

Research Article

Accumulation and human health risk analysis of arsenic, lead, and mercury in three fishes caught from an oil-polluted-creek in Niger Delta, Nigeria

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Abstract

Bodo Creek, like other aquatic habitats in the Niger Delta region of Nigeria, is reportedly contaminated by heavy metals due to crude oil pollution. It is unknown whether edible fishes caught from these sources pose any health risk to human consumers. The present study evaluated concentrations of arsenic (As), mercury (Hg), and lead (Pb) in different organs of three edible fishes caught from Bodo Creek, and assessed the health risks of consuming these fishes by both children and adult populations. Samples of *Ethmalosa fimbriata*, *Crenimugil seheli* and *Macolor niger* were collected from artisanal fishermen fishing in Bodo Creek, and taken to the laboratory for heavy metal analysis. The results showed that concentrations of arsenic, lead, and mercury recovered from organs of these fishes differed, with gills having significantly ($p < 0.05$) higher concentrations than muscle and liver. In terms of health risks, estimated daily intakes of Pb were higher in *E. fimbriata*, followed by *M. niger*, and the least values were recorded in *C. seheli* for both children and adult consumers. Target hazard quotients of these metals were below the risk level of 1 in both children and adult consumers of the three studied fishes. However, hazard index for children consumers of *E. fimbriata* was greater than 1. The values of incremental long-term cancer risk analysis for the studied metals were greater than the USEPA recommended safe limit of 0.000001. Based on these results, consumption of these fishes may pose future health risks for both children and adult populations.

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Introduction

Crude oil is a complex mixture of hydrocarbon and non-hydrocarbon compounds (including heavy metals) found in subsurface deposits worldwide. In Nigeria, crude oil exploitation began in the late 1960s and has since then become major foreign exchange earnings, accounting for over 80 percent of the country's foreign exchange market value (World Bank, 2020). The Niger Delta is the major oil producing region of Nigeria, which is located by the Atlantic Coast where River Niger divides into numerous tributaries. However, crude oil exploration activities in this region have resulted in oil spillage and its numerous problems, including contamination of water bodies which poses danger to aquatic life, and humans that depend on them for food, as well as destruction of mangroves and aquatic habitats (UNDP, 2006). Numerous studies have shown elevated levels of heavy metals in areas where oil spillage occurred (Horsfall and Spiff, 2002; Howard *et al.*, 2006).

Exposure to heavy metals needs to be continuously monitored as they are known to have serious health implications for humans. For example, arsenic exposure affects virtually all human organs and systems including the cardiovascular, dermatologic, nervous, hepatobiliary, renal, gastro-intestinal, and respiratory systems (Tchounwou *et al.*, 2003). According to ATSDR (2010), lead is the most systemic toxicant that affects several organs in the body, including the kidneys, liver, central nervous system, hematopoietic system, endocrine system, and reproductive system. Lead, if absorbed by a pregnant mother, can

readily be transferred to the developing fetus (Ong *et al.*, 1985) causing reduced birth weight and preterm delivery, and neuro-developmental abnormalities in offspring (Huel *et al.*, 1992). Similarly, all forms of mercury are toxic and their effects include gastrointestinal toxicity, neurotoxicity, and nephrotoxicity (Tchounwou *et al.*, 2003). According to El Zlitne *et al.* (2022), feeding on fish containing methyl mercury and other heavy metals for longer periods deleteriously affects human health by suppressing their immune system, leading to increased susceptibility to bacterial and viral infection, frequency of cancer development, frequency of renal and hepatic failure, and development of critical cases of endocrine disruption.

Previous studies have shown marine and brackish water food fish as significant contributors to human intake of some metal contaminants due to their presence in the aquatic environment and their accumulation in the flesh of both finfishes and shellfishes (Silva *et al.*, 2007; Camargo *et al.*, 2009). Some aquatic organisms have also been reported to have capacity to accumulate heavy metals many times the concentration present in the water, making potential human health hazards associated with the consumption of contaminated aquatic food higher than the ingestion of contaminated water (WHO, 2011). According to Osakwe *et al.* (2014), concentrations of metals in an organism vary from organ to organ. Specific findings on the trend of metal accumulation in different organs of food fish species caught in polluted environments is necessary to elucidate the suitability of consumption of

different parts of fish/other aquatic organisms, thereby serving as an effective strategy toward ecological protection for public health risks of fish consumers.

