

Concentrations and Risk Evaluation of Selected Heavy Metals in *Amaranthus* (L.) Leaf Cultivated in Katsina State, North West Nigeria

ABSTRACT

Bioaccumulation of seven heavy metals (Cr, Cd, Fe, Ni, Mn, Pb and Zn) in *Amaranthus* leaf cultivated in Katsina state Nigeria were measured using atomic absorption spectrometer. The health risks to the local inhabitants from the consumption of the *Amaranthus* leaf were evaluated based on the Target Hazard Quotient. The possibility of cancer risks in the *Amaranthus* (L.) leaf through the intake of carcinogenic heavy metals was estimated using the Incremental Lifetime Cancer Risk. The target hazard quotient was (THQ)>1, indicating that the *Amaranthus* leaf cultivated may pose a non-carcinogenic risk for all the studied metals. Hazard index (HI) was low. The incremental cancer risk (ILCR) for Cd violated the threshold risk limit ($>10^{-4}$) and ILCR for Pb reached the moderate risk limit ($>10^{-3}$) in all the studied samples in adults, While in children ILCR for both Pb in samples from Dabai, Daura, Funtua, Matazu and Zango and Cd for all samples have reached the moderate risk limit ($>10^{-3}$), while the ILCR for Pb in samples from Birchi, Dutsinma, Kafur, Katsina and Malunfashi are beyond the moderate risk level ($>10^{-2}$). The study suggests that consumption of *Amaranthus* leaf cultivated in Katsina may contribute to the population cancer burden.

Keywords: *Amaranthus*, Heavy metals, Katsina, health risk index, cancer risk, Nigeria

1. INTRODUCTION

Amaranthus sp. is an erect erect, many-branched annual herb growing up to 1.5 m. The stem is smooth, robust, cylindrical and often reddish. The leaves are simple and alternate, glabrous or with sparse hairs on the main veins below, often diamond-shaped, long petiolate, up to 9 cm. They are dotted with numerous translucent spots; the venation is well marked. The leaf axils bear pairs of fine and slender spines. The plant occurs on roadsides, villages, disturbed areas, along the livestock holding areas. It is a weed of crops. It prefers rich soil in organic matter and nitrogen [1]. Agricultural foods are usually adulterated with pollutants, especially heavy metals by direct and indirect industrial activities, automobile exhaust, excess metal-based fertilization, and pesticide application. In contrast, some heavy metals (As, Cd, and Pb) have no known beneficial role in human metabolism and are considered as chemical carcinogens even at very low levels of exposure [1]. Among the food system, vegetables are the most exposed food to environmental pollution due to aerial burden [2]. Vegetables play important roles in human nutrition and health, particularly as sources of vitamin C, thiamine, niacin, pyridoxine, folic acid, minerals, and dietary fibre [3]. Prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases [1, 4].

In Katsina State Nigeria, there is limited information on the levels of heavy metals in locally cultivated leafy vegetables. This work therefore seeks to bridge that gap by providing information especially to the Katsina state populace on the levels of heavy metals of these most consumed vegetables. Information will further be provided on the heavy metals

composition of the sources of these vegetables and the extent to which they are contaminated with these heavy metals for future studies and effective comparative analysis. The objective of this study, therefore, was to evaluate human exposure to some heavy metals through the consumption of some locally cultivated leafy vegetables in Katsina State, Nigeria.

2. MATERIAL AND METHODS

2.1 STUDY AREA AND SAMPLE COLLECTION

The study was carried out during 2017 in Katsina State, Nigeria located between latitude 12°15'N and longitude of 7°30'E in the North West Zone of Nigeria, with an area of 24,192km² (9,341 sq meters). Katsina State has two distinct seasons: rainy and dry. The rainy season begins in April and ends in October, while the dry season starts in November and ends in March. This study was undertaken during the dry season. The average annual rainfall, temperature, and relative humidity of Katsina State are 1,312 mm, 27.3°C and 50.2%, respectively (5). The study was conducted within some catchment areas that cultivate Amaranthus located within the 3 senatorial zones constitute to make up the state (Katsina senatorial zone: Birchi, Dutsinma and Katsina; Daura senatorial zone: Daura, Ingawa and Zango; Funtua senatorial zone: Dabai, Funtua, Kafur, Malunfashi and Matazu). Sampling for this work was carried out by dividing the catchment areas into five (5) locations. In each of the locations, the plot where the vegetables are cultivated was subdivided into twenty (20) sampling areas. Samples were collected from each of the areas and combined to form bulk sample, from which a representative sample was obtained. The samples were code-named and stored in glass bottles with tight covers to protect them from moisture and contamination. It was then stored in the refrigerator at 40°C until ready for use

2.2 IDENTIFICATION OF SAMPLE

The samples were identified in the herbarium of the department of biology in Umaru Musa Yar'adua University, Katsina, Nigeria.

2.3 SAMPLE PREPARATION

Amaranthus leaf samples were washed with tap water thoroughly to remove dust particles, soil, unicellular algae etc. The edible parts of the samples were further washed with distilled water and finally with deionized water. The washed vegetables were dried with blotting paper followed by filter paper at room temperature to remove surface water. The vegetables were immediately kept in desiccators to avoid further evaporation of moisture from the materials. The vegetables were then chopped into small pieces and oven dried at (55± 1)°C for hours in a Gallenkamp hotbox oven (CHF097XX2.5) and then blended in an electric blender. The resulting powder was kept in airtight polythene packet at room temperature before digestion and metals analyses.

