Health Risk Assessment for Carcinogenic and Non Carcinogenic Heavy Metal Exposure from Hibiscus Leaf Cultivated in Katsina State, North West Nigeria

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Authors' contributions

Author AIY designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors AJA, AN, KIM, AU and AI performed the statistical analysis and manage the analysis of the study. Authors IUM, SAY, SY and JAM managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

This study was conducted to determine the heavy metals concentration in Hibiscus leaves cultivated in Katsina State Nigeria. The objectives were mainly to detect the presence of heavy metals in the cultivated Hibiscus leaves in the study area, compare the concentration of heavy metals in samples in relation to the permissible limits specified by WHO/FAO/USEPA Standards. Samples of cultivated Hibiscus leaves were collected in the year 2017 from the selected area.

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Analysis for the concentration of these heavy metals; Cr, Cd, Fe, Ni, Mn, Pb and Zn was conducted by the use of AAS (by Atomic Absorption Spectrophotometry) method. The health risks to the local inhabitants from the consumption of the samples were evaluated based on the Target Hazard Quotient (THQ). The possibility of cancer risks for the adult and children population through intake of carcinogenic heavy metals in the samples was estimated using the Incremental Lifetime Cancer Risk (ILCR). Results from this study shows that with the exception of Pb with mean concentration of (0.508-0.978), the mean concentration (mg/kg) range values of Cr (0.135-0.261), Cd (0.042-0.051), Fe (0.684-0.978) and Zn (1.048-1.208) in the samples were generally lower than the USEPA, WHO/FAO maximum permissive limits. The results have indicated that the estimated daily intake (EDI) of the heavy metals were lower than the tolerable daily intake limit set by the USEPA in both samples and the health risk index (HRI) for all the heavy metals were <1. The target hazard quotient (THQ) for the samples for both the adult and children population was in the decreasing order of Zn>Pb>Fe>Cr>Cd. ILCR for Cd is below the threshold risk limit (>10^{-5}) 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 and Cd in samples from Dabai, Daura, Funtua, Matazu and Zango 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 were 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: Funtua > Daura > Dutsinma > Katsina > Malunfashi > Matazu > Zango > Ingawa > Kafur > Dabai > Birchi for both adult and children, cumulative cancer risk (∑ILCR) of all the studied Hibiscus samples have reached the moderate risk limit (>10^{-3}) in adult. While in children with the exception of the sample from Birchi, Dabai and Kafur which are within the moderate cancer risk (>10^{-3}) limit, all other samples were beyond the moderate cancer risk (>10^{-2}) limit. 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.

Keywords: Hibiscus; heavy metals; Katsina; health risk index and cancer risk.

1. INTRODUCTION

Heavy metals are environmental contaminants capable of causing human health problems. If excess amount is ingested through food they are non-biodegradable and persistent, have long biological half-lives and can be bio-accumulated through biological chains [1]. Heavy metal toxicity may occur due to contamination of irrigation water, the application of fertilizer and metal based pesticides, industrial emission, harvesting process, transportation, storage or sale. Crops and vegetables grown in soils contaminated with heavy metals have greater accumulation than those grown in uncontaminated soils [2]. The toxicity of heavy metals most commonly involves the brain and kidney but other manifestations can occur in some other parts of the body for example arsenic is clearly capable of causing cancer, hypertension can result in individuals exposed to lead and renal toxicity in individual exposed to cadmium [3].

