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LEVELS OF SELECTED HEAVY METALS IN FRUITS AND VEGETABLES SOLD IN MAJOR MARKETS IN KOGI STATE, NORTH CENTRAL NIGERIA

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ABTSRACT

The study investigated the concentrations of cadmium, copper, lead, nickel and zinc in some fruits and vegetables sold in major markets in Kogi State, North Central Nigeria. The levels of metals were obtained by atomic absorption spectrophotometry. Average concentrations of Cd, Cu, Pb, Ni and Zn obtained were 0.02±0.01, 0.03±0.02, 0.59±0.15, 0.02±0.02, and 0.04±0.02 (mg/kg) respectively. Corresponding levels in vegetables were not significantly different (P>0.05). Values of Cu were highest among the metals with average concentrations of 0.59±0.15 mg/kg and 0.69±0.13 mg/kg in fruits and vegetables respectively. The levels of the metals compared with those reported for similar studies elsewhere in the world as current levels do not pose danger to human health.

Key words: Fruits, Vegetables, Heavy metals, Kogi State, Health risk.

INTRODUCTION

Fruits and vegetables are important sources of mineral elements. The consumption of vegetables and fruits as food offers fast means of providing adequate vitamins, minerals and fiber. Vegetables used as food include those used in making soups or served as integral parts of the main sources of a meal (Ihekoronye and Ngoddy, 1985). Leafy vegetables are used to increase the quality of soups and also for their dietary purposes (Sobukola *et al*., 2007). Vegetables have been reported to contain substances such as cellulose, semi-cellulose and pectin (Sobukola and Dairo, 2007). These substances in vegetables are said to contribute to their texture and firmness (Sobukola and Dairo, 2007). In human diet, fresh fruits and vegetables offer a lot of nutrients that are of health benefits to human. Their importance results from the fact that they contain water, fibre, vitamins and minerals such as calcium, iron, sulphur and potassium (Sobukola *et al*., 2007). To stay healthy in life one needs to take good diet, and to complement our carbohydrate and proteins in other to have a balanced diet, fruits and vegetables are important. They play a vital role in the maintenance of health and the prevention and treatment of various diseases (Mello, 2003).

When food is undergoing digestion, acid substances are often times formed and vegetables act as neutralizing agents for these acidic substances formed during digestion (Yusuf, and Oluwole, 2009). Food is needed in large quantity to meet up with the ever increasing population. The increasing population has also resulted in increase in human activities. To meet up with food challenges and population explosion especially in developing countries has led to the application of modern technologies. This has resulted in some areas pollution of soils, water and contamination of the human food chain. The uptake of heavy metals by plants grown in polluted soils has been studied to a considerable extent (Sukreeyapongse*et al*., 2002; Yusuf, and Oluwole, 2009). Heavy metal contamination in vegetables cannot be underestimated as these foodstuffs are important components of human diet. Heavy metal contamination of the food items is one of the most important aspects of food quality assurance (Radwan and Salama, 2006; Khan*et al*., 2008).

Human beings are encouraged to consume more vegetables and fruits, which are a good source of vitamins, minerals, fibers and also beneficial to their health (Khairiah *et al*., 2004; Chojnacha *et al*., 2005). However, these plants contain both essential and toxic metals over a wide range of concentrations. It is well known that plants take up metals by absorbing them from contaminated soils as well as from deposits on parts of the plants exposed to the air from polluted environments (Khairiah *et al*., 2004; Chojnacha *et al*., 2005). Heavy metals are among the major contaminants of food supply and may be considered as one of the most important problems to our environment (Zaidi *et al*., 2005). Such problem is getting more serious all over the world especially in developing countries. Heavy metals, in general, are not biodegradable, have long biological half-lives and have the potential for accumulation in the different body organs leading to unwanted side effects (Jarup, 2003; Sathawara *et al*., 2004). Kogi State is a major food producing State in Nigeria and most of the fruits and vegetables are from major commercial farms in the state and a few like onions and apple are obtained outside the state. But limited data are available on heavy metal levels in fruits and vegetables sold in major markets in the state. The aim of this study is to determine the level of heavy metals (Cd, Cu, Ni, Pb and Zn) in selected fruits and vegetables sold in major markets in Kogi State, North Central Nigeria and to inform authorities if levels pose threat to human health for actions to be taken.

