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Risks Assessment of Copper (Cu), Lead (Pb), Mercury (Hg) and Zinc (Zn): A case study of *Tilapia guineensis* in Lagos Lagoon

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ABSTRACT

This study determined the daily intake of copper (Cu), lead (Pb), zinc (Zn) and mercury (Hg) in order to assess both carcinogenic and non-carcinogenic risks caused by the heavy metals in *Tilapia guineensis* of Lagos Lagoon. The heavy metals were determined using flame atomic absorption spectrophotometer. The metal distribution was Zn > Cu > Pb > Hg with values of 0.0248±0.04, 0.0093±0.01, 0.0005±0.00 and 0.000±0.00 mg/kg respectively. Daily Intake of Metal was in the order of Zn (0.02) > Cu (0.01) > Pb (0.00) ≥ Hg (0.00), while Target Hazard Quotient decreased in the order of Pb (3.82×10^{-4}) > Cu (9.54×10^{-5}) > Zn (2.49×10^{-6}) > Hg (0.00). The values for Health Risk Index were 0.016 (Cu), 0.00 (Hg), 0.006 (Zn) and 0.034 (Pb), while that for the Health Quotients were 0.400 (Pb), 0.187 (Cu), 0.066 (Zn) and 0.00 (Hg). Similarly, all Hazard Index were less than one. This showed that the consumption of the fish from the study area had no non-carcinogenic health implication of Hg, Zn, Cu and Pb. However, the Target Cancer Risk for Pb (1.38×10^{-8}) indicated minimum cancer risks for the consumers. The ecological risks quotients also revealed that *T. guineensis* of Lagos Lagoon constituted no ecological risk to the environment since the ERQs were less than one (i.e. ERQ < 1).

Keywords: Food security; environmental pollution; food safety; heavy metals; lagoon; carcinogenic risk
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Introduction

The high contents of essential materials for growth and development (e.g. protein, lipid etc.) contained in fishes makes them important components of meals across the globe (Olawusi-Peters & Adejugbagbe, 2020). However, increased aquatic pollution as results of agricultural runoffs, domestic wastes, industrial discharges, accidental oil spills and mine drainages indicate that consumption of fish could seriously expose man to heavy metals as well as other hazardous substances. Most metals in the aquatic environment are bio-accumulative in nature (Orosun *et al.,* 2016) either passively from water or by facilitated uptake. Metals cause great danger to the various components of the food chain in any environment (Abubakar *et al*., 2014). Generally, bio-accumulation of heavy metals in fish depends on physico-chemical properties of water, metabolism and feeding patterns of fish, time of exposure, environmental or ecological conditions and intrinsic factors such as bioavailability of each metal in the environment, the process of storage, uptake and excretion mechanisms (Ajibare *et al*., 2018).

Fish that have bio-accumulated heavy metals may pose chronic health problems to its consumers. For example, there is a link between Lead (Pb), cardiovascular, muscular, and renal problems. Lead may also cause reproductive disorders in adults as well as developmental and cognitive deficiencies in children (Omobepade *et al.,* 2020). Copper (Cu) on the other hand is an essential mineral needed for various body activities and functions in humans, but may cause kidney, liver and brain disorders when concentration is above the recommended limit (Ajibare *et al*., 2018). Due to various health effects associated with heavy metals and other toxic substances stated above, global research interest has increased in the area of food safety with respect to accumulation of these hazardous substances in food including fish.

Different studies have shown that fish is a major source of dietary heavy metals and that its consumers may be exposed to adverse health as a result of the consumption (Orosun *et al.,* 2016). However, fish consumption does not always represent health risk. Thus, this study determined the Daily Metal Intake (DIM), Health Risk Index (HRI), Hazard Index (HI), Health Quotient (HQ), Target Hazard Quotient (THQ) and Target Cancer Risk (TR) of Zn, Hg, Cu, and Pb via consumption of *Tilapia guineensis* of Lagos lagoon in order to evaluate the health risks posed by the metals to its consumers.