Vegetables play important roles in human nutrition and health, particularly as sources of vitamin C, thiamine, niacin, pyridoxine, folic acid, minerals, and dietary fiber [4]. Sorrel (Hibiscus sabdariffa, Linn.), is an annual crop native to tropical Africa [5] where it is known by different synonyms and vernacular names [6,7]. In Africa, the crop is used for both food and non-food applications [8,9]. For food applications, the tender stem, leaves and calyxes are used as vegetable in the preparation of soups and sauces, the calyxes are specially prepared into a textural form suitable for use as a meat substitute. ``Zobo'', a refreshing drink near akin to black currant is made from the calyx. Also, the kernels are roasted and consumed as such, though slightly bitter [10] and in Northern Nigeria, the seeds are cooked and fermented into a soup condiment known as ``Daddawar botso''. "Kwadon Yakuwa" a salad prepared from boiled Hibiscus leaves mixed with groundnut cake and spices is a delicacy of the inhabitants of Katsina state. For non-food applications, the flower and fleshy fruit are used in pharmaceutical infusions to relieve symptoms of bronchitis and coughs [10].

In Katsina State Nigeria, there is limited information on the levels of heavy metals in locally cultivated leafy vegetables. This work there-fore 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.

2. MATERIALS 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 24,192 km^2 (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 rainy season. The average annual rainfall, temperature, and relative humidity of Katsina State are 1,312 mm, 27.3°C and 50.2%, respectively [11]. The study was conducted within some catchment areas that cultivate hibiscus 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 4°C until ready for use.

2.2 Identification of Sample

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

2.3 Sample Preparation

Leafy vegetable 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

0.5 g of each sample was weighed and ashed at 550°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 (HN0$_3$) 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 [12] 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:

$$DIM = \frac{C_{metal} * C_{factor} * D_{intak}}{B_{weight}}$$

Where, C_{metal}, C_{factor}, D_{intake} and B_{weight} represent the heavy metal concentrations in the vegetable samples, the conversion factor, the daily intake of the sample and the average body weight, respectively. The conversion factor (CF) of 0.085 [13] was used for the conversion of the samples to dry weights. The average daily intake of the samples was 0.527 kg person^{-1} d^{-1} [14] and the average body weight for the adult and children population was 60 kg [15] and 24 kg [16] 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 or vegetable were evaluated by computing the target hazard quotient (THQ) using following equation [17].

$$THQ = CDI/R_fD$$

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 [18]. 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) [19,20].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 [21]. It is calculated as follows:

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

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 [22]. 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 [23].

2.6 Cancer Risks

The possibility of cancer risks in the studied samples through intake of carcinogenic heavy metals was estimated using the Incremental Lifetime Cancer Risk (ILCR) [24].

$$ILCR = CDI \times CSF$$

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 (17). ILCR value in the samples represents the probability of an individual's lifetime health risks from carcinogenic heavy metals' exposure [25]. The level of acceptable cancer risk (ILCR) for regulatory purposes is considered within the range of 10^{-6} to 10^{-4} [18]. 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 [24].

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

Where EDI is the estimated daily intake of metal via consumption of millet; 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 × ED_{tot}), and 60 years life time for carcinogenic effect [17]. 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 [24].

$$\sum ILCRn = ILCR_1 + ILCR_2 + \cdots + ILCR_n$$

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

3. RESULTS AND DISCUSSION

3.1 Results

The present study investigated the presence of heavy metals in Hibiscus 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 Zn (range: 1.048-1.208), followed by Pb (range: 0.508-0.982), Fe (range: 0.684-0.978) and Cr (range: 0.135-0.261). While Cd has the lowest concentration (range: 0.042-0.051 ppm) with the heavy metals Mn and Ni being below detection level (BDL)

The results for the estimated daily intake (EDI) of the heavy metals on consumption of the cultivated millet were given in Table 2. From the table the estimated daily intake of the heavy metals (Pb, Zn, Cd, Cr and Fe) were lower than the tolerable daily intake limit set by the USEPA [26] in both samples and the health risk index (HRI) for all the heavy metals were <1 (Table 3).

