



## Human Health Risk Assessment in Relation to Heavy Metals in The Atlantic Mackerel (*Scomber scombrus*, L. 1758) Sold in Some Major Markets in Benin City, Nigeria

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### ABSTRACT

The study determined the concentrations of Zinc (Zn), Copper (Cu), Lead (Pb), Cadmium (Cd) and Chromium (Cr) in the Atlantic Mackerel (*Scomber scombrus*) of mean length  $25.54 \pm 1.35$  cm and mean weight  $195.22 \pm 1.15$  g, from selected markets in Benin city, in order to evaluate their suitability for human consumption, employing Atomic Absorption Spectrometric technique. The summary statistics revealed that the mean concentrations (dry weight) of the aforesaid heavy metals in fish ranged from 0.05 mg/kg (Cd) to 15.93 mg/kg (Zn). The mean concentration of Zn ranged from 13.33 mg/kg at *Oba* market to 19.22 mg/kg at *Ramat* market while for Cu, the range was from 7.453 mg/kg (*Oba* market) to 8.670 mg/kg (*Uselu* market). The mean concentration of Pb ranged from 0.03 mg/kg at *Uselu* market to 0.15 mg/kg at *Ramat* market while for Cr, the range was from 2.33 mg/kg (*Oba* market) to 3.05 mg/kg (*Uselu* market). The mean concentration of Cd ranged from 0.03 mg/kg at *Oba* market to 0.06 mg/kg at *Ramat* and *Uselu* markets. The estimated daily intake (EDI) values for heavy metals in fish ranged from 0.002 mg/person/day for Cd to 0.64 mg/person/day for Zn. The study revealed the mean concentrations of Zn, Cu, Pb and Cr in *S. scombrus* were well below established thresholds for heavy metals in fishery products. However the toxic/hazard quotient (TQ) values revealed that Cd was the metal that presented a potential risk/hazard to the fish consuming public.

### Introduction

Heavy metals have been recognized as pollutants of environmental concern owing to their toxicity both to wildlife and man. In addition, they have been regarded as a threat to ecological integrity. [1] In this realm, studies abound on the concentrations of heavy metals in various environmental matrices in Nigeria. For example, water, [2] sediment, [3, 4, 5] finfish, [6] shellfish [7] and flora. [8] Most of these studies recognized the contribution of anthropogenic impact to the levels of environmental pollution. The Atlantic Mackerel (*Scomber scombrus*, Linnaeus, 1758), is widely consumed in Nigeria and is one of the most popular species of frozen fish in fish markets which is imported into the country typically in packs of 20 kg net weight from European countries, such as Norway. Distribution of such fish is usually from large refrigerated rooms (cold rooms), to fish mongers who buy from such sources for onward sale to the public. *S. scombrus* is commonly sold in fish markets in Benin city. This study principally ascertained the concentrations of Zn, Cu, Pb, Cd and Cr in the scombrid fish species, from selected markets in Benin city, in order to evaluate their suitability for human consumption, against the backdrop that such markets are located along busy vehicular routes which may conceivably expose fish to atmospheric deposition of chemical elements such as heavy metals. Possible sources of Zn are batteries, refuse, pesticides, alloys, dyes, fossil fuels, electroplating and metallurgical processes while sources of Cu include paints, copper pipes, copper wires, fertilizers and pesticides. Sources of Pb are batteries, alloys, solders, fossil fuels, plastics and pesticides while sources of Cd include batteries, fossil fuels, fertilizers, plastics, alloys and paints. Some sources of Cr include paints, textiles, automobile parts, fertilizers and alloys. [2]

Comparisons of heavy metal levels in *S. scombrus* have been made with reference to the Food and Agriculture Organization of the United Nations (FAO) and Commission Regulation (EC) maximum limits for heavy metals in fishery products.

### Materials and Methods

#### Description of study area

The study was conducted in Benin City, Edo state, Nigeria, gridlocked within Latitude  $6^{\circ} 20' 0''$  N and Longitude  $5.80^{\circ}$  E (Figure 1). The city is located within the equatorial region, having two climatic regimes. The wet season spans from April to October while the dry season spans from November to March. Annual temperature ranges from  $21^{\circ}\text{C}$  to  $34^{\circ}\text{C}$  with humidity ranging from 68% to 96%. The population of the city according to the 2006 National census is One million, one hundred and forty-seven thousand, one hundred and eighty-eight (1,147,188) while the vegetative belt is within the tropical rain forest. [9] The city is approximately 40 km north of the Benin River and it is situated 320 km by road east of Lagos state. Benin is the centre of Nigeria's rubber industry and palm oil processing. [10] The city plays host to several cottage industries, elementary and secondary schools, hospitals and tertiary institutions of learning, including the University of Benin. The markets selected for the study were *Uselu* market, *Oba* market, *Ramat* market and *New Benin* market. The Atlantic Mackerel (Plate 1) can readily be found in the aforesaid markets where it is referred to as "*scubia*" in the local parlance. *Uselu* market is located along the busy *Uselu-Ugbowo* road of the city while *Oba* market is situated at the Ring road axis. *Ramat* market is located along the very busy *Benin-Agbor* road while *New Benin* market is situated at the intersection of *New Lagos* and *Mission* roads of the city.

