

Occurrence and Concentration of Heavy Metals in Garden Egg, Tomatoes, Cucumber and Watermelon from Two Major Markets in Edo State, Nigeria

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Abstract This study was conducted to analyze the concentration of some heavy metals in selected vegetable fruits sold in Uselu and Ologbo markets. Using Atomic Absorption Spectrophotometry (AAS) the heavy metal concentration of Cd, Zn, Pb, Cu, Mn, Fe, Co, Cr Ni in Watermelon, Garden Egg, Tomato and Cucumber was studied. Trace element as Fe and Zn showed high concentration occurrence. The heavy metal concentration of Fe in Cucumber and watermelon from both markets was found to be 4.57 ± 0.32 mg/kg and 4.39 ± 0.32 mg/kg respectively exceeding the permissible limit of food and agricultural organization and world health organization. Pb recorded the lowest value of 0.13 ± 0.09 mg/kg in garden egg and 0.28 ± 0.18 mg/kg in watermelon, Analysis of Variance (ANOVA) showed a significant difference ($P < 0.05$) in the concentration of heavy metals (Fe, Pb, Cd, Mn, Cu, and Zn) in both stations. Fruits sample used for the study is contaminated with Fe and Zn which serves as a serious health concern.

Keywords Heavy metals, Fruits, Health risk, WHO limits, Contamination, Permissible limits

1. Introduction

Compared to water, heavy metals are elements with relatively high density. With the belief that there exists a relationship between heaviness and toxicity, metalloids such as arsenic, which can induce toxicity at a low level of exposure can also be regarded as heavy metal (Wang et al., 2007). There has been increased ecological and public health concerns globally linked to heavy metal contamination of recent. There has been increased human exposure due to its increased industrial, agricultural, domestic and technological applications. Reported sources of heavy metals in the environment include industrial, agricultural, pharmaceutical, domestic wastes, and atmospheric sources. Dogheim and Alla, (2004) reported agricultural, domestic wastes, industrial discharges, pharmaceutical, cosmetics and atmospheric sources as the main source of heavy metals in the environment. Environmental pollution is common in point source areas such as mining, foundries and smelters and other metal-based industrial operation (Hawkens, 1997). Although heavy metals are naturally occurring elements

found throughout the earth's crust, most environmental contamination and human exposure arise from human activities such as mining and smelting operations, industrial production and use, and domestic activities such as mining operations, domestic and agricultural use by metals and metal-containing compound (Jamali et al., 2007). Environmental contamination can also occur through metal corrosion atmospheric deposition, soil erosion of metal ions and leaching of heavy metals, sediment resuspension and metal evaporate from water source to soil and groundwater. Natural phenomena such as weathering, tsunamis and volcanic eruption have also been reported to significantly contribute to heavy metal pollution in the environment. Other sources of heavy metal pollutions are industrial sources such as metal processing in refineries, coal burning in power plants, petroleum combustion, nuclear power stations and high-tension lines, plastics, textiles microelectronics, wood preservation and paper processing plant. Heavy metal can enter into the human body through inhalation, ingestion and dermal contact absorption (Adelekan and Abegunde, 2011; Rajaganapathy et al., 2011). They also accumulate in soils, plants and aquatic biota (Obodai et al., 2011; Wuana and Okieimen, 2011).

The aim of this work is therefore to determine the heavy metal concentration in fruits from two major markets in Edo State, Nigeria if these fruits are safe for human consumption.

