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Lead, Cadmium, and Mercury Concentration Levels in Some Commercial Fish From the Arabian Sea and the Gulf of Aden During the Years 2020–2022

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Cover Page Footnote

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Lead, Cadmium, and Mercury Concentration Levels in Some Commercial Fish From the Arabian Sea and the Gulf of Aden During the Years 2020–2022

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Abstract: This study was conducted to determine and evaluate the heavy metal concentrations levels: lead (Pb), cadmium (Cd), and mercury (Hg) in the meat of some commercial fish species: Grouper (*Epinephelus areolatus*), Emperor (*Lethrinus nebulosus*) and Cuttlefish (*Sepia Pharaonis*). A total of 84 fish samples were caught and collected from different sites in the Gulf of Aden and the Arabian Sea between January 2020 and December 2022. Some of them were sold in the cities markets of Hadhramout governorate- Yemen, and some of them were exported abroad. All samples were analyzed at the laboratories of the Faculty of Environmental Sciences and Marine Biology- Hadhramout University, where atomic absorption spectrometer was used in the process of analyzing elements, following the generally accepted scientific methods. The results of the study showed that the mean concentrations of lead, cadmium, and mercury in the Grouper samples were (0.1225, 0.0101, and 0.0098 ppm) respectively. In Emperor the mean concentrations of lead, cadmium, and mercury reached (0.1228, 0.0108, and 0.0084 ppm) respectively, while in Cuttlefish, lead, cadmium, and mercury reached (0.1220, 0.0143, and 0.0063 ppm) respectively. All these values remained even lower than the minimum permissible limits, whether according to Yemeni or European standards. The results of one-way analysis of variance (ANOVA) did not show any significant difference in lead concentrations among study's fish ($P>0.05$), whereas, it indicated that there were significant differences between cadmium and mercury. Among years, (ANOVA) did not indicate any significantly difference ($P>0.05$) in cadmium concentration, whereas, there were significant difference ($P<0.05$) in lead and mercury. This study concluded by providing some important recommendations to local authorities and competent scientific centers in this field.

Keywords: Cadmium, Fish, Lead, Mercury, Yemen

1. Introduction:

Lead (Pb), cadmium (Cd), and mercury (Hg) and other elements such as chromium and arsenic are considered heavy metals and that have been the most common heavy metals that induced human poisonings [1]. Heavy metals are defined on the basis of some criteria, such as density and atomic weight. If we take the density standard as a basis, then heavy metals are a group of elements that have density values greater than 5g/cm^3 [2]. Heavy metals are usually

found in trace amounts in the earth's crust, ranging from a few parts per trillion (ppt) for noble metals to up to 5% for iron. They could be encountered in their elemental, metallic form. Weathering and erosion led them to leach into the soil, rivers, and groundwater [3].

The industrial progress has increased the heavy metal emissions from anthropogenic activities. And as a consequence, a series of incidents related to heavy metal pollution such as mercury pollution inducing Minamata

disease, cadmium pollution which caused Itai-Itai disease, and poisoning induced by lead and [4].

Many researches and studies were carried out [5, 6, 7, 8, and 9] that dealt with heavy metals in details, their sources, concentrations, importance, and negative effects on human health and biota in general. The researchers noted that presence of varying concentrations of heavy metals can bioaccumulate, biomagnify, and may lead to health problems to consumers. Also they have indicated that some heavy metals which were found in marine organisms, caused cancer, heart and blood vessel diseases, kidneys, nervous system and bone diseases. All of this, certainly creates many problems in the fishery sector, especially for those countries that consider fish wealth one of the pillars of the economy.

The fishery sector in the Republic of Yemen is considered as one of the main important sectors of the Yemeni economy and actively participates in the process of sustainable development. The latest data published by the Ministry of Fisheries in 2012 showed that the fish sector contributed about 3% of the gross domestic product, and constituted the second largest source of export revenues after oil [10].

Yemen has a maritime border of about 2,500 kilometers, rich with different fish species and various marine life, which are among the finest types of fish in the region. The total fish exports for the year 2004 reached 61,823 tons, with an estimated value of about 83 million dollars [11]. Compared to 2007, in which fish exports of Yemen Republic were about 81 thousand tons, in the end of 2008 exports increased to 105 thousand tons with a total value of about 235 million US dollars [11]. According to the Ministry of Fisheries, different species of fish as: Emperor, Grouper, Blacktip trevally, Yellowfin tuna, Longtail tuna, Cuttlefish, and Shrimp were exported to many countries around the world [12]. 86 companies are still working in the field of export, 18 of which have obtained European licenses, the most important of which were Al-Tamimi, Broome, Gulf of Aden and Ben Sodon [12].