Furthermore, Zabbey and Babatunde (2015) showed that Bodo Creek, like most aquatic ecosystems in the Niger Delta of Nigeria, is heavily polluted by crude oil and consequently contaminated by heavy metals. Despite these pollutions, artisanal fishermen continue to fish in these areas while the fish caught are being sold for human consumption. The goal of this research is therefore to assess, in comparison to the internationally permissible levels, the concentrations and health risks associated with arsenic, lead, and mercury in three food fish species caught from the Bodo Creek in the Niger Delta region of Nigeria.

Materials and methods

Study area

The study area, Bodo Creek, is located east of the city of Port Harcourt, Rivers State, and emptied into the coast of the Guinea Gulf. The creek extends across four local government areas (Khana, Gokana, Eleme, and Tai). The map of Bodo Creek is presented in Figure 1. Fish sample collection was done between May 2021 and October 2021 at the Gokana axis (portion enclosed in the map) from artisanal fishermen fishing in the creek. The Gokana portion of Bodo Creek is located between latitude 4°37' north of the equator and longitude 7°16' east of the meridian.

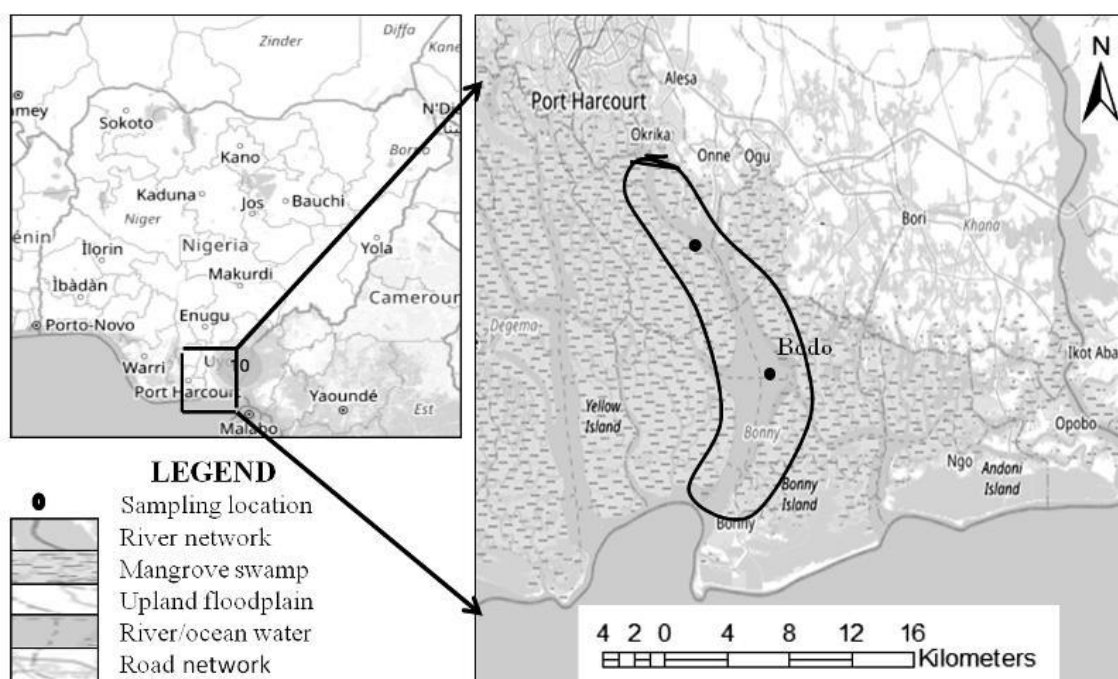


Figure 1: Map of Bodo Creek in Niger Delta, Nigeria.