MATERIALS AND METHODS

Sampling

A total of 210 samples of fruits and vegetables were purchased from major markets in Kogi State, Nigeria in 2011. The markets were selected from the three senatorial zones of the state including the state capital Lokoja. In each market, seven vendors were identified and samples were collected at three different sampling times. The sampled fruits include pineapple (*Ananascomosus* Schult), banana (*Musa sapientum* L) apple (*Malussylvestria*), pawpaw (*Carica papaya* L), watermelon (*Citrulluslanatus*), orange (*Citrus sinensis* L) and cashew (*Anacardiumoccidentale*). The vegetables are bitter leaf (*Vernoniaamygdalna* L), cabbage (*Brassica oleracae*L), tomato (*Lycopersiconesculentum* Mill), eggplant (*Sonalummelongena* L), carrot (*Daucuscarota* L), spinach (*Amaranthushybriduln*L) and onions (*Allium cepa* L). Edible portions of the samples were used for analysis.

Sample Preparation and Treatment

Sub-samples (1 kg, each) were taken at random from the composite sample (10 kg) and were processed for analysis by the wet digestion method. In the laboratory, the leaf samples were washed with distilled water, cut into small pieces with stainless steel

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knife, and air-dried for 7 days. The samples were placed in clean acid-washed porcelain crucible and oven-dried at 60^{o}C for 2 hours until they were brittle and crisp. The dried samples were ground using acid washed mortar and pestle. The ground samples were sieved with a plastic sieve of 2.0 mm mesh size to obtain fine powder. These were finally stored in screw capped plastic containers and labeled appropriately for analysis.

Procedure for Digestion: Metals in dry samples were extracted with a 3:1 mixture of HNO_3 and $HClO_4$. A 0.5g powdered fruit and vegetable sample was weighed into a 100 ml beaker. A10ml acid mixture of HNO_3 and $HClO_4$ was added to the sample in the beaker, and allowed to stand for 3 min. The mixture was heated at 70^{o}C until a transparent solution was obtained (digestion complete). The sample was then cooled, diluted and filtered into a 25mL standard flask and two 5 ml portions of distilled water were used to rinse the beaker and the contents filtered into the 25 mL volumetric flask using Whatman No. 41 filter paper. The filtrate was allowed to cool to room temperature before dilution was made to the mark and the content mixed thoroughly by shaking. A sample blank digestion was also carried out. The digested plant samples and blank were analyzed for Cd, Cu, Ni, Pb, and Zn using a Buck Scientific flame atomic absorption spectrophotometer (Model 200A).

Instrument Calibration and Sample Reading: The equipment was calibrated with commercial stock standards of the metals to be run. Working standard solutions were prepared by diluting a volume of a stock standard solution of 1000ppm concentration of each of the metals to required concentrations in standard volumetric flasks. These were used to calibrate the AAS. The absorbance values of blank and working standard solutions were measured using a Buck Scientific flame atomic absorption spectrophotometer (Model 200A). The spectrophotometer of 0.2nm slit width was operated in the air-acetylene flame mode. The lamps for heavy metals (Cd, Cd, Cu, Ni, Pb, Zn) were operated at wavelengths of 228.8, 324.7, 232.0, 217.0 and 312.9 respectively. A blank reading was subtracted from those of analyte samples and the corrected absorbance values obtained.

Ash Content Determination: Ash was obtained by the incineration of 2.0g samples in a muffle furnance (LMF4 from Carbolite, Bamford, Sheffield England) at 600^{0}C for 2 hours.

Determination of moisture content: Moisture content was determined by heating 2.0g portions of each of the samples in an oven (Plus 11 Sanyo Gallenkamp PLC, England) at 105^{0}C until a constant weight was obtained.

Quality Assurance

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. Samples were generally carefully handled to avoid contamination. Glassware was properly cleaned, and the reagents were of analytical grade. Double distilled deionised water was used throughout the study. Reagent blank determinations were used to correct the instrument readings. Stock standard solutions for the atomic absorption analyses were prepared from Analar R grade salts. Working standards were made from the stock by dilution of measured aliquots. For validation of the analytical procedure, a recovery study was carried out by spiking and homogenizing several already analyzed samples with varied amounts of standard solutions of the metals (Onianwa*et al*., 2001). The spiked samples were processed for the analysis by the wet digestion method and reanalyzed as described above. The average recoveries obtained were 97.5±6.4%, 89.0±8.9%, 92±13%, 93.7±1.9% and 97.9±5.8% for Cd, Cu, Pb, Ni and Zn, respectively.