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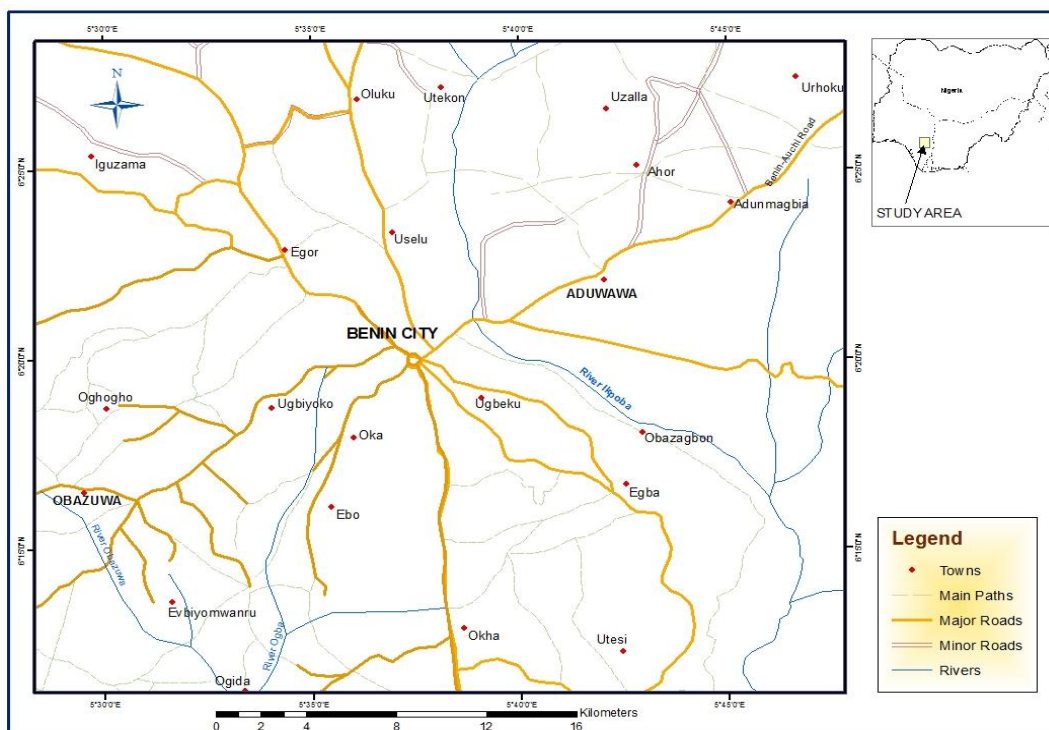


Figure 1. Map of Study Location

2. Materials and Methods

This study was carried out in Uselu and Ologbo market located in Benin City, Edo State, southwestern Nigeria (Figure 1) which has Latitude of $6^{\circ} 20' 21.0660''$ N and Longitude of $5^{\circ} 37' 2.8092''$ E.

Sampling Technique

The samples were collected from November 2015 to May 2016. The sampling stations were visited monthly for the study period. Fruits samples were collected in the early hours of the morning between 9 am and 12 pm each sampling day starting from Uselu market and ending at Ologbo market.

Sample Collection

A total of 216 samples consisting of four different fruits including watermelon (*Citrullus lanatus* Thunb), cucumber (*Cucumis sativa*), garden egg (*Solanum melongena*), and tomato (*Lycopersicum esculentus*) were collected and analyzed monthly in Cherish Lab. Ibadan, from each station Uselu market and Ologbo for a total of six months. In each station, three replicates of each fruit were collected per month making a total of 18 samples per fruit for the duration of the study. Different samples collected were stored using clean polythene bags and transported to the laboratory for analysis.

Sample Preparation

To remove suspended particles, fruits were washed using distilled water. Samples were cut using a clean knife. Samples for analysis were dried to a constant weight using the oven-dry method at 105°C for 1 hour 30 min to obtain the moisture content (AOAC, 2003). Samples were ground into

a fine powder using a commercial blender after drying and stored in polyethylene bags until used. 1g of each sample was weighed into a 100ml digestion tube, 5ml of HCl was mixed into a separate beaker, 2ml of concentrated H_2SO_4 and 20ml of concentrated HNO_3 in a conical flask under a fume hood. The content was mixed and heated gently at 180°C - 220°C for about 30min on a hot plate. The content was continuously heated until dense white fumes appear. It was finally heated for about 30min and then allowed to cool before making it up to the mark in a 50ml volumetric flask (Sobukola et al., 2007).

Heavy Metal Determination/Analysis

After fruit samples extraction, heavy metals concentration was determined. Fruit samples digestion was carried out by taking 10ml of the sample and adding 4ml Perchloric acid, 20ml concentrated nitric acid and 2ml concentrated tetraoxosulphate VI acid. This was digested using aluminum block digester 110. The mixture was heated until white fumes evolved and a clear solution obtained. After digestion, the samples were allowed to cool and then transferred to a 100ml volumetric flask. This was made up to mark with distilled water and thoroughly mixed. The samples were then allowed to stand overnight (in place of centrifuge) to separate insoluble materials. They were then filtered through Whatman No.42 filter paper. Deionized water was added to make up the 100 ml mark. Samples were then mixed properly and transferred into polythene bottles. A blank was also prepared as above. Iron (Fe), copper (Cu), lead (Pb), manganese (Mn), Nickel (Ni) and chromium (Cr) cadmium (Cd) zinc (Zn) and mercury (Hg) were determined using Atomic Absorption Spectrophotometry (AAS) PG, 550

Model. Calibration was done with the standard for each metal under investigation at individual wavelengths.