Materials and Methods

Study Area

The Lagos lagoon (Latitude N 06^{o} 31. 048' and Longitude E 003^{o} 24. 473') is an open, shallow and tidal lagoon, with a surface area of about 208 km^2. The lagoon is located in the South-Western region of Nigeria (Onyema, 2008). The lagoon experiences more discernable brackish condition in the dry season, while low brackish/freshwater conditions are observed in the wet season due to increased river inflow (Onyema, 2008).

Collection and Identification of Samples

36 samples of *Tilapia guineensis* were collected fortnightly from June to August, 2011, using cast net. The samples were identified according to Olaosebikan & Raji (2013) and transported in ice chest to the Marine Science Laboratory of the University of Lagos. The samples were refrigerated at -4^{o}C before the determination of heavy metals.

Determination of heavy metals

The thawed fish samples were oven dried at 105°C for 72 hours to achieve constant dry weight. They were pulverized, homogenized and digested according to Javed *et al*. (2015). Dried tissue (1.0 g) of the sample was weighed into a digestion vessel. About 10 ml acidic mixture of HNO_3/$HClO_4$ in a ratio of 2:1 was then added to the sample. It was stirred with a glass rod so that it would be evenly distributed in the acid. The beaker was then placed on the digestion block in a fume cupboard for 2 hours at 150°C for digestion. The digested samples were then filtered into a 25 ml volumetric flask and made to mark with deionized water. The determination of heavy metals (Cu, Pb, Zn and Hg) in the muscles of the fish was done with flame atomic absorption spectrophotometer (FAAS) according to Javed *et al*. (2015).

For quality control/assurance, double distilled water was used throughout the study. In addition, glass wares were thoroughly prewashed with detergent, soaked in 20% nitric acid and were further rinsed using double distilled water. Samples were run in triplicates in order to ensure the accuracy of all the instruments. Moreover, to check instrumental drift, a reagent blank and a standard were done after every three samples.

Statistical Analysis

The concentration of Cu, Pb, Zn and Hg obtained in this study was subjected to descriptive statistics to determine the means and standard deviations using SPSS 20.0. The ecological and potential health risks of heavy metal consumption through *T. guineensis* were assessed based on the Ecological Risk Quotient (ERQ), Health Risk Index (HRI), Health Quotient (HQ), Hazard Index (HI), Target Hazard Quotient (THQ) and Target Cancer Risk (TR) for individual adult.

Ecological Risk Quotient (ERQ) – which numerically evaluates the associated risks in quantification and interpretation of the concentration of chemicals in aquatic environment was calculated as:

$$\mathrm{ERQ} = \frac{\text{Environmental Concentration (mg/kg)}}{\text{Recommended Limit (mg/kg)}}$$

Ogungbile *et al.,* 2022)

NOTE: Values of ERQ below 1 are unlikely to result in any negative ecological effects and would normally be considered as acceptable.

Health Risk Index (HRI) – which gives quantitative information on risk posed by each contaminant to the health of the fish consumers was calculated as:

$$\mathrm{HRI} = \frac{\text{Daily Intake of Metal (DIM)}}{\text{Reference Oral Dose (RfD)}}$$

(Olawusi-Peters & Adejugbagbe, 2020)

Where,

$$\mathrm{DIM} = \frac{\text{M} \times \text{Conversion Factor} \times \text{Daily intake of fish}}{\text{Average body weight}}$$

(Isibor & Imoobe, 2017)

M is the concentration of metal in sample (mg/kg), Conversion Factor is 0.085, daily fish intake of 48 g/day was estimated as the fish consumption rate in Nigeria (Omobepade *et al.*, 2020), while the average body weight of consumers was 60 kg (Olawusi-Peters & Adejugbagbe, 2020). Reference Oral Doses (RfD) was 0.040, 0.300, 0.004, and 0.0003 mg/kg/day for Cu, Zn, Pb and Hg respectively (USEPA, 2011; Olawusi-Peters & Adejugbagbe, 2020).

Health Quotient – which estimates the hazard heavy metal could have on the human population in their later life) was determined as:

$$\mathrm{HQ} = \frac{\mathrm{W} \times \mathrm{M}}{\mathrm{RfD} \times \mathrm{B_o}}$$

(Omobepade *et al.*, 2020)

Where,

W is the dry weight of the fish consumed per/day, M is concentration of metal in the edible parts of fish (mg/kg), RfD is the reference oral dose.