Until now, it is believed that the fish resources in Yemen has not been affected by pollution due to the absence of sources of pollution from heavy industries or the chemical and petrochemical industries, whose wastes are often thrown into the sea. However, the irresponsible pollution of the territorial waters due to passing ships and throwing their wastes into the marine environment along costal line of Yemen, is considered as a challenge that faces the country. It is known that Yemen does not have the capabilities to monitor its coastline extending over long distances. With the global increase in heavy metals in the environment and the threat they pose to human health in recent years [4], ensuring that fish stocks are safe and free of pollutants, especially heavy metals, is considered an important vital issue.

1.1 Study objectives:

Determination of lead (Pb), cadmium (Cd), and mercury (Hg) concentration levels (bioaccumulation) in some species of exported commercial fish, including grouper (*Epinephelus areolatus*), emperor (*Lethrinus nebulosus*), and cuttlefish (*Sepia pharaonis*), between 2020 and 2022.

- Comparison of the results with Yemeni and European standards and previous studies.
- Development of proposals and recommendations that will preserve the environment from pollution.

The importance of this study lies on the fact that it will clarify the state of the marine waters of the country, as well as it will give a clear picture of the quality of fish resources and marine organisms and their pollution with heavy metals.

2. Materials and methods:

2.1 Tools preparation:

All tools, glassware and polyethylene containers were cleaned and dried using a standardized procedure to remove any contaminants that could interfere with the analysis. The procedure consisted of soaking the glassware in a solution of concentrated hydrochloric acid (HCl) (Gainland Chemical Company, UK) and distilled water for 2 hours, followed by washing with 10% nitric acid (HNO₃) (Sigma-Aldrich, Germany) and distilled water. The polyethylene containers were washed with deionized water and dried in an oven.

2.2 Sampling plan:

A total of 84 meat samples were collected from three commercial fish species: Grouper (*Epinephelus areolatus*), Emperor (*Lethrinus Lentjan*) and Cuttlefish (*Sepia Pharaonis*) from many sites in the Gulf of Aden and the Arabian Sea between January 2020 and December 2022. The fish were caught with the help of local fishermen using a variety of methods, including bottom trawling, gillnetting, and long lining. The samples were immediately placed in polyethylene bags and stored in an ice box. After they were brought to the laboratory at Faculty of Environmental Sciences and Marine Biology - Hadramout University, the fish were washed with deionized water and cut into fillets (for Grouper& Emperor) and mantle (for Cuttlefish) [13]. The fillets and mantle were then weighed and placed in individual polyethylene bags. The bags were labeled with the species, date, and location of collection, and then kept in a freezer at -20°C until analysis [13].

2.3 Samples Preparing:

The fish fillets and mantle were homogenized in a blender and then dried in an oven at 80°C until the sample weight was constant. Dried samples were weighing and transferred to a digestion flask and a digestion solution was added. The digestion solution consisted of 25% nitric acid (HNO₃ 65%), 75% hydrochloric acid (HCl 37%), and 1% hydrogen peroxide (H₂O₂ 30%). The flasks were heated on a hot plate until the samples were completely digested. The digested samples were cooled and transferred to a volumetric flask. The samples were diluted to the mark with deionized water [14].

2.4 Samples analysis:

The levels of heavy metals in the samples were analyzed using an atomic absorption spectrometer (AAS) from The Buck Scientific Company, UAS Made, model (flame system 210 and combination batch cold vapor generator). The AAS is a device that measures the amount of a particular element in a sample by measuring the amount of light absorbed by the element. Lead and cadmium were analyzed using flame atomic absorption spectrometry (FAAS) by method [14],

while mercury was analyzed using cold vapor atomic absorption spectrometry (CVAAS) by method [15].

2.5 Statistical Analysis:

Statistical analyses were employed to assess data on the heavy metals accumulated in fish tissues comparing various species and between years. One-way analysis of variance (ANOVA) and Post Hoc tests (LSD) were the methods used for study. For statistical confidence, $\alpha = 0.05$ was chosen. Using SPSS version 20, a statistical analysis of the data was carried out.

3. The Results and discussion:

Tables (1-3) show all the results of analyzing fish samples. It should be noted that in some months we were unable to obtain the results (not available-NA) of the fish studied samples for several reasons, including the samples not arriving at the laboratory due to the conditions in the country (Yemen is in war), the testing device being out of service, or the unavailability of chemicals in the markets. It has also been observed in recent years that some types of fish become available in certain months and disappear in the same months in other years.