Sample collection and preparation

Three commercially important food fish species from the study area (*Ethmalosa fimbriata*, *Crenimugil seheli* and *Macolor niger*) were collected from artisanal fishermen at Gokana for heavy metal analysis. The fish were identified using appropriate identification charts (Adesulu and Sydenham, 2007; Froese and Pauly, 2017). Ten samples of each fish species (weighing >250 g) were collected from different fishermen, wrapped with clean nylon materials and transported in an ice box to the laboratory. The fish were dissected at the laboratory and the organs (gills, liver, and muscle) were harvested for analysis.

Digestion of Samples and Heavy Metal Analysis

The methods used in the study followed the procedure of AOAC (2008). Briefly, a total volume of 100 ml of H₂SO₄, HNO₃, and HCl in ratio of 4:4:2 was mixed. 10 g of each fish sample was weighed and put into a conical flask. 2 ml of the mixed acid was added to each of the samples in the conical flask. The sample was digested in a fume cupboard with a hot plate until white fumes appeared. The residue was filtered, after it was allowed to cool, by using a 0.45µm Whitman filter paper and was then transferred to a 100 mL volumetric flask. The remaining volume in 100 ml mark of the flask was made up with distilled water. Following sample digestion, an atomic absorption spectrophotometer (AAS, Perkin Elmer Analyst 400 model) was employed for determining the proportion of arsenic (Ar), lead (Pb) and mercury (Hg) in each sample. Analytical blank was

prepared, and series of calibration solutions of known amounts of these metals (standards) were made. The blank and standards were atomized in turn and their responses measured. A calibration graph was plotted for each of the solutions, after which the sample solutions were atomized and measured. Various metal concentrations from the sample solution were determined from the calibration, based on the absorbance obtained for the known samples.

Risk assessment of the analyzed metals

The health risk of a metal/contaminant is usually assessed based on numerical expressions of the risk levels (including comprehensive analysis for quantifying and interpreting the values). The recommended tolerant limits of each metal are usually compared with the analyzed values to determine the risk, but further evaluation must be carried out to classify the risks as either carcinogenic or non-carcinogenic health hazards (Wongsasuluk *et al.*, 2014). The present study further evaluated the carcinogenic and non-carcinogenic health risks (for both children and adult populations) resulting from the consumption of toxic metals via fishes caught from oil polluted Bodo River, using Target Hazard Quotients (THQ), Hazard Index (HI), and the Incremental Lifetime Cancer Risk (ILCR).

Determination of estimated daily intake (EDI)

Estimated Daily Intake (EDI) for fish consumers was determined as

$$EDI \text{ (mg kg}^{-1}\text{day}^{-1}\text{)} = \frac{EFr \times ED \times FIR \times M}{BW \times ATn}$$

where EFr is exposure frequency; ED is exposure duration; FIR is the fish ingestion rate; M is the metal concentration analyzed from the fish; BW is the body weight of the consumer and ATn is average time. The input parameters for these values for both children and adult consumers are presented in Table 1. It was assumed that all the

organs analyzed in this study (gill, muscle, and liver) are readily consumed by both children and adult populations in the studied area, thus mean values of the studied metals from these organs were used for the risk estimation.

Table 1: Input parameters for estimating health risk via fish consumption in the studied area.

Parameters	Children	Adults
Body weight (BW, kg)	25	60
Exposure duration (ED, years)	10	30
Exposure frequency (EFr, days/year)	365	365
Fish ingestion rate (FIR, mg/kg)	150	300
Average Time for carcinogens (ATn, days/year)	365×70	365×70
Average Time for non-carcinogens (ATn, days/year)	365×ED	365×ED

Target hazard quotient (THQ)

THQ value (which is a dimensionless index of risk associated with long-term exposure to a toxic contaminant) is based upon comparison with reference upper safe limits (Abdi and Kazemi 2015). THQ was calculated as

$$THQ = \frac{EFr \times ED \times FIR \times M}{RfD \times BW \times ATn} \times 10^{-3}$$

Where, EF, ED, FIR, BW, M and ATn are described earlier. RfD is the oral reference dose (the values for analyzed metals are presented in Table 2).