The statistical analysis of the data was carried out with the aid of the General Linear Models statistical package, using appropriate tool, such as the Analysis of Variance. Confidence level was accepted at 95% and P>0.05 was considered not significant.

RESULTS

The overall average result of level of pH, moisture and ash contents in fruit and vegetable samples are given in Table 1. The observed pH in fruit samples ranged from 1.47±0.02 in lime orange to 10.1±0.12 in watermelon. Corresponding pH level in vegetables ranged from 2.91±0.12 in onions to 9.44±0.08 in tomato. The moisture content of the fruits samples ranged from 77.7±0.02% in banana to 95.3±0.03 in watermelon while corresponding values in vegetables ranged from 83.4±0.11% in eggplant to 94.4±0.02% in tomato. The ash content in fruits ranged from 94.3±0.03% in watermelon to 96.0±0.02% in orange. Vegetables had values that ranged from 90.7±0.02% in pumpkin to 96.6±0.02% in carrot. The mean concentrations of heavy metals determined (Cd, Cu, Pb, Ni and Zn) in fruits are given in Fig 1 while in vegetables are given in Fig 2. The metal concentrations determined were based on sample dry weight. The results showed that the mean concentrations of Cd in fruits were between 0.01±0.02mg/kg in apple to 0.03±0.14 mg/kg in pawpaw. The highest concentration of 0.05 mg/kg was observed in pawpaw. Corresponding mean levels in vegetables were between 0.02±0.02 mg/kg in onions to 0.04±0.08 mg/kg in spinach. The Cd levels do not range widely in the fruits and vegetables. A similar result was observed for Pb, Ni and Zn.

Table 1: Physicochemical parameters of selected fruits and vegetables

Fruits	pH	Moisture content (%)	Ash content (%)
Pineapple	3.28±0.02	88.4±0.01	95.5±0.02
Pawpaw	3.52±0.03	91.2±0.04	95.8±0.03
Orange	4.79±0.11	92.5±0.02	96.0±0.02
Lime Orange	1.47±0.02	91.0±0.03	95.9±0.01
Apple	2.09±0.01	87.5±0.11	95.9±0.02
Watermelon	10.1±0.12	95.3±0.03	94.3±0.03
Banana	3.87±0.04	77.7±0.02	95.5±0.01
Cashew	3.300.02	92.2±0.03	95.5±0.01
Vegetables			
Onions	2.91±0.12	90.8±0.01	92.7±0.03
Bitter leaf	5.98±0.01	92.2±0.02	91.4±0.01
Spinach	7.78±0.04	89.0±0.03	93.1±0.04
Pumpkin	5.42±0.11	85.4±0.03	90.7±0.02
Cabbage	4.10±0.21	92.5±0.04	94.0±0.02
Egg plant	4.52±0.05	83.4±0.11	95.3±0.01
Carrot	5.02±0.03	90.1±0.04	96.6±0.02
Tomato	9.44±0.08	94.4±0.02	94.5±0.02

