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### Effectiveness of Cleanup and Dredging Operations on Some Heavy Metal Residues in Fish Flesh From Manzala Lake, Egypt

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### Abstract

BACKGROUND: The current study aimed to evaluate the effectiveness of cleanup and dredging operations in Manzala Lake on the levels of mercury (Hg), arsenic (As), lead (Pb), and cadmium (Cd) in fish muscles and compare them with the concentrations previously recorded in the same lake before treatment.

METHODS: A total of 180 fish samples, including 60 each of Nile tilapia (*Oreochromis niloticus*), flathead grey mullet (*Mugil cephalus*), and African catfish (*Clarias gariepinus*), were analyzed using atomic absorption spectrophotometry. RESULTS: The mean concentrations (mg/kg) of Hg, As, Pb, and Cd in tilapia were  $0.01 \pm 0.03$ ,  $3.16 \pm 0.35$ ,  $1.01 \pm 0.16$ , and  $0.12 \pm 0.02$ , respectively; in mullet were  $0.15 \pm 0.03$ ,  $4.25 \pm 0.47$ ,  $0.87 \pm 0.15$ , and  $0.05 \pm 0.01$ , respectively; and in catfish were  $0.29 \pm 0.05$ ,  $4.74 \pm 0.38$ ,  $0.95 \pm 0.15$ , and  $0.06 \pm 0.01$ , respectively. The heavy metal values in the majority of the tested fish samples exceeded the permissible limits established by Egyptian and international standards. The pollution levels of Hg, As, Pb, and Cd in Manzala Lake did not decrease after the recent cleanup and dredging operations. CONCLUSION: This finding indicates that the fish species caught from Manzala Lake after the recent cleanup and dredging operations may carry a potential risk to public health. Consequently, monitoring the effectiveness of cleanup

and dredging is necessary to evaluate pollution levels over time and protect human health.

Keywords: Dredging operations, Fish muscles, Heavy metal residues, Maximum permissible limits

### 1. Introduction

**F** ish is a nutritive and healthy food because it offers low cholesterol content, rich in proteins of high biological value, poly unsaturated fatty acids, B-complex vitamins, and several important minerals, including cobalt, copper, iron, zinc, sodium, magnesium, potassium, phosphorous, fluorine, and iodine [1]. The consumption of fish is increasing worldwide, which may be attributed to its low price, ease of digestion, and simplicity of preparation, in addition to reducing the risk of cardiovascular diseases such as stroke and asthma [2]. Flathead gray mullet, Nile tilapia, and African catfish are the most common species among Egyptian fisheries and are widely consumed in Egypt. However, fish are considered to be the main source of heavy metal contamination originating from agricultural and industrial wastes, untreated sewage effluents, and mining activities [3].

Toxic contaminants, particularly lead (Pb), cadmium (Cd), mercury (Hg), and arsenic (As), are found in water from agricultural, industrial, and mining activities [4]. Pollution from heavy metals in aquatic systems has become a global challenge because these elements are virtually non-degradable and can continuously accumulate for several years into the surrounding environment, including sediments and water, and bio accumulate in fish flesh to levels that may have undesirable effects on fish [5].

The over accumulation of toxic metals in aquatic organisms poses a major public health concern. The

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ingestion of fish contaminated with toxic metals may result in potential risks for consumers, such as dermatitis and cardiovascular, liver, and kidney diseases, in addition to teratogenicity, carcinogenic, and immunosuppressive effects due to bio magnification over time [6,7]. Contamination of fish by heavy metals can be a serious threat to human health worldwide, partly because of increased treatment failures. To minimize the health risks associated with metals through the consumption of fish, international guidelines have been established to ensure the safe ingestion of fish [8].

Egypt is considered among the most important countries in fish production due to its large coastal lagoons, where many inland fisheries are located, in addition to a long coastline extending along the Red Sea to the east and the Mediterranean Sea to the north. The country has various fisheries centers, including Nasser and Manzala Lake, in addition to The Nile River, with various irrigation canals.