Table 1. Heavy metal concentration (mg/kg) in cultivated hibiscus leaf samples from Katsina State

Location	Heavy Metal						
	Mn	Zn	Pb	Cd	Ni	Fe	Cr
Birchi	BDL	1.138000 ±0.000700	0.651000 ±0.000200	0.043000 ±0.000100	B DL	0.978000 ±0.001000	0.261000 ±0.000900
Dabai	BDL	1.116000 ±0.000200	0.821000 ±0.000300	0.047000 ±0.000300	B DL	0.924000 ±0.000700	0.174000 ±0.001300
Daura	BDL	1.114000 ±0.000400	0.957000 ±0.001600	0.046000 ±0.000300	B DL	0.917000 ±0.003100	0.135000 ±0.000700
Dutsinma	BDL	1.108000 ±0.000100	0.926000 ±0.000400	0.051000 ±0.000100	B DL	0.862000 ±0.000200	0.152000 ±0.000700
Funtua	BDL	1.172000 ±0.013000	0.982000 ±0.000100	0.045000 ±0.000300	B DL	0.834000 ±0.000200	0.160000 ±0.001100
Ingawa	BDL	1.135000 ±0.000300	0.508000 ±0.000200	0.043000 ±0.000200	B DL	0.951000 ±0.001400	0.168000 ±0.000600
Kafur	BDL	1.105000 ±0.000200	0.672000 ±0.000300	0.042000 ±0.000200	B DL	0.684000 ±0.000400	0.153000 ±0.000800
Katsina	BDL	1.048000 ±0.000600	0.923000 ±0.000200	0.046000 ±0.000100	B DL	0.905000 ±0.002500	0.142000 ±0.000600
Malunfashi	BDL	1.118000 ±0.000200	0.918000 ±0.000300	0.047000 ±0.000200	B DL	0.917000 ±0.000600	0.157000 ±0.000800
Matazu	BDL	1.125000 ±0.000200	0.825000 ±0.000100	0.048000 ±0.000200	B DL	0.868000 ±0.000300	0.145000 ±0.000600
Zango	BDL	1.208000± 0.000200	0.816000±0.0 00100	0.047000± 0.000300	B DL	0.926000±0.0 00200	0.158000± 0.000500

Values are expressed as Mean ± Standard

Table 2. Daily intake of heavy metal in adults from consuming cultivated hibiscus from Katsina State

Location	Heavy metal				
	Zn	Pb	Cd	Fe	Cr
Birchi	0.000850	0.000486	0.000032	0.000730	0.000195
Dabai	0.000833	0.000613	0.000035	0.000690	0.000130
Daura	0.000832	0.000715	0.000034	0.000685	0.000101
Dutsinma	0.000827	0.000691	0.000038	0.000644	0.000113
Funtua	0.000875	0.000733	0.000034	0.000623	0.000120
Ingawa	0.000847	0.000379	0.000032	0.000710	0.000125
Kafur	0.000825	0.000502	0.000031	0.000511	0.000114
Katsina	0.000782	0.000689	0.000034	0.000676	0.000106
Malunfashi	0.000835	0.000685	0.000035	0.000685	0.000114
Matazu	0.000839	0.000616	0.000036	0.000648	0.000108
Zango	0.000900	0.000609	0.000033	0.000691	0.000118

The non-cancer risks (THQ) of the investigated heavy metals through the consumption of vegetables and fruits 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.

Tables 6 and 7 shows Incremental Life Time Cancer Risk in both Adults and from Consuming Cultivated Hibiscus Leaves from Katsina State. The sampling area trend of risk for developing cancer as a result of consuming the studied samples showed: Funtua > Daura > Dutsinma > Katsina > Malunfashi > Matazu > Zango > Ingawa > Kafur > Dabai > Birchi for both adult and children.