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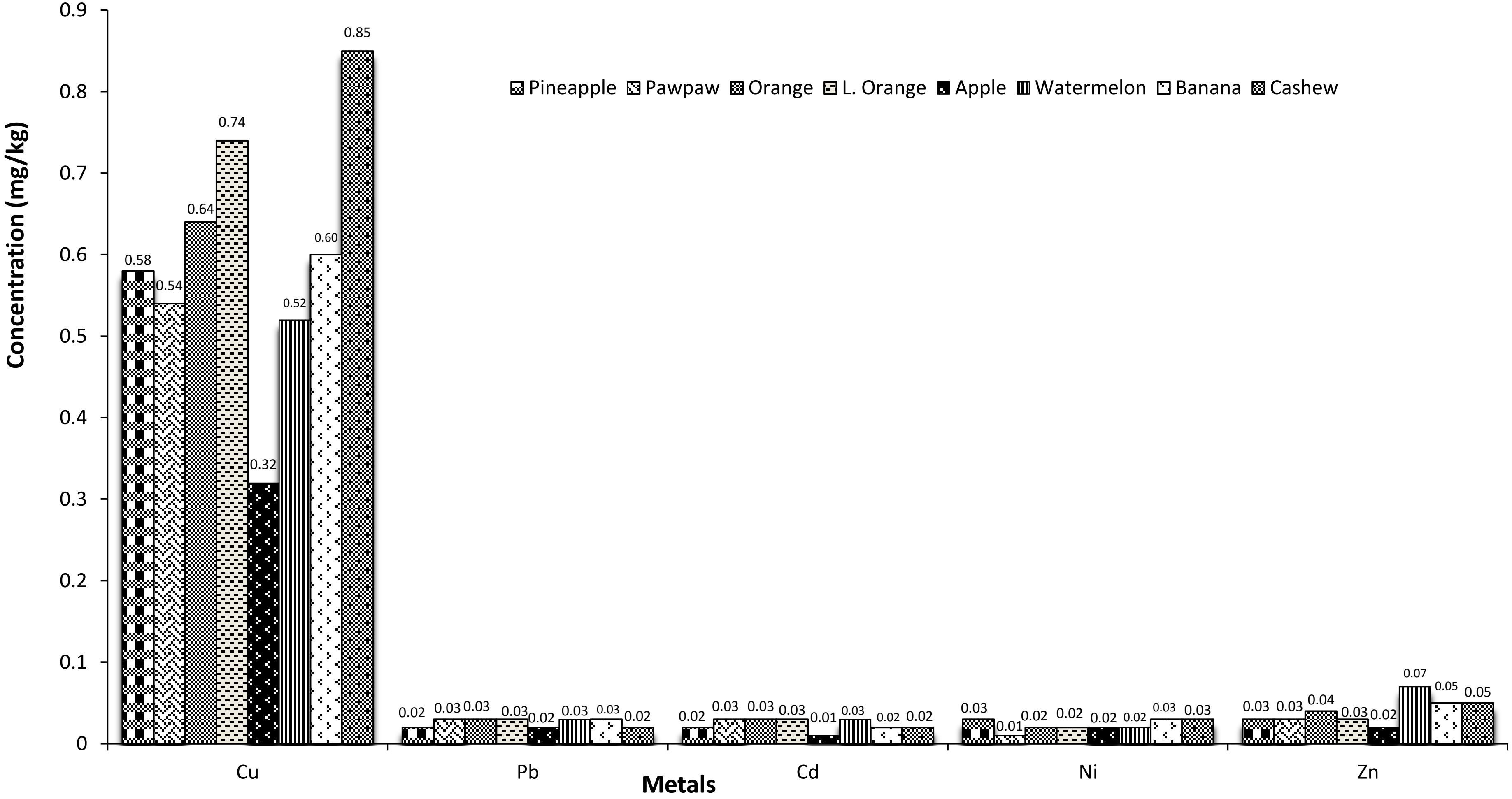