Target Hazard Quotient (THQ) – which is a dimensionless quantity that defines the exposure duration and the non-carcinogenic risk within the period was calculated as:

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

(Orosun *et al.*, 2016)

Where FIR is the fish ingestion rate (48 g/person/day); EF is the metal Exposure Frequency (350 days/year); ED is the metal Exposure Duration (54 years, equivalent to the average life expectancy of the Nigerian population)

(Omobepade *et al*., 2020); BW is the average Body Weight (60 kg) (Olawusi-Peters *et al.,* 2019); and ATn is the average Exposure Time for non-carcinogens (19710). If the THQ value is greater than 1, the exposure is likely to cause obvious adverse effects (USEPA, 2011).

Hazard Index (HI) – which is expressed as the sum of the target hazard quotients was calculated as:

$$HI = \sum_{n-1}^{n} THQ$$ (Núñez *et al*., 2018)

Where,

n = number of heavy metals examined, THQ = unit THQ of *ith* heavy metal.

NOTE: Values below one for Target Hazard Quotient (THQ), Health Quotient (HQ), Health Risk Index (HRI) and Hazard Index (HI) are not likely to cause any chronic systemic adverse effects in a given lifetime of exposure and is generally considered as acceptable.

Target Cancer Risk (TR) – which indicates the carcinogenic risk (USEPA, 2011) was calculated as:

$$TR = \frac{M \times FIR \times CPSo \times EF \times ED}{Bw \times ATn} \times 10^{-3}$$

M, FIR, ED, EF, BW and ATn are as explained earlier. CPSo is the Carcinogenic Potency Slope oral (mg/kg bw-day−1). TR values for intake of only Pb was calculated because Hg, Cu and Zn do not have carcinogenic effects (USEPA, 2012). Carcinogenic Potency Slope oral (CPSo) for Pb as obtained from the integrated risk information system database was 0.009 (USEPA, 2012). According to New York State Department of Health (NYSDOH, 2007), the TR values are classified as $TR \geq 10^{-1}$ = 'Very High'; 10^{-3} to 10^{-1} = 'High'; 10^{-4} to 10^{-3} = 'Moderate'; $TR \leq 10^{-6}$ = 'Low'.

Results and Discussion

Concentration of heavy metals

The mean concentrations of Mercury (Hg), Zinc (Zn), Copper (Cu), and Lead (Pb) in *T. guineensis* of Lagos lagoon, Nigeria are presented in Table 1. Metal distribution in the fish was in the order of Zn > Cu > Pb > Hg. The concentrations of Zn ranged from 0.006±0.00 to 0.0575±0.07 mg/kg with a mean of 0.0248±0.04 mg/kg. Cu content in the fish ranged from 0.0070±0.00 to 0.0135±0.01 mg/kg with mean of 0.0093±0.01 mg/kg. The average concentration of Pb was 0.0005±0.00 mg/kg with a range of 0.000±0.00 mg/kg to 0.0015±0.00 mg/kg, while Hg was not detected (ND) and was regarded as 0.000 ± 0.00 mg/kg throughout the study.

The Daily Intake of Metals (DIM), Health Risk Index (HRI), Health Quotient (HQ), Target Hazard Quotient (THQ) and Target Cancer Risk (TR) of Zn, Hg, Pb, and Cu through consumption of *T. guineensis* in Lagos lagoon is presented in Table 2. The trend of the DIM was in the order of Zn > Cu > Pb ≥ Hg with values of 0.02, 0.01, 0.00 and 0.00 respectively. The HRI of the fish species analysed for the metals were all less than 1 (HRI < 1) with values of 0.016, 0.00, 0.006 and 0.034 for Cu, Hg, Zn and Pb respectively. This indicated that no negative health effect would arise as a result of the consumption of the fish. The health quotient (HQ) for Pb (0.400), Cu (0.187), Zn (0.066) and Hg (0.00) indicated that there would be no health risk in the later life of the consumers.