Table1. Concentrations of Pb, Cd, and Hg in Grouper during the years 2020-2022, ppm

| Year | 2020 | | | 2021 | | | 2022 | | |
|---------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| | Pb | Cd | Hg | Pb | Cd | Hg | Pb | Cd | Hg |
| January | 0.108 | 0.019 | 0.008 | 0.092 | 0.011 | 0.018 | 0.111 | 0.008 | 0.005 |
| February | 0.083 | 0.008 | 0.011 | 0.111 | 0.005 | 0.013 | 0.132 | 0.009 | 0.013 |
| March | 0.131 | 0.011 | 0.003 | 0.099 | 0.011 | 0.019 | 0.142 | 0.013 | 0.005 |
| April | 0.162 | 0.013 | 0.008 | 0.102 | 0.007 | 0.018 | 0.147 | 0.010 | 0.005 |
| May | 0.192 | 0.013 | 0.005 | NA | NA | NA | 0.133 | 0.017 | 0.005 |
| June | 0.125 | 0.012 | 0.006 | 0.095 | 0.013 | 0.005 | 0.137 | 0.012 | 0.011 |
| July | 0.172 | 0.013 | 0.008 | 0.122 | 0.008 | 0.005 | NA | NA | NA |
| August | 0.185 | 0.009 | 0.014 | 0.065 | 0.006 | 0.014 | NA | NA | NA |
| September | 0.188 | 0.015 | 0.019 | 0.089 | 0.008 | 0.012 | 0.119 | 0.005 | 0.012 |
| October | 0.089 | 0.016 | 0.010 | 0.159 | 0.015 | 0.014 | 0.100 | 0.010 | 0.014 |
| November | NA | NA | NA | NA | NA | NA | 0.086 | 0.009 | 0.005 |
| December | NA | NA | NA | NA | NA | NA | 0.076 | 0.011 | 0.008 |
| Average | 0.144 | 0.012 | 0.009 | 0.104 | 0.009 | 0.013 | 0.118 | 0.009 | 0.008 |
| Average Pb over the three years | | | | | | 0.121 | | | |
| Average Cd over the three years | | | | | | 0.010 | | | |
| Average Hg over the three years | | | | | | 0.010 | | | |

NA – not available- In the sense that this species of fish was not available for analysis at that time

Table2. Concentrations of Pb, Cd, and Hg in Emperor during the years 2020-2022, ppm

| Year | 2020 | | | 2021 | | | 2022 | | |
|-----------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| | Pb | Cd | Hg | Pb | Cd | Hg | Pb | Cd | Hg |
| January | 0.111 | 0.007 | 0.005 | NA | NA | NA | 0.105 | 0.008 | 0.013 |
| February | 0.132 | 0.008 | 0.005 | 0.131 | 0.015 | 0.016 | 0.081 | 0.013 | 0.009 |
| March | 0.188 | 0.015 | 0.008 | 0.151 | 0.008 | 0.017 | 0.088 | 0.013 | 0.005 |
| April | 0.131 | 0.017 | 0.008 | 0.078 | 0.012 | 0.005 | 0.132 | 0.008 | 0.011 |
| May | NA | NA | NA | 0.166 | 0.007 | 0.015 | 0.145 | 0.008 | 0.005 |
| June | 0.183 | 0.017 | 0.003 | 0.152 | 0.014 | 0.005 | NA | NA | NA |
| July | 0.085 | 0.015 | 0.005 | 0.106 | 0.005 | 0.005 | 0.132 | 0.011 | 0.009 |
| August | 0.133 | 0.014 | 0.005 | 0.129 | 0.008 | 0.012 | NA | NA | NA |
| September | NA | NA | NA | NA | NA | NA | NA | NA | NA |
| October | 0.111 | 0.012 | 0.005 | NA | NA | NA | 0.154 | 0.011 | 0.008 |

| Year | 2020 | | | 2021 | | | 2022 | | |
|---------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| | Pb | Cd | Hg | Pb | Cd | Hg | Pb | Cd | Hg |
| November | 0.156 | 0.018 | 0.006 | 0.097 | 0.009 | 0.014 | 0.100 | 0.008 | 0.014 |
| December | 0.152 | 0.016 | 0.008 | 0.102 | 0.018 | 0.015 | 0.089 | 0.010 | 0.008 |
| Average | 0.138 | 0.013 | 0.006 | 0.124 | 0.011 | 0.012 | 0.105 | 0.009 | 0.008 |
| Average Pb over the three years | | | | 0.122 | | | | | |
| Average Cd over the three years | | | | 0.011 | | | | | |
| Average Hg over the three years | | | | 0.008 | | | | | |

NA – not available- In the sense that this species of fish was not available for analysis at that time

