

Health Risk Assessment of Heavy Metal Concentrations in Some Commonly Sold Fruits in Lafia City Modern Market

Matthew Olaleke Aremu¹✉, Halima Eshi Ibrahim¹,
Jude Chinedu Onwuka¹, Augustine Uche Abel¹,
Yahaya Yakubu Ishaleku¹

¹Department of Chemistry, Federal University of Lafia, PMB 146, Nasarawa State, Nigeria

Abstract: Levels of nickel, zinc, arsenic, copper, lead, cadmium, chromium, iron, mercury and selenium were determined in mango (*Magnifera indica*), cucumber (*Cucumis sativus*) and pawpaw (*Carica papaya*) sold in Modern Market (Central Market) of Lafia City, Nasarawa State, Nigeria. The study also assessed the potential health risks from the consumption of these fruits. The samples were randomly collected at two different times for one month interval, processed and analyzed for heavy metals using atomic absorption spectrophotometer (AAS). The result showed that the mean metal concentrations recorded for mango, cucumber and pawpaw in the decreasing order were: Cd < As < Pb < Cr < Hg < Se < Ni < Zn < Cu < Fe, Cd < As < Hg < Pb < Cr < Se < Ni < Cu < Zn < Fe and Hg < Ni < Cd < As < Pb < Cr < Se < Zn < Cu < Fe, respectively. Generally, the highest mean metal concentration in all the fruit samples was Fe. The average daily intake (ADI) values indicate that all the heavy metals in the fruit samples are below the permissible limits as endorsed by WHO/FAO. Also, hazard quotient (HQ) and hazard indices (HI) values do not exceed unity. Therefore, none of the fruit samples under investigation poses any health risks to the consumers as their HI was less than 1.

Keywords: Mango, Cucumber, Pawpaw, ADI, HQ, HI

Introduction

Heavy metals are generally referred to as metals and metalloids having densities greater than 5 g cm⁻³ [1]. Heavy metals are harmful because of their non-biodegradable in nature, long biological half-lives, and potential to accumulate in the body. Prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans, disrupting numerous biochemical processes and leading to cardiovascular, nervous, kidney and bone diseases [2, 3]. Some heavy metals, such as Cd, Cr, and Pb, are nonessential and can cause adverse human health effects [4]. Other heavy metals, such as (Zn, Ni, Fe and Cu) are micronutrients for human beings, but high intake of these metals may affect health. Heavy metals are the main contaminants of food supply as they are considered a vital concern in food safety and quality assurance and can be regarded as a major problem in the environment (soil, water, air) [5, 6].

Fruits are essential and beneficial for maintaining health, preventing and treating several diseases [5]. These fruits comprise both essential and toxic heavy metals with a wide variety of concentrations [7]. Protective antioxidants and phytonutrients in fruits include; flavonoids, anthocyanins and phenylpropanoids are critical best functioning of human immune system, protecting against communicable and non-communicable diseases such as cancer and other degenerative diseases [8]. Fruits

such as mango, pawpaw and cucumber are among the important diets in Lafia, the capital of Nasarawa State. It is known that consumption of these fruits regularly could improve health practices. Fruits contamination by heavy metals is one of the major problems that arise from the huge uses of fertilizers, pesticides and other chemicals due to the pursuit for fast economic development by industrial and new agriculture practices [9]. Therefore, the main objective of this study was to determine the concentrations of Ni, Zn, As, Cu, Pb, Cd, Cr, Fe, Hg and Se in mango (*Magnifera indica*), cucumber (*Cucumis sativus*) and pawpaw (*Carica papaya*) sold in Modern Market (Central Market) of Lafia City, Nasarawa State, Nigeria, and to consider health risk assessment of the heavy metals in the fruit samples.

Materials and Methods

Sample Collection

The three different fruits of cucumber (*Cucumis sativus*), mango (*Magnifera indica*) and pawpaw (*Carica papaya*) were collected from Lafia Modern Market at two different times within an interval of one month. The samples were bought from hawkers inside the market. The fruits in this market mainly come from villages within Nasarawa state and neighbouring states such as Plateau, Benue and Kogi.





Fig. 1: Map Showing Location (Lafia City) Where the Samples were Collected

Sample Preparation and Treatment

The samples were cleaned thoroughly using tap and rinse three times with distilled water to remove surface dirt. The clean samples were cut into small slices after peeling (cucumber was not peeled) and open air dried on paper for about 3 h to eliminate excess moisture. After that, each sample was weighed, dried in an oven at 70°C for several hours until constant weight is achieved. The samples were pounded into powder form using mortar and pestle, and later ground into fine powder that could pass through a 0.2 mm mesh sieve using food blender before stored in clean and dry polyethylene bag for further analysis.