Statistical Analysis

Basic statistical measurement of central tendency and dispersion was used to characterize stations in terms of heavy metal concentration. Inter-stations comparisons were carried out to test for a significant difference in the heavy metals concentration using parametric analysis of variance (ANOVA), for each fruit sample was performed separately using variable such as station (Ogbeibu, 2014). The significant difference among the station means values were determined using Kruskal Wallis Test (KWT) to test for the hypothesis. Graphical representations were done in Microsoft Word.

3. Result and Discussion

Tables 1 and 2 show the concentrations of heavy metals in fruits frequently consumed in Edo State, Nigeria. The values are given as mean \pm SE and the results are means of three replicates. The heavy metal concentration is determined based on plants' dry weight. Heavy metals affect the nutritive contents of agricultural products and also have a harmful effect on humans. National and international regulations on food quality set the maximum permissible limit of toxic metals in human food; hence an essential aspect of food quality should be to regulate the concentrations of heavy metals in food (Radwan and Salama, 2006; Sobukola et al., 2008).

Cadmium is a non-essential element in foods and natural waters and primarily it accumulates in the kidneys and liver

(Divrikli et al., 2006). In all the samples analyzed from the two markets, the concentration was observed to be above the permissible limit of 0.02 mg/kg as reported by WHO (2001), which might be a treat for human consumption. It was highest in garden egg with a value of 0.820 mg/kg and 0.705 mg/kg respectively and lowest in watermelon and cucumber with a value of 0.640mg/kg and 0.574mg/kg respectively.

Al Jassir et al. (2005) reported that levels of Cd were higher in the garden rocket vegetable species for both washed and unwashed samples.

Zinc is one of the most essential metals for normal growth and development in humans (Divrikli et al., 2006). Zinc deficiency is of growing concern in developing countries. In these countries, Zinc deficiencies have been attributed to the large consumption of bread made without yeast (Divrikli et al., 2006). Excess Zinc can also be harmful, and cause Zinc toxicity. Such toxicity levels have been seen to occur at ingestion greater than 225mg/kg of Zinc.

The concentration of Zinc in all the samples analyzed from the two markets (Table 1 and 2), was observed to be below the WHO permissible limit of 1.50mg/kg as reported by WHO (2001) except for garden egg with a higher value of 5.35 and 1.546 mg/kg respectively. Kihampa et al. (2011) reported a high Zinc concentration ranging between 18.16 and 122.88mg/kg in *Amaranthus* species. Radwan and Salama (2006) and Onianwe et al. (2001) have also reported Zn levels of 5.35 and 7.40 mg/kg; 2.38 and 2.20 mg/kg; as well as 5.59 and 1.50 mg/kg for watermelon, orange and banana, respectively. Zinc (Zn) accumulation in high amounts can cause eminent health problems, such as stomach cramp, skin irritation, vomiting, nausea and anemia (Wuana and Okieimen, 2011).

Table 1. Concentrations of heavy metals in the fruits from Uselu market

Parameters	Garden egg	Tomatoes	Cucumber	Watermelon	WHO/FAO Acceptable Limit (mg/Kg)	Significance
Cd(mg/kg)	0.845 \pm 0.19	0.820 \pm 0.30	0.660 \pm 0.32	0.640 \pm 0.170	0.02	p>0.05
Zn (mg/kg)	5.35 \pm 0.130	1.380 \pm 0.100	1.21 \pm 0.23	1.220 \pm 0.190	1.50	p>0.05
Pb(mg/kg)	0.020 \pm 0.010	0.010 \pm 0.003	0.214 \pm 0.143	0.268 \pm 0.134	0.50	p>0.05
Cu(mg/kg)	0.331 \pm 0.135	0.305 \pm 0.080	0.307 \pm 0.086	0.260 \pm 0.071	2.00	p>0.05
Mn(mg/kg)	0.760 \pm 0.100	0.527 \pm 0.081	0.520 \pm 0.000	0.783 \pm 0.121	5.00	p>0.05
Fe(mg/kg)	4.053 \pm 0.240	4.231 \pm 0.1698	4.40 \pm 0.277	4.392 \pm 0.306	1.00	p>1.00