Hazard index (HI)

The total potential non-carcinogenic health impact that could result due to exposure to a mixture of the analyzed heavy metals in each fish species was computed using Hazard Index (HI). The HI was expressed (as described by Di Leo *et al.* (2010) as the arithmetic sum of the target hazard quotients (THQ) as follows:

$$HI = THQ(\text{arsenic}) + THQ(\text{mercury}) + THQ(\text{lead})$$

According to Guerra *et al.* (2012), there is the possibility that non-carcinogenic impacts may occur in fish consumers when $HI > 1$, while the exposed person is unexpected to experience evident harmful health impacts when $HI < 1$.

Incremental lifetime cancer risk (ILCR)

The Incremental Lifetime Cancer Risk (ILCR) is defined as incremental probability of a person developing any type of cancer over a lifetime as a result of twenty-four hours per day exposure to a given daily amount of a carcinogenic element for a period of seventy years (Gržetić and Ghariani, 2008). In this study, the lifetime cancer risks of the analyzed metals were calculated as

$$ILCR = EDI \times CSF$$

Where EDI is the estimated daily intake (as described and calculated above) and CSF is the cancer slope factor (the values

for each analyzed metal are presented in Table 2). The permissible limits of 10^{-6} mg/kg/day are considered to be safe for a single carcinogenic element (Tepanosyan *et al.*, 2017).

Statistical analysis

The values of toxic metals obtained from different fish species were analyzed using

one-way ANOVA in SPSS version 20 and the values were considered significant at $p > 0.05$. Metal concentrations were recorded as mean \pm standard deviation and plotted as bar charts in the Microsoft Excel 2010 version. Mean values of the metal concentrations, derived from examined organs of each fish species, were employed for calculations of health risk assessments.

Table 2: Guidelines for toxicological properties of the toxic metals analyzed in this study.

Properties	Toxic Metals			References
	As	Hg	Pb	
Permissible limit in fish (mg/kg)	0.2	0.2	0.5	FAO/WHO, 2021
Ingestion/oral reference dose (RfD)	3.0×10^{-4}	3.0×10^{-4}	3.5×10^{-3}	USEPA 2011
Ingestion cancer slope factor (CSF)	1.5	NA	8.5×10^{-3}	USEPA 2011

Results

Concentrations of arsenic, lead, and mercury recovered from three commercially important fish species caught from Bodo River are presented in Figures 2 to 4. For *Ethmalosa fimbriata*, lead was found to be significantly higher ($p < 0.05$) in gills than in muscle and liver (Fig. 2).

Similarly, insignificantly higher values in gills than in muscles and liver were observed in both arsenic and mercury concentrations. Gills and muscles showed heavy metal accumulation in this sequence Pb>As>Hg, while the liver showed Pb>Hg>As sequence.

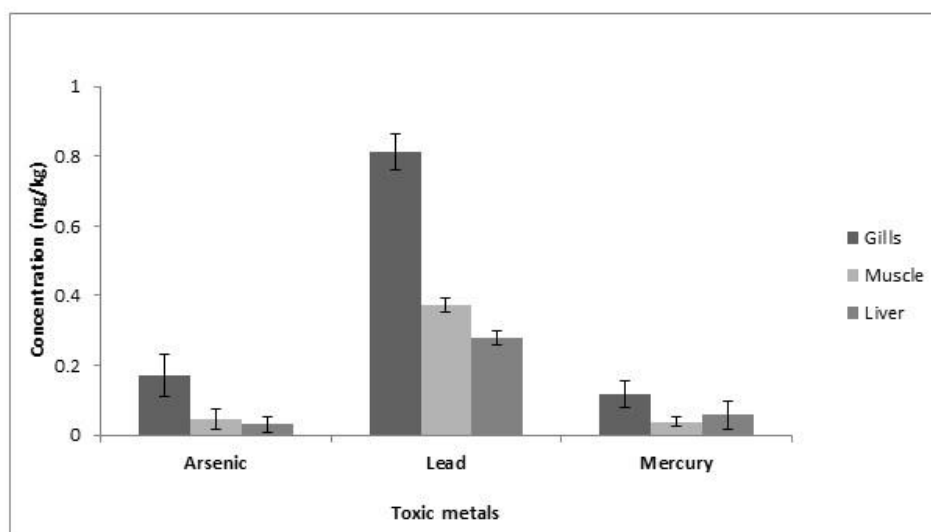


Figure 2: Concentrations of heavy metals in different organs of *Ethmalosa fimbriata* caught from Bodo Creek, Error bars indicate standard deviations of 10 fish samples.