Table3. Concentrations of Pb, Cd, and Hg in Cuttlefish during the years 2020-2022, ppm

| Year | 2020 | | | 2021 | | | 2022 | | |
|---------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| | Pb | Cd | Hg | Pb | Cd | Hg | Pb | Cd | Hg |
| January | 0.187 | 0.010 | 0.007 | 0.088 | 0.018 | 0.009 | 0.152 | 0.015 | 0.006 |
| February | NA | NA | NA | NA | NA | NA | 0.155 | 0.015 | 0.009 |
| March | NA | NA | NA | NA | NA | NA | 0.178 | 0.018 | 0.008 |
| April | NA | NA | NA | NA | NA | NA | 0.146 | 0.015 | 0.008 |
| May | NA | NA | NA | NA | NA | NA | 0.142 | 0.008 | 0.006 |
| June | 0.165 | 0.013 | 0.005 | 0.111 | 0.015 | 0.005 | 0.133 | 0.010 | 0.008 |
| July | 0.100 | 0.017 | 0.008 | 0.100 | 0.017 | 0.008 | 0.172 | 0.013 | 0.008 |
| August | 0.142 | 0.017 | 0.005 | 0.142 | 0.017 | 0.005 | 0.133 | 0.015 | 0.005 |
| September | 0.089 | 0.017 | 0.005 | 0.098 | 0.015 | 0.005 | 0.100 | 0.017 | 0.008 |
| October | 0.133 | 0.015 | 0.006 | 0.133 | 0.015 | 0.006 | NA | NA | NA |
| November | 0.088 | 0.015 | 0.005 | 0.088 | 0.015 | 0.005 | 0.087 | 0.014 | 0.005 |
| December | 0.078 | 0.009 | 0.005 | 0.078 | 0.009 | 0.005 | 0.075 | 0.013 | 0.005 |
| Average | 0.123 | 0.014 | 0.006 | 0.105 | 0.015 | 0.006 | 0.134 | 0.014 | 0.007 |
| Average Pb over the three years | | | | 0.120 | | | | | |
| Average Cd over the three years | | | | 0.014 | | | | | |
| Average Hg over the three years | | | | 0.006 | | | | | |

NA – not available- In the sense that this species of fish was not available for analysis at that time

Yemeni Organization for Standardization, Metrology and Quality Control has set the permissible standards for heavy metals in frozen fish [16]. It indicated that the permissible limit for lead is 1 ppm, while cadmium and mercury are 0.5 ppm each. European standards were more detailed. Where the permissible limit for heavy metals depends on the species of fish, so the permissible limit for lead was 0.3 ppm (for muscle meat of fish) to 1 ppm (for Bivalve mollusks). Cadmium ranged from 0.05 (for the most of muscle meat of fish) to 0.3 ppm (for Bivalve mollusks and Cephalopods). Mercury ranged from 0.5 to 1 ppm (for wide range of fishery products and of muscle meat of fish) [17]. It is clear that there are differences between the Yemeni and the European standards, as the European standards are more stringent than the Yemeni ones regarding lead and cadmium. For mercury, Yemeni standards are lower. Therefore, the minimum standard would be taken into account when analyzing the results. The reasons

for using European standards is that these fish species are often exported to European markets.

3.1 Lead (Pb):

The results of the analysis showed that the lead concentrations in the studied samples Grouper, Emperor and Cuttlefish ranged between 0.065- 0.192 ppm, with average value 0.1215 ppm over the three years 2020-2022 (Table1-3). The lowest lead concentration was 0.065 ppm in August of the year 2021 and was found in Grouper sample, while the highest concentration was 0.192 ppm in May of the year 2020, and also was found in the same type of fish.

Yemeni standards set the permissible limit for lead in frozen fish at 1 ppm, while the European standards are 0.3 ppm. Therefore, the European standard was taken for comparison because we previously indicated that the lower standard, whether Yemeni or European, will be the decisive factor in judging the extent of samples contamination. So when the results were compared, whether individual or the

average concentrations, with European standards, we found that they remained within the permissible limits (Figure-1). This is a good indicator for Yemeni marine waters and confirms that it is free from lead contamination. Also these results were consistent with other studies [18, 19, and 20].

It should only be noted here that the results of this study, although within the permissible limits, indicate that, when compared to the results of studies Al-Qadasy et al. (2018) [18], Obaidat et al. (2015) [19] and Al-Dohail et al. (2020) [20], an increase in lead concentrations was observed by 18%, 17%, and 98%, respectively. The reason might be the pollution of seas and oceans with various pollutants due to increasing human activities in recent years. Here, it is important to continuously monitor and periodically examine

marine samples and ensure that they are free of lead. It is known that Lead is a toxic metal and has the ability to accumulate and has negative effects on living organisms even in very low concentrations [21]. Lead levels can decrease in water due to absorption of suspended solids and sedimentation to the bottom, and due to its ability to accumulate in aquatic organisms [22]. WHO, in 2016 reported that about 1.3 million people die annually as a result of exposure to some chemical pollutants, one of these pollutants is lead [23]. About 540,000 people die annually as a result of exposure to lead, millions are left disabled and then die by the long-term health consequences of lead exposure [23].

