

Original Research Article

CONCENTRATIONS AND RISK EVALUATION OF SELECTED HEAVY METALS IN AMARANTHUS 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 samples were evaluated based on the Target Hazard Quotient. The possibility of cancer risks in the samples through intake of carcinogenic heavy metals was estimated using the Incremental Lifetime Cancer Risk. The results of the study have indicated that the studied samples pose non carcinogenic risk for all the studied metals [target hazard quotient (THQ) > 1]. 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 the studied samples in Katsina state is of public health concern as they may contribute to the population cancer burden

Comment [P1]: Use Amaranthus leaf

Comment [P2]: After risk a full-stop required

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Suggestion:
The target hazard quotient was (THQ) > 1, indicating that the Amaranthus leaf cultivated may pose non carcinogenic risk for all the studied metals .

Comment [P4]: Use Amaranthus leaf & reconstruct the sentence.

Suggestion:
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

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 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 fiber (3). 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 causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases (1; 4).

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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 consumption of some locally cultivated leafy vegetables in Katsina State, Nigeria

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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 that 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 were 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. They were then stored in the refrigerator at 40C until ready for use

2.2 IDENTIFICATION OF SAMPLE

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

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 hours in a Gallenkamp hotbox oven (CHF097XX2.5) and then blended in an electric blender. The resulting powder was kept in air tight polythene packet at room temperature before digestion and metals analyses.

2.4 HEAVY METALS DETERMINATION

5 g of each Sample was dried at 800C for 2 hours in a Gallenkamp hotbox oven (CHF097XX2.5) and then blended in an electric blender. 0.5 g of each sample was weighed and ashed at 5500C 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 (HN03) 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).

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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:

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

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 of the 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).

$$THQ = CDI / R_f D$$

CDI is the chronic daily heavy metal intake (mg/kg/day) obtained from the previous section and R_fD 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:

$$HI = THQ_1 + THQ_2 + \dots + THQ_n$$

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 $HI < 1$ and in a level of concern when $1 < HI < 5$ (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).

$$ILCR = CDI \times CSF$$

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 is considered within the range of 10^{-6} to 10^{-4} (12). The CDI value

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was calculated on the basis of the following equation and CSF values for carcinogenic heavy metals were used according to the literature (18).

$$CDI = (EDI \times EFr \times ED_{tot})/AT$$

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, average lifetime for Nigerians; AT is the period of exposure for non-carcinogenic effects ($EFr \times ED_{tot}$), and 60 years life time 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).

$$\sum ILCR_n = ILCR_1 + ILCR_2 + \dots + ILCR_n$$

Where, $n = 1, 2, \dots, n$ 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 analysed in the sampled seeds is similar to that reported for heavy metals in beans and some beans products from some selected markets in Katsina state, Nigeria (21).

Lead was detected in all the samples, with 100% 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

179 EU and 0.05 mg/kg limit set by USEPA (22). The high percentage of
180 samples which were in violation of the maximum permissible limits of Pb set
181 by WHO EU and US EPA is a cause for public health concern considering
182 the frequency of exposure. The Pb concentration range for the leaf samples
183 in this study is lower than that reported for leafy vegetables from Kaduna
184 state Nigeria (23) and that reported for beans samples from Italy, Mexico,
185 India, Japan, Ghana and Ivory Coast with a Pb concentration range of
186 4.084- 14.475ppm (24) and the result of Pb in homegrown vegetables near
187 a former chemical manufacturing facility in Tarnaveni, Romania (25). But the
188 results are higher than that reported for the concentration of Pb from Kano
189 (millet and sorghum) and Kaduna (cereals) states, Nigeria (26; 27). The
190 value was still higher than the range (0.116 to 0.390) reported by Ahmed
191 and Mohammed in food stuff from Egypt in 2005 (28), the range (0.007 to
192 0.032 mg/kg) reported by Okoye et al., (29) in a study conducted in South
193 east of Nigeria in 2009 in wheat flours, the results for Pb in leafy vegetables
194 and the result for Pb in cucumber from Awka, Anambra state Nigeria (30).
195 This difference has earlier been attributed to differences in anthropogenic
196 activities that introduce metals into the soil in the areas where these
197 samples were grown or even deposition of Pb on the surface of these
198 vegetables during production, transport and Marketing or by emissions from
199 Vehicles and industries (31).