2.4 HEAVY METALS DETERMINATION

5 g of each Sample was dried at 800°C for 2 hours in Gallenkamp hotbox oven (CHF097XX2.5) and then blended in an electric blender. A total of 0.5 g of each sample was weighed and ashed at 5500°C for 24 hours in an electric muffle furnace (Thermolyne FB131DM Fisher Scientific). The ash was diluted with 4.5 ml concentrated hydrochloric acid (HCl) and concentrated nitric acid (HNO₃) mixed at ratio 3:1 the diluent is left for some minutes for proper digestion in a beaker. 50 ml of distilled water was added to the diluents to make up to 100 ml in a volumetric flask. The levels of heavy metals (Pb, Zn, Ni, Cd, Cr, Mn and Fe) were determined using AA210RAP BUCK Atomic Absorption Spectrometer flame emission spectrometer filter GLA-4B Graphite furnace (East Norwalk USA), according to standard methods (6) and the results were given in (mg/kg).

2.5 HEAVY METAL HEALTH RISK ASSESSMENT

2.5.1 DAILY INTAKE OF METALS (DIM)

The daily intake of metals was calculated using the following equation:

$$DIM = \frac{C_{\text{metal}} \times C_{\text{factor}} \times D_{\text{met}}}{B_{\text{weight}}}$$

Where, C_{metal} , C_{factor} , D_{intake} and B_{weight} represent the heavy metal concentrations in the samples, the conversion factor, the daily intake of the food crops and the average body weight, respectively. The conversion factor (CF) of 0.085 [7] was used for the conversion of the samples to dry weights. The average daily intake rate was 0.527 kg person⁻¹ d⁻¹ [8] and the average body weight for the adult and children population was 60 kg [9] and 24 kg [10] respectively; these values were used for the calculation of HRI as well.

2.5.2 NON-CANCER RISKS

Non-carcinogenic risks for individual heavy metal in samples were evaluated by computing the target hazard quotient (THQ) using the following equation (11).

$$\text{THQ} = \text{CDI} / \text{R}_D$$

CDI is the chronic daily heavy metal intake (mg/kg/day) obtained from the previous section and R_D is the oral reference dose (mg/kg/day) which is an estimation of the maximum permissible risk on human population through daily exposure, taking into consideration a sensitive group during a lifetime [12]. The following reference doses were used (Pb = 0.6, Cd = 0.5, Zn = 0.3, Fe = 0.7, Ni = 0.4, Mn = 0.014, Cr = 0.3) [13, 14]. To evaluate the potential risk to human health through more than one heavy metal, chronic hazard index (HI) is obtained as the sum of all hazard quotients (THQ) calculated for individual heavy metals for a particular exposure pathway (15). It is calculated as follows:

$$\text{HI} = \text{THQ}_1 + \text{THQ}_2 + \dots + \text{THQ}_n$$

Where, 1, 2 ..., n are the individual heavy metals or vegetable and fruit species.

It is assumed that the magnitude of the effect is proportional to the sum of the multiple metal exposures and that similar working mechanism linearly affects the target organ [16]. The calculated HI is compared to standard levels: the population is assumed to be safe when $\text{HI} < 1$ and in a level of concern when $1 < \text{HI} < 5$ (17).

2.6 CANCER RISKS

The possibility of cancer risks in the studied millet samples through intake of carcinogenic heavy metals was estimated using the Incremental Lifetime Cancer Risk (ILCR) [18].

$$\text{ILCR} = \text{CDI} \times \text{CSF}$$

Where, CDI is chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure to the chemical carcinogen.

The US EPA ILCR is obtained using the cancer slope factor (CSF), which is the risk produced by a lifetime average dose of 1 mg/kg BW/day and is contaminant specific [11]. ILCR value in millet represents the probability of an individual's lifetime health risks from carcinogenic heavy metals exposure [19]. The level of acceptable cancer risk (ILCR) for regulatory purposes are considered within the range of 10^{-6} to 10^{-4} (12). The CDI value was calculated on the basis of the following equation and CSF values for carcinogenic heavy metals were used according to the literature [18].

$$\text{CDI} = (\text{EDI} \times \text{EFr} \times \text{ED}_{\text{tot}}) / \text{AT}$$

where EDI is the estimated daily intake of metal via consumption of the samples; EFr is the exposure frequency (365 days/year); ED_{tot} is the exposure duration of 60 years, the average lifetime for Nigerians; AT is the period of exposure for non-carcinogenic effects ($\text{EFr} \times \text{ED}_{\text{tot}}$), and 60 years lifetime for carcinogenic effect (11). The cumulative cancer risk as a result of exposure to multiple carcinogenic heavy metals due to consumption of a particular type of food was assumed to be the sum of the individual heavy metal increment risks and calculated by the following equation (18).

$$\sum \text{ILCR}_n = \text{ILCR}_1 + \text{ILCR}_2 + \dots + \text{ILCR}_n$$

Where, n = 1, 2..., n is the individual carcinogenic heavy metal

3. RESULTS AND DISCUSSION

Contamination of dietary substances by chemicals and non-essential elements such as heavy metals is known to have a series of adverse effects on the body of humans and animals (20). The present study investigated the presence of heavy metals in *Amaranthus* leaf which is a major component of the diet among the population in Katsina state, Nigeria. A total of 11 samples were analyzed for the presence of heavy metals in this study. As shown in Table 1, among the heavy metals evaluated, the highest concentration (mg/kg) was observed for Fe (range: 0.836-3.795), followed by Mn (range: 0.255-3.665), Zn (range: 1.227-1.421), Pb (range: 0.827-1.019) and Cr (range: 0.116-0.351). While Cd has the lowest concentration (range: 0.043-0.058). The results for the heavy metals were analysed in the sampled seeds are similar to that reported for heavy metals in beans and several beans products from selected markets in Katsina state, Nigeria [21].