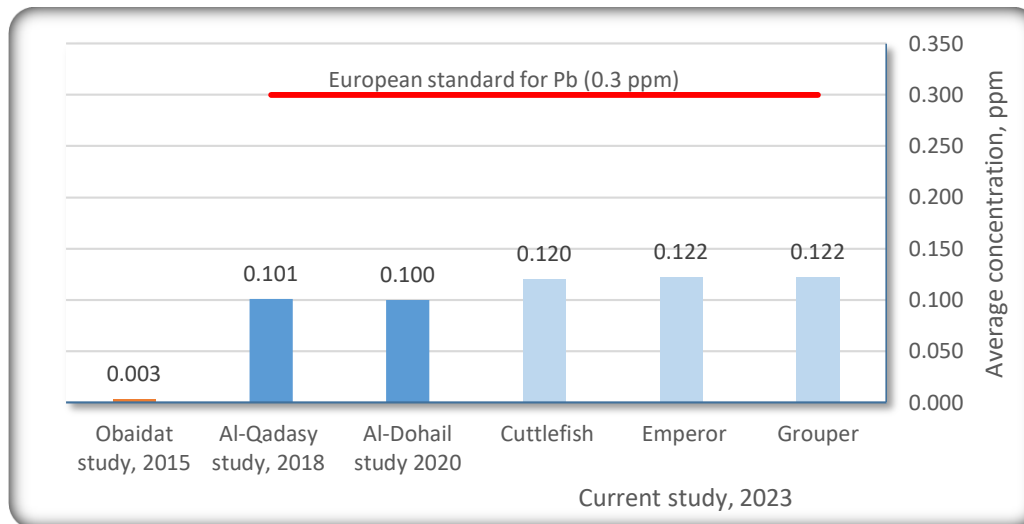


Figure 1. Pb concentrations in fish species and their comparison with standards and other studies

3.2 Cadmium (Cd):

The results of the analysis showed that the cadmium concentrations in the studied samples (Grouper, Emperor and Cuttlefish) ranged between 0.005 and 0.019 ppm, with average value 0.0117 ppm (Table 1-3). The lowest lead concentration was about 0.005 ppm in February 2021 and September 20 in Grouper samples, and also was in July 2021 in Emperor, while the highest concentration was 0.019 ppm in January 2020 in Grouper. All results were still very low and under European permissible standards (Figure-2).

The results of the current study agreed when compared to the previous study conducted by Al-Qadasy et al. (2018) [18]. And despite the noticeable increase in the results of Al-Qadasy's study over the current study by about 73%, it remains within the minimum limits according to European standards. Also, although the results of the Al-Dohail et al. (2020) [20] study exceeded the minimum limit (Figure 2), an increase of 88% over the average results of the current study, they remain within the minimum limits due to the fact that the fish sample in the Al- Dohail's study was cuttlefish, in which

the permissible limits for cadmium concentrations are considered 1 ppm. The results of and Obaidat et al. (2015) [19] study did not agree with the results of this study. Firstly, its results exceeded the minimum limits according to European standards by 58%, and secondly, compared to the average results of the current study, they were high by 88%.

We don't know the main reason for this. But possibly the human factor was behind the increase in cadmium pollution at that time. In fact, it is known that Cadmium (Cd), a toxic and nonessential transition metal, is a major threat to common organisms in aquatic ecosystems and has been widely recognized as a major pollutant in waters worldwide [24]. The kidney and liver are the most sensitive organs to Cd toxicity, exposure to low Cd concentration in a prolonged time impaired male and female reproduction systems.

It also affected pregnancy and showed bigger adverse effects on female neonates [4]. Usually cadmium pollution happens as a result of throwing factory waste into water surfaces, which annually reach more than 1000 tons, or may leak out from pipes and pipe connections that made of plastic which contains cadmium [25].