#### Collection of fish samples

Fish samples were purchased from the markets fortnightly between November 2015 and April 2016. They were transported to the laboratory within 24 hours inside a Thermolineo® ice chest in order to retain their integrity. A total of 12 fishes were obtained from each market

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but only 40 fish samples of similar weight and size (representing 10 from each market) were eventually used for the study.

**Laboratory procedures**

All reagents used were of analytical grade (BDH, Poole, England). All glassware was washed with detergent and rinsed with distilled water before they were soaked in potassium permanganate solution overnight. They were then rinsed with distilled water before use to ensure high precision results. Fish samples were weighed whole in grams using an ATOM A-110C® electronic compact scale while their total lengths (in centimeters) were recorded using a translucent plastic ruler. The musculature of fish was excised with a stainless-steel dissecting lancet and oven dried at a temperature of 80 ° C for 48 hours in a Surgifield-Uniscope® (SM 9023 model) laboratory oven. Each dried sample was ground separately using a porcelain mortar and pestle and kept in sealed plastic vials prior to digestion. Digestion was carried out using the double acid method. [11] The validity of the optimized digestion procedure was assured by spiking the samples with a standard of known concentration of the analyte metals by subtracting blank values that were similarly prepared. Standard metal solutions were used to test the recovery of the metals in the sample to specified levels. The digests were stored in 100 ml plastic reagent bottles for two weeks before Atomic absorption spectrophotometric (AAS) analysis. Fish digests were analyzed for Pb, Cr, Cu and Cd by means of an Atomic Absorption Spectrophotometer (Unicam® 696 series) equipped with solar software using air acetylene flame. Concentrations of metals in fish were expressed in mg/kg (Dry weight).

**Estimated daily intake (EDI) of heavy metals by man**

The daily intake of metals was calculated in order to estimate the daily loading of metals into the body system of man via the consumption of fish. [12]

$$EDI = \frac{40g/person/day \times HM (mg/kg)}{1000g/kg}$$

where: 40g/person/day = Estimated consumption of fishery products in Niger Delta

HM = Mean concentration of heavy metal in fish species.

EDI = mgHM/person/day

**Estimated annual intake (EAI) of heavy metals by man**

Borne out of the EDI, the estimated annual intake (EAI) of heavy metals was calculated;