Note: n=3, CI=95%

Table 2. Concentrations of heavy metals in the fruits from Ologbo market

Parameters	Garden egg	Tomatoes	Cucumber	Watermelon	WHO/FAO Acceptable Limit (mg/Kg)	Significance
Cd(mg/kg)	0.816 \pm 0.192	0.705 \pm 0.245	0.574 \pm 0.184	0.769 \pm 0.159	0.02	p>0.05
Zn (mg/kg)	1.546 \pm 0.132	1.348 \pm 0.129	1.406 \pm 0.103	1.428 \pm 0.114	1.50	p>0.05
Pb(mg/kg)	0.133 \pm 0.057	0.402 \pm 0.226	0.310 \pm 0.103	0.279 \pm 0.184	0.50	p>0.05
Cu(mg/kg)	0.431 \pm 0.103	0.438 \pm 0.091	0.319 \pm 0.083	0.385 \pm 0.92	2.00	p>0.05
Mn(mg/kg)	0.784 \pm 0.156	0.510 \pm 0.103	0.604 \pm 0.148	0.599 \pm 1.82	5.00	p>0.05
Fe(mg/kg)	4.127 \pm 0.215	3.978 \pm 0.203	4.574 \pm 0.324	4.396 \pm 0.318	1.00	p>0.05

Note: n=3, CI=95%

The concentrations of Pb that exceeded WHO established permissible limit of 0.5 mg kg^{-1} , were not observed in all the samples in this study. The highest levels of Pb in were observed in watermelon (0.268 mg/kg) and Tomatoes (0.402 mg/kg). Pb being a harmful body poison can enter into the human system through air, water and food and cannot be eliminated through fruits and vegetable washing (Divrikli *et al.*, 2003). The traces of lead found in the fruit sample might be linked to the concentration in the polluted area and also the consequence of road traffic and lead emission from petrol in the area where these fruits were cultivated and sold (Zhen, 2008). The level of Pb reported in this study is comparable to those reported for apple (0.19 and 0.76 mg/kg); watermelon (0.30 mg/kg); orange (0.15 mg/kg) and banana (0.02 mg/kg) by Radwan and Salama (2006) and Parveen *et al.* (2003).

Copper is the third most used metal in the world (VCI, 2011). Copper is an essential micronutrient required in the growth of both plants and animals. The concentration of Cu in all the samples analyzed from the two markets was lower than the WHO permissible limit of 0.50 mg/kg . The highest levels of Cu were observed in garden egg (0.331 mg/kg) and Tomatoes (0.438 mg/kg). The results obtained were observed to be lower compared to other published results. 1.22 and 2.13 mg/kg ; 1.27 and 2.13 mg/kg and 2.51 and 0.95 mg/kg have been reported for the concentration of Cu in watermelon, orange and banana by Radwan and Salama (2006) and Onianwa *et al.* (2001), respectively.

Mn is essential for the normal bone structure, reproduction and Mn play a very essential role in the functioning of the central nervous system. Mn deficiency will lead to reproductivity failure in both males and females (Saraf and Samant, 2013). The concentration of Manganese (Mn) in all the analyzed fruits samples from the two markets were below the WHO permissible limits of 5.0 mg/kg . This result is not in agreement with that obtained by Tsoumbaris and Tsoukali-Papadopolou (1994) who reported high concentration (25.83 – 70.76 mg/kg) of Mn in cereals and fruits respectively in Greece. And also Abdulla *et al.* (1990) recorded a value of 151.68 – 227 mg/kg for fruits and vegetables grown on refuse dumpsite. Since the level of Manganese in all fruit samples analyzed in this study was lower than the stipulated permissible limit, the continuous consumption of these fruit does not have any health effects (Viard *et al.*, 2004; Sheng *et al.*, 2014).

Iron (Fe) is an important element in humans and plays a significant role in the formation of hemoglobin, oxygen and electron transfer in the human body. In all the samples analyzed from the two markets, Fe concentration was observed to be above the permissible limit of 0.80 mg/kg as reported by WHO and FAO (2010). Cucumber had the highest value of 4.40 mg/kg and 4.574 mg/kg respectively. This result is in an agreement with the study of Kalagbor and Eresiya (2014) that reported Fe level of 28.6 mg/kg for avocado pear, 19.00 mg/kg for orange, 29.60 mg/kg for pawpaw and 25.70 mg/kg for pineapple. The increased content of Iron in this study can be attributed to the Iron

materials used for simple and mechanized farming practices and during transportation of these fruits.

4. Conclusions

The result of this study showed that the four analyzed edible fruits commonly consumed by people in Edo state, from the two different markets, were contaminated with different levels of heavy metals. The results also revealed that the heavy metal concentration of some of the fruits analyzed was higher than WHO acceptable limits which can pose a serious health challenge while some are below the limit making them suitable for human consumption.

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