Manzala Lake is an important fishery resource in Northeast Egypt and provides more than 30% of the commercial fish in the country. Human activities, including industrial waste, sewage discharge, and road networks, pose serious threats to the water quality of lake [9]. Manzala Lake receives polluted water from the Hadous, Bahr El-Baqar, El-Serw, Mataria, Ramsis, and Faraskur drains, with a total drainage of ~4000 million m<sup>3</sup>/year. Therefore, in 2019, the Egyptian government issued directives for cleanup and dredging Manzala Lake, including the removal of sediments and weeds, development of sparks that connect the lake to the Mediterranean Sea using the latest giant dredgers, and disposal of pollutants and increasing depth, making them better at the global level. The lake development process will be completed in 2021.

Dredging removes large amounts of contaminated sediments from aquatic environments; however, this is not always accompanied by a corresponding reduction in risk [10]. Several studies have reported that sediment dredging is a successful method for controlling heavy-metal pollution in freshwater [11–13]. However, sediment dredging may negatively affect freshwater ecosystems [10,14,15].

The heavy metal content in fish captured from Manzala Lake before cleanup and dredging operations have been previously reported by El-Moselhy, Saeed and Shaker, Elkady et al., Sallam et al. [16–19]. To date, no information is available regarding heavy metal levels in fish caught from Manzala Lake, Egypt, after dredging operations. Hence, the present study was conducted for the first time in Manzala Lake after dredging operations, with the specific goal of investigating the levels of Pb, Cd, As, and Hg in Nile tilapia (*Oreochromis niloticus*), flathead grey mullet (*Mugil cephalus*), and African catfish (*Clarias gariepinus*) caught from Manzala Lake, Egypt, after the recent cleanup and dredging operations, to evaluate the effectiveness of dredging operations in controlling heavy metal pollution.

### 2. Materials and methods

#### 2.1. Study area

Manzala Lake is the largest brackish lake in Egypt, with an area of ~404.69 Km<sup>2</sup> with 1.2-1.5 m depth and 60 km length. It is located in northeastern Egypt in the Nile River Delta, bordered by longitudes of  $31^{\circ}50'$  and  $32^{\circ}25'E$  and latitudes of  $31^{\circ}10'$  and 31°40'N. It connects four governorates, Dakahlia from the southwest, Port Said from the northeast, Damietta from the west, and Sharkia from the south, and shares its northern borders with the Mediterranean Sea and Suez Canal from the east. The importance of the lake stems from the fact that it is home to different types of fish such as tilapia, mullet family, bagrus, catfish, crabs, and shrimp. The lake receives gradual flows of pollutants from many drains, such as Ramsis, Hadous, El-Serw, Faraskour, and Bahr El-Bagr drains which cause pollution (Fig. 1).

### 2.2. Samples collection

In total, 180 fresh fish samples of three different species (60 each of Nile tilapia, Flathead grey mullet, and African catfish) were collected from fishermen at various sites in Manzala Lake after the recent cleanup and dredging operations, from September to December 2022, and used for analysis of Pb, Cd, As, and Hg residues. Fish of the same species have similar weights and sizes. Fish samples were individually packed in clean plastic bags, marked, stored in a cooler, and transported without delay to the Food Safety Hygiene and Technology Laboratory, Faculty of Veterinary Medicine, Mansoura University for digestion.

### 2.3. Reagents

The digestion reagents used were ultrapure grade, including perchloric acid 70%, hydrochloric acid 37%, hydrogen peroxide 30%, and nitric acid 65% (Merck, Darmstadt, Germany). All materials used for the preparation and digestion of samples were



Fig. 1. Egypt map showing Manzala Lake.

immersed in diluted nitric acid (10%) for 2 h, rinsed with deionized water, and dried on a clean bench to avoid element contamination.

#### 2.4. Samples digestion

Preparation and digestion procedures were performed as described by Sallam et al. [19]. Briefly, 2 gm of muscle beneath the dorsal fin, with its covered skin, of each sample was excised using a scalpel and forceps. Muscle tissue was cut with a knife and placed in a previously washed tube with 10% nitric acid containing a mixture of 8 ml concentrated nitric acid 65% and 4 ml concentrated perchloric acid (70%). The tubes were then closed and the samples were digested overnight at 53 °C using a water bath. The digest was filtered into a clean bottle using Whatman filter paper No. 42. The filtrate was then diluted with deionized water to 50 ml, marked with fish species, number, and stored until measurement of the toxic metals. The reagent blanks, one blank for each reagent, were prepared without samples and subjected to the same procedure as for sample digestion.