$$EAI = EDI * 365 \text{ days} = X \text{ mgHM/person/year}$$

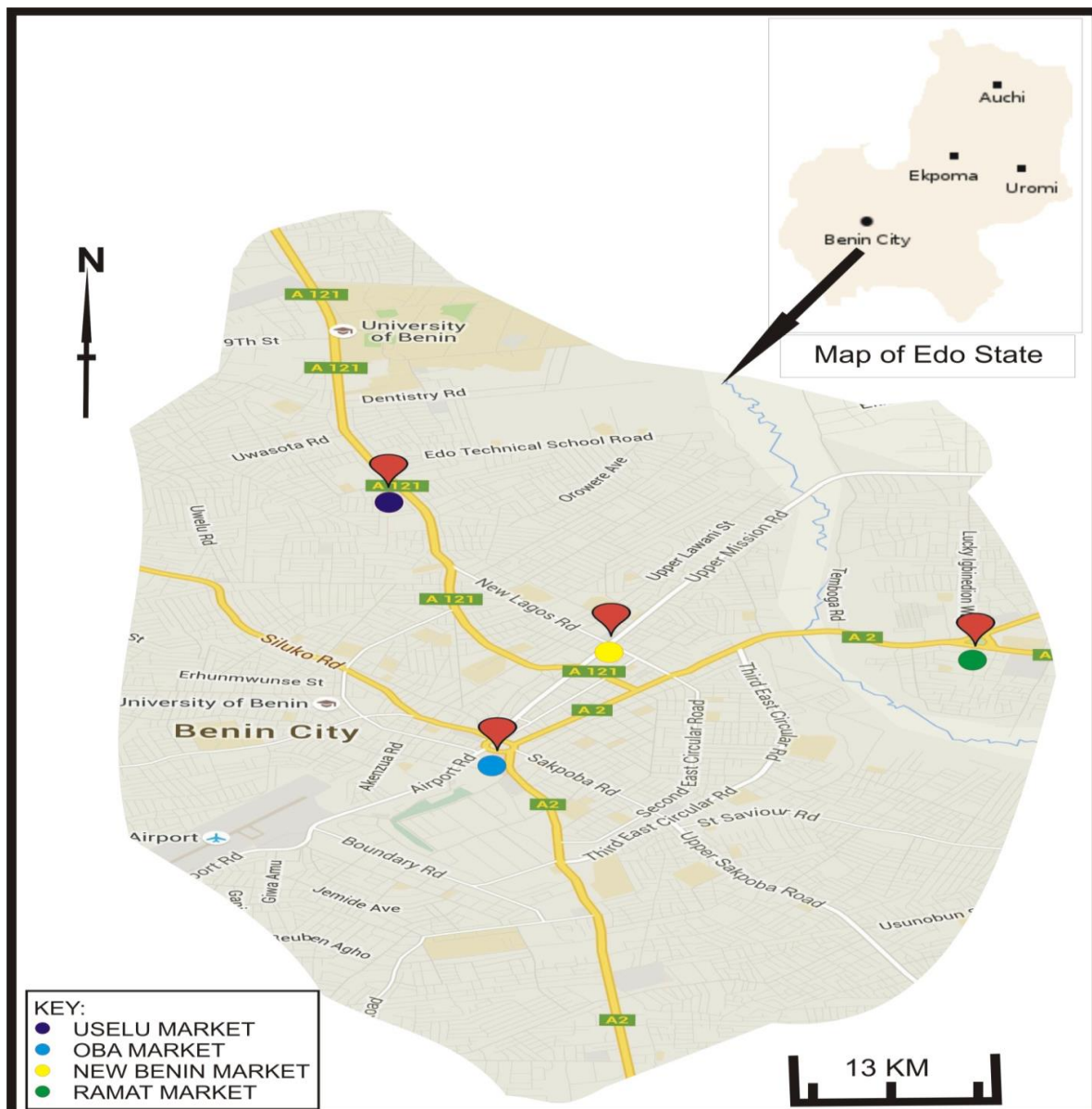
**Calculation of theoretical maximum daily intake (TMDI) for heavy metals**

The TMDI is used for making a first estimate of heavy metal residue intake. It is calculated by multiplying the established maximum limit by the estimated average daily regional consumption for each food item and then summing the products. [13]

$$TMDI = \sum ML^1 * F^1$$

where: ML = Maximum limit for a given food;

F = Per capita/ regional food consumption.



**Figure 1:** Map of the study area  
Source: Google maps (2016)

**Evaluation of margin of exposure (MOE) for heavy metals**

The MOE is usually applied in cumulative risk assessments. It is essentially a margin between a reference dose for a heavy metal and the calculated or actual exposure for the same metal. <sup>[14]</sup>

$$\text{MOE} = \frac{\text{Reference dose}}{\text{Calculated or actual exposure}}$$

**Calculation of total toxicity of mixtures (TTM) index for heavy metals**

Whether or not a mixture of metals in a particular medium exceeds the quality guideline value for that medium, was determined by applying the TTM index. <sup>[15]</sup>

$$\text{TTM} = \sum (C^i/GV^i)$$

where:  $C^i$  = Concentration of the 'i<sup>th</sup>' component of mixture

$GV^i$  = Guideline value for the 'i<sup>th</sup>' component

$\text{TTM} > 1$  = The mixture has exceeded the Guideline value

**Toxicity/hazard quotient (TQ) for heavy metals**

The Toxicity/hazard quotient (TQ) for chemical elements is a comparison of the measured concentration of site-related elements in ecological matrices with specific health-based criteria. <sup>[16]</sup>

$$\text{TQ} = \frac{\text{Concentration of heavy metal in fish sample}}{\text{Health based criteria}}$$

**Statistical analysis**

Statistical software (GENSTAT® version 13.3 for Windows) was used for analyzing data. One-way analysis of variance (ANOVA) was used to test for significant differences between mean values of heavy metals at 5% probability level. Duncan Multiple Range Test (DMRT) was used to separate significant means. Microsoft Excel (for Windows 2010), was used for all graphical presentations.

**Results and Discussion**

As presented in Table 1, the summary statistics revealed that the mean concentrations of the investigated heavy metals in fish ranged from 0.05 mg/kg (Cd) to 15.93 mg/kg (Zn). In Table 2, the mean concentration of Zn ranged from 13.33 mg/kg at *Oba* market to 19.22 mg/kg at *Ramat* market while the mean concentration of Cu, ranged from 7.453 mg/kg (*Oba* market) to 8.670 mg/kg (*Uselu*). The mean concentration of Pb ranged from 0.03 mg/kg at *Uselu* market to 0.15 mg/kg at *Ramat* market while the mean concentration of Cr, ranged from 2.33 mg/kg (*Oba* market) to 3.05 mg/kg (*Uselu*). The mean concentration of Cd ranged from 0.03 mg/kg at *Oba* market to 0.06 mg/kg at *Ramat* and *Uselu* markets. There were no significant differences ( $P > 0.05$ ) in the mean concentrations of metals in fish between markets.

