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# Heavy Metals Contamination of Some Food Materials from Markets in South Eastern Nigeria

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***Authors' contributions***

*This work was carried out in collaboration among all authors. Author CAA designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors RNO and AEC managed the analyses of the study. Authors CLA and JUA managed the literature searches. All authors read and approved the final manuscript.*

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## ABSTRACT

This research work aimed at assessing the metal composition of the cotyledons of *Citrullus colocynthis* (egusi) and *Irvingia wombolu* (ogbono) as well as cassava chips sold in open markets in the five states of the South-Eastern Nigeria. These food materials were sampled from 135 stores and markets in three waves (Wet season, Harmattan and Dry season) between February, 2015 and March, 2016 in each of the five states in south eastern Nigeria (Enugu, Anambra, Imo, Abia and Ebonyi). The food materials were analyzed for Zinc, Lead, Copper and Iron using the Atomic Absorption Spectroscopy (AAS) method. The concentrations of these metals were not higher than the maximum permissible limit set by NAFDAC in Nigeria except for Lead in Enugu State ((0.042±0.068 mg/g). The highest quantity of Lead was detected in *Citrullus colocynthis* (0.039±0.006 mg/g) while the least concentration was detected in Cassava chips (0.009±0.005 mg/g). Lead was detected in samples collected across all the seasons (Wet season, Dry season and Harmattan). The high concentration of Lead in the sampled food materials from Enugu state across all the waves of sample collection that violated the permissible limits of lead set by WHO

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and NAFDAC is of public health concern. Lead exposure has been shown to cause severe health challenges thus regular monitoring of these toxic heavy metals from foods sold in markets is essential, to prevent their excessive build-up in the food chain. The inference of this study reveals that these food materials sold in the open market are not completely safe for consumption as a result of lead contamination due to poor handling and processing. Findings from this study will be of great help to all stakeholders in this area of interest such as researchers and environmental regulators, relevant government agencies saddled with the responsibility of ensuring the safety of foods consumed in Nigeria.

*Keywords: Metals; contamination; South Eastern Nigeria; food materials; Egusi; Ogbono; cassava chips; lead.*

## 1. INTRODUCTION

Food contamination as a result of improper handling and poor processing is a major problem in the Sub-Saharan Africa. The numerous and emerging issues of food security have become a global concern. Contamination of these food materials is one of the most important aspects of food quality assurance [1]. Contamination of foods is oftentimes caused by heavy metals as a result of industrial growth, advances in agricultural chemicalization and urban activities of human beings [2].

It is a common practice in Nigeria to sell food materials in an open market without proper aseptic handling and processing conditions [3,4,5]. This has led to a wide acceptability of food materials sold in the open market among majority of the Nigerian populace which cut across social, cultural, ethnic, geographical and political class. This is rampant because these food materials are usually cheap and easily accessible. Undoubtedly, it is presumed that there are serious environmental hazards associated with this ugly practice [6].

Consumption of foods contaminated with heavy metals can cause serious health challenges in man by depleting some essential nutrients in the body [7]. Therefore an understanding of the quality of Egusi, Ogbono and Cassava chips sold in the open market in South Eastern Nigeria which are bought and consumed by the masses is a prerequisite for remediation of this illnesses caused by food contamination. Unfortunately, there is a paucity of information on the quality of these food materials sold in the south eastern Nigerian markets. It is on this premise that this research is conducted. No doubt, the inference of this study will furnish the public especially the nutritionists with information on the quality of these food materials sold in the South Eastern Nigerian markets and also provide basis for decision making both to the government, researchers, development organizations and all stakeholders concerned with food safety and food security.

## 2. MATERIALS AND METHODS

### 2.1 Study Area

This study was conducted in the South-Eastern Nigeria. South-Eastern Nigeria with a population of 30 million people [8] has an area of 16,000 sq mi (40,000 $km^2$) and is located within the latitude 5-7 degrees north and longitude 6-8 degrees east. Geographically, South-Eastern Nigeria extends from latitudes $4°40^{l}$ to $7°20^{1}$ north latitude, and $6°00^{1}$ to $8°20^{1}$ east longitude. The culture area occupies about 50 000 $km^2$ of Nigeria's total area of 923 768 $km^2$.