200 The concentration of Cd (mg/kg) range from 0.043 to 0.058 in Amaranthus
201 leaf samples is similar to that reported for Romaine lettuce and cabbage in a
202 study conducted in Zhejiang China (32) and the Cd values for cabbage and
203 cucumber from Awka, Anambra state Nigeria (30). But these values are
204 higher than the range (0.002 to 0.004 mg/kg) reported by Edem et al., (33)
205 in Wheat flours in 2009. The reported Cd values in the present study are
206 lower than that reported for various beans samples from Europe, Asia and
207 parts of West Africa (24), that reported in a study for the Cadmium
208 concentration range for both unprocessed and processed bean samples
209 from Katsina state Nigeria (21) and for locust beans from Odo-Ori market
210 Iwo, Nigeria (34). The values are also lower than those obtained by Okoye
211 et al., (29) in Cereals in South eastern Nigeria (0.007 to 0.23mg/kg) in 2009,
212 Ahmed and Mohammed (28) in Cereal products (0.091-0.143mg/kg) in
213 2005, Orisakwe et al., (35) in Owerri (0.00 to 0.24mg/kg) in 2012 and Dahiru
214 et al., (26) in Kano (0.11 to 0.28mg/kg) in 2013. These differences could be

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Suggestion:

The percentage of Pb in this study is higher than maximum permissible limits set by WHO, EU & USEPA and have caused public health concerns considering the frequency of exposure.

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 to 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 evaluate heavy metals in millet from Kaduna, Nigeria (27). The result is similar to that reported for market sold beans from Katsina, Nigeria (21), but is 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 obtain 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).

The result for the heavy metal Mn concentrations in the present study is lower than the result of Mn levels in tumeric (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, Bangladesh to evaluate heavy metals in spices and results of evaluation of heavy metals in various foods reported in other studies (24; 29), but is similar to that reported by

249 Yaradua et al,. (21) in a study on Mn levels in beans from Katsina states
 250 Nigeria.

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

Comment [P11]: To two decimal places

Loca tion	Heavy Metal						
	Mn	Zn	Pb	Cd	Ni	Fe	Cr
Birc hi	0.411000	1.421000	0.864000	0.043000	B	0.836000	0.116000
	±0.00040	±0.00030	±0.00020	±0.00020	D	±0.01200	±0.00030
	0	0	0	0	L	0	0
Daba i	0.827000	1.392000	0.827000	0.046000	B	1.325000	0.254000
	±0.00020	±0.00020	±0.00030	±0.00020	D	±0.00030	±0.00020
	0	0	0	0	L	0	0
Daur a	0.348000	1.257000	0.953000	0.051000	B	0.924000	0.137000
	±0.00020	±0.00020	±0.00040	±0.00130	D	±0.00020	±0.00020
	0	0	0	0	L	0	0
Dutsi nma	0.826000	1.321000	0.921000	0.043000	B	1.267000	0.158000
	±0.00160	±0.00030	±0.00130	±0.00060	D	±0.00020	±0.00030
	0	0	0	0	L	0	0
Funt ua	0.421000	1.480000	0.857000	0.051000	B	1.913000	0.164000
	±0.00030	±0.00020	±0.00030	±0.00020	D	±0.00130	±0.00020
	0	0	0	0	L	0	0
Inga wa	0.318000	1.251000	0.811000	0.042000	B	0.937000	0.258000
	±0.00100	±0.00040	±0.00010	±0.00060	D	±0.00030	±0.00080
	0	0	0	0	L	0	0
Kafu r	0.481000	1.263000	0.952000	0.046000	B	0.892000	0.173000
	±0.00040	±0.00020	±0.00020	±0.00030	D	±0.00020	±0.00020
	0	0	0	0	L	0	0
Katsi na	0.267000	1.382000	0.893000	0.058000	B	1.512000	0.351000
	±0.00020	±0.00020	±0.00030	±0.00020	D	±0.00060	±0.00020
	0	0	0	0	L	0	0
Malu nfash hi	3.665000	1.259000	1.019000	0.052000	B	0.970000	BDL
	±0.00030	±0.00100	±0.00030	±0.00020	D	±0.00180	
	0	0	0	0	L	0	
Mata zu	0.255000	1.227000	0.843000	0.044000	B	3.795000	0.297000
	±0.00080	±0.00040	±0.00030	±0.00020	D	0.000400	±0.00180