As shown in Table 3, the mean concentration of Zn in fish ranged from 12.00 mg/kg in December to 21.99 mg/kg in February while the mean concentration of Cu in fish ranged from 6.51 mg/kg in December to 8.75 mg/kg in January. The mean concentration of Pb in fish ranged from 0.08 mg/kg in November/January to 0.14 mg/kg in February while the mean concentration of Cr in fish ranged from 1.69 mg/kg in December to 3.345 mg/kg in November. The mean concentration of Cd in fish ranged from 0.00 mg/kg in December to 0.50 mg/kg in April. There were significant differences ( $P < 0.05$ ) in the mean concentrations of the metals between months, with the exception of Cd.

The estimated daily intake (EDI) values for heavy metals in fish ranged from 0.002 mg/person/day for Cd to 0.64 mg/person/day for Zn while the estimated annual intake (EAI) ranged from 0.73 mg/person/year (Cd) to 234 mg/person/year (Zn) as shown in Figure 2. The theoretical maximum daily intake (TMDI) value was 3614 mg/person/day (Figure 3). The margin of exposure (MOE) for heavy metals ranged from 1.0 for Cd to 3.82 for Cu (Figure 4) while the total toxicity of mixtures (TTM) for the study was 2.21 (Figure 5). The toxicity/hazard quotient (TQ) values for heavy metals peaked at 1.0 for Cd (Figure 6) while the heavy metal quota in fish ranged from 0.18 % for Cd to 59.84 % for Zn, as presented in Figure 7.

**Table 1:** Summary statistics for heavy metals (mg/kg) in *Scomber scombrus*

Metal	Mean	Minimum	Maximum	SD
Zn	15.93	5.33	40.00	4.32
Cu	7.85	3.73	10.12	0.77
Pb	0.10	0.00	0.32	0.06
Cd	0.05	0.00	0.25	0.05
Cr	2.69	0.58	5.00	0.53