The lead was detected in all the samples, with the majority of samples seen to be higher than 0.01 mg/kg which is the maximum permissible limit set by WHO/FAO and also the maximum allowable concentration of 0.02 mg/kg by EU and 0.05 mg/kg limit set by USEPA (22). The percentage of Pb in this study is higher than the maximum permissible limits set by WHO, EU & USEPA and have caused public health concerns considering the frequency of exposure. The Pb concentration range for the leaf samples in this study was lower than that reported for leafy vegetables from Kaduna state Nigeria (23) and that reported for beans samples from Italy, Mexico, India, Japan, Ghana and Ivory Coast with a Pb concentration range of 4.084- 14.475ppm (24) and the result of Pb in homegrown vegetables near a former chemical manufacturing facility in Tarnaveni, Romania (25). But the results are higher than that reported for the concentration of Pb from Kano (millet and sorghum) and Kaduna (cereals) states, Nigeria (26; 27). The value was still higher than the range (0.116 to 0.390) reported by Ahmed and Mohammed in foodstuff from Egypt in 2005 (28), the range (0.007 to 0.032 mg/kg) reported by Okoye et al., (29) in a study conducted in Southeast of Nigeria in 2009 in wheat flours, the results for Pb in leafy vegetables and the result for Pb in cucumber from Awka, Anambra state Nigeria (30). This difference was earlier been attributed to differences in anthropogenic activities that introduce metals into the soil in the areas where these samples were grown or even deposition of Pb on the surface of these vegetables during production, transport and Marketing or by emissions from Vehicles and industries (31).

The concentration of Cd (mg/kg) ranged from 0.043 to 0.058 in *Amaranthus* leaf samples are similar to be reported for Romaine lettuce and cabbage in a study conducted in Zhejiang China (32) and the Cd values for cabbage and cucumber from Awka, Anambra state Nigeria (30). But these values are higher than the range (0.002 to 0.004 mg/kg) reported by Edem et al., (33) in Wheat flours in 2009. The reported Cd values in the present study are lower than that reported for various beans samples from Europe, Asia and parts of West Africa (24), reported in a study for the Cadmium concentration range for both unprocessed and processed bean samples from Katsina state Nigeria (21) and for locust beans from Odo-Ori market Iwo, Nigeria (34). The values are also lower than those obtained by Okoye et al., (29) in Cereals in Southeastern Nigeria (0.007 to 0.23mg/kg) in 2009, Ahmed and Mohammed (28) in Cereal products (0.091-0.143mg/kg) in 2005, Orisakwe et al., (35) in Owerri (0.00 to 0.24mg/kg) in 2012 and Dahiru et al., (26) in Kano (0.11 to 0.28mg/kg) in 2013. These differences could be due to differences in the concentration of the metal in the soils where these vegetables were grown.

The mean values obtained for the heavy metal (Cr) is similar with the results of Gomaa et al., (36) for Cr in cucumber (0.16 mg/kg) from Egypt and the mean Cr concentration for market sold beans from Katsina, Nigeria (21). But the results are lower than the Cr concentrations reported for homegrown vegetables near a former chemical manufacturing facility in Tarnaveni, Romania (25)

The Fe values for the present study are higher than the range reported by Edem et al., (33) in Calabar (0.002 to 0.004mg/kg) in 2009. These values are also too low to provide for the Recommended Daily allowance for Fe in both adult male (10mg/day) and female (15mg/day) from a nutritional point of view (27). In the present study, the mean Fe concentration in the millet samples is higher than that reported in a study that evaluated heavy metals in millet from Kaduna, Nigeria (27). The result is similar to that reported for market sold beans from Katsina, Nigeria (21), but lower to that reported in a study in eastern Nigeria (Okoye et al., 2009) and that recorded by Zahir et al., (37) in a study conducted in Pakistan and the results for the study conducted by Di Bella et al., (24).

The heavy metal Zn values obtained in this study is similar to that reported in some studies on heavy metal evaluation in millet sorghum and various food crops (26; 38), but are higher than the range (0.04 to 0.19mg/kg) reported by Edem et al., (33) in 2009 but far below the range reported by Ahmed and Mohammed (28) in 2005 (4.893 to 15.450 mg/kg) and that reported in a study conducted by Sulyman et al. (39). These values can also not provide for the required daily allowance for Zn which is 11mg/day for men and 8mg/day for women (27).

Mn concentrations in the present study is lower than the result of Mn levels in turmeric (76 mg/kg) , red chilli (74.02 mg/kg) and coriander (52.91 mg/kg) reported by Das et al. (40) in their study conducted in Chittagong Metropolitan City,

175 Bangladesh to evaluate heavy metals in spices and results of evaluation of heavy metals in various foods reported in
 176 other studies (24, 29), but is similar to that reported by Yaradua et al., (21) in a study on Mn levels in beans from Katsina
 177 states Nigeria.