Determination of Moisture Content

Moisture content was determined according to the standard methods of Association of Official Analytical Chemists [10]. Porcelain crucibles were cleaned and dried in the oven at 100°C for 1 h to achieve a constant weight. Sample were cooled in a desiccator and then weighed. 2 g of sample was placed in each crucible and dried in the oven at 100°C until constant weight was achieved. The crucible together with the samples were cooled in a desiccator and weighed. The relationship in Equation (1) was used to determine the moisture content.

$$\% \text{ Moisture content} = \frac{W_2 - W_3}{W_2 - W_1} \times \frac{100}{1} \quad (1)$$

Where:

W_1 = weight of crucible

W_2 = weight of crucible + sample before drying

W_3 = weight of crucible + sample after drying

Mineral analysis

The standards of Ni, Zn, As, Cu, Pb, Cd, Cr, Fe, Hg and Se solutions of 0.2, 0.4, 0.6, 0.8 and 1.0 mgL⁻¹ were prepared from each of the metal solutions of 1000 mgL⁻¹ stock solutions. The filtrates of the

digested samples were analysed by atomic absorption spectrophotometer (AAS). The detection limit of the metals in the sample was 0.000 mgL⁻¹ by means of the UNICAM 929, London, AAS powered by the solar software. The optimal analytical range was 0.1 to 0.5 absorbance units with coefficient of variation from 0.9% to 2.21%.

Average Daily Intake (ADI)

The average daily intake (ADI) of a heavy metal was calculated as a product of average fruit daily consumption per person percentage of dry weight of fruits and average heavy metal concentration per dry weight fruit as shown in the following equation [11];

$$ADI = A_{V_{\text{consumption}}} \times \% DW_{\text{fruit}} \times C_{\text{heavy metal}}$$

Where ADI is average daily intake of heavy metal per person per day (mg/person/day), $A_{V_{\text{consumption}}}$ is average daily consumption of fruit per person per day (g/day), $\% DW_{\text{fruit}}$ is percentage of dry weight of fruit ($\% DW = 100 / 100 - [\% \text{ moisture}]$) and $C_{\text{heavy metal}}$ is average of heavy metal concentration of dry weight of fruit (mg/g). The average daily consumption of fruits suggested by WHO guideline in human diet is 300 to 350 g per person [12]. An average of weight of person was considered to be 60 kg [13].

Hazard Quotient (HQ)

Hazard quotient (HQ) is a proportion of the probable exposure to an element/ chemical and level at which no negative impacts are expected. When the quotient is 1, it signifies there are potential health risks due to exposure [14]. The HQ is calculated as a fraction of determined dose to the reference dose as shown in the following equation;

$$HQ = ADI / R_f D$$

Where ADI is the average heavy metal intake per day (mg/kg/day) and $R_f D$ is the oral reference dose of the metal (mg/kg/day), $R_f D$ is an approximation of daily tolerable exposure to which a person is expected to have without any significant risk of harmful effects during a lifespan. $R_f D$ for the following metals are:

Ni = 0.02, Zn = 0.30, Cu = 0.04, Pb = 0.004, Cd = 0.0005, Pb = 0.004, Cr = 0.003, Fe = 0.70, Mn = 0.14 mg/kg/day [13].

Hazard Index (HI)

Hazard index (HI) is a vital index that assesses overall likely impacts that can be posed by exposure to more than one contaminant. When the HI is greater than 1, this suggests that there are significant health effects from consuming pollutants contained in a foodstuff. The HI is calculated as an arithmetic sum of the hazard quotients for each pollutant as shown in the following equation:

$$HI = \text{Sum of } ADI_{Ni} / R_f D_{Ni} + ADI_{Zn} / R_f D_{Zn} + ADI_{Cu} / R_f D_{Cu} + ADI_{Pb} / R_f D_{Pb} + ADI_{Cd} / R_f D_{Cd} + ADI_{Cr} / R_f D_{Cr} + ADI_{Fe} / R_f D_{Fe}$$

Results and Discussion

The result of concentrations of heavy metals in mango (*Magnifera indica*) samples is shown in Table 1. The values of Ni, Cd and Se were not at detectable range of AAS for second sampling. The levels of concentration of Ni, Cu, Pb, Cd, Fe, Hg and Se are

higher in first sampling compared with that of second sampling and CV% varies from 10.88 in Hg to 141.42 in (Ni, Cd and Se). The mean metal concentrations recorded for mango are in the decreasing order of Cd < As < Pb < Cr < Hg < Se < Ni < Zn < Cu < Fe.