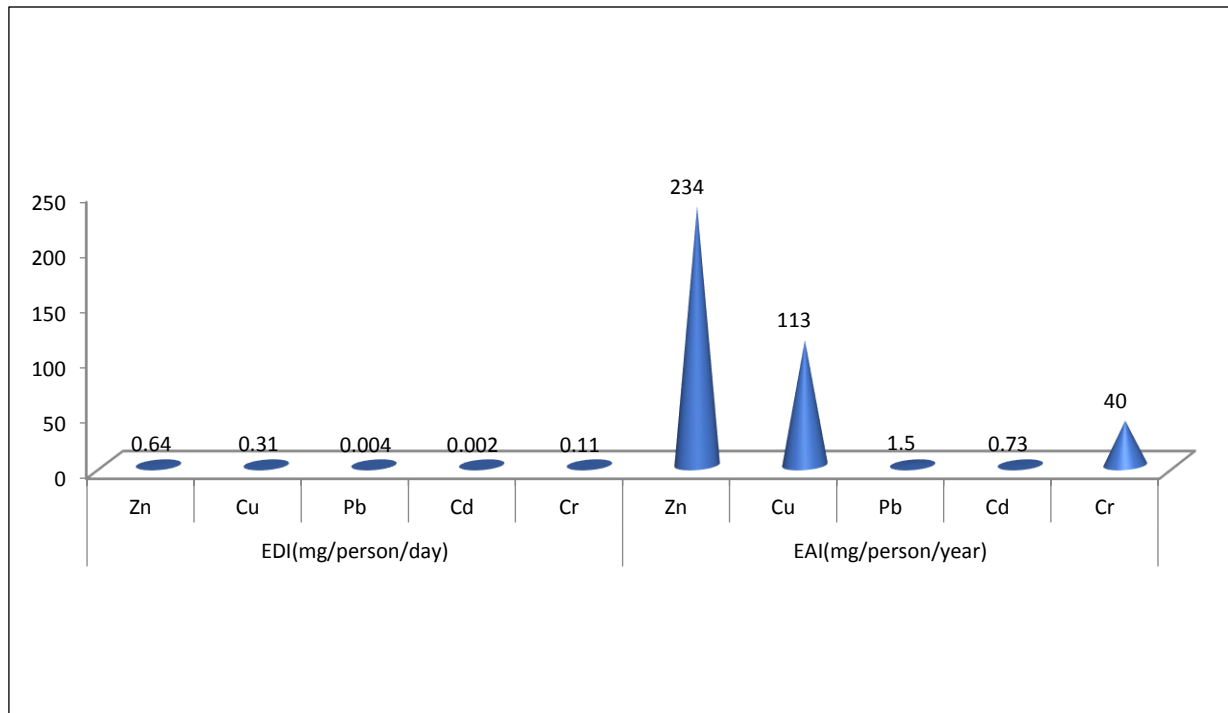


**Plate 1:** Atlantic Mackerel (*Scomber scombrus*)

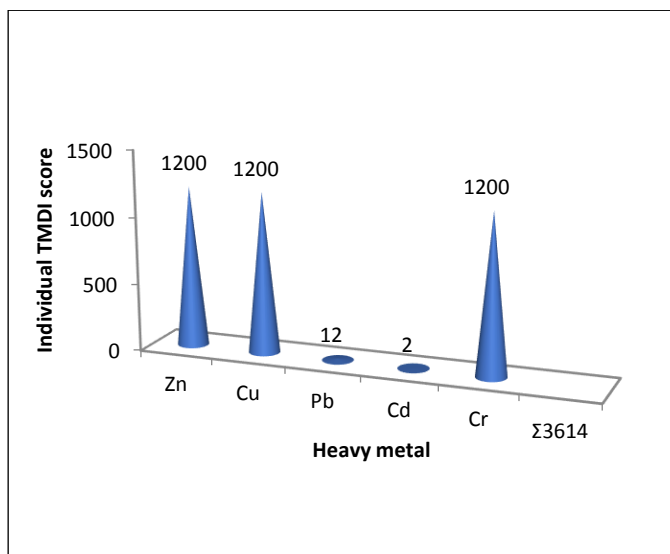
The lowest heavy metal quota (7.5%) and highest quota (37.5%) were calculated for Pb at *Uselu* and *Ramat* markets respectively (Figure 8).

The mean concentrations of heavy metals in fish varied amongst the investigated markets and months. For example, the mean concentrations of Zn, Pb and Cd were highest at *Ramat* market while the mean concentrations of Cu, Cr, Cd and Zn were lowest at *Oba* market. Quota-wise Zn, Pb and Cd were dominant in fish at *Ramat* market while Cu and Cr were dominant in fish at *Uselu* market. Similarly, the mean concentrations of Zn, Cu, Cr and Cd were lowest in the month of December while the mean concentrations of the investigated metals were higher in the dry months with the exception of Cd. It was observed that there were no significant differences ( $P > 0.05$ ) in the mean concentrations of the metals between markets, indicating that the fish could have been sourced from the same distributor or source. However, there were significant differences ( $P < 0.05$ ) in the mean concentrations of the metals between the months, with the exception of Cd. This observation may have been due to seasonal variations which could invariably influence the levels of metals in fish. The observed heavy metal profile in fish was  $\text{Zn} > \text{Cu} > \text{Cr} > \text{Pb} > \text{Cd}$ . The amount and proportion of metals in fish reflect the dynamics of bioavailability and bioaccumulation. Fish will only take up contaminants including metals from their aquatic media when such xenobiotics are bioavailable. <sup>[17]</sup> In addition, when the rate of retention of metals in the fish body is greater than the rate of elimination, bioaccumulation will manifest. <sup>[18]</sup> It has been reported that marine species such as fish are able to readily absorb metals and their bodies regulate to accommodate their presence. <sup>[19]</sup> Such metals are stored in fatty issue and will bioaccumulate if fish is exposed to further contamination. The EDI and EAI values for heavy metals were dominated by Zn and were the least for Cd. This observation can be attributed to the fact that Zn had the highest proportion of the metals in fish while Cd had the least. In order words, the heavy metal rank profile in fish dictates the trend in which EDI and EAI values would follow. The individual TMDI scores recorded for metals in this study were highest for Zn, Cu and Cr and the least for those of Pb and Cd. This observation can be attributed to the fact that the established thresholds for Zn, Cu and Cr in fish are much higher than that of Pb and Cd. For example, the Food and Agriculture Organization of the United Nations, <sup>[20]</sup> have set maximum limits of 30 mg/kg for some essential elements in fish while the Commission Regulation, EC [21], has set maximum limits of 0.30 mg/kg for Pb and 0.05 mg/kg for Cd. However, the overall TMDI value recorded for the study was 3614 mg/person/day, taking into account the per capita consumption and maximum limits for metals in fish. The per capita consumption of fish in the Niger Delta has been reported to be 40 grams. <sup>[12]</sup> This same figure was adopted since Benin city is an integral part of the Niger Delta region. The MOE values followed a rank profile of  $\text{Cr} > \text{Cu} > \text{Pb} > \text{Zn} > \text{Cd}$ . From this finding, Cr is the metal that has the potential for future risk and should thus be closely monitored.