178 **Table 1 Heavy Metal Concentration (mg/kg) in Cultivated Amaranthus leaf Samples from Katsina State**

Location	Heavy Metal						
	Mn	Zn	Pb	Cd	Ni	Fe	Cr
Birchi	0.411000 ±0.000400	1.421000 ±0.000300	0.864000 ±0.000200	0.043000 ±0.000200	BD L	0.836000 ±0.01200	0.116000 ±0.000300
Dabai	0.827000 ±0.000200	1.392000 ±0.000200	0.827000 ±0.000300	0.046000 ±0.000200	BD L	1.325000 ±0.000300	0.254000 ±0.000200
Daura	0.348000 ±0.000200	1.257000 ±0.000200	0.953000 ±0.000400	0.051000 ±0.001300	BD L	0.924000 ±0.000200	0.137000 ±0.000200
Dutsinma	0.826000 ±0.001600	1.321000 ±0.000300	0.921000 ±0.001300	0.043000 ±0.000600	BD L	1.267000 ±0.000200	0.158000 ±0.000300
Funtua	0.421000 ±0.000300	1.480000 ±0.000200	0.857000 ±0.000300	0.051000 ±0.000200	BD L	1.913000 ±0.001300	0.164000 ±0.000200
Ingawa	0.318000 ±0.001000	1.251000 ±0.000400	0.811000 ±0.000100	0.042000 ±0.000600	BD L	0.937000 ±0.000300	0.258000 ±0.000800
Kafur	0.481000 ±0.000400	1.263000 ±0.000200	0.952000 ±0.000200	0.046000 ±0.000300	BD L	0.892000 ±0.000200	0.173000 ±0.000200
Katsina	0.267000 ±0.000200	1.382000 ±0.000200	0.893000 ±0.000300	0.058000 ±0.000200	BD L	1.512000 ±0.000600	0.351000 ±0.000200
Malunfashi	3.665000 ±0.000300	1.259000 ±0.001000	1.019000 ±0.000300	0.052000 ±0.000200	BD L	0.970000 ±0.001800	BDL
Matazu	0.255000 ±0.000800	1.227000 ±0.000400	0.843000 ±0.000300	0.044000 ±0.000200	BD L	3.795000 0.000400	0.297000 ±0.001800
Zango	0.351000±0.00 0300	1.211000±0.00 0300	0.721000±0.00 0200	0.043000±0.00 0300	BD L	2.360000±0.00 0300	0.160000±0.00 1300

179 Values are expressed as Mean ± Standard

180
 181
 182 The degree of heavy metal toxicity to humans depends on daily consumption rate (41). The results for the estimated daily
 183 intake (EDI) of the heavy metals on consumption of the samples were given in Tables 2 and 3. From the tables with the
 184 exception of the heavy metal Pb the estimated daily intake of the heavy metals (Zn, Cd, Cr, Fe and Mn) in adults and
 185 children were lower than the tolerable daily intake limit set by the USEPA (42) in all the samples.

192 **Table 2 Daily Intake of Heavy Metal in Adults from Consuming Cultivated Amaranthus from Katsina State**

Location	Heavy metal					
	Mn	Zn	Pb	Cd	Fe	Cr
Birchi	0.000307	0.001061	0.000645	0.000032	0.000624	0.000289
Dabai	0.000394	0.001039	0.000618	0.000034	0.000989	0.000190
Daura	0.000260	0.000939	0.000712	0.000038	0.000690	0.000391
Dutsinma	0.000617	0.000986	0.000688	0.000032	0.000946	0.000118
Funtua	0.000314	0.001105	0.000640	0.000038	0.001428	0.000120
Ingawa	0.000237	0.000934	0.000606	0.000031	0.000700	0.000187
Kafur	0.000359	0.000943	0.000711	0.000034	0.000666	0.000127
Katsina	0.000199	0.001032	0.000667	0.000043	0.001129	0.000261
Malunfashi	0.002736	0.000940	0.000815	0.000039	0.000127	BDL
Matazu	0.000190	0.000916	0.000629	0.000033	0.002833	0.000217
Zango	0.000262	0.000904	0.000538	0.000032	0.001762	0.000119

193
 194 **Table 3 Daily Intake of Heavy Metal in Children from Consuming Cultivated Amaranthus from Katsina State**

Location	Heavy metal					
----------	-------------	--	--	--	--	--

	Mn	Zn	Pb	Cd	Fe	Cr
Birchi	0.000767	0.002652	0.001613	0.000080	0.001560	0.000217
Dabai	0.001544	0.002598	0.001544	0.000086	0.002566	0.000474
Daura	0.000646	0.002346	0.001751	0.000095	0.001725	0.000256
Dutsinma	0.001542	0.002466	0.001719	0.000080	0.002365	0.000295
Funtua	0.000786	0.002762	0.001560	0.000086	0.003571	0.000306
Ingawa	0.000594	0.002346	0.001514	0.000078	0.001749	0.000482
Kafur	0.000898	0.002544	0.001590	0.000086	0.001665	0.000323
Katsina	0.000495	0.002560	0.001617	0.000012	0.002822	0.000655
Malunfashi	0.000684	0.002350	0.001902	0.000097	0.001811	BDL
Matazu	0.000476	0.002290	0.001573	0.000082	0.001083	0.000554
Zango	0.000655	0.002260	0.001346	0.000080	0.004405	0.000299

The non-cancer risks (THQ) of the investigated heavy metals through the consumption of samples for both adults and children inhabitants of the study area were determined and presented in Tables 4 and 5. The THQ has been recognized as a useful parameter for evaluating the risk associated with the consumption of metal-contaminated foods (43). THQ is interpreted as either greater than 1 or less than 1 where $THQ > 1$ shows human health risk concern (44). Bhalkhair and Ashraf (9) in their study have put forward the suggestion that the ingested dose of heavy metals is not equal to the absorbed pollutant dose in reality because a fraction of the ingested heavy metals may be excreted, with the remainder being accumulated in body tissues where they can affect human health. The risk level of Target Hazard Quotient ($THQ < 1$) was observed for all the evaluated heavy metals for both adults and children. It indicates that the intake of heavy metals through consumption of the samples does not pose a considerable non-cancer risk. The THQ for the samples was in the decreasing order $Mn > Zn > Pb > Fe > Cr > Cd$, in all the samples respectively. In both adults and children the highest THQ for the heavy metal Mn from the sample from Dabai, while the lowest was for the heavy metal Cd from the sample from Funtua. The sequence of risk was the same for both adults and children although the children had higher THQ values in all cases. Similar observations have been reported previously by Mahfuza et al. [45], Micheal et al. [11] and Liu et al. [18].