Table 3. Daily intake of heavy metal in children from consuming cultivated hibiscus from Katsina State

Location	Heavy metal				
	Zn	Pb	Cd	Fe	Cr
Birchi	0.002105	0.001215	0.000080	0.001825	0.000487
Dabai	0.002083	0.001533	0.000088	0.001725	0.000325
Daura	0.002079	0.001786	0.000086	0.001712	0.000252
Dutsinma	0.002069	0.001728	0.000095	0.001609	0.000284
Funtua	0.002188	0.001832	0.000084	0.001557	0.000299
Ingawa	0.002118	0.001508	0.000080	0.001775	0.000314
Kafur	0.002062	0.001254	0.000078	0.001277	0.000286
Katsina	0.001956	0.001723	0.000086	0.001689	0.000265
Malunfashi	0.002087	0.001713	0.000088	0.001712	0.000293
Matazu	0.002100	0.001840	0.000090	0.001620	0.000271
Zango	0.002247	0.001523	0.000088	0.001728	0.000295

Table 4. Heavy metal target hazard quotient and health risk index in children from consuming cultivated hibiscus from Katsina State

Location	Target Hazard Quotient Heavy Metal					Health Risk Index (HRIs)
	Zn	Pb	Cd	Fe	Cr	
Birchi	0.007018	0.002026	0.000161	0.002608	0.001624	0.013435
Dabai	0.006943	0.002554	0.000175	0.002464	0.001083	0.013219
Daura	0.006931	0.002977	0.000172	0.002445	0.000840	0.013364
Dutsinma	0.006894	0.002881	0.000190	0.002298	0.000946	0.013209
Funtua	0.007292	0.003055	0.000168	0.002224	0.000995	0.013699
Ingawa	0.007061	0.002514	0.000161	0.002536	0.001045	0.013316
Kafur	0.006875	0.002090	0.000157	0.001824	0.000952	0.011898
Katsina	0.006520	0.002871	0.000172	0.002413	0.000883	0.012860
Malunfashi	0.006956	0.002856	0.000176	0.002445	0.000977	0.013409
Matazu	0.006999	0.002566	0.000179	0.002314	0.000902	0.012961
Zango	0.007391	0.002538	0.000166	0.002369	0.000983	0.013357

Table 5. Heavy metal target hazard quotient and health risk index in adults from consuming cultivated hibiscus from Katsina State

Location	Target Hazard Quotient Heavy Metal					Health Risk Index (HRIs)
	Zn	Pb	Cd	Fe	Cr	
Birchi	0.002832	0.001643	0.000064	0.001043	0.000650	0.006232
Dabai	0.002773	0.001022	0.000070	0.000986	0.000433	0.005283
Daura	0.002772	0.001191	0.000069	0.000992	0.000359	0.005384
Dutsinma	0.002757	0.001152	0.000076	0.000919	0.000378	0.005283
Funtua	0.002917	0.002222	0.000067	0.000890	0.000398	0.006493
Ingawa	0.002825	0.000632	0.000064	0.001014	0.000418	0.004953
Kafur	0.002750	0.000836	0.000052	0.000730	0.000381	0.004749
Katsina	0.002608	0.001148	0.000069	0.000965	0.000353	0.005744
Malunfashi	0.002782	0.001142	0.000070	0.000978	0.000378	0.005351
Matazu	0.002750	0.001027	0.000072	0.000926	0.000361	0.005830
Zango	0.003000	0.001015	0.000066	0.000962	0.000393	0.005436

Table 6. Incremental life time cancer risk in adults from consuming cultivated hibiscus leaves from Katsina State

Location	ILCR		∑ILCR
	Pb	**Cd**	
Birchi	3.061957E-03	5.457500E-05	3.123145E-03
Dabai	3.861547E-03	5.965100E-05	3.921198E-03
Daura	4.501224E-03	5.838100E-05	4.559605E-03
Dutsinma	4.355416E-03	6.472700E-05	4.420143E-03
Funtua	4.618807E-03	5.711300E-05	4.675920E-03
Ingawa	3.389363E-03	5.457500E-05	3.443938E-03
Kafur	3.160735E-03	5.330500E-05	3.214040E-03
Katsina	4.341304E-03	5.838100E-05	4.371421E-03
Malunfashi	4.317786E-03	5.965100E-05	4.377437E-03
Matazu	3.880365E-03	6.092100E-05	3.941286E-03
Zango	3.838303E-03	5.584300E-05	3.893878E-03