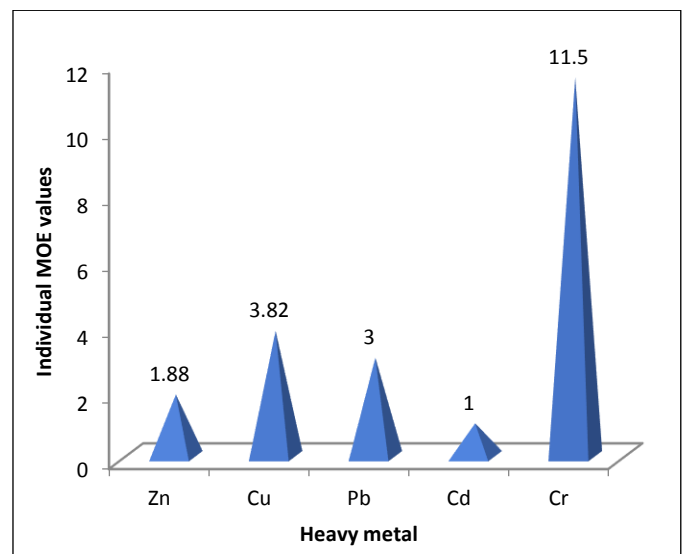




**Figure 2:** Estimated daily intake (EDI) and estimated annual intake (EAI) values for heavy metals



**Figure 3:** Theoretical maximum daily intake (TMDI) for heavy metals



**Figure 4:** Margin of exposure (MOE) for heavy metals

The TQ values revealed that Cd is the metal that presents a risk/hazard to the fish consuming public as a TQ value of 1 was calculated. This observation is further confirmed by the fact that the mean concentration of the metal in fish fell on exactly the EC maximum limit of 0.05 mg/kg, as previously highlighted. In a study on frozen fish in Ibadan city, Nigeria, the presence of Cd, Cu and Hg was reported in *S. scombrus*.<sup>[22]</sup> The aforesaid study revealed that the heavy metals investigated in the major organs and flesh of fish were all above the limits recommended by World Health Organization, thereby revealing their unfitness for human consumption. Mean concentrations of 0.07 mg/kg and 2.81 mg/kg for Cu and Zn respectively were reported in *S. scombrus* obtained from Gwagwalada market, Abuja, Nigeria while Pb was not detected in such fish.<sup>[23]</sup> The heavy metal content in *S. scombrus* from markets in Ile- Ife, Nigeria was reported for Pb, Cr and Cd to be 0.44 mg/kg, 0.33 mg/kg and 0.08 mg/kg respectively.<sup>[24]</sup>

From the TTM value of 2.21, it is suggestive that the maximum limits or guideline values for heavy metals in fish were partially exceeded owing to the fact that Cd is the metal that presented the greatest risk in the study. Apart from the metal load already in fish, additional loading of metals can come from atmospheric deposition. This is particularly so against the backdrop that the fish markets covered in this study are all situated along busy vehicular routes and are therefore conceivably exposed to automobile exhaust fumes that are spewed into the atmosphere. As earlier noted, fossil fuels are sources of heavy metals especially Cd and Pb. At elevated concentrations Cd is acutely toxic and can cause severe renal damage with renal failure. Cadmium also causes acute gastroenteritis which closely mimics the gastroenteritis caused by micro-organisms. The half-life of cadmium in the body is several decades; hence, it is important to avoid exposure. Cadmium poisoning is very difficult to treat due to rapid and irreversible uptake by the kidneys. Immediate medical attention is usually sought when Cd poisoning is suspected.<sup>[25, 26]</sup>

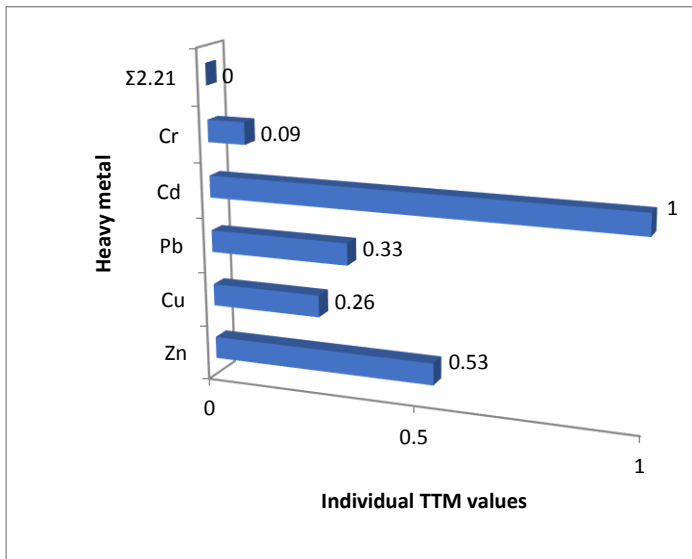
**Table 2:** Mean heavy metal concentration (mg/kg) in *Scomber scombrus* by market