### 2.5. Heavy metals analysis

The digested samples were analyzed for toxic metal residues (Pb, Cd, Hg, and As) at the Central Laboratory, Faculty of Veterinary Medicine, Zagazig University, Egypt, according to the Association of Official Analytical Chemistry [20] using an Atomic Absorption Spectrophotometer provided with flame acetylene flow (Norwalk, CT, USA) for analysis of Cd and Pb, while As and Hg were detected by a flameless (cold vapor technique) Atomic Absorption Spectrophotometer. Calibration standard solutions were used at different concentrations of 10, 2.5, 0.25, 0.05, 0.01, and 0.005 µg/l for Pb, 20, 5, 0.5, 0.1, 0.02, and 0.01 µg/l for Cd and As, 5, 2, 0.5, 0.1, 0.05, and  $0.01 \mu g/l$  for Hg. The heavy metals were determined using calibration curves. The elemental content was calculated according to Eq. (1).

Element (wet weight mg/kg) = D/W × R (1)

where D = Prepared sample dilution, W=Sample wet weight, and R=Reading of metal value (mg/L) from the Atomic Absorption Spectrophotometer. Heavy metal levels in the blank samples were also

| Species              | Hg              | As              | Pb                  | Cd              |  |  |  |
|----------------------|-----------------|-----------------|---------------------|-----------------|--|--|--|
| Nile tilapia         |                 |                 |                     |                 |  |  |  |
| Min                  | 0.03            | 0.14            | 0.05                | 0.01            |  |  |  |
| Max                  | 0.55            | 5.81            | 2.95                | 0.40            |  |  |  |
| Mean $\pm$ SE        | $0.01 \pm 0.03$ | $3.16\pm0.35$   | $1.01 \pm 0.16^{a}$ | $0.05\pm0.01$   |  |  |  |
| Flathead grey mullet |                 |                 |                     |                 |  |  |  |
| Min                  | 0.04            | 0.15            | 0.02                | 0.01            |  |  |  |
| Max                  | 0.40            | 9.36            | 2.90                | 0.42            |  |  |  |
| Mean $\pm$ SE        | $0.15\pm0.03$   | $4.25 \pm 0.47$ | $0.87 \pm 0.15$     | $0.12\pm0.02$   |  |  |  |
| African catfish      |                 |                 |                     |                 |  |  |  |
| Min                  | 0.10            | 1.40            | 0.02                | 0.01            |  |  |  |
| Max                  | 1.58            | 8.88            | 2.95                | 0.21            |  |  |  |
| $Mean \pm SE$        | $0.29 \pm 0.05$ | $4.74 \pm 0.38$ | $0.95\pm0.15$       | $0.06 \pm 0.01$ |  |  |  |

Table 1. Heavy metal residues (mg/kg) in muscle tissues of different fish species (n = 60 for each species).

calculated and subtracted from each analyzed sample.

#### 2.6. Statistical analysis

The obtained results were analyzed by one-way analysis of variance (ANOVA) to assess the differences in mean values of metal levels among the three fish species examined using the COSTAT computer software program https://cohortsoftware. com/costat.html. A P value less than 0.05 indicates a significant difference.

#### 3. Results and discussion

3.1. Heavy metals values in flesh samples of three different fish species caught from Manzala lake after the recent cleanup and dredging operations

Heavy metal pollution in aquatic systems has become a global challenge because these elements are virtually nondegradable, where they become dangerous for many aquatic organisms and pose a major concern to public health through the consumption of fish. The mean  $\pm$  SE concentrations of heavy metals in the muscle samples of the three fish species analyzed in this study are presented in Table 1.