Furthermore, the non-cancer risk for each sample was expressed as the cumulative HI, which is the sum of individual metal THQ. All the studied samples showed the risk level ($HI < 1$) with the highest in the sample from Dabai and lowest in the sample from Matazu. It suggests that the inhabitants of Katsina state might not be exposed to non-carcinogenic health risk through the intake of heavy metals.

Table 4 Heavy Metal Target Hazard Quotient and Health Risk Index in Adults from Consuming Cultivated Amaranthus from Katsina State

Location	Target Hazard Quotient						Health Risk Index (HRIs)
	Mn	Zn	Pb	Cd	Fe	Cr	
Birchi	0.021918	0.003536	0.001075	0.000064	0.000892	0.000289	0.027774
Dabai	0.028104	0.003464	0.001029	0.000069	0.001413	0.000632	0.034711
Daura	0.018560	0.003128	0.001181	0.000076	0.000986	0.000391	0.024322
Dutsinma	0.024048	0.003288	0.001146	0.000064	0.001351	0.000393	0.030290
Funtua	0.022451	0.003683	0.000640	0.000062	0.002040	0.000119	0.028995
Ingawa	0.016958	0.000311	0.001009	0.000063	0.000999	0.000622	0.026942
Kafur	0.025650	0.003143	0.001185	0.000069	0.000951	0.000423	0.028401
Katsina	0.014239	0.003439	0.001111	0.000087	0.001613	0.000871	0.033310
Malunfashi	0.019544	0.003133	0.001358	0.000078	0.000181	BDL	0.026564
Matazu	0.013596	0.003054	0.001049	0.000069	0.004046	0.007217	0.018181
Zango	0.018718	0.003014	0.000898	0.000064	0.002517	0.000398	0.026649

Table 5 Heavy Metal Target Hazard Quotient and Health Risk Index in Children from Consuming Cultivated Amaranthus from Katsina State

Location	Target Hazard Quotient						Health Risk Index (HRIs)
	Mn	Zn	Pb	Cd	Fe	Cr	
Birchi	0.054794	0.008841	0.002688	0.000161	0.002229	0.000722	0.069434
Dabai	0.110254	0.008660	0.002573	0.000172	0.003666	0.001580	0.126906
Daura	0.046128	0.007821	0.002918	0.000190	0.002464	0.000853	0.060373
Dutsinma	0.110120	0.008219	0.002865	0.000161	0.003373	0.000983	0.125726
Funtua	0.056127	0.009208	0.002666	0.000152	0.005200	0.001020	0.074135
Ingawa	0.042395	0.007821	0.002523	0.000157	0.002498	0.001605	0.056999
Kafur	0.064126	0.008480	0.002650	0.000172	0.002378	0.001076	0.079083
Katsina	0.035382	0.008598	0.002778	0.000217	0.004032	0.002187	0.048377
Malunfashi	0.048862	0.007833	0.003170	0.000194	0.002586	BDL	0.050240
Matazu	0.033996	0.007634	0.002622	0.000164	0.001548		0.035193
Zango	0.046795	0.007534	0.002243	0.000161	0.006293	0.001000	0.064025

Cd and Pb are classified by the IARC as being carcinogenic agents (46). Chronic exposure to low doses of Cd, and Pb could, therefore, result in many types of cancers (1). The computed ILCR and cumulative incremental lifetime cancer risk (Σ ILCR) for Cd, and Pb are presented in Tables 6 and 7. US-EPA recommended the safe limit for cancer risk is below about 1 chance in 1,000,000 lifetime exposure ($ILCR < 10^{-6}$) and threshold risk limit ($ILCR > 10^{-4}$) for chance of cancer is above 1 in 10,000 exposure where remedial measures are considerable and moderate risk level ($ILCR > 10^{-3}$) is above 1 in 1,000 where public health safety consideration is more important (19; 47). ILCR for Cd violated the threshold risk limit ($>10^{-4}$) and ILCR for Pb reached the moderate risk limit ($>10^{-3}$) in all the studied samples in adults, While in children, ILCR for Cd in samples from Dabai, Ingawa, Matazu and Zango and Cd for all samples have reached the moderate risk limit ($>10^{-3}$), while the ILCR for Pb in samples from Birchi, Daura, Dutsinma, Kafur, Katsina Funtua and Malunfashi are beyond the moderate risk level ($>10^{-2}$). The sampling trend of risk for developing cancer as a result of consuming the studied samples showed: Malunfashi > Kafur > Daura > Dutsinma > Katsina > Birchi > Funtua > Matazu > Dabai > Ingawa > Zango for both adult and children (Tables 6 and 7).