Fig 1: Average levels (mg/kg)of Cu, Pb, Cd, Ni and Zn in different fruit types

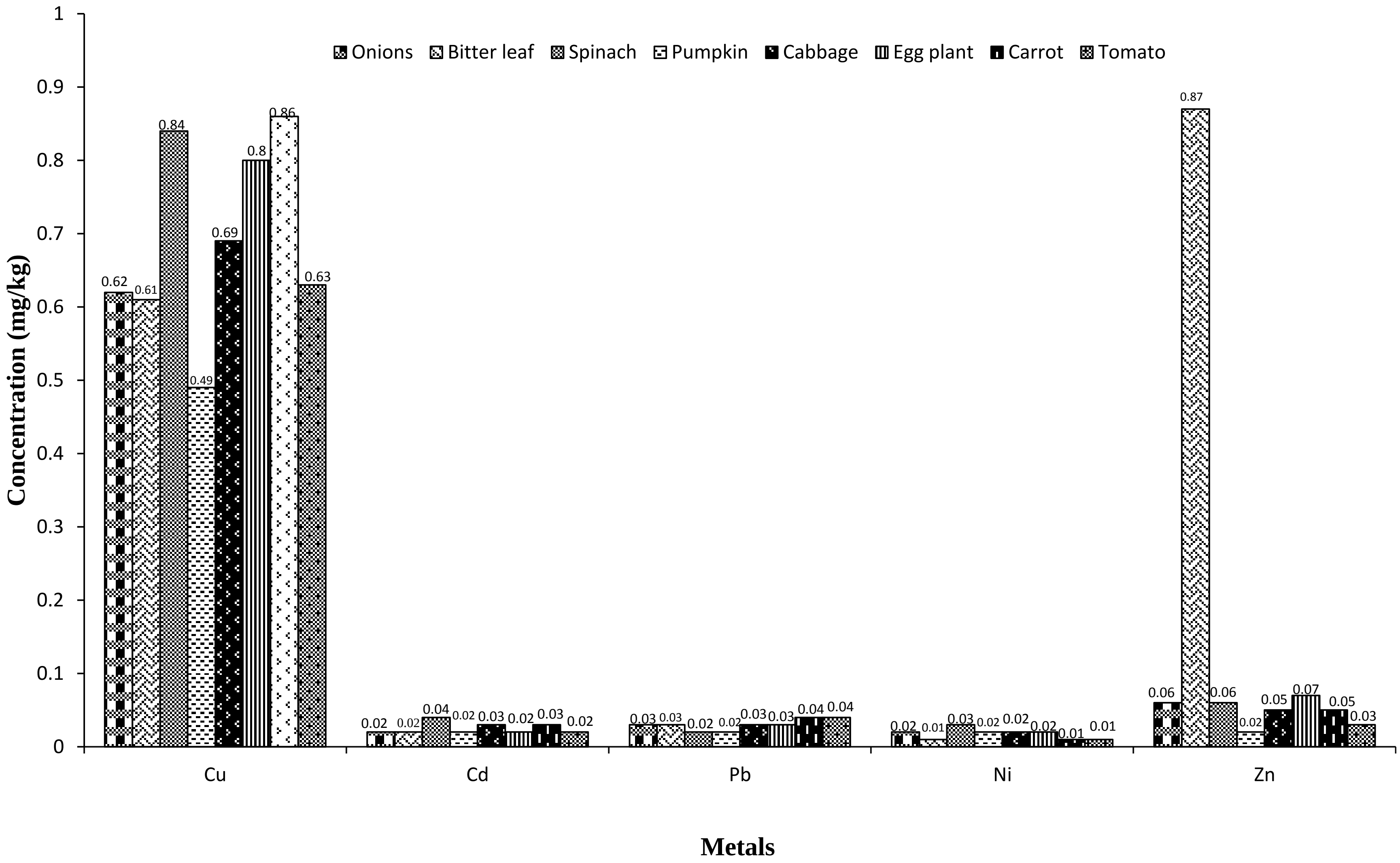

Fig 2: Average levels (mg/kg) of Cu,Cd, Pb and Zn in some vegetable types

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Table 2: Comparison of Cd and Pb levels (mg/kg) in some Nigerian foods with levels in similar foods in other countries

Food	This study		Nigeria[a]		Egypt[b]		Ethiopia[c]		Pakistan[d]		Greece[e]	
	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb
Pineapple	0.02	0.02	0.004	0.13	-	-	-	-	-	-	-	-
Pawpaw	0.03	0.03	0.003	0.07	-	-	-	-	-	-		-
Orange	0.03	0.03	0.01	0.11	0.04	0.05	-	-	-	-	0.001	-
Lime Orange	0.03	0.03	-	-	-	-	-	-	-	-	-	-
Apple	0.01	0.02	0.04	0.11	0.05	0.19	-	-	0.14	0.76	0.0003	-
Watermelon	0.03	0.03	0.004	0.11	0.02	0.03	-	-	-	-	0.0004	-
Banana	0.02	0.03	0.01	0.12	0.02	0.05	-	-	-	-	0.001	-
Cashew	0.02	0.02	-	-	-	-	-	-	-	-	-	-
Vegetables												
Onions	0.02	0.03	-	-	0.02	0.14	-	-	0.07	0.06	0.0003	-
Bitter leaf	0.02	0.03	0.06	0.14	-	-	-	-	-	-	-	-
Spinach	0.04	0.02	-	-	0.11	0.34	-	-	-	-	0.05	-
Pumpkin	0.02	0.02	0.09	0.21	-	-	-	-	-	-	-	-
Cabbage	0.03	0.03	0.06	0.13	-	-	-	-	-	-	-	-
Egg plant	0.02	0.03	-	-	0.02	0.21	-	-	0.31	1.30	0.03	-
Carrot	0.03	0.04	-	-	0.01	0.18	0.07	0.08	-	-	0.01	-
Tomato	0.02	0.04	-	-	0.01	0.26	-	-	0.33	1.56	0.02	-

a Sobukola and Dairo, 2007
bRadwan and Salama, 2006
cRahlenbeck *et al*, 1999
d Parveen *et al*., 2003
e Karavoltsos *et al*., 2002