### 2.2 Design of the Study

This experiment was a factorial experiment laid in a Complete Randomized Design (CRD). Samples were collected in three waves (Wet season, Harmattan and Dry season) in the fives states that make up the South Eastern Nigeria (Enugu, Abia, Anambra, Imo and Ebonyi). The independent variables were wave of collection/sampling period, plant materials and location. The dependent variable was the heavy metal concentration.

### 2.3 Sample Collection

The method of Adetunji et al. [9] was adopted in sampling food materials; the food materials sampled were *I. wombolu, C. colocynthis* and cassava chips. Food samples that were locally sourced, sun dried, not sorted and had been stored for at least four months in their respective state of sampling were collected in dry season (February- April), wet season (May- August) and Harmattan (November- January) from designated

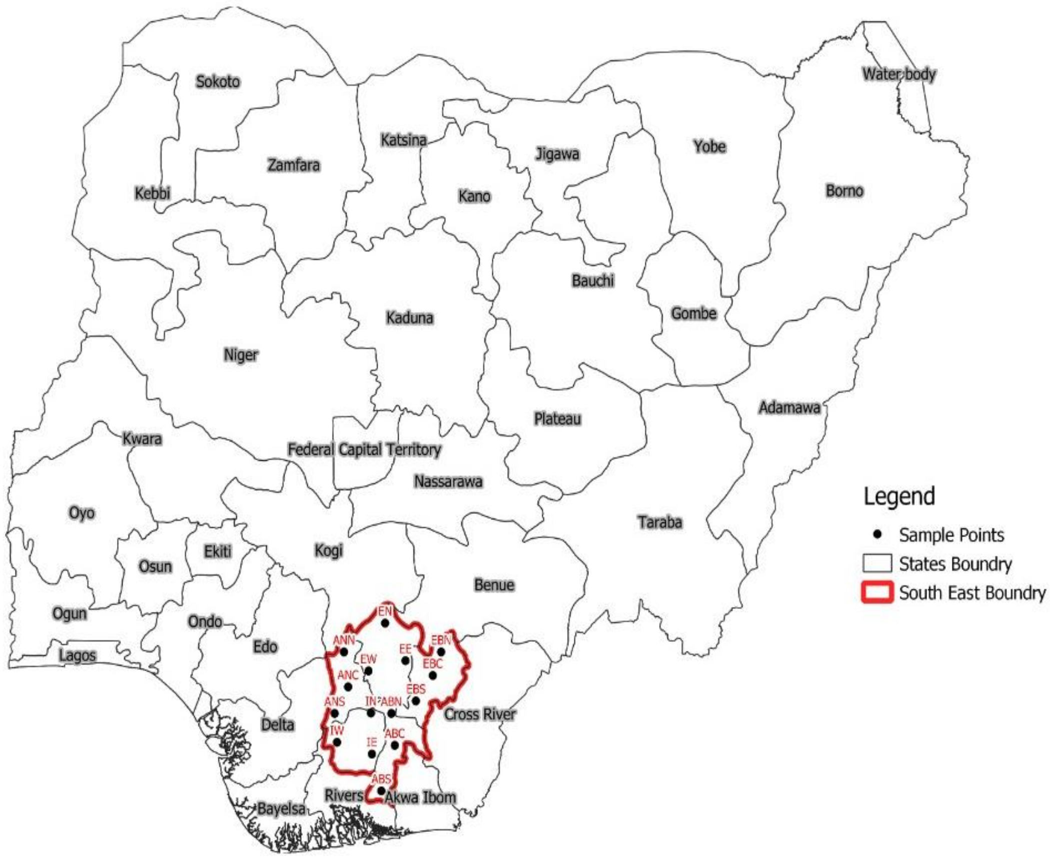


**Fig. 1. South Eastern state of Nigeria with sampling points**

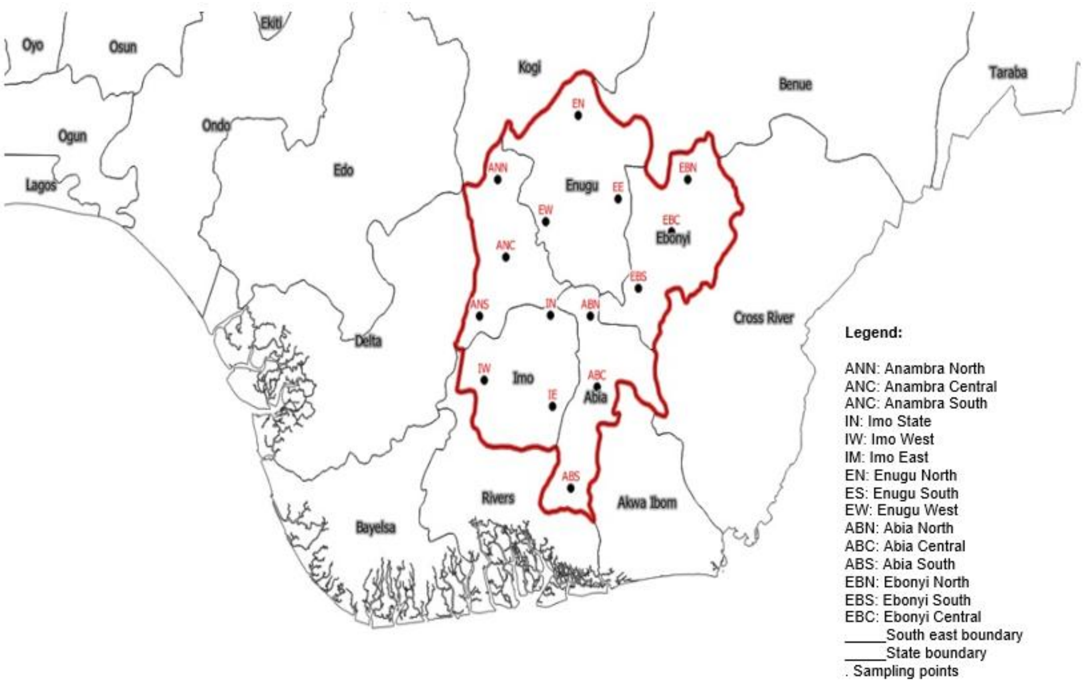


**Fig. 2. South Eastern Nigeria region with senatorial zones in each of the states**

markets and stores in the three senatorial districts of each state. Using simple random sampling technique a total of three markets from each senatorial zone were sampled, 5 g each of sample was collected from three different sampling points from each market. Each of the plant materials purchased from all senatorial zones within a state were pooled together to form a single bulk, from where the secondary sample was made. Five grams working sample was drawn at random from the mixed samples. These working samples represented plant materials from the states in which they were collected. Hence a total of five samples per plant materials at each analysis time (dry season, wet season and harmattan) were used for the experiment.

## 2.4 Determination of Concentration of Elements in the Food Samples across the States of Collection

### 2.4.1 Sample digestion

One gram (1 g) of each prepared sample was weighed into a 100 ml beaker. Thirty mills of aqua-regia (a mixture of nitric acid and hydrochloric acid in the ratio 1:3) was measured in a 100 ml measuring cylinder and added into the weighed samples. Ten drops of hydrogen peroxide was added to each of the preparation to increase the complexing power of the mineral acids. The samples in beakers were placed on a digital laboratory heating mantle under fume cupboard and heated at 1000$^oC$ until the samples completely digest. Each digest was allowed to cool and diluted with 50 ml of distilled-deionised water. They were filtered into 100 ml volumetric flask using whatman filter paper (125 mm). The digests were made up to the 100 ml mark using distilled-deionised.