Table 1: Mean Concentrations (mg/100 g dw) of Heavy Metals in Mango (*Magnifera indica*) Samples

Metal	Time of Sampling		Mean	SD	CV%
	1 st	2 nd			
Ni	0.0619	ND	0.03095	0.04377	141.4214
Zn	0.0387	0.0562	0.04745	0.012374	26.07875
As	0.0006	0.0006	0.0006	0	0
Cu	0.0624	0.0485	0.05545	0.009829	17.72549
Pb	0.0011	0.0009	0.001	0.000141	14.14214
Cd	0.0005	ND	0.00025	0.000354	141.4214
Cr	0.0009	0.0012	0.00105	0.000212	20.20305
Fe	0.0965	0.0517	0.0741	0.031678	42.75086
Hg	0.0014	0.0012	0.0013	0.000141	10.87857
Se	0.0327	ND	0.01635	0.023122	141.4214

CV = Coefficient of Variation; SD = Standard Deviation; ND = Not Detected

Table 2 shows the result of concentrations of heavy metals in cucumber obtained from Lafia Modern Market. The values of Ni, Cd and Se were not at detectable range of AAS in the second sampling. The concentrations of Zn, Se and Cu are essential to human and are required in trace amount. However, elevated concentration could be harmful. Generally, low level of these metals were found in both mango and pawpaw. Cr and Cd were recorded low in concentrations while As, Cu, Pb were reported to be high in concentration which are more toxic. The

presence of harmful elements such as Cd, Pb, and As in the fruits may be attributed to the environmental factors on the soils where the crops were grown. The levels of concentration of Ni, As, Cu, Cd, Fe, Hg and Se are higher in first sampling compared with that of second sampling and CV% varies from 6.65 in Zn to 141.42 in (Ni, Cd and Se). The mean metal concentrations recorded for mango are in the decreasing order of Cd < As < Hg < Pb < Cr < Se < Ni < Cu < Zn < Fe.

Table 2: Mean Concentrations (mg/100 g dw) of Heavy Metals in Cucumber (*Cucumis sativus*) Samples

Metal	Time of Sampling		Mean	SD	CV%
	1 st	2 nd			
Ni	0.03163	ND	0.015815	0.022366	141.42
Zn	0.03627	0.03985	0.03806	0.002531	6.65
As	0.00046	0.00041	0.000435	3.54E-05	8.13
Cu	0.0399	0.03277	0.036335	0.005042	13.88
Pb	0.00034	0.00072	0.00053	0.000269	50.70
Cd	0.00036	ND	0.00018	0.000255	141.42
Cr	0.00082	0.00103	0.000925	0.000148	16.05
Fe	0.06890	0.0411	0.055	0.019658	35.74
Hg	0.0009	0.00007	0.000485	0.000587	121.01
Se	0.02654	ND	0.01327	0.018767	141.42

CV = Coefficient of Variation, SD = Standard Deviation, ND = Not Detected

The concentrations of heavy metals in pawpaw fruit are presented in Table 3. The mean concentrations (mg/100 g) obtained were Ni (0.000915), Zn (0.03735), As (0.00535), Cu (0.0429), Pb (0.0054), Cd (0.0021), Cr (0.00655), Fe (0.06155), Hg (0.0005) and Se (0.01235). The CV% varies from 4.62 in Cu to 141.42 in (Ni, Cd and Se). The mean metal concentrations recorded for pawpaw were in the decreasing order of Hg < Ni < Cd < As < Pb < Cr < Se < Zn < Cu < Fe. The concentrations of Ni, Cd and

Se were not at detectable range of AAS in the second sampling. Generally, the highest mean metal concentration in all the samples was Fe (Tables 1 to 3). This was due to the level of iron in the soils where all the crops were grown. The observation here confirms that most soils contain appreciable quantities of iron [15]. Iron is an essential nutrient for humans, hence, appropriate content of iron in food crops is essential for good health.