In *Crenimugil seheli* (Fig. 3), arsenic values were insignificantly higher in gills than in muscle and liver. Lead concentrations were significantly higher in gills than in muscle and liver, while mercury was higher in gills and liver than in muscle. The sequence of metal accumulation in gills, muscles and liver were in the order of Hg>Pb>Ar, Ar>Hg>Pb and Hg>Ar>Pb, respectively. In *Macolor niger* (Fig. 4), lead concentration in gill was significantly higher than in other examined organs, while the highest values of arsenic and mercury were found in gills and liver, respectively. The order of metal

accumulation in gills and muscles were Pb>Ar>Hg, Pb>Ar>Hg, while that of liver was Hg>Ar>Pb.

The health risks that may result from consumption of three studied fishes caught from Bodo Creek are presented in Tables 3 and 4.

In the present study, arsenic, lead, and mercury were recovered from *E. fimbriata*, *C. Seheli*, and *M.niger* caught from Bodo Creek.

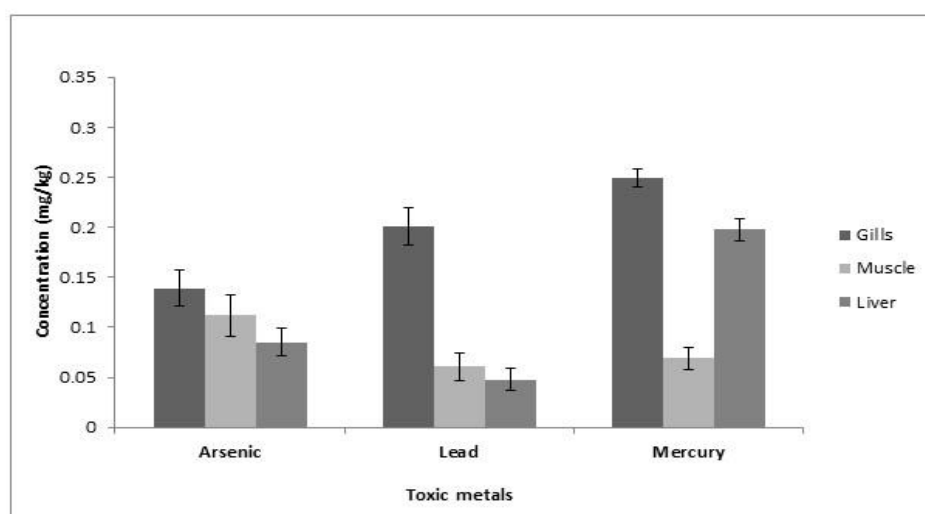


Figure 3: Concentrations of heavy metals in *Crenimugil seheli* caught from Bodo Creek, Error bars indicate standard deviations of 10 fish samples.