### 3.1.1. Mercury concentrations in tilapia, mullet, and catfish

Mercury residues varied from 0.03 to 0.55 mg/kg in tilapia samples, 0.04–0.40 mg/kg in mullet samples,

and 0.16–1.58 mg/kg in catfish with mean levels of 0.01  $\pm$  0.03 mg/kg, 0.15  $\pm$  0.03 mg/kg, and 0.29  $\pm$  0.05 mg/kg, respectively (Table 1). Catfish had a significantly higher Hg content than tilapia and mullet (P < 0.05) (Table 2). Mercury concentrations among different fish species were in the following order: catfish greater than mullet greater than tilapia, which may be attributed to larger fish accumulating higher levels of Hg than smaller fish [21].

Lower values of Hg were reported in mullet  $(0.0145 \ \mu g/g)$  and catfish  $(0.017 \ \mu g/g)$  samples collected from Manzala Lake, Egypt [19]. In addition, Elkady et al. [18] detected lower Hg levels  $(0.004 \ \mu g/g)$  in *O. niloticus* caught from the same lake, which is an indication of Hg pollution levels in Manzala Lake. A lower value of Hg, ranging from 0.0011 to 0.048  $\mu g/g$ , has also been observed in different fish species from the Yangtze River, China [22]. Conversely, a much higher Hg level of 3.15  $\mu g/g$  was found in fish caught from the Persian Gulf of Iran [23].

### 3.1.2. Arsenic concentrations in tilapia, mullet, and catfish

Arsenic values in tilapia and mullet ranged from 0.14 to 5.81 and 0.15–9.36 mg/kg with a mean of  $3.16 \pm 0.35$  and  $4.25 \pm 0.47$  mg/kg, respectively, and varied from 1.04 to 8.88 mg/kg with a mean of  $4.74 \pm 0.38$  mg/kg in catfish samples (Table 1). A significant difference was found only between tilapia and catfish.

Similar to our results, As levels in the range of  $0.84-10.2 \,\mu$ g/g with a mean of  $4.3 \,\mu$ g/g, were recorded in five different fish species collected from the Aegean Sea [23]. Conversely, much lower As levels of 0.511, 0.621, and 0.518  $\mu$ g/g were found in tilapia, mullet, and catfish, respectively, caught from Manzala Lake, Egypt [19] and in *O. niloticus* (0.014) collected from the same Manzala Lake, Egypt [18]. Likewise, lower As levels in the range of 0.156–0.834  $\mu$ g/g were detected in different fish species collected from the Persian Gulf, Iran [24], and 0.012–0.029  $\mu$ g/g in fish from the Yangtze River, China [22].

### 3.1.3. Lead concentrations in tilapia, mullet, and catfish

Lead was detected in all muscle samples of tilapia, mullet, and catfish with a range of 0.05-2.95, 0.02 to

Table 2. Mean  $\pm$  SE (standard error) of heavy metal residues (mg/kg wet weight) in muscle tissues of the different fish species examined.

|                      | , ,    | 0 0                     |                       |                     |                     |
|----------------------|--------|-------------------------|-----------------------|---------------------|---------------------|
| Species              | Number | Hg                      | As                    | Pb                  | Cd                  |
| Nile tilapia         | 60     | $0.01 \pm 0.03^{a}$     | $3.16 \pm 0.35^{a}$   | $1.01 \pm 0.16^{a}$ | $0.05 \pm 0.01^{a}$ |
| Flathead grey mullet | 60     | $0.15 \pm 0.03^{a}$     | $4.25 \pm 0.47^{ab}$  | $0.87 \pm 0.15^{a}$ | $0.12 \pm 0.02^{b}$ |
| African catfish      | 60     | $0.29 \pm 0.05^{\rm b}$ | $4.74\pm0.38^{\rm b}$ | $0.95 \pm 0.15^{a}$ | $0.06 \pm 0.01^{a}$ |

\* Mean  $\pm$  SE bearing different superscript letters (<sup>a</sup> and <sup>b</sup>) in the same column for each species are significantly different (P < 0.01).

2.90, and 0.02-2.95 mg/kg, respectively, although Nile tilapia showed the highest mean  $\pm$  SE value (1.01  $\pm$  0.16 mg/kg) compared with mullet (0.87  $\pm$  0.15 mg/kg), and catfish (0.95  $\pm$  0.15 mg/kg), such differences were not significant (Tables 1 and 2).