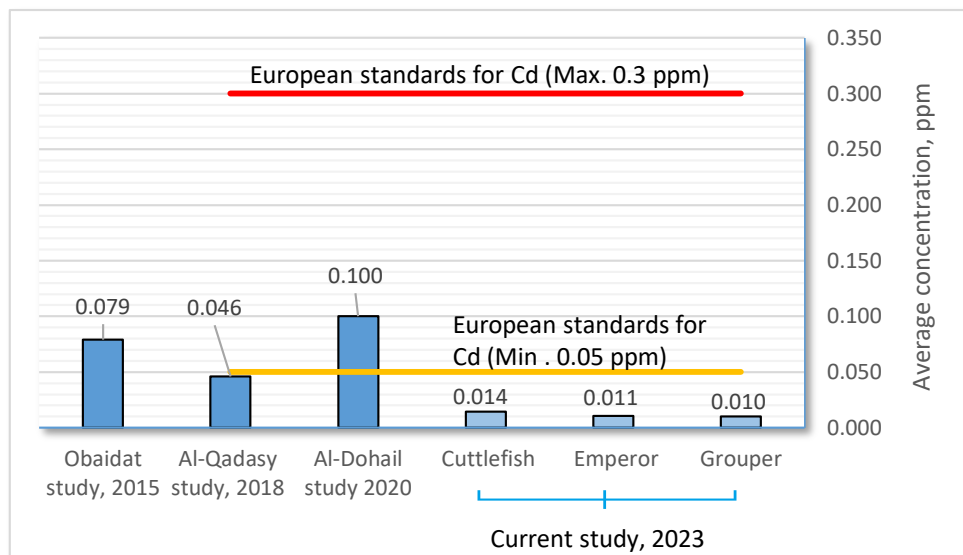


Figure 2. Cd concentrations in fish species and their comparison with standards and other studies

3.3 Mercury (Hg):

The results of mercury concentrations in the studied samples (Grouper, Emperor and Cuttlefish) ranged between 0.003 and 0.019 ppm, with average value 0.0067 (Table 1-3). The lowest mercury concentration was 0.003 ppm in March of the year 2020 in Grouper and in June 2020 in Emperor, while the highest concentration was 0.019 ppm in September 2020 and March 2021, and also in Grouper.

Yemeni standards set the permissible limit for mercury in frozen fish at 0.5 ppm, while the European standards gave a wide range depending on the species of fish, and ranged from 0.5 to 1 ppm. In order to get a clear picture of the results and judge them, 0.5 ppm was taken as a reference for comparison. From the (Figure-3) we can see that the general mercury concentrations over the three years 2020-2022 in all samples were even under the permissible limits whether by Yemeni and minimum European standards.

The results of this study agreed with the previous study conducted by Al-Qadasy et al. (2018) and Obaidat et al. (2015) (Figure-3). Despite the slight increase indicated by Al-Qadasy's study compared to the current study (about 86%), the results of this study, in comparison with Obaidat's study, were also slightly higher. And with all this, all results were also below the permissible limits standards. This is very important to ensure that mercury concentrations remain within permissible limits, since mercury is known to be toxic to humans and ecosystems. For example, mercury can build up in the bodies of fish in the form of methyl mercury (organic mercury) which is very poisoning and largely linked to eating seafood, mainly fish [26]. As for when it reaches humans bodies, Hg is converted into methyl mercury, which may lead to serious implications including neurological disorders, reproductive abnormalities, kidney failure, emotional instability, gingivitis and tremors [27].

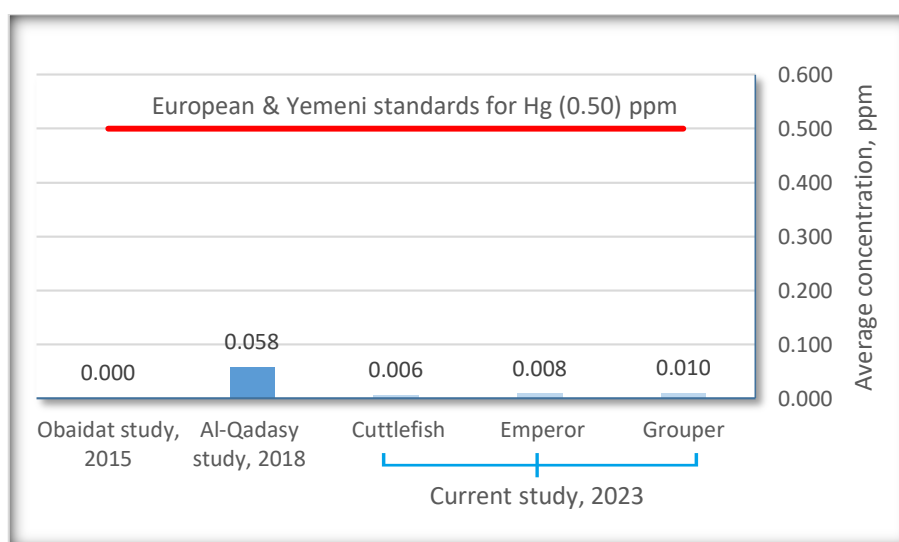


Figure 3. Hg concentrations in fish species and their comparison with standards and other studies

3.4 One way analysis variance (ANOVA) of heavy metals between the study's fish:

The results of the ANOVA (Table-4) showed that the bioaccumulation of lead metal during the study period was high in Emperor fish (0.1228 ±0.038 ppm) and low in Cuttlefish (0.1220 ±0.034 ppm). Although, considerable variation in bioaccumulation of lead (Pb) metal was observed between three different fish species, ANOVA did not indicate significant variation ($p > 0.05$) among them ($F=0.004$, sig. =0.996). In contrast, Results of this study showed that bioaccumulation of cadmium (Cd) metal was high (0.0143 ±0.003 ppm) in Cuttlefish compared with Grouper and Emperor fishes (0.0101 ±0.005 and 0.0108 ±0.005 ppm respectively).