## 2.5 Determination of Concentration of Elements in the Food Samples across the States of Collection

The digested samples were analyzed for metals (lead, copper, zinc and iron) using Atomic Adsorption Spectrophotometer (AAS) manufactured by Buck Scientific, 210VGP, USA. Values were reported in milligram per gram.

## 3. RESULTS

## 3.1 Concentrations of Heavy Metals in Food Samples

Values for concentrations of heavy metals in samples is presented in Table 1. For the comparison of metals in food samples, the highest concentration of lead was detected in *C. colocynthis* (0.039±0.006 mg/g) this was significantly (P>0.05) higher than 0.009±0.005 mg/g and 0.008±0.0004 mg/g recorded from cassava chips and *I. wombolu* respectively. There was no significant difference in the concentration of Zinc across all the food samples, although the values recorded ranged between 0.163±0.025 mg/g-0.223±0.022 mg/g, for copper the concentration was higher in *C. colocynthis* (0.199±0.02 mg/g) and *I. wombolu* (0.11±0.054 mg/g), Iron was lowest in *I. wombolu* (0.218±0.093 mg/g) this was significantly lower than values recorded in other food materials at P>0.05 level of significance, the highest in Iron was Egusi with value of 0.596±0.025 (Table 1).

## 3.2 Concentration of Heavy Metals in Cassava Chips

Results for concentrations of heavy metals in samples is presented in Table 2. For the concentration of metal cassava chips, lead was not detected in all the samples across the states except in Enugu (0.042±0.098 mg/g). For zinc, there was significant difference in its concentration across the states, the highest concentration was detected in Enugu state (0.305±0.332 mg/g) this was significantly higher than values recorded from other states, next was Abia state (0.252±0.071mg/g), values recorded from Anambra (0.057±0.011 mg/g) and Ebonyi (0.050±0.033 mg/g) were significantly lower than values obtained from other schools. Copper was not detected from samples collected from Ebonyi and Imo state, the highest value of copper was detected from Enugu (0.148±0.081 mg/g) this was significantly higher than 0.01±0.0270 mg/g and 0.031±0.094 mg/g recorded from Abia and Anambra states. The highest concentration of iron was recorded from sample collected from Abia state (0.735±0.010 mg/g), this was significant different from 0.214±0.063 mg/g obtained from Enugu, relatively lower concentration of iron was recorded in samples collected from Anambra (0.116±0.018 mg/g) and Imo state (0.151±0.006 mg/g). With respect to the wave of collection, lead was not detected during dry season but was detected during harmattan (0.022±0.077 mg/g) and wet season (0.003±0.013 mg/g), zinc was highest during wet season with a value of 0.294±0.306mg/g, this was significantly (P>0.05) higher than 0.144±0.075 mg/g observed during harmattan which is on the same hand significantly higher than 0.049±0.039ppb recorded during dry season. Copper was relatively higher during harmattan (0.063±0.038 mg/g) and wet season (0.047±0.014 mg/g), there was significant difference in the concentration of iron across the wave of collection, the highest value of 0.442±0.092 mg/g was recorded during harmattan, next to this was 0.207±0.041 mg/g observed during dry season while the least concentration of 0.195±0.047 mg/g was recorded during wet season (Table 2).

### 3.3 Concentration of Heavy Metals in *Citrullus colocynthis*

Results for concentrations of heavy metals in samples is presented in Table 3. With respect to state of collection, lead was only detected in Enugu state with a value of 0.193±0.233 mg/g. Zinc was detected in samples from all the states, highest in Abia with a value of 0.373±0.055mg/g, although this was not significantly different from 0.315±0.003 mg/g and 0.242±0.234 mg/g detected from Enugu and Imo state respectively, lower value of zinc was detected from Anambra (0.081±0.093 mg/g) and Ebonyi state (0.080±0.053 mg/g).Copper was more in Abia (0.150±0.040 mg/g) and Ebonyi state (0.103±0.057 mg/g) and least in Imo state (0.005±0.011 mg/g). For iron, more concentrations were detected from samples collected from Enugu state with a value of 2.362±1.920 mg/g, this was significantly