Market	Zn	Cu	Pb	Cr	Cd
Ramat	19.22±1.33	7.48±1.54	0.15±0.07	2.53±1.07	0.06± 0.02
New Benin	15.59±2.45	7.78±0.97	0.10 ±0.09	2.72±0.84	0.05±0.01
Oba	13.33±0.98	7.45±1.24	0.12±0.05	2.33±0.97	0.03 ± 0.01
Uselu	15.59±1.67	8.67±0.85	0.03±0.01	3.05±0.85	0.06 ±0.01

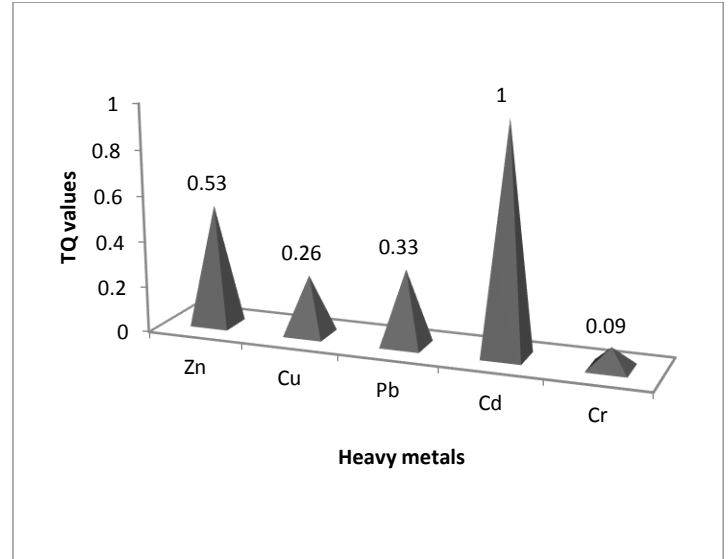
**Table 3:** Mean heavy metal concentration (mg/kg) in *Scomber scombrus* by months

Month	Zn	Cu	Pb	Cr	Cd
November	14.83±2.21 <sup>b</sup>	6.72±1.45 <sup>b</sup>	0.08±0.001 <sup>b</sup>	3.34±1.05 <sup>a</sup>	0.09±0.001 <sup>a</sup>
December	12.00±1.97 <sup>b</sup>	6.51±1.65 <sup>b</sup>	0.10±0.002 <sup>b</sup>	1.69±0.92 <sup>b</sup>	0.00±0.00 <sup>a</sup>
January	14.78±2.65 <sup>b</sup>	8.75±2.21 <sup>a</sup>	0.08±0.001 <sup>b</sup>	2.78±0.76 <sup>a</sup>	0.03±0.001 <sup>a</sup>
February	21.99±2.25 <sup>a</sup>	8.45±0.78 <sup>a</sup>	0.14±0.001 <sup>a</sup>	2.08±0.55 <sup>b</sup>	0.08±0.001 <sup>a</sup>
March	18.88±1.27 <sup>a</sup>	8.04±1.07 <sup>a</sup>	0.13±0.002 <sup>a</sup>	3.12±0.34 <sup>a</sup>	0.06±0.001 <sup>a</sup>
April	13.91±2.28 <sup>b</sup>	8.63±1.28 <sup>a</sup>	0.09±0.001 <sup>a</sup>	2.95±0.75 <sup>b</sup>	0.50±0.002 <sup>a</sup>

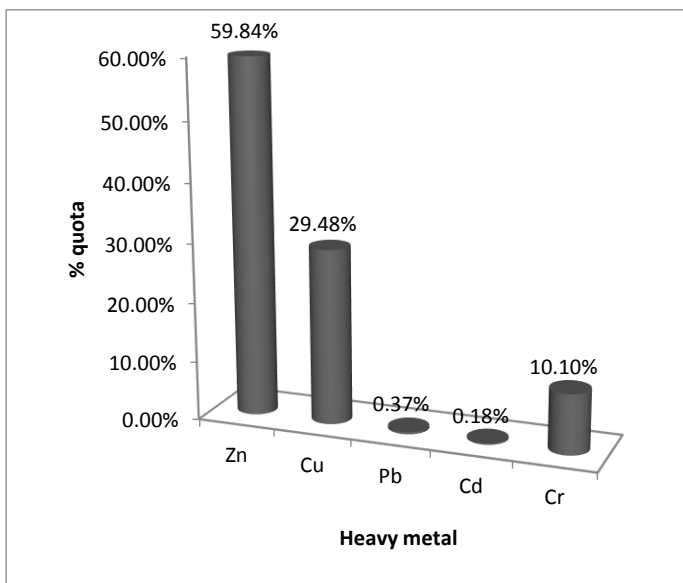
Means with similar superscripts are not significantly different (P > 0.05). Vertical comparisons only