The trend of THQ values for consumers decreased in the order of Pb > Cu > Zn > Hg with values of 3.82×10^{-4}, $9.54 \times 10^{-}$

^{5}, 2.49×10^{-6} and 0.00 respectively. This also showed that no health implication may arise due to consumption of the fish. The target cancer risk for Pb was 1.38×10^{-8} which indicated low cancer risks for the consumers of the fish. The ecology risk quotient (ERQ) in this study revealed no ecological risk from the bio-accumulation of Cu, Hg, Zn and Pb since the values were lesser than 1 (ERQ < 1) for Cu (0.003), Hg (0.000), Zn (0.001) and Pb (0.000). The Hazard Index (HI) of heavy metals in *T. guineensis* of Lagos lagoon as presented in Figure 1 shows that no systemic adverse health risk would be experienced by the consumers of the fish since all values were less than 1 (HI < 1).

TABLE 1

Concentrations of heavy metals (Copper, Mercury, Zinc and Lead) in Tilapia guineensis from Lagos lagoon

Month	Cu (mg/kg)	Hg (mg/kg)	Zn (mg/kg)	Pb (mg/kg)
June	0.0075 ± 0.00	ND	0.0060 ± 0.00	0.0000 ± 0.00
July	0.0070 ± 0.00	ND	0.0110 ± 0.00	0.0000 ± 0.00
August	0.0135 ± 0.01	ND	0.0575 ± 0.07	0.0015 ± 0.00
Grand Mean	0.0093 ± 0.01	ND	0.0248 ± 0.04	0.0005 ± 0.00
FEPA (2003), WHO (2007)	3.00		30.00	2.00

TABLE 2

Risks assessment of Copper, Mercury, Zinc and Lead in Tilapia guineensis from Lagos lagoon

Index	Cu	Hg	Zn	Pb
Daily Metal Intake	0.001	0.000	0.002	0.000
Health Risk Index	0.016	0.000	0.006	0.034
Health Quotient	0.187	0.000	0.066	0.400
THQ	9.54E-05	0.000	2.49E-06	3.82E-04
TR	*	*	*	1.38E-08
Ecological Risk Quotient	0.003	0.000	0.001	0.000

*Cannot be computed because Carcinogenic Potency Slope oral (CPSo) has not been established

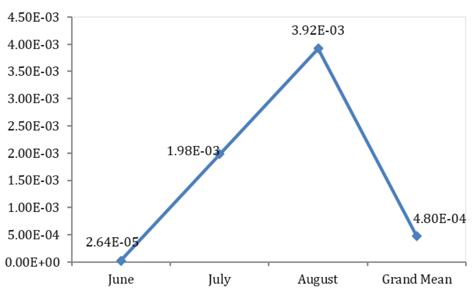

Fig. 1: Hazard Index (HI) of *Tilapia guineensis* from Lagos lagoon

The concentration of copper (Cu) observed in the tissues of *Tilapia guineensis* in this study was below the Federal Environmental Protection Agency (FEPA 2003) guideline of 3 mg/kg, while the mean concentrations of lead (Pb) and mercury (Hg) were below the permissible limit of 2 mg/kg recommended by FEPA (2003) and World Health Organisation (WHO, 2007). Furthermore, when compared with the 30mg/kg recommended as maximum limit for

Zn in fish (WHO, 2007), the observed concentrations were low. The observed trend of metal bioaccumulation in the fish was similar to the observations of Adedeji & Okocha (2011) and Ajibare *et al*. (2018) who reported decreasing order of Zn > Fe > Mn > Cu > Pb > Cd in *Macrobrachium macrobrachion, Macrobrachium vollenhovenii* and *Nematopalaemon hastatus* from Epe Lagoon, Asejire River and coastal waters of Ondo State, Nigeria respectively.