**Table 1. Comparison of heavy metals in the food samples**

| Food samples | Metal concentration (mg/g) | | | |
|---|---|---|---|---|
| | Lead | Zinc | Copper | Iron |
| Cassava chip | 0.009±0.005[b] | 0.163±0.025[a] | 0.038±0.004[c] | 0.281±0.926[b] |
| *C.colocynthis* | 0.039±0.006[a] | 0.218±0.085[a] | 0.199±0.020[a] | 0.596±0.025[a] |
| *I. wombolu* | 0.008±0.004[b] | 0.223±0.022[a] | 0.110±0.054[b] | 0.218±0.093[b] |
| p-value | 0.022 | 0.526 | 0.008 | 0.042 |

*Results are Mean ± Standard Deviation of triplicate determinations*
*Means with the same letter in a column are not significantly different (p>0.05)*

**Table 2. Concentration of heavy metals in cassava chips**

| States | Wave of collection | Average metal concentrations (mg/g) | | | |
|---|---|---|---|---|---|
| | | Lead | Zinc | Copper | Iron |
| Abia | (Total) | 0.000±0.000[b] | 0.252±0.071[b] | 0.01±0.0270[b] | 0.735±0.010[a] |
| Anambra | | 0.000±0.000[b] | 0.057±0.011[d] | 0.031±0.094[b] | 0.116±0.018[d] |
| Ebonyi | | 0.000±0.000[b] | 0.050±0.033[d] | 0.000±0.000[c] | 0.191±0.037[c] |
| Enugu | | 0.042±0.098[a] | 0.305±0.332[a] | 0.148±0.081[a] | 0.214±0.063[b] |
| Imo | | 0.000±0.000[b] | 0.148±0.141[c] | 0.000±0.000[c] | 0.151±0.006[d] |
| Total | HM | 0.022±0.077[a] | 0.144±0.075[b] | 0.063±0.038[a] | 0.442±0.092[a] |
| | DS | 0.000±0.000[c] | 0.049±0.039[c] | 0.004±0.009[c] | 0.207±0.041[b] |
| | WS | 0.003±0.013[b] | 0.294±0.306[a] | 0.047±0.014[b] | 0.195±0.047[c] |

*Results are Mean ± Standard Deviation of triplicate determinations*
*Means with the same letter in a column are not significantly different (p>0.05)*
*HM is Harmattan; DS is Dry season; WS is Wet season*

**Table 3. Concentration of heavy metals in *Citrullus colocynthis***

| States | Wave of collection | Average metal concentrations (mg/g) | | | |
|---|---|---|---|---|---|
| | | Lead | Zinc | Copper | Iron |
| Abia | (Total) | 0.000±0.000[b] | 0.373±0.055[a] | 0.150±0.040[b] | 0.137±0.212[c] |
| Anambra | | 0.000±0.000[b] | 0.081±0.093[b] | 0.053±0.099[c] | 0.086±0.040[d] |
| Ebonyi | | 0.000±0.000[b] | 0.080±0.053[b] | 0.103±0.057[b] | 0.084±0.077[d] |
| Enugu | | 0.193±0.233[a] | 0.315±0.003[a] | 0.685±0.004[a] | 2.362±1.920[a] |
| Imo | | 0.000±0.000[b] | 0.242±0.234[a] | 0.005±0.011[d] | 0.309±0.291[b] |
| Total | HM | 0.044±0.129[b] | 0.159±0.096[b] | 0.256±0.039[a] | 0.709±0.106[a] |
| | DS | 0.067±0.176[b] | 0.047±0.021[c] | 0.102±0.021[b] | 0.579±0.237[b] |
| | WS | 0.005±0.021[a] | 0.448±0.053[a] | 0.240±0.032[a] | 0.499±0.198[b] |

*Results are Mean ± Standard Deviation of triplicate determinations*
*Means with the same letter in a column are not significantly different (p>0.05)*
*HM is Harmattan; DS is Dry season; WS is Wet season*

