daily, large amounts of wastewater enter the water sources directly or through the municipal wastewater network. Also, herbicides and phosphate fertilizers used in the agricultural industry contain large amounts of Cd, and fungicides and chemical fertilizers used in rice fields contain large amounts of the heavy metal As. Pollution of these metals through rainfall, atmospheric pollution, non-treatment of industrial wastewater, and their direct discharge into the rivers leading to the bay can be the cause of maximum accumulation and pollution of metals in the sampling area [5]. It seems that the reason for the fact that the contamination of mullet with heavy metals is more than the other studied species is the location of Mullet at higher bay level, because environmental pollution and

industrial and municipal wastewater are more concentrated in these areas.

In conclusion, the highest accumulation of evaluated heavy metals in all three investigated species belonged to the estuary area, and the reason could be that more surge pollution from rivers enters the bay than the wetlands and channel mouth. Due to the enclosure of the Caspian Sea and consequently the Gorgan bay, the shelf life of various incoming pollutants is very long, and the purification of pollutants is slow; consequently, the Gorgan bay basin is ecologically sensitive and vulnerable for aquatic animals. According to the results, it seems that Pb would be a concern of aquatic health and consequently human health in the future. Furthermore, mullet is the most influencing species by heavy metal pollution, due to its feeding habitat and niche occupying. Identifying the correlation among sediment, water, and edible fish muscle concentration is recommended for further research to help provided public health.

Author Contribution Toktam Bagheri: methodology, Ali Misaghi: formal analysis, Ali Taheri Mirghaed: investigation, Abolfazl Kamkar: data curation. Aliakbar Hedayati: writing-review and editing.

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Data Availability Data will be made available upon request.

Declarations

Ethics Approval All experimental procedures related to the fish were approved by the Animal Care and Ethical Committee, which were in accordance with ethical standards in Laboratory animal-guideline for ethical review of animal welfare.

Consent to Participate Not applicable.

Consent for Publication Not applicable.

Conflict of Interest The authors declare no competing interests.

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