The difference in bioaccumulation of cadmium (Cd) between particular fish species was significant ($p < 0.05$) ($F = 9.027$; Sig. = 0.001), and the post hoc tests (LSD) revealed the significantly different in cadmium concentration between grouper and Emperor compared to Cuttlefish, However, the post hoc tests (LSD) did not show significant difference between Grouper compared with Emperor fish. The results of the study, also showed that the bioaccumulation of mercury metal was high (0.0098 ±0.005 ppm) in Grouper and low (0.0063 ±0.002 ppm) in Cuttlefish. The results of one way analysis variance (ANOVA) revealed a significant differences in bioaccumulation of mercury metal between three different fish ($P < 0.05$) ($F = 10.676$; Sig = 0.000).

Table 4. Results of (ANOVA) with (LSD) tests of mean bioaccumulation of metals (ppm) in study's fish species (Grouper, Emperor and Cuttlefish)

| Metal | Type of fish | | | F | Sig |
|--------------|----------------------------|----------------------------|----------------------------|--------|-------|
| | Grouper | Emperor | Cuttlefish | | |
| Lead (Pb) | 0.1225 ±0.035 ^a | 0.1228 ±0.038 ^a | 0.1220 ±0.034 ^a | 0.004 | 0.996 |
| Cadmium (Cd) | 0.0101 ±0.005 ^a | 0.0108 ±0.005 ^a | 0.0143 ±0.003 ^b | 9.027 | 0.001 |
| Mercury (Hg) | 0.0098 ±0.005 ^a | 0.0084 ±0.005 ^a | 0.0063 ±0.002 ^b | 10.676 | 0.000 |

Mean ±SD. indicated n=84.

Means followed by the different letters are significantly different ($P < 0.05$).

Also Post Hoc tests (LSD) showed a significant differences in mercury concentration between Grouper and Emperor compared to Cuttlefish. Whereas, it indicates did not any significantly different between bioaccumulation of mercury in Grouper and Emperor compared to Cuttlefish. This study confirms the concentration of heavy metal in fish tissues depends on fish species. This view was further supported by the findings of previous study by [28] in which the levels of the heavy metals varied significantly among fish species and organs. These variation in bioaccumulation of heavy metals between different fish species may be mainly due to change eating habits. Previous studies by [29] and [30] also confirm the importance of eating habits in the estimation of bioaccumulation of heavy metals in different fish. Different fish have different ability to accumulate heavy metals. This may be linked to the lifespan of fish in the water body and the difference in physiological metabolism.

3.5 One way analysis variance (ANOVA) of heavy metals in fish tissues among years:

The bioaccumulation of heavy metals in fish tissues were measured during three years. The obtained results from this study showed that annually variation in bioaccumulation of heavy metals in fish (Table-5). The lowest and the highest bioaccumulation of lead metal observed in fish tissues during

2020 (1303 ±0.031 ppm) and 2021 (0.1109 ±0.027 ppm).

The post hoc tests (LSD) revealed the significant difference in lead concentration among them. Cadmium concentrations during three years (2020, 2021, 2022) were recorded (0.0119 ±0.005, 0.0113 ±0.005 and 0.0118 ±0.004) respectively. While one way analysis variance (ANOVA) did not indicate significant variation ($p > 0.05$) among them ($F=0.102$, sig. =0.903). In contrast, bioaccumulation of mercury metal in fish tissues during three years (2020, 2021, 2022) were determined (0.0067 ±0.003, 0.0101 ±0.005 and 0.0083 ±0.004) respectively, one way analysis variance (ANOVA) did show significant variation ($p < 0.05$) among them ($F=49.93$, sig. =0.009). The post hoc tests (LSD) revealed the significant difference in mercury concentration between 2020 compared to 2021, however, the (LSD) did not show significant difference between 2020 and 2021 compared with 2022. Annually variation of bioaccumulation of heavy metals in fish tissues may be mainly due to the different levels of water heavy metals during annuals. Previous study by [31] showed significant differences between water heavy metals of months. Also there are several studies confirm positive relationship between level of heavy metals in water and their bioaccumulation in fish tissues.