Table 3: Comparison of Cu, Ni and Zn levels (mg/kg) in some Nigerian fruits with levels in similar foods in other countries

Fruits	This study			Nigeria[f]			Nigeria[g]			Egypt[h]			Pakistan[i]		
	Cu	Ni	Zn	Cu	Ni	Zn	Cu	Ni	Zn	Cu	Ni	Zn	Cu	Ni	Zn
Pineapple	0.58	0.03	0.03	0.80	-	0.97	0.02	0.13	0.05	-	-	-	-	-	-
Pawpaw	0.54	0.01	0.03	0.64	-	2.80	0.003	0.11	0.05	-	-	-	-	-	-
Orange	0.64	0.02	0.04	2.13	-	2.20	0.002	0.12	0.04	1.27	-	2.38	-	-	-
Lime Orange	0.74	0.02	0.03	-	-	-	-	-	-	-	-	-	-	-	-
Apple	0.32	0.02	0.02	0.25	-	2.05	0.003	0.12	0.05	1.47	-	1.38	0.50	-	2.05
Watermelon	0.52	0.02	0.07	2.13	-	7.40	0.04	0.14	0.05	1.22	-	5.38	-	-	-
Banana	0.60	0.03	0.05	0.95	-	1.50	0.01	0.13	0.05	2.51	-	5.59	-	-	-
Cashew	0.85	0.03	0.05	-	-	-	-	-	-	-	-	-			
Vegetables															
Onions	0.62	0.02	0.06	7.30	-	17.3	-	-	-	1.49	-	11.4	0.09	-	0.83
Bitter leaf	0.61	0.01	0.87	0.20	-	2.87	0.02	0.02	0.01	-	-	-	-	-	-
Spinach	0.84	0.03	0.06	-	-	-	-	-	-	4.48	-	20.9	-	-	-
Pumpkin	0.49	0.02	0.02	0.07	-	3.53	0.02	0.24	0.10	-	-	-	-	-	-
Cabbage	0.69	0.02	0.05	0.41	-	2.00	0.07	0.16	0.05	-	-	-	-	-	-
Egg plant	0.80	0.02	0.07	5.47	-	9.67	-	-	-	1.41	-	11.5	3.14	-	3.52
Carrot	0.86	0.01	0.05	0.40	-	1.70	-	-	-	1.51	-	8.03	-	-	-
Tomato	0.63	0.01	0.03	0.36	-	1.00	-	-	-	1.83	-	7.69	2.31	-	2.45

fWapnir and Balkman, 1991
g Sobukola and Dairo, 2007
h Radwan and Salama, 2006
I Parveen *et al*., 2003

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DISCUSSION

Results indicated that all fruits and vegetable determined were acidic except watermelon and tomato with pH of 10.1 and 9.44 respectively. Lime orange was more acidic (1.47) followed by apple and onions with pH of 2.09 and 2.91 respectively. The ash content reflects the level of inorganic elements present in the fruits and vegetables, and they are of nutritional benefits.

The differences in levels of Cd in fruits and vegetables were not significant (P>0.05). Similar average Cd concentration of 0.03 mg/kg was observed in pawpaw, orange, lime orange and watermelon in fruits while in spinach 0.04 mg/kg was observed. The observed Cd levels in fruits in this study were slightly higher than the 0.2 mg/kg FAO/WHO regulatory standard limit for Cd in fruits (FAO/WHO, 2001). Cadmium is a very toxic heavy metal that has no beneficial functions in the human body. It accumulates principally in the kidneys and liver (Divrikli et al., 2006). The levels of Pb and Ni in fruits and vegetables were similar to observed Cd concentrations.