Table 3: Mean Concentrations (mg/100 g dw) of Heavy Metals in Pawpaw (*Carica papaya*) Samples

Metal	Time of Sampling		Mean	SD	CV%
	1st	2 nd			
Ni	0.00183	ND	0.000915	0.001294	141.42
Zn	0.0311	0.0436	0.03735	0.008839	23.66
As	0.0058	0.0049	0.00535	0.000636	11.90
Cu	0.0443	0.0415	0.0429	0.00198	4.62
Pb	0.0010	0.0098	0.0054	0.006223	115.23
Cd	0.0042	ND	0.0021	0.00297	141.42
Cr	0.0109	0.0022	0.00655	0.006152	93.92
Fe	0.0752	0.0479	0.06155	0.019304	31.36
Hg	0.0001	0.0009	0.0005	0.000566	113.14
Se	0.0247	ND	0.01235	0.017466	141.42

CV = Coefficient of Variation, SD = Standard Deviation, ND = Not Detected

The moisture content of the three samples is displayed in Table 4. Cucumber has the highest mean value of 97.20% and pawpaw with the least mean value of 90.70%. The CV% varies from 0.01 in cucumber to 0.65 in mango sample.

Table 4: Moisture Content (%) of the Samples

Fruit	Time of Sampling				
	1 st	2 nd	Mean	SD	CV%
Cucumber	97.20	97.19	97.20	0.01	0.01
Mango	92.34	93.19	92.77	0.60	0.65
Pawpaw	90.70	90.90	90.80	0.14	0.15

CV = Coefficient of Variation, SD = Standard Deviation

Estimating heavy metal exposure levels is indispensable in determining organism health risk

(Singh *et al.*, 2010). Among the various routes of human exposure to heavy metal, food chains, especially fruits and vegetables, are the most significant. The average daily intakes (ADI) of seven metals (Ni, Zn, Cu, Pb, Cd, Cr, and Fe) in the studied samples were estimated according to the mean concentration of each metal in each fruit. The ADI and the permitted maximum tolerable daily intake (PMTDI) of studied metals from the consumption of three fruits samples are shown in Table 5. The ADI values indicate that all the fruit samples' heavy metals are below the permissible limits as endorsed by [12, 13].

Table 5: Average daily intake (ADI) of heavy metals (mg/person/day) in the fruits

Metal	Cucumber	Mango	Pawpaw	PMTDI
Ni	2.4×10^{-5}	1.2×10^{-4}	4.5×10^{-6}	0.3
Zn	5.8×10^{-5}	1.9×10^{-4}	1.9×10^{-4}	15.0
As	na	na	na	na
Cu	5.5×10^{-5}	2.2×10^{-4}	2.1×10^{-4}	2.0
Pb	8.0×10^{-7}	3.9×10^{-6}	2.7×10^{-5}	0.21
Cd	2.7×10^{-7}	9.8×10^{-7}	1.0×10^{-5}	0.046
Cr	1.4×10^{-6}	4.1×10^{-6}	3.3×10^{-5}	0.2
Fe	8.3×10^{-5}	2.9×10^{-4}	3.1×10^{-4}	15.0
Hg	na	na	na	na
Se	na	na	na	na

PMTDI = Permitted maximum tolerable daily intake (FAO/WHO, 1996; WHO/FAO, 2013); na = Not available

Nickel is involved in a variety of bodily functions, including enzyme functions. In general, it occurs more frequently in plants than in animal bodies. A trace amount of Ni activates some enzyme systems, but at higher concentrations, it is toxic [16]. Ni disrupts calcium metabolism, which can lead to cancer in the human body. It has also been suggested that high levels of nickel may impair iron absorption or utilization when iron status is low [17]. The ADI values of Ni in the samples tested varied between 4.5×10^{-6} (in pawpaw) and 1.2×10^{-4} (in mango) mg/person/day. Zinc is a critical component of the human physiological system. It is required for enzymatic function and contributes to the synthesis of DNA, protein, and insulin. Zn is also necessary for normal cell function, such as protein synthesis, carbohydrate metabolism, cell growth, and cell division [18]. The estimated ADI value of zinc is 2.0×10^{-4} (cucumber) and 6.0×10^{-4} (cucumber and

pawpaw) mg/person/day in all the three fruit samples, below the PMTDI limit as recommended by [12]. However, Zn is toxic to humans when the tolerable limit is exceeded. According to report of some researchers, chronic exposure to Zn and Cu is linked to Parkinson's disease [19].