Table 5. Results of (ANOVA) with (LSD) tests of bioaccumulation of metals (ppm) in fish species among years (2020, 2021 and 2022)

| Metal | Periods (year) | | | F | Sig |
|--------------|----------------------------|----------------------------|-----------------------------|-------|-------|
| | 2020 | 2021 | 2022 | | |
| Lead (Pb) | 0.1303 ±0.031 ^a | 0.1109 ±0.027 ^b | 0.1245 ±0.044 ^{ab} | 5.14 | 0.046 |
| Cadmium (Ca) | 0.0119 ±0.005 ^a | 0.0113 ±0.005 ^a | 0.0118 ±0.004 ^a | 0.102 | 0.903 |
| Mercury (Hg) | 0.0067 ±0.003 ^a | 0.0101 ±0.005 ^b | 0.0083 ±0.004 ^{ab} | 49.93 | 0.009 |

Mean ±SD. indicated n=84.

Means followed by the different letters are significantly different ($P < 0.05$).

4. Conclusions and recommendations:

1. Lead, cadmium and mercury concentration levels (bioaccumulation) in studied fish species during the years 2020-2022 were safe and remained within the permissible limits according to the Yemeni and European standards.
2. Despite the increase in the levels of lead concentration levels in the current study compared to previous studies, they remained within the permissible limits.
3. Although the previous studies showed high levels of cadmium concentrations, and even the Odaidat's study exceeded the minimum European and Jordanian standards for the permissible limits, however, the current study showed very low concentration levels. In fact, the reason of the cadmium levels increase in previous studies is not known, but the human factor could be the main reason.
4. Regarding the mercury, the current results and also previous studies showed that the levels of its concentration levels remained within the permissible limits according to the Yemeni and European standards.
5. It has been concluded that, although officially in Yemen the fishing season is closed from July to October, authors obtained fish samples in some months of the closing season, and this is bad news that indicates the lack of commitment by fishermen to regulations and laws, and the absence of the role of the relevant bodies in this regard.
6. From the perspective of author's' management, the variations in the importance of heavy metals in respect to yearly bioaccumulation will be significant in the Gulf of Aden and Arabian Sea regions.
7. The fluctuation in the concentrations of heavy metals in fish, despite their presence within permissible limits, requires the relevant authorities, including universities, colleges, and scientific centers, to continuously monitor and respond quickly to any changes that occur in regional waters and marine life.
8. Local and national authorities must provide all forms of support and assistance to implement research studies for early detection of pollution.

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دراسة مستويات تراكيز الرصاص والكاديوم والزنك في بعض الأسماك التجارية من خليج عدن

وبحر العرب خلال الأعوام 2020 - 2022

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الملخص: أجريت هذه الدراسة لتحديد وتقييم مستويات تراكيز المعادن الثقيلة: الرصاص (Pb) ، الكاديوم (Cd) ، والزنك (Hg) في لحوم بعض أنواع الأسماك التجارية: الهامور (Epinephelus areolatus) ، الجحش (Lethrinus nebulosus) والحبار (Sepia Pharaonis). تم جمع 84 عينة من الأسماك من مواقع مختلفة في خليج عدن وبحر العرب خلال المدة من يناير 2020 إلى ديسمبر 2022. تم بيع بعضها في أسواق مدن محافظة حضرموت- اليمن، وتم تصدير ما بقي منها إلى الخارج. حللت جميع العينات في مختبرات كلية علوم البيئية والأحياء البحرية- جامعة حضرموت، حيث تم استخدام مطياف الامتصاص الذري في عملية تحليل العناصر باتباع الطرائق العلمية المتعارف عليها. أظهرت نتائج الدراسة أن متوسط تراكيز الرصاص والكاديوم والزنك في عينات الهامور بلغ 0.1225، 0.0101، و0.0098 جزءا في المليون على التوالي. في الجحش بلغ متوسط تركيزات الرصاص والكاديوم والزنك 0.1228 و0.0108 و0.0084 جزءا في المليون على التوالي، بينما في الحبار وصل الرصاص والكاديوم والزنك إلى 0.1220 و0.0143 و0.0063 جزءا في المليون على التوالي. تظل كل هذه القيم أقل حتى من الحدود الدنيا المسموح بها سواء حسب المعايير اليمنية أو الأوروبية. لم تظهر نتائج تحليل التباين الأحادي (ANOVA) أي فرق معنوي في تراكيز الرصاص بين أسماك الدراسة ($P>0.05$) ، في حين أشارت إلى وجود فروق معنوية بين الكاديوم والزنك في. بين السنوات، لم يشر تحليل التباين (ANOVA) إلى وجود اختلاف معنوي ($P>0.05$) في تركيز الكاديوم، بينما كان هناك فرق معنوي ($P<0.05$) في تركيز الرصاص والزنك. خلصت هذه الدراسة بتقديم بعض التوصيات المهمة للسلطات المحلية والجهات العلمية المختصة في هذا المجال.

الكلمات المفتاحية: الرصاص ، الكاديوم ، الزنك، الأسماك، اليمن.