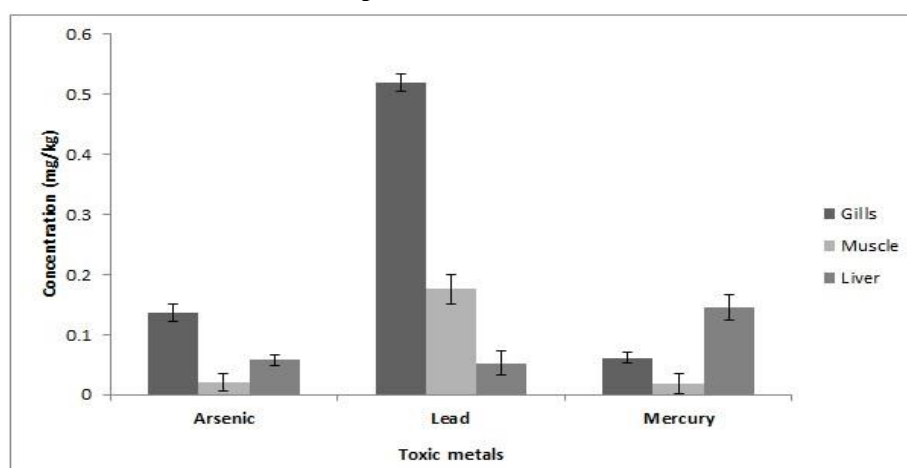


Figure 4: Concentrations of heavy metals in *Macolor niger* caught from Bodo Creek, Error bars indicate standard deviations of 10 fish samples.

Discussion

The results showed that estimated daily intake (EDI) of lead (Pb) were higher in *E. fimbriata*, followed by *M. niger* and the least values were recorded in *C. seheli* for both children and adult consumers. Target hazard quotient (THQ) of arsenic, mercury, and lead was below the risk level of 1 in both children and adult consumers of the three studied fishes. However, relatively higher values of THQ were observed in children than adult consumers, with values for lead doubled in children when

compared to adult consumers of *E. fimbriata*. Consequently, hazard index (HI) for children consumers of *E. fimbriata* was greater than 1.

For carcinogenic risk analysis, values of ILCR for all studied metals were greater than the recommended safe limit of 0.000001, indicating serious cancer risks for both children and adult populations consuming the three fishes in the future.

Table 3: Estimates for different health risks that may be associated with consumption of the three fish species caught from Bodo Creek, Niger Delta, Nigeria.

Health risk	Consumer group	Toxic metal	Values for different fish species		
			<i>Ethmalosa fimbriata</i>	<i>Crenimugil seheli</i>	<i>Macolor niger</i>
EDI (mg/kg/day)	Adults	Arsenic	0.412	0.560	0.358
		Lead	2.438	0.517	1.247
		Mercury	0.350	0.862	0.272
	Children	Arsenic	0.494	0.672	0.430
		Lead	2.926	0.620	1.496
		Mercury	0.420	1.034	0.326
THQ	Adults	Arsenic	0.137	0.187	0.119
		Lead	0.495	0.148	0.356
		Mercury	0.117	0.287	0.091
	Children	Arsenic	0.165	0.224	0.143
		Lead	0.836	0.177	0.427
		Mercury	0.140	0.345	0.109
ILCR	Adults	Arsenic	0.02646	0.03599	0.02305
		Lead	0.00150	0.00204	0.00135
		Mercury	ND	ND	ND
	Children	Arsenic	0.010585	0.014399	0.00921
		Lead	0.003553	0.000753	0.00182
		Mercury	ND	ND	ND

EDI: Estimated Daily Intake, THQ: Target hazard quotient, ILCR: Incremental Lifetime Cancer Risk, ND: not detected

Table 4: Hazard Index for adult and children populations consuming the three fish species caught in Bodo Creek, Nigeria.

Fish Consumer group	Fish Species	Hazard Index (HI)
Adult	<i>Ethmalosa fimbriata</i>	0.748
	<i>Crenimugil seheli</i>	0.621
	<i>Macolor niger</i>	0.566
Children	<i>Ethmalosa fimbriata</i>	1.141
	<i>Crenimugil seheli</i>	0.746
	<i>Macolor niger</i>	0.679

Previous reports showed that Bodo Creek was heavily polluted and contaminated with heavy metals due to crude oil exploration in the area (Zabbey and Babatunde, 2015; Ezemonye *et al.*, 2019). Heavy metals are known to be detrimental to human health (El Zlitne *et al.*, 2022), so their presence beyond recommended limits in fish may invalidate the benefits of fish consumption and endanger public health.

An earlier report by Yousafzai *et al.* (2010), stated that the accumulation of heavy metals in fish could be influenced by variation in age, season, and gender, and this might correlate with feeding habits in different seasons and areas. The connection between feeding habits, foraging behavior, and heavy metal concentration is also well established as being higher for omnivorous and herbivorous when compared to carnivorous fishes. Feeding on different food chains is considered to have greater chances of heavy metal bioaccumulation (Yousafzai and Shakoori, 2006).