Moreover, cumulative cancer risk (Σ ILCR) of all the studied samples have reached a moderate risk limit ($>10^{-3}$) in adult. While in children with the exception of the sample from Zango which is within the moderate cancer risk ($>10^{-3}$), all other samples were beyond the moderate cancer risk ($>10^{-2}$). Further, among all the studied samples, Amaranthus sample from Malunfashi has the highest chances of cancer risks ($ILCR 5.197458 \times 10^{-3}$ in adults; $ILCR 1.343792 \times 10^{-2}$ in children) and the sample from Zango has the lowest chances of cancer risk ($ILCR 3.445776 \times 10^{-3}$ in adults; $ILCR 9.681865 \times 10^{-3}$ in children). These risk values indicate that consumption of the sample from Malunfashi would result in an excess of 52 cancer cases in adults per 10,000 people exposure and 13 cancer cases per 1,000 people exposure while consumption of the sample from Zango would result in an excess of 35 cancer cases in adults and 97 cancer cases in children per 10,000 people exposure (US-EPA). Prompt action is needed to control the excess use of heavy metal-based fertilizer and pesticides and also emission of heavy metals exhaust from automobiles should be checked to save the population from cancer risk

Table 6 Incremental Life Time Cancer Risk in Adults from Consuming Cultivated Amaranthus in Katsina State

Location	ILCR		Σ ILCR
	Pb	Cd	
Birchi	4.063802E-03	5.457500E-04	4.118337E-03
Dabai	3.889771E-03	5.838100E-04	3.948153E-03
Daura	4.482405E-03	6.472700E-04	4.547132E-03
Dutsinma	4.331000E-03	5.457500E-04	4.386473E-03
Funtua	4.030872E-03	6.472700E-04	4.095599E-03
Ingawa	3.814517E-03	5.330500E-04	3.867822E-03
Kafur	4.517774E-03	5.838100E-04	4.576155E-03
Katsina	4.200197E-03	7.361100E-04	4.273808E-03
Malunfashi	5.131488E-03	6.599700E-04	5.197458E-03

Matazu	3.965024E-03	5.584300E-04	4.020867E-03
Zango	3.391201E-03	5.457500E-04	3.445776E-03

Table 7 Incremental Life Time Cancer Risk in Children from Consuming Cultivated Amaranthus from Katsina State

Location	ILCR	Σ ILCR	
	Pb	Cd	
Birchi	1.015951E-02	1.203855E-03	1.136336E-02
Dabai	9.724434E-03	1.287855E-03	1.101229E-02
Daura	1.102964E-02	1.427835E-03	1.245749E-02
Dutsinma	1.082975E-02	1.203855E-03	1.203361E-02
Funtua	1.007719E-02	1.287855E-03	1.136505E-02
Ingawa	9.536290E-03	1.175865E-03	1.071216E-02
Kafur	1.001840E-02	1.287855E-03	1.121277E-02
Katsina	1.050051E-02	1.623750E-03	1.066288E-02
Malunfashi	1.198210E-02	1.455825E-03	1.343792E-02
Matazu	9.912571E-03	1.231860E-03	1.114443E-02
Zango	8.478010E-03	1.203855E-03	9.681865E-03

4. CONCLUSION

This study determines the heavy metals concentration in Amaranthus leaf samples from Katsina state Nigeria. Results from this study has shown that with the exception of Pb the concentration of Mn, Zn, Cd, Cr and Fe in the samples were generally lower than the USEPA, WHO/FAO maximum permissive limits. The results have indicated that the estimated daily intake of the heavy metals was lower than the tolerable daily intake limit set by the USEPA. The risk level of Target Hazard Quotient (THQ < 1) was observed for all the evaluated heavy metals for both adults and children. ILCR for Cd violated the threshold risk limit ($>10^{-4}$) and ILCR for Pb reached the moderate risk limit ($>10^{-3}$) in all the studied samples in adults. While in children, ILCR for Cd in samples from Dabai, Ingawa, Matazu and Zango and Cd for all samples have reached the moderate risk limit ($>10^{-3}$), while the ILCR for Pb in samples from Birchi, Daura, Dutsinma, Kafur, Katsina Funtua and Malunfashi are beyond the moderate risk level ($>10^{-2}$). The sampling area trend of risk for developing cancer in the studied samples showed: Malunfashi > Kafur > Daura > Dutsinma > Katsina > Birchi > Funtua > Matazu > Dabai > Ingawa > Zango for both adult and children, cumulative cancer risk (Σ ILCR) of all the studied samples have reached the moderate risk limit ($>10^{-3}$) in adult. While in children with the exception of the sample from Zango which is within the moderate cancer risk ($>10^{-3}$), all other samples were beyond the moderate cancer risk ($>10^{-2}$). Further, among all the studied samples, Amaranthus sample from Malunfashi have the highest chances of cancer risks (ILCR 5.197458×10^{-3} in adults; ILCR 1.343792×10^{-2} in children) and sample from Zango have the lowest chances of cancer risk (ILCR 3.445776×10^{-3} in adults; ILCR 9.681865×10^{-3} in children). The study suggests that consumption of the studied Amaranthus leaf samples in Katsina state is of public health concern as they may contribute to the population cancer burden.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

- Jarup L, Hazards of heavy metal contamination British Medical Bulletin, 2003; 68, 167–182
- Yeasmin NJ, Ashrafu I, Shawkat A. Transfer of metals from soil to vegetables and possible health risk assessment. Springer plus, 2013; 2(1): 385
- Siegel KR, Ali MK, Srinivasiah A, Nugent RA, Narayan KMV. Do we produce enough fruits and vegetables to meet global health need? PloS one, 2014; 9, e104059
- WHO Cadmium environmental health criteria World Health Organization, 1992 pp: 134