Zang	0.351000	1.211000	0.721000	0.043000	L	2.360000	0.160000
o	±0.00030	±0.00030	±0.00020	±0.00030	B	±0.00030	±0.00130
	0	0	0	0	D	0	0
					L		

Values are expressed as Mean ± Standard

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

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

Comment [P12]: To two decimal places

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

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

Comment [P13]: To two decimal places

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	0.00065	0.00226	0.00134	0.00008	0.00440	0.00029

The non-cancer risks (THQ) of the investigated heavy metals through the consumption of the 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 (>1) or less than 1 (<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. Risk level of Target Hazard Quotient ($THQ < 1$) was observed for all the evaluated heavy metals for both adults and children. It indicates that intake of these heavy metals through consumption of the samples does not poses 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 was 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 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.

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

Comment [P14]: To two decimal places

Location	Target Hazard Quotient						Health Risk Index (HRIs)
	Heavy Metal						
	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

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

Comment [P15]: To two decimal places

	Target Hazard	Health Risk
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Location	Quotient Heavy Metal						Index (HRIs)
	Mn	Zn	Pb	Cd	Fe	Cr	
Birchi	0.0547 94	0.0088 41	0.0026 88	0.0001 61	0.0022 29	0.0007 22	0.0694 34
Dabai	0.1102 54	0.0086 60	0.0025 73	0.0001 72	0.0036 66	0.0015 80	0.1269 06
Daura	0.0461 28	0.0078 21	0.0029 18	0.0001 90	0.0024 64	0.0008 53	0.0603 73
Dutsinma	0.1101 20	0.0082 19	0.0028 65	0.0001 61	0.0033 73	0.0009 83	0.1257 26
Funtua	0.0561 27	0.0092 08	0.0026 66	0.0001 52	0.0052 00	0.0010 20	0.0741 35
Ingawa	0.0423 95	0.0078 21	0.0025 23	0.0001 57	0.0024 98	0.0016 05	0.0569 99
Kafur	0.0641 26	0.0084 80	0.0026 50	0.0001 72	0.0023 78	0.0010 76	0.0790 83
Katsina	0.0353 82	0.0085 98	0.0027 78	0.0002 17	0.0040 32	0.0021 87	0.0483 77
Malunfashi	0.0488 62	0.0078 33	0.0031 70	0.0001 94	0.0025 86	BDL	0.0502 40
Matazu	0.0339 96	0.0076 34	0.0026 22	0.0001 64	0.0015 48		0.0351 93
Zango	0.0467 95	0.0075 34	0.0022 43	0.0001 61	0.0062 93	0.0010 00	0.0640 25

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 into many types of cancers (1). The computed ILCR and cumulative incremental lifetime cancer risk (Σ ILCR) for Cd, and Pb through the samples 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 area 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 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 has the highest chances of cancer risks (ILCR 5.197458×10^{-3} in adults; ILCR 1.343792×10^{-2} in children) and the sample from Zango has the lowest chances of cancer risk (ILCR 3.445776×10^{-3} in adults; ILCR 9.681865×10^{-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 should be needed to control the excess use of heavy metal-based fertilizer and pesticides and also emission of heavy metal 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 from 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

Comment [P16]: To two decimal places

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

Comment [P17]: To two decimal places

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 the heavy metal Pb the concentration values 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 were lower than the tolerable daily intake limit set by the USEPA in both samples. 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 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, cumulative cancer risk (\sum 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 has the highest chances of cancer risks (ILCR 5.197458×10^{-3} in adults; ILCR 1.343792×10^{-2} in children)

and the sample from Zango has 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

Comment [P18]: Full-stop required

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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