The Pb content in this study was consistent with the levels ranged from 0.33 to 0.93 and  $0.33-0.86 \mu g/g$  detected in fish samples collected from the Black and Aegean Sea [25] and Aegean, Marmara, Mediterranean seas [26], respectively.

On the other hand, Ei-Moselhy [16], Elkady et al. [18] and Sallam et al. [19] detected lower Pb values of 0.13, 0.022 and 0.704 µg/g in *O. niloticus* collected from Manzala lake, Egypt, respectively, as well as in Mullet (0.67 µg/g) and catfish (0.75 µg/g) from the same lake Sallam et al. [19]. Much higher Pb contents in the range of 2.89–3.41 µg/g were detected in five fish species collected from Kutubdia Channel of the northern Bay, Bangladesh [27]. The Pb pollution was possibly due to industrial discharge from the battery.

### 3.1.4. Cadmium concentrations in tilapia, mullet, and catfish

Cadmium levels ranged from 0.01 to 0.40, 0.01 to 0.42, and 0.01–0.21 mg/kg in Nile tilapia, flathead grey mullet, and African catfish, respectively. The mean  $\pm$  SE value of Cd in mullet fish (0.12  $\pm$  0.02 mg/kg) was significantly higher than that of tilapia (0.05  $\pm$  0.01 mg/kg) and catfish (0.06  $\pm$  0.01 mg/kg) (Tables 1 and 2).

Nearly similar mean Cd levels of 0.03 and 0.024  $\mu$ g/g were found in the muscle of *O. niloticus* caught from Manzala Lake, Egypt [16,19], respectively, as well as in catfish (0.02) collected from the same lake [19]. In contrast, lower Cd concentrations (0.003 0.02 and 0.002  $\mu$ g/g) have been reported in fish muscle from Taihu Lake in China [28] and the Adriatic Sea in Croatia [29], as well as in the muscle of mullet fish (0.006) collected from Manzala Lake, Egypt [19]. The Cd contamination was possibly due to industrial and mining activities [30].

The data in this study showed that the least metal detected in the muscle of the fish species examined was Cd, which may be attributed to the high tendency of Cd to accumulate in organs such as the kidney and liver [4] and a rare metal in the earth's crust [31].

These variations in element values among fish from different countries could be attributed to nutritional habits, pollution rate in the collection area [32], metabolic activity of fish size, geography, season [33] and metal capability to undergo bioaccumulation in fish [34].

## 3.2. Comparison of heavy metal levels in the examined fish with the Egyptian and international standards

A comparison of the metals detected with Egyptian and international standards is shown in Table 3. Among the 180 fish samples examined, Hg levels in 3.4% (2/60), 3.4% (2/60), and 20% (12/60) of tilapia, mullet, and catfish, respectively, were above the legal limit proposed by the Egyptian standards [35]. Conversely, Hg levels in all mullet and catfish samples did not exceed the maximum permissible limit, whereas 16% of tilapia were above the MPLs [19]. Dietary intake of Hg may induce behavioral problems in young children, learning disabilities, and development of intelligence impairment [36].

Our findings indicated that of the 180 fish samples, 70% (42/60), 80% (48/60), and 86.6% (52/60) of tilapia, mullet, and catfish, respectively, had As contents that exceeded the limit recommended by Food Standards Australia New Zealand [37]. These results are inconsistent with those reported by Salam et al. [19], who found that As concentrations in all fish samples (tilapia, mullet, and catfish) caught from the same lake were within the legal limit of 1.5  $\mu$ g/g.