**Figure 5:** Total toxicity of mixtures (TTM) index for heavy metals



**Figure 6:** Toxicity/hazard quotient (TQ) for heavy metals



**Figure 7:** Heavy metal quota in *Scomber scombrus*

**Conclusion**

The study revealed the mean concentrations of Zn, Cu, Pb and Cr in *Scomber scombrus* were well below established maximum limits for heavy metals in fishery products. However the TQ values revealed that Cd is the metal that presents a risk/hazard to the fish consuming public. The investigated scombriid fish must thus be consumed with caution in order to prevent Cadmium poisoning. In this realm, it is further advocated that Health Inspection Officers from the Ministry of Health and Environmental Monitoring Officers from the Ministry of Environment, be mandated to jointly spot check fishery products in these markets, in order to ensure and promote the fitness of such products for human consumption. Frozen fish should also be checked for levels of heavy metal contamination at the point of importation since the experimental fish was an imported species.

**Conflict of interest**

The authors declare no conflict of interest.

**Acknowledgement**

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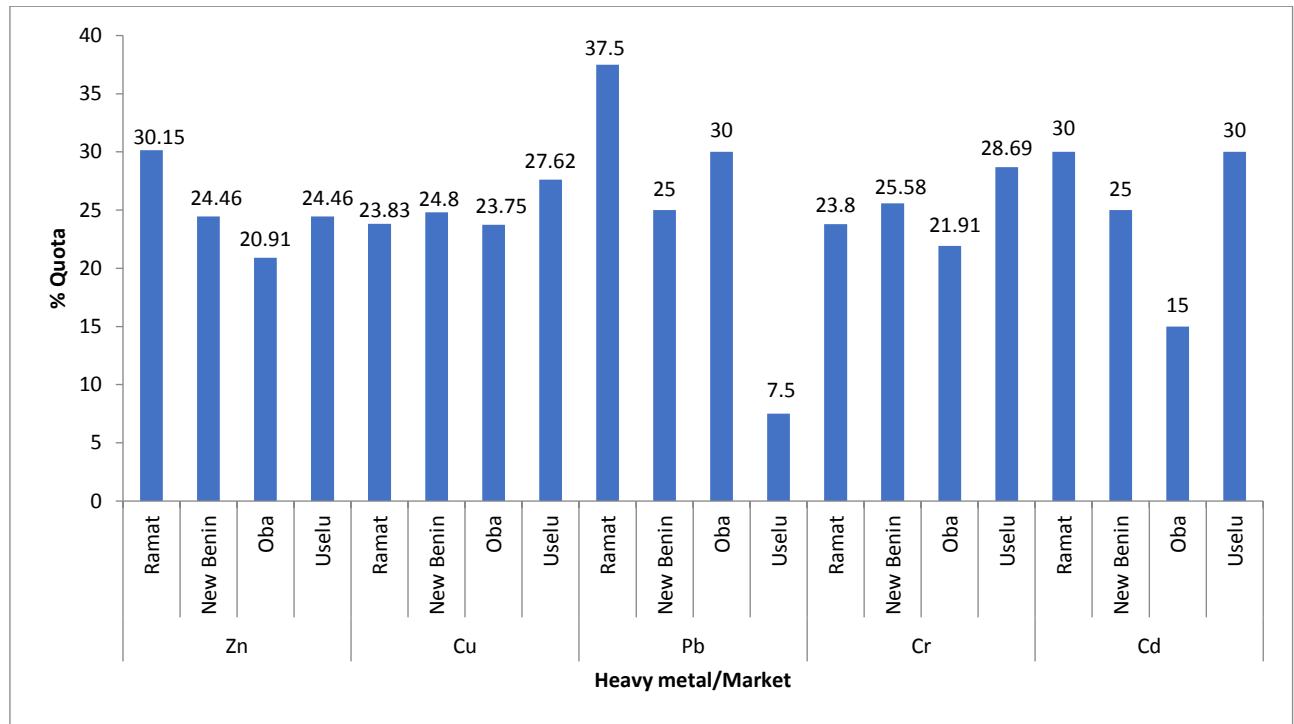


Figure 8: Heavy metal quota in *Scomber scombrus* by market

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