**Table 4. Concentrations of metals in *Irvingia wombolu***

| States | Wave of collection | Average metal concentrations (mg/g) | | | |
|---|---|---|---|---|---|
| | | Lead | Zinc | Copper | Iron |
| Abia | (Total) | 0.000±0.000$^{b}$ | 0.078±0.082$^{d}$ | 0.011±0.021$^{c}$ | 0.235±0.032$^{b}$ |
| Anambra | | 0.000±0.000$^{b}$ | 0.394±0.001$^{a}$ | 0.077±0.139$^{b}$ | 0.118±0.033$^{c}$ |
| Ebonyi | | 0.000±0.000$^{b}$ | 0.141±0.048$^{c}$ | 0.058±0.118$^{b}$ | 0.053±0.059$^{d}$ |
| Enugu | | 0.042±0.068$^{a}$ | 0.289±0.071$^{b}$ | 0.394±0.451$^{a}$ | 0.430±0.047$^{a}$ |
| Imo | | 0.000±0.000$^{b}$ | 0.210±0.036$^{b}$ | 0.011±0.023$^{c}$ | 0.255±0.061$^{b}$ |
| Total | HM | 0.023±0.006$^{a}$ | 0.303±0.081$^{a}$ | 0.186±0.025$^{a}$ | 0.207±0.046$^{b}$ |
| | DS | 0.003±0.010$^{b}$ | 0.073±0.016$^{c}$ | 0.007±0.019$^{c}$ | 0.078±0.002$^{c}$ |
| | WS | 0.000±0.000$^{c}$ | 0.291±0.041$^{b}$ | 0.138±0.079$^{b}$ | 0.371±0.075$^{a}$ |

*Results are Mean ± Standard Deviation of triplicate determinations*
*Means with the same letter in a column are not significantly different (p>0.05)*
*HM is Harmattan; DS is Dry season; WS is Wet season*

(P>0.05) higher than concentrations of iron recorded from other states, Abia state was next in iron (0.137±0.212 mg/g) while the least concentration of iron was detected from Anambra and Ebonyi state with concentrations of 0.086±0.040 mg/g and 0.084±0.077 mg/g respectively. With respect to wave of collection, lead was detected from all the samples across the waves, highest during dry season (0.067±0.176 mg/g) and lowest during wet season (0.005±0.021 mg/g), the highest concentration of zinc was obtained during wet season 0.448±0.053 mg/g, next to this was 0.159±0.096 mg/g obtained during harmattan and the least concentration of zinc was detected from samples collected during dry season 0.047±0.021 mg/g. Copper was detected more in sample collected during harmattan and wet season with values of 0.256±0.039 mg/g and 0.240±0.032 mg/g respectively these were not significantly different from each other but significantly higher than 0.102±0.0219 mg/g recorded during dry season, there was no significant difference in the concentration of iron across the wave of collection, although the highest values of 0.709±0.106 mg/g was recorded during harmattan while the least value of 0.499±0.198 mg/g was obtained during wet season (Table 3).

## 3.4 Concentration of Heavy Metals in *Irvingia wombolu*

Results for concentrations of heavy metals in samples is presented in Table 4. Lead was not detected in any of the states except in Enugu (0.042±0.068 mg/g). The values of zinc were not significantly different from eachother across the state except in Abia state with value of 0.078±0.082 mg/g this was significantly lower than values from other states. For copper, 0.394±0.451 mg/g was obtained from Enugu; this was significantly higher than 0.077±0.139 mg/g and0.058±0.118 mg/g recorded from Anambra and Ebonyi state respectively, samples from Abia state and Imo state gave a lower concentration of copper with values of 0.011±0.021 mg/g and 0.011±0.023 mg/g. Iron was more from sample collected from Enugu state (0.430±0.047 mg/g) next were 0.255±0.061 mg/g and 0.235±0.032 mg/g recorded from Imo and Abia state respectively, Ebonyi was the least in iron with the concentration of 0.053±0.059 mg/g. With respect to wave of collection, there was significant difference in the concentration of lead across the wave of collection, the highest value of lead was detected harmattan (0.023±0.006 mg/g) this was significantly higher than 0.003±0.010 mg/g recorded during dry season, lead was not detected during wet season, the value of zinc across the wave of collection ranged between 0.073±0.016mg/g and 0.303±0.081 mg/g but there was not significant difference among the values at P>0.05 level of probability, high concentrations of copper were detected during harmarttan and wet season with values of 0.186±0.025mg/g and 0.138±0.079 mg/g respectively, these were significantly higher than 0.007±0.019 mg/g obtained during dry season. There was significant difference in the concentration of iron across the three waves of collection; the highest value of iron was recorded during wet season with values of 0.371±0.075 mg/g while the least in iron was dry season (0.078±0.002 mg/g) (Table 4).

