Heavy Metals and Health Risk Burden, How Safe is the Consumer Population?: A Study on the Heavy Metal Content of Soybeans Cultivated in Katsina State, North West Nigeria

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Authors' contributions

This work was carried out in collaboration among all authors. Author AIY designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors JIB, LS, AJA, AN, UB and AU performed the statistical analysis and manage the analysis of the study. Authors ZAS, AA, MMM, AIY and IAY managed the literature searches. All authors read and approved the final manuscript.

Original Research Article

ABSTRACT

The level of heavy metals in soybeans samples were evaluated to ascertain their likely health risks to the Katsina State populace. Atomic absorption spectrometry method was employed for the evaluation, while the three senatorial zones of the state were used for sampling. Using methods adapted from the United States Environmental Protection Agency (US EPA), the health risks to the

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population from the heavy metals in the samples were determined. The results have revealed the mean concentration (mg/kg) in decreasing order of concentrations to be for Zn (range: 1.204-1.432), Pb (range: 0.658-0.998), Fe (range: 0.563-0.687), Cr (range: 0.128-0.151) and Cd (range: 0.041-0.046), with the concentrations of Mn and Ni below the level of detection. The computed non-carcinogenic health risks from consumption of the samples were below 1 (THQ and HI>1). The cancer risk to the population lies beyond the threshold limit with the heavy metal Pb being the major contributor to the violation. It may be presumed that the Soybean samples in the study may add to the cancer manifestations in the population.

1. INTRODUCTION

Toxicities arising from pollution by heavy metals are worrisome, due to their unique ability to persist in environmental components (soil, water and air) leading to their bioaccumulation and biomagnification in food components and can results in deleterious devastation consequences that may take the form of carcinogenesis, mutations, and neurological problems [1-3].

"Of recent, exposures to environmental toxic metals have become a global public health concern owing to their potential bioaccumulation and their deleterious health effects in humans" [4-7]. "Environmental exposure to heavy metals is a well-known risk factor for cancers [8] as chronic exposure to high levels of toxic metals has been associated with higher risk of cancers of the bladder, kidney, liver, pancreas, lung, and skin [9] and many visceral organs diseases". "Because of heavy metals influence on disease systems and organs heavy metals exposure may also lead to cardiovascular, nervous, urinary and reproductive disease, as well as aggravation of pre-existing symptoms and disease" [10]. "Evidence suggests that these toxic metals may have adverse effects on these outcomes even at lower concentrations [7], which might be prevalent in many parts of the world".

Research in heavy metals in foods locally produced in Katsina State and their possible health risks to the population is in its infancy, a situation that results into paucity of information on the contribution of these notable pollutants to the disease burden of the Katsina State population.

This work contributes to the monitoring of heavy metal exposure in soybeans cultivated in Katsina State, Nigeria and possible carcinogenic and non-carcinogenic risk to the population.

2. MATERIALS AND METHODS

2.1 Study Area

The study was conducted in Katsina State, North western part of Nigeria covering an area that span 24,192 km^2 (9,341 m^2) and globally positioned at latitude 12^{0}15'N and longitude of 7$^°$30'E [11]. The month of April heralds the beginning of the rainy season which ends in October, with the dry season starting in November and ending in March. The average annual rainfall is 1,321 mm, an average temperature of 27.3°C, and a relative humidity of 50.2%. The soil is alluvial flood plain that varies base on location, having good water holding capacity and low in contaminants [11]. Sampling was conducted in five local government areas (Matazu; Funtua; Dabai/Danja; Malumfashi; Kafur) located in the southern part of Katsina State (Fig. 1).

2.2 Sampling

The catchment areas for sampling for this work were divided into five (5) locations, which were subdivided further into twenty (20) sampling sites. Sampling (1kg per sample) was done in each of the site and thereby combined to form bulk sample, with a representative sample (1 kg) obtained from the bulk. Glass Bottles (code named) were used to store the samples for protection against contamination and moisture at a temperature of 4$^°$C prior to analysis.

2.3 Sample Preparation

After cleaning of the samples by removing all impurities and drying at room temperature, 300 g of the sample was grinded to a fine powder and stored under the same condition as the duplicate portion prior to analysis.