- 289 5. Katsina State investor's handbook, Yaliam Press Ltd 2016: 12-15
- 290 6. A.O.AC Official Methods of Analysis 18th Edition Association of Official Analytical Chemists, 1995 U.S.A
- 291 7. Jan FA, Ishaq M, Khan S, Ihsanullah I, Ahmad I, Shakirullah M, A comparative study of human health risks via
292 consumption of food crops grown on wastewater irrigated soil (Peshawar) and relatively clean water irrigated soil (lower
293 Dir). J. Hazard. Mater, 2010; 179: 612–6219. Balkhaira KS, Ashraf MA, Field accumulation risks of heavy metals in soil
294 and vegetable crop irrigated with sewage water in western region of Saudi Arabia. Saudi Journal of Biological Sciences
295 2015; 23 (1): S32-S44
- 296 8. Balkhaira KS, Ashraf MA, Field accumulation risks of heavy metals in soil and vegetable crop irrigated with sewage
297 water in western region of Saudi Arabia. Saudi Journal of Biological Sciences 2015; 23 (1): S32-S44
- 298 9. Orisakwe OE, Mbagwu HOC, Ajaezi GC, Edet UW, Patrick U, Uwana PU. Heavy metals in sea food and farm
299 produce from Uyo, Nigeria Levels and health implications. Sultan Qaboos Univ Med J. 2015; 15(2): e275–e282.
- 300 10. Ekhaton OC, Udowelle NA, Igbiri S, Asomugha RN, Igweze ZN, Orisakwe OE. Safety Evaluation of Potential Toxic
301 Metals Exposure from Street Foods Consumed in Mid-West Nigeria. Journal of Environmental and Public Health Volume
302 2017, Article ID 8458057
- 303 11. Micheal B, Patrick O, Vivian T. Cancer and non-cancer risks associated with heavy metal exposures from street foods:
304 Evaluation of roasted meats in an urban setting. Journal of Environment Pollution and Human Health, 2015; 3, 24–30
- 305 12. Li S, Zhang Q. Risk assessment and seasonal variations of dissolved trace elements and heavy metals in the Upper
306 Han River, China. Journal of Hazardous Materials, 2010; 181, 1051–1058
- 307 13. Li PH, Kong S, Geng CM, Han B, Sun RF, Zhao RJ, Bai ZP, Assessing the hazardous risks of vehicle inspection
308 workers' exposure to particulate heavy metals in their work places. Aerosol and Air Quality Research, 2013; 13, 255–265
- 309 14. United States Environmental Protection Agency. EPA Human Health Related Guidance, OSWER, 2002; 9355 (pp. 4–
310 24). Washington, DC: United States Environmental Protection Agency
- 311 15. NFPCSP Nutrition Fact Sheet; Joint report of Food Planning and Nutrition Unit (FMPU) of the ministry of Food of
312 Government of Bangladesh and Food and Agricultural Organization of the United Nation (FAO) September 14, 2011; 1–2,
313 National Food Policy Plan of Action and Country Investment Plan, Government of the People's Republic of Bangladesh
- 314 16. The Risk Information System, 2007; Retrieved from http://rais.oml.govt/tox/rap_toxp.shtml.
- 315 17. Guerra F, Trevizam AR, Muraoka T, Marcante NC, Canniatti-Brazaca SG, Heavy metals in vegetables and potential
316 risk for human health. Scientia Agricola, 2012; 69, 54–60.10.1590/S0103-90162012000100008
- 317 18. Liu X, Song Q, Tang Y, Li, W, Xu J, Wu J, Wang F, Brookes, PC. Human health risk assessment of heavy metals in
318 soil–vegetable system: A multi-medium analysis. Science of the Total Environment, 2013; 463–464, 530–540
- 319 19. Pepper IL, Gerba CP, Brusseau ML. Environmental and pollution Science: Pollution Science Series, 2012; pp. 212–
320 232. Academic Press
- 321 20. D'Souza C, Peretiatko R. The nexus between industrialization and environment: A case study of Indian enterprises.
322 Environ. Manage. Health **2002**, 13, 80–97.
- 323 21. Yaradua AI, Alhassan AJ, Shagumba AA, Nasir A, Idi A, Muhammad and Kanadi A.M. Evaluation of heavy metals in
324 beans and some beans product from some selected markets in Katsina state Nigeria. Bayero Journal of Pure and Applied
325 sciences, 2017; <http://dx.doi.org/10.4314/bajopas.v10i1.1S>
- 326 22. Landrigan PJ, Fuller R, Acosta NJR, Adeyi O, Arnold R, Basu N, Zhong M, The Lancet Commission on pollution and
327 health. The Lancet, 2017; [https://doi.org/10.1016/S0140-6736\(17\)32345-0](https://doi.org/10.1016/S0140-6736(17)32345-0)
- 328 23. Mohammed SA, Folorunsho JO, Heavy metals concentration in soil and Amaranthus retroflexus grown on irrigated
329 farmlands in Makera Area, Kaduna, Nigeria. Journal of Geography and Regional Planning, 2015; Vol. 8(8), pp. 210 - 217

330 24. Di Bella G, Clara N, Giuseppe D B, Luca R, Vincenzo L T, Angela G P` and Giacomo D. Mineral composition of some
331 varieties of beans from Mediterranean and Tropical areas. *International Journal of Food Sciences and Nutrition* vol., 2016;
332 67, no. 3, 239–248

333 25. Mihaileanu RG, Neamtiu IA, Fleming M, Pop C, Bloom MS, Roba C, Surcel M, Stamatian F, Gurzau E. Assessment of
334 heavy metals (total chromium, lead, and manganese) contamination of residential soil and homegrown vegetables near a
335 former chemical manufacturing facility in Tarnaveni, Romania. *Environmental Monitoring Assessment*, 2019; 191:8
336 <https://doi.org/10.1007/s10661-018-7142-0>

337 26. Dahiru MF, Umar AB, Sani MD. Cadmium, Copper, Lead and Zinc Levels In Sorghum And Millet Grown In The City Of
338 Kano And Its Environs. *Global Advanced Research Journal of Environmental Science and Toxicology* (ISSN: 2315-5140).
339 2013; Vol 2(3), 082-085

340 27. Babatunde OA, Uche E, A comparative evaluation of the heavy metals content of some cereals sold in Kaduna, North
341 west Nigeria. *International Journal of Scientific & Engineering Research*, 2015; Volume 6, Issue 10, 485 ISSN 2229-5518

342 28. Ahmed KS, and Mohammed AR. Heavy Metals (Cd, Pb) and Trace Elements (Cu, Zn) Contents Of Some Food Stuffs
343 from Egyptian Markets. *Emir J. Agric.sci*, 2005; 17(1):34-42.