Copper is an essential element in the human body because it maintains central nervous system health, proper metabolic processes, pigmentation, and anaemia prevention [20]. Because of the nature of the role, copper plays, continuing low Cu levels are harmful to humans [21]. When people with Wilson's disease are overexposed to copper, they are at a high risk of developing health problems [2]. When Cu levels in the human body exceed safe levels, it causes hypertension, sporadic fever, coma, anaemia, liver and kidney damage, as well as stomach and intestine irritation [14]. Lead is a non-essential element whose

high concentration in the human body can cause nephrotoxicity, neurotoxicity, and liver, lung, and spleen damage [22]. Pb has no known any biochemical functions and is a serious cumulative body poison that enters the body through the air, water, and food. It is not possible to remove it by washing fruits and vegetables [16]. Lead is emitted into the air by car exhaust, metal smelting, battery manufacturing, painting outside buildings, and other factories that use lead. An increase in the amount of Pb in the soil may increase the availability of Pb in vegetables and fruits [23]. This study showed that among three samples of fruits, maximum ADI of Pb was found in pawpaw with about 2.7×10^{-5} mg/person/day and minimum in cucumber with 8.0×10^{-7} .

Cadmium is a non-essential nutrient that accumulates in the kidneys and liver [16]. Higher cadmium concentrations may cause severe diseases such as tubular growth, kidney damage, cancer, diarrhoea, and incurable vomiting. Cadmium ADI values in the fruit samples were found to be 2.0×10^{-2} , 1.4×10^{-3} , and 5.0×10^{-4} mg/person/day in pawpaw, mango, and cucumber, respectively. The order of contribution for the Fe intake is pawpaw > mango > cucumber. The observed levels of ADI were below the PMTDI recommended by [12]. Excessive Fe in the human body can result in siderosis in the liver, pancreas, thyroid, pituitary, adrenal glands, and heart, depending on the chemical form. It is also reported that doses with Fe content larger than 20 mg/person/day may cause stomach upset, constipation, and blackened stools [13]. The presence of Cr in the diet is critical due to its active role in lipid metabolism and insulin function. The ADI for Cr in cucumber was found to be 1.4×10^{-6} mg/person/day and 3.3×10^{-5} mg/person/day in pawpaw. Chromium is also an essential micronutrient for plants, but in high concentrations, it can be toxic to plants, animals, and humans [24]. Cr toxicity affects many human organs, including the liver, kidney, lungs, and spleen, resulting in severe biochemical defects and cancer.

Table 6: Hazard quotient and Hazard Index for each Studied Sample

Metal	Hazard Quotient		
	Cucumber	Mango	Pawpaw
Ni	1.0×10^{-3}	5.0×10^{-3}	2.0×10^{-4}
Zn	2.0×10^{-4}	6.0×10^{-4}	6.0×10^{-4}
As	na	na	na
Cu	1.4×10^{-3}	5.5×10^{-3}	5.3×10^{-3}
Pb	2.0×10^{-4}	1.0×10^{-3}	6.8×10^{-3}
Cd	5.0×10^{-4}	2.0×10^{-3}	2.0×10^{-2}
Cr	5.0×10^{-4}	1.4×10^{-3}	1.1×10^{-2}
Fe	1.0×10^{-4}	4.0×10^{-4}	4.0×10^{-4}
Hg	na	na	na
Se	na	na	na
HI = \sum HQ	3.9×10^{-3}	1.6×10^{-2}	4.4×10^{-2}

na = Not available

The result showed that the hazard quotient (HQ) values of all the heavy metals in the fruit samples were less than 1 (Table 6). When HQ exceeds 1, it indicates that exposure may have a negative impact on health [25]. This study revealed no apparent health risk to the local population from consuming individual metals found in the fruit samples. Estimation of hazard index (HI), which takes care of the chemical mixtures, is fundamental in assessing the effects of all the heavy metals in a fruit sample. The result in Table 6 showed that the HI was highest in pawpaw (4.4×10^{-2}) followed by mango (1.6×10^{-2}) and then cucumber (3.9×10^{-3}). When the HI exceeds unity, it suggests that consumers may experience potential health hazards due to dietary intake of heavy metals in the sample [25, 26]. The study indicates that none of the fruit samples under investigation poses any health risk to the consumers as their HI was less than one.

Conclusion

The results revealed that the highest mean heavy metal concentration in all the fruit samples was Fe. The average daily intake (ADI) values indicate that all the heavy metals in the fruit samples are below the permissible limits as endorsed by WHO/FAO. Also, hazard quotient (HQ) and hazard indices (HI) values do not exceed unity. Therefore, none of the fruit samples under investigation poses any health risks to the consumers as their HI was less than 1. However, no matter how low levels of heavy metals are present in fruits or any foodstuffs, their presence is not desirable. This study therefore, recommends regular monitoring of heavy metals in fruits and foodstuffs so as to prevent excessive accumulation in food chain.

Conflict of Interest

The authors declare that there is no conflict of interest reported in this study.

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