Table 7. Incremental life time cancer risk in children from consuming cultivated hibiscus leaves from Katsina State

Location	ILCR		∑ILCR
	Pb	**Cd**	
Birchi	7.654903E-03	1.203855E=03	8.858758E-03
Dabai	7.655506E-03	1.315845E-03	8.971351E-03
Daura	1.125306E-02	1.278550E-03	1.253161E-02
Dutsinma	1.088854E-02	1.427835E-03	1.231637E-02
Funtua	1.154703E-02	1.259850E-03	1.280688E-02
Ingawa	9.501017E-03	1.203855E-03	1.070487E-02
Kafur	7.901838E-03	1.175850E-03	9.077688E-03
Katsina	1.085326E-02	1.287855E-03	1.214112E-02
Malunfashi	1.079447E-02	1.315845E-03	1.211031E-02
Matazu	9.700916E-03	1.343850E-03	1.104477E-02
Zango	9.595089E-03	1.315845E-03	1.091093E-02

3.2 Discussion

The results for the heavy metals analysed in the sampled leaves are similar to that reported for heavy metals in beans and some beans products from some selected markets in Katsina state, Nigeria [27]. 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 EU and 0.05 mg/kg limit set by USEPA [28]. The violation of the maximum permissible limits of Pb set by WHO, EU, and US EPA is a cause for public health concern considering the frequency of exposure. But the results are higher than that reported for the concentration of Pb in cereals from Kano and Kaduna states, Nigeria [29,30]. The value was still higher than the range (0.116 to 0.390) reported by Ahmed and Mohammed in Egypt [31] in 2005 and the range (0.007 to 0.032 mg/kg) reported by Okoye et al.[32] in a study conducted in South east of

Nigeria in 2009 The Pb concentration range for the leaf samples in this study is lower than that reported for leafy vegetables from Kaduna state Nigeria [33] and that reported for beans samples from Italy, Mexico, India, Japan, Ghana and Ivory Coast with a Pb concentration range of 4.084-14.475 ppm [34] and the Pb result for homegrown vegetables near a former chemical manufacturing facility in Tarnaveni and Romania [35]. The concentration of Cd (mg/kg) range from 0.042 to 0.059 in the leaf samples, these values were higher than the range (0.002 to 0.004 mg/kg) reported by Edem et al.in Wheat flours in 2009 [36]. The Cd concentration range for the samples in this study is lower than that reported for various beans samples from Europe, Asia and parts of West Africa [34], for locust beans from Odo-Ori market Iwo, Nigeria [37] and the Cd concentrations reported for cabbage (0.140 mg/kg) from Awka, Anambra state Nigeria [38]. The values are also lower than those obtained by Okoye et al.[32] in Cereals in South eastern Nigeria (0.007 to 0.23mg/kg) in 2009. Ahmed

and Mohammed [31] in Cereal products (0.091-0.143 mg/kg) in 2005, Orisakwe et al. [39] in Owerri (0.00 to 0.24 mg/kg) in 2012 and Dahiru et al. [29] in Kano (0.11 to 0.28 mg/kg) in 2013. But the results are similar that reported in a study for the Cadmium concentration range for both unprocessed and processed bean samples from Katsina state Nigeria [27]. These differences could be due to differences in the concentration of the metal in the soils where these vegetables were grown.

The Fe values for the present study are higher than the range reported by Edem et al. [36] 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 [30]. In the present study, the mean Fe concentration in the leaf samples is higher than that reported in a study that evaluate heavy metals in millet from Kaduna, Nigeria [30] the Fe concentration reported for cabbage (0.360 mg/kg) from Awka, Anambra state Nigeria [38]. The result is similar to that reported for market sold beans from Katsina, Nigeria [27], but is lower to that reported in a study in eastern Nigeria [32] and that recorded by Zahir et al. [40] in a study conducted in Pakistan and the results for the study conducted by Di Bella et al. [34].