344 29. Okoye COB, Odo IS, Odika IM. Heavy metals content of grains commonly sold in markets in south-east Nigeria. *Plant*
345 *Products Research Journal*, 2009; vol. 13, SSN 1119-2283

346 30. Sab-Udeh SS, Okerulu IO, Determination of Heavy Metal Levels of some Cereals and Vegetables sold in Eke-Awka
347 Market Awka, Anambra State, Nigeria. *Journal of Natural Sciences and Research*, 2017; Vol 7, No 4

348 31. Gottipolu RR, Flora SJ, Riyaz B. Environmental Pollution-Ecology and human health: P. Narosa publishing house,
349 2012; New Delhi India. 110 002, 166-223

350 32. Pan X.-D, Wu P-G, Jiang X-G. Levels and potential health risk of heavy metals in marketed vegetables in Zhejiang,
351 China. *Sci. Rep.*, 2016; 6, 20317; doi: 10.1038/srep20317

352 33. Edem CA, Grace I, Vincent O, Rebecca E, Matilda O. A Comparative Evaluation Of Heavy Metals In Commercial
353 Wheat Flours Sold In Calabar –Nigeria. *Pakistan Journal of Nutrition*, 2009; 8, 585-587

354 34. Olusakin PO, Olaoluwa DJ, Evaluation of Effects of Heavy Metal Contents of Some Common Spices Available in Odo-
355 Ori Market, Iwo, Nigeria. *J Environ Anal Chem.*, 2016; 3:174. doi:10.41722380-2391.1000174

356 35. Orisakwe EO, John KN, Cecilia NA, Daniel OD, Onyinyechi B. Heavy Metals Health Risk assessment for Population
357 Consumption of Food Crops and Fruits in Owerre-Southern Nigeria. *Chem. Cent.*, 2012; 6, 77

358 36. Gomaa NA, Mohamed BMA, Essam MS, Ahmed SMF, Estimated heavy metal residues in Egyptian vegetables in
359 comparison with previous studies and recommended tolerable limits. *J. Biol. Sci.*, 2018; 18:135-143

360 37. Zahir E, Naqvi II, Mohi Uddin SH. Market basket survey of selected metals in fruits from Karachi city (Pakistan),
361 *Journal of Basic and Applied Sciences* 2009; 5(2):47-52

362 38. Yahaya MY, Umar RA, Wasagu RSU, Gwandu HA, Evaluation of some heavy metals in food crops of Lead polluted
363 sites of Zamfara State Nigeria, *International Journal of Food Nutrition and Safety*, 2015; 6(2): 67-73

364 39. Sulyman YI, Abdulrazak S, Oniwapele YA, Ahmad, A Concentration of heavy metals in some selected cereals
365 sourced within Kaduna state, Nigeria. *IOSR Journal of Environmental Science, Toxicology and Food Technology (IOSR-
366 JESTFT)* e-ISSN: 2319-2402,p- ISSN: 2319-2399. 2015; Volume 9, Issue 10 Ver. II, PP 17-19

367 40. Das PK, Halder M, Mujib ASM, Islam F, Mahmud ASM, Akhter S, Joardar JC. Heavy Metal Concentration in Some
368 Common Spices Available at Local Market as Well as Branded Spicy in Chittagong Metropolitan City, Bangladesh. *Curr*
369 *World Environ.* 2015;10(1). doi : <https://dx.doi.org/10.12944/CWE.10.1.12>

41. Singh A, Sharma RK, Agrawal M, Marshall FM. Health risk assessment of heavy metals via dietary intake of foodstuffs from the wastewater irrigated site of a dry tropical area of India, Food Chem. Toxicol., 2010; 48, 611–619, doi:10.1016/j.fct.2009.11.041
42. SEPA limits of pollutants in food. State environmental protection administration, 2005 China GB2762
43. Agbenin JO, Danko M, Welp G. Soil and vegetable compositional relationships of eight potentially toxic metals in urban garden fields from northern Nigeria J. Sci. Food Agric., 2009; 89 (1), pp. 49–54
44. Bassey SC, Ofem OE, Essien NM, Eteng MU. Comparative Microbial Evaluation of Two Edible Seafood *P. palludosa* (Apple Snail) and *E. radiata* (Clam) to Ascertain their Consumption Safety. J Nutr Food Sci 2014; 4: 328. doi: 10.4172/2155-9600.1000328
45. Mahfuza SS, Rana S, Yamazaki S, Aono T, Yoshida S. Health risk assessment for carcinogenic and noncarcinogenic heavy metal exposures from vegetables and fruits of Bangladesh. Cogent Environmental Science, 2017; 3: 1291107 <http://dx.doi.org/10.1080/23311843.2017.1291107>
46. Tani, F. H., & Barrington, S. (2005). Zinc and copper uptake by plants under two transpiration rates. Part I. Wheat (*Triticum aestivum* L.). *Environmental Pollution*, 138(3), 538-547.
47. Tchounwou PB, Yedjou CG, Patlolla AK, Sutton DJ, Heavy metal toxicity and the environment. Molecular Clinical and Environmental Toxicology, 2014; 101, 133–164