Copper levels were generally higher in the fruits and vegetables than the corresponding cadmium, lead, nickel and zinc levels in this study. However, these levels of Cu in fruits and vegetables are within 20.0 mg/kg FAO/WHO regulatory standard limits. In fruits the mean concentrations of Cu ranged from 0.32 mg/kg in apple to 0.85 mg/kg in cashew. Corresponding levels in vegetables ranged from 0.49 mg/kg in pumpkin to 0.86 mg/kg in carrot. Copper is an important element in human diet. Akinyele and Osibanjo (1982) reported that Cu is necessary for body pigmentation in addition to Fe, the maintenance of a healthy central nervous system, prevention of anaemia, and as well function as a biocatalyst and is interrelated with the function of Zn and Fe in the body.

Zn like Cu is an essential element in human diet. Very low concentration of Zn can cause problems. However, too much Zn is also harmful to human health (Agency for Toxic Substances and Diseases Registry, 2004). Excessive zinc intake can be harmful as it suppresses copper absorption (Wapnir and Balkman, 1991). Adverse effects of severely high zinc intake include nausea, vomiting, loss of appetite, stomach pains, headaches, and diarrhea (Joseph and Megan Ware, 2015) In this study, the mean levels of Zn ranged from 0.02 mg/kg in apple to 0.07 mg/kg in watermelon and in vegetables mean levels ranged from 0.02 mg/kg in pumpkin to 0.87 mg/kg in bitter leaf. The difference in mean Zn level in bitter leaf and the other vegetables determined was significant. However mean levels do not vary significantly among the vegetables and in fruits.

The levels of metals (Cd and Pb) found in this study of fruits and vegetables in major markets of Kogi State, North Central Nigeria were generally comparable to levels found in similar food items in some other parts of the world (Table 2). The average Cd concentrations in fruits in this study are not significantly different (p>0.05) from those obtained in a previous Nigerian study (Onianwa *et al*., 2001) and in Egypt (Radwan and Salama, 2006). However, this study results were higher than those obtained in Greece (Karavoltsos, 2002) and the difference was significant but in vegetables the differences were not significant. The level of metals in apple in this study is significantly lower (P<0.05) than obtained (Parveen*et al*., 2003) in

Pakistan. The levels of Cd in vegetables were lower than obtained in other parts of Nigeria (Onianwa *et al*., 2001) and significantly lower than obtained in Pakistan (Parveen *et al*., 2003) and in Egypt (Radwan and Salama, 2006).

Lead levels in fruits and vegetables in this study were lower than reported levels in previous work in Nigeria (ATSDR, 2009), in Egypt (Radwan and Salama, 2006) and in Pakistan (Parveen *et al*., 2003). The levels of Pb in eggplant and tomato reported (Parveen *et al*., 2003) in Pakistan are significantly higher th an obtained in this study.

The mean levels of Cu were similar to those obtained by other study in Nigeria (Karavoltsos, 2002), but lower than levels obtained (Sobukola, *et al*., 2010) in south west Nigeria in previous studies. However, the mean levels of Cu in Onions (0.62 mg/kg) and in eggplant (0.80 mg/kg) in this study are significantly lower ($P<0.05$) than previous work reported (Onianwa *et al*., 2001) as given in Table 3. The levels in fruits and vegetables in this study are significantly lower than levels reported in Egypt (Radwan and Salama, 2006).

The present study mean levels of Zn were similar to reported levels in south west Nigeria (Sobukola, *et al*., 2010) in previous study. However, they are significantly lower ($P<0.05$) than those obtained (Onianwa *et al*., 2001) in other parts of Nigeria. Similarly, the average levels of Zn in this study were lower than those reported in Egypt (Radwan and Salama, 2006) and Pakistan (Parveen*et al*., 2003).

The mean levels of Ni in fruits and vegetables in this study are slightly lower than the level obtained in southwest Nigeria (Sobukola, *et al*., 2010). Nickel also plays some role in body functions including enzyme functions. It activates some enzyme systems in trace amount but at higher concentration its toxicity becomes more prominent (Divrikli, 2006).

The results compared favourably with those from previous studies in Nigeria and other countries with similar food items. This study provides data from North Central Nigeria on heavy metals in some Nigeria fruits and vegetables. The concentrations of Cd and Pb were slightly above FAO (2001) regulatory standard and this makes the fruits and vegetables of concern and may pose health challenge. In view of the health risks posed by heavy metals in their mechanism in human body, there is need to monitor continuously various ways in which they can enter the body especially through food items.

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