The daily intake of metals (DIMs) gives information on the relative bio-availability of metal but does not consider possible metabolic ejection of the metals. It however tells the probable ingestion rate of a given metal (Olawusi-Peters *et al*., 2019). This study revealed that Daily Intake of Metals through fish for Cu (0.001), Zn (0.02), Pb (0.00) and Hg (0.00) were significantly lower than the recommended Oral Reference Dose (RfD) for the metals (USEPA, 2011). According to New York State Department of Health (NYSDOH, 2007), if the ratio of DIM of heavy metal to its RfD was equal to or less than the RfD, then there is minimum risk. However, if it is greater than up to five times the RfD, then there is low risk. Also, if it is greater than up to 10 times the RfD, then there is moderate risk. But, if it is greater than 10 times the RfD, it indicates high risk. Therefore, since the concentrations of the observed metals were lower than the RfD, there was minimum or no potential health hazard to the consumers (NYSDOH, 2007). Similarly, the HRI for Cu, Hg, Zn and Pb in *Tilapia guineensis* were all less than 1, indicating that the consumption of *Tilapia guineensis* from Lagos lagoon posed minimum or negligible health risk to the consumers. This observation was similar to the findings of Abubakar *et al*. (2014), Isibor & Imoobe (2017), Olawusi-Peters *et al*. (2019), Omobepade *et al*. (2020) and Olawusi-Peters & Adejugbagbe (2020) who reported no (or negligible) health risks on different fishes and shellfishes in their independent researches.

The results of Hazard Quotient (HQ) showed values below 1 for all the recorded metals. This observation was similar to the findings of Omobepade *et al*. (2020) who observed HQ values of less than 1 in *Nematopalaemon hastatus* of coastal waters of Ondo state, Nigeria and the authors concluded that the metals would not pose any serious health hazards on the consumers in the later life. Moreover, the Target Hazard Quotient (THQ) is commonly used to assess the non-carcinogenic risks associated with prolonged exposure to dietary metals (Orosun *et al*., 2016). THQ does not measure risk, but it shows the extent of concern and its value should not exceed 1. THQ values greater than 1 means potential (non-carcinogenic) risks to the exposed population. Also, NYSDOH (2007) reported that if THQ is greater than one but less than five (i.e. 1 < THQ < 5), the risk is low; if THQ is greater than five but less than 10 (5 < THQ < 10), the risk is moderate; however, if THQ is greater than 10 (THQ > 10), the risk is high. The THQs observed for the metals in this study were all lesser than 1, and this suggests that health effects associated with the metals were unlikely to occur.

The Hazard Index (which is the numerical sum of THQs) values of Zn, Hg, Pb and Cu ranged from 2.64×10^{-5} to 3.92×10^{-3}, suggesting no significant health risks for the consumers since the recorded values were all less than 1. According to Abubakar *et al*. (2014), HI should not exceed 1 otherwise it implied significant health risks to consumers. Similarly, carcinogenic risks describe the incremental probability of an individual to develop cancer during a lifetime exposure to potential carcinogens of any level. Therefore, the Target Cancer Risks (TR) of less or equal to 10^{-6} (TR $\leq 10^{-6}$) as observed for Pb in this study is considered as low or no carcinogenic risk (NYSDOH, 2007).

Also, the results of ecological risk quotient showed that no observed metal posed ecological risk to the environment. However, given that Cu, Zn, and Pb were found in the fish throughout the study, there may be a potential ecological risk if indiscriminate effluent discharge is not controlled. Prolonged exposure to these heavy metals can have detrimental ecological effects such as high mortality of juvenile fish, reduced breeding potential of adult fish, discomfort, disorientation, inability to find food, decrease in organism's fitness, imbalance, and failure of multi-system organs, among others. Additionally, the ecological impact could include reproductive consequences such as spawning blockage, deformities in newly-hatched fry, decreased egg production per female, decreased young survivability, and other effects at low levels of Cu, as previously observed by Ajibare *et al*. (2018).

Conclusion and Recommendation

This study has revealed that the Daily Intake (DIM) of Zn, Hg, Pb and Cu through consumption of *T. guineensis* in Lagos lagoon, Nigeria, was within the recommended limits. The Target Hazard Quotient (THQ), Hazard Index (HI), Health Quotient (HQ), and Health Risk Index (HRI) all showed that there was no probable non-carcinogenic health risk for fish consumers. The consumption of *T. guineensis* in the study area was not related with any carcinogenic risk, according to the Target Cancer Risk (TR), which quantifies the degree of exposure to carcinogenic risk. The Ecological Risk Quotient (ERQ) indicated that T. *guineensis* was not ecologically risky to the environment; consequently, its consumption does not pose any carcinogenic and non-carcinogenic risk to any population. In conclusion, since Zn, Cu, and Pb were found in the studied fish species, there is need for routine monitoring of heavy metals in the Lagos lagoon in order to ensure food security and safety in and around the study area.

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