## 4. DISCUSSION

All the metals tested were detected in all the food materials sampled at different concentrations. Some were present at concentrations above the permissible levels while some at concentrations below the permissible level. The concentration of lead in all the food samples were not significantly different from each other but they were all higher than the maximum permitted concentration (0.002 mg/g) of lead [10] in food materials consumed by man. This is in tandem with the results of some researchers, Nkansah and Amoako [11] reported that there was high level of lead in food materials (spices) sampled from markets. Chukwujindu et al., [12] recorded that lead concentration in food materials were above the guideline value for lead. The results of some other researchers do not agree with the findings of this research, for instance Aiwonegbe and Ikhuoria, [13] reported that the lead concentration in food materials tested fell under safe range of concentration (below 0.02 ug/g), while the result of a similar study carried out on locally produced wild rice in Kaduna by [14] showed that the mean lead (Pb) content of the wild rice samples was below the permissible level. Concentrations of Iron, Zinc and copper observed in this research work were all below the permissible level, this is in contrast to the documentation of some researchers who carried out similar work, Izah et al. [15] reported that metal concentrations in food samples, including iron, mercury, tin, antimony, cadmium, zinc, copper, chromium, lead, and manganese, seldom exceed the maximum contaminant level recommended by the Standard Organization of Nigeria (SON) and the World Health Organization (WHO), these differences could be linked to the different handling technique and other climatic and environmental factors such as air pollution.

The concentration of lead in the food materials also varies across the state, with Enugu State having the highest concentration. The knowledge of concentrations of metals (both heavy and essential) in food is of significant scientific interest because some of these elements are essential for human health (e.g., Cu, Fe, Mn, Zn) while others, if present even at low concentrations, can be toxic (e.g., Cd, Pb, Hg, As) [16,17]. The so-called essential metals can be toxic depending on their concentration. Therefore the high level of lead from Enugu State might have resulted from accumulation of lead through air pollution and from industrial emission. Zukowska [18] reported that air, soil and water pollution are contributing to the presence of harmful elements, such as cadmium, lead and mercury in foodstuff. The occurrences of heavy metals-enriched ecosystem components, firstly, arise from rapid industrial growth, advances in agricultural chemicalization, or the urban activities of human beings. These agents have led to metal dispersion in the environment and, consequently, impaired health of the population by the ingestion of victuals contaminated by harmful elements.

## 5. CONCLUSION

In this research, the high concentration of lead in the sampled food materials from Enugu that violated the permissible limits of lead set by WHO is of public health concern. Among possible target organs of heavy metals, are soft tissues such as the kidney and liver and the central nervous system appear to be specially sensitive [19]. Lead exposure has been shown to cause severe anemia, permanent brain damage, neurological disorders, reproductive problems, diminished intelligence and a host of other diseases. According to the Agency for Toxic Substances and Disease Registry, a division of the U.S. Public Health Service, the major exposure of lead to the general population in food is through foods such as fruits, vegetables and grains [20]. Thus regular monitoring of these toxic heavy metals from foods sold in markets is essential, to prevent their excessive build-up in the food chain.

## CONSENT

It is not applicable.

## ETHICAL APPROVAL

It is not applicable.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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