The heavy metal Zn concentration range obtain in this study is similar to that reported in some studies that evaluate Zn in millet, sorghum and various food crops from Kano and Zamfara [29, 41], but are higher than the range (0.04 to 0.19 mg/kg) reported by Edem et al. [35] in 2009 in wheat flours, but far below the range reported by Ahmed and Mohammed [31] in 2005 (4.893 to 15.450 mg/kg) in various foodstuff in a study in Egypt and that reported in a study conducted by Sulyman et al. [42].These values can also not provide for the required daily allowance for Zn which is 11mg/day for men and 8mg/day for women [30]. The degree for heavy metal toxicity to humans depends on daily consumption rate [43].

The results for the estimated daily intake (EDI) of the heavy metals on consumption of the cultivated millet were given in Table 2. From the table the estimated daily intake of the heavy metals (Pb, Zn, Cd, Cr and Fe) were lower than the tolerable daily intake limit set by the USEPA (43) in both samples and the health risk index (HRI) for all the heavy metals were <1 (Table 3).

The non-cancer risks (THQ) of the investigated heavy metals through the consumption of vegetables and fruits 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 [44]. THQ is interpreted as either greater than 1 (>1) or less than 1 (<1), where THQ >1 shows human health risk concern [45]. Bhalkhair and Ashraf [14] 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. In both adults and children the highest THQ was for the heavy metal Zn from the sample from Zango, while the lowest was for the heavy metal Cd from the sample from Kafur. The THQ for the samples was in the decreasing order Zn>Pb>Fe>Cr>Cd, in all the samples respectively. 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. [46], Micheal et al. [17] and Liu et al. [24].

Cd and Pb are classified by the IARC as being carcinogenic agents [47]. Chronic exposure to low doses of Cd, and Pb could therefore result into many types of cancers [48]. The computed ILCR and cumulative incremental lifetime cancer risk (∑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 [25,49]. ILCR for Cd is below the threshold risk limit (>10^{-5}) 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 were 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: Funtua > Daura > Dutsinma > Katsina > Malunfashi > Matazu > Zango > Ingawa > Kafur > Dabai > Birchi for both adult and children (Tables 6 and 7).

Moreover, cumulative cancer risk (∑ILCR) of all the studied Hibiscus samples have reached the moderate risk limit ($>10^{-3}$) in adult. While in children with the exception of the sample from Birchi, Dabai and Kafur which are within the moderate cancer risk ($>10^{-3}$) limit, all other samples were beyond the moderate cancer risk ($>10^{-2}$) limit. Further, among all the studied samples, the sample from Funtua has the highest chances of cancer risks (ILCR 4.675920 × 10^{-3} in adults; ILCR 1.280688 × 10^{-2} in children) and the sample from Birchi has the lowest chances of cancer risk (ILCR 3.123145 × 10^{-3} in adults; ILCR 8.858758 × 10^{-3} in children). These risk values indicate that consumption of the sample from Funtua would result in an excess of 47 cancer cases in adults per 10,000 people exposure and 13 cancer cases per 1,000 people exposure while consumption of the sample from Birchi would result in an excess of 31 cancer cases in adults and 89 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

4. CONCLUSION

This study determines the heavy metals concentration in hibiscus leaf samples from Katsina state Nigeria. Results from this study have shown that with the exception of the heavy metal Pb the concentration values of Mn, Zn, Cd and Fe in the samples were generally lower than the USEPA, WHO/FAO maximum permissive limits. Risk level of Target Hazard Quotient (THQ < 1) was observed for all the evaluated heavy metals for both adults and children. ILCR for Cd is below the threshold risk limit ($>10^{-5}$) and ILCR for Pb reached the moderate risk limit ($>10^{-3}$) in all the studied samples in adults, While in children ILCR for 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 were 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: Funtua > Daura > Dutsinma > Katsina > Malunfashi > Matazu > Zango > Ingawa > Kafur > Dabai > Birchi for both adult and children. 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.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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