The three fish species examined in this study exhibit different feeding habits. Blay and Eyeson (1982) reported that *E. fimbriata* is primarily a plankton feeder, as these authors observed protozoa, crustacea, molluscan larvae, and a considerable amount of detritus in the stomach of marine samples while the stomachs of the estuarine fish contained mainly phytoplankton, protozoa, sand grains and organic detritus. Similarly, the diet of *C. seheli* comprised large amounts of detritus, benthic invertebrates, green filamentous macroalgae and microalgae (Cardona *et al.*, 2015). *M. niger* is mainly carnivorous, feeding primarily on fishes and crustaceans (Longenecker *et al.*, 2014). From foregoing

point of view, omnivorous nature of *E. fimbriata* could make it more prone to heavy metal bioaccumulation as compared to herbivorous *C. seheli* and carnivorous *M. niger*, as observed in this study.

The concentration of arsenic, lead, and mercury recovered from the examined organs (gills, muscle, and liver) were observed to differ greatly, with gills having higher metal concentrations than muscle and liver. A similar observation was made by Bervoets *et al.* (2001) who reported that fish gills tend to accumulate more concentration of heavy metals than liver and muscles. The reason for a higher concentration of the studied metals in gills may be because gills are the first contact points of waterborne pollutants. The extremely branched morphology of gill tissues and the movement of water through it may result in maximum accumulation of heavy metal (Ruiz-Picos and López-López, 2012). Heavy metals do not contact directly with muscle tissues, as compared to gill tissues which are completely exposed to an aquatic environment. Although edible parts of the fish include gills for most people, it may be necessary to avoid eating gill portions of the studied fishes to avoid being exposed to unacceptable levels of heavy metals.

The results of this study further showed that lead concentrations in the gills of *E. fimbriata* and *M. niger* were greater than FAO's recommended limit of 0.5mg/kg. Similarly, lead in both gills and liver of *C. seheli* was higher than FAO's recommended limit of 0.2mg/kg (FAO/WHO, 2021). Concentrations of arsenic, lead, and mercury in the muscles of the three fishes were within the

recommended limits. However, mean values of the three organs (which were used for further calculations) for each fish species were below their recommended values.

In terms of health risks analysis, EDI of lead (Pb) was higher in *E. fimbriata*. THQs were below risk level of 1 in both children and adult consumers of the three studied fishes, but HI for children consumers of *E. fimbriata* was greater than 1, implying health risks. It should be noted that Pb seems to be responsible for the observed HI value, as it contributed more than half to the HI. The findings of this research were in agreement with the report of Ezemonye *et al.* (2019) as they reported that EDI values estimated for Pb in fish were higher than other analyzed metals, while THQs of Pb were high in fish with a value of 1.93 which implied some likely health risks due to the presence of Pb. For carcinogenic risk analysis, values of ILCR for all studied metals were greater than the recommended safe limit of 0.000001 (FAO/WHO, 2021), indicating serious cancer risks for both children and adult populations consuming the three fishes in the future. The results obtained in this study underscore the need for periodic bio-monitoring of the coastal environments to prevent contaminations during oil exploration activities and guarantee safe consumption of fish.

The present study assessed the concentration of arsenic, lead, and mercury in relation to health risks these metals may pose to fish consumers. Findings in the study indicated that, except for Pb content in the gills of *E. fimbriata*, metal concentrations recovered from each fish species were within recommended limits.

However, the consumption pattern of eating whole fish including gills may be deleterious as calculated ILCR values indicated cancer risks for both children and adult populations. It is therefore advisable that fish gills should be discarded to avoid consuming food with high levels of heavy metal since fish gills contain a higher proportion of the studied metals. There is a need to implement appropriate policies that prohibit indiscriminate spillage of crude oil into the environment and accelerate mop-up strategies whenever spillage occurs to prevent environmental pollution.

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