Our results showed that Pb concentrations in 70% (42/60), 66.6% (40/60), and 73.4% (44/60) of tilapia, mullet, and catfish exceeded the MPL set by Egyptian standards [35]. Similar observations were

Table 3. Comparison of heavy metal concentrations in fish muscles (mg/kg wet weight) with the national and international maximum permissible limit (MPL).

| Heavy<br>metals | MPL               | Nile tilapia  | Nile tilapia |               | Flathead grey mullet |               | African catfish |  |
|-----------------|-------------------|---------------|--------------|---------------|----------------------|---------------|-----------------|--|
|                 | (mg/kg)           | Within MPL    | Exceeded MPL | Within MPL    | Exceeded MPL         | Within MPL    | Exceeded MPL    |  |
| Hg              | 0.5 <sup>a</sup>  | 96.6% (58/60) | 3.4% (2/60)  | 96.6% (58/60) | 3.4% (2/60)          | 80% (48/60)   | 20% (12/60)     |  |
| As              | 2 <sup>b</sup>    | 30% (18/60)   | 70% (42/60)  | 20% (12/60)   | 80% (48/60)          | 13.4% (8/60)  | 86.6% (52/60)   |  |
| Pb              | 0.3 <sup>a</sup>  | 30% (18/60)   | 70% (42/60)  | 33.4% (20/60) | 66.6% (40/60)        | 26.6% (16/60) | 73.4% (44/60)   |  |
| Cd              | 0.05 <sup>a</sup> | 70% (42/60)   | 30% (18/60)  | 36.6% (22/60) | 63.4% (38/60)        | 66.6% (40/60) | 33.4% (20/60)   |  |

<sup>a</sup> Egyptian Standards (2010).

<sup>b</sup> Food Standards Australia and New Zealand Standard, 2011 (FSANZ, 2011)).

reported by Sallam et al. [19] who found that the Pb concentrations in tilapia, mullet, and catfish were 78%, 58%, and 72% above the MPL of 1.5  $\mu$ g/g. Pb is a toxic metal that can induce hypertension, hematological effects, kidney dysfunction, and negative effects on the reproductive, nervous, gastrointestinal, and immune systems [38].

Regarding the MPLs of Egyptian standards [35], the Cd levels in 30% (18 samples), 63.4% (38 samples), and 33.4% (20 samples) of tilapia, mullet, and catfish, respectively, exceeded the allowable limit of 0.05  $\mu$ g/g. Our results were inconsistent with those reported by Sallam et al. [19], who found that the Cd concentration in 6% of each tilapia and catfish were within the MPL (0.05  $\mu$ g/g), while all samples of mullet fish did not exceed the legal limit. Cd intake is associated with skeletal damage, renal damage, and digestive disorders [39].

Dredging removes large amounts of contaminated sediments from aquatic environments; however, this is not always accompanied by a corresponding reduction in risk [10]. Conversely, Ding et al. [13] found that dredging of sediment was an effective technique for reducing the heavy metal content of freshwater.

The aforementioned results demonstrate that the pollution levels of Hg, As, Pb, and Cd in Manzala Lake increased after the recent cleanup and dredging operations compared with previous studies conducted in the same lake before dredging operations. This may be attributed to the dredging of sediment, which can have short-term negative on freshwater ecosystems, effects including increased pollutant concentrations in the water, increased pollutant concentrations in fish tissue adjacent to the dredging activity [10], incomplete decontamination, leaching of heavy metals into the water, and suspension and loss of polluted sediments [15,40]. Heavy metals can be released from stable sulfide precipitates, with the sediments being suspended in the water column under the strong oxidation conditions created during sediment dredging [41]. However, monitoring the cleanup and dredging effects is necessary to evaluate pollution levels over time.

### 4. Conclusion

It can be concluded that most fish samples examined in the current study contained Hg, As, Pb, and Cd levels higher than the maximum permissible limits for the fish reference standard, indicating that fish caught from Manzala Lake after recent cleanup and dredging operations can pose a potential risk to public health. Consequently, monitoring the effectiveness of cleanup and dredging is necessary to evaluate pollution levels over time and protect human health.

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### Authors' contributions

SA, AZ, and HR conceived of and designed the study. We conducted the following experiments and analyzed the results: SA, AZ, and HR. SA and AZ drafted the manuscript.

### Data availability statement

All relevant data are within the paper and its Supporting Information files.

### Ethics approval and consent to participate

This protocol was performed by following the animal ethics guidelines and approved by Medical Research Ethics Committee of Mansoura University with code number (M/140).

### **Conflicts of interest**

The authors declare that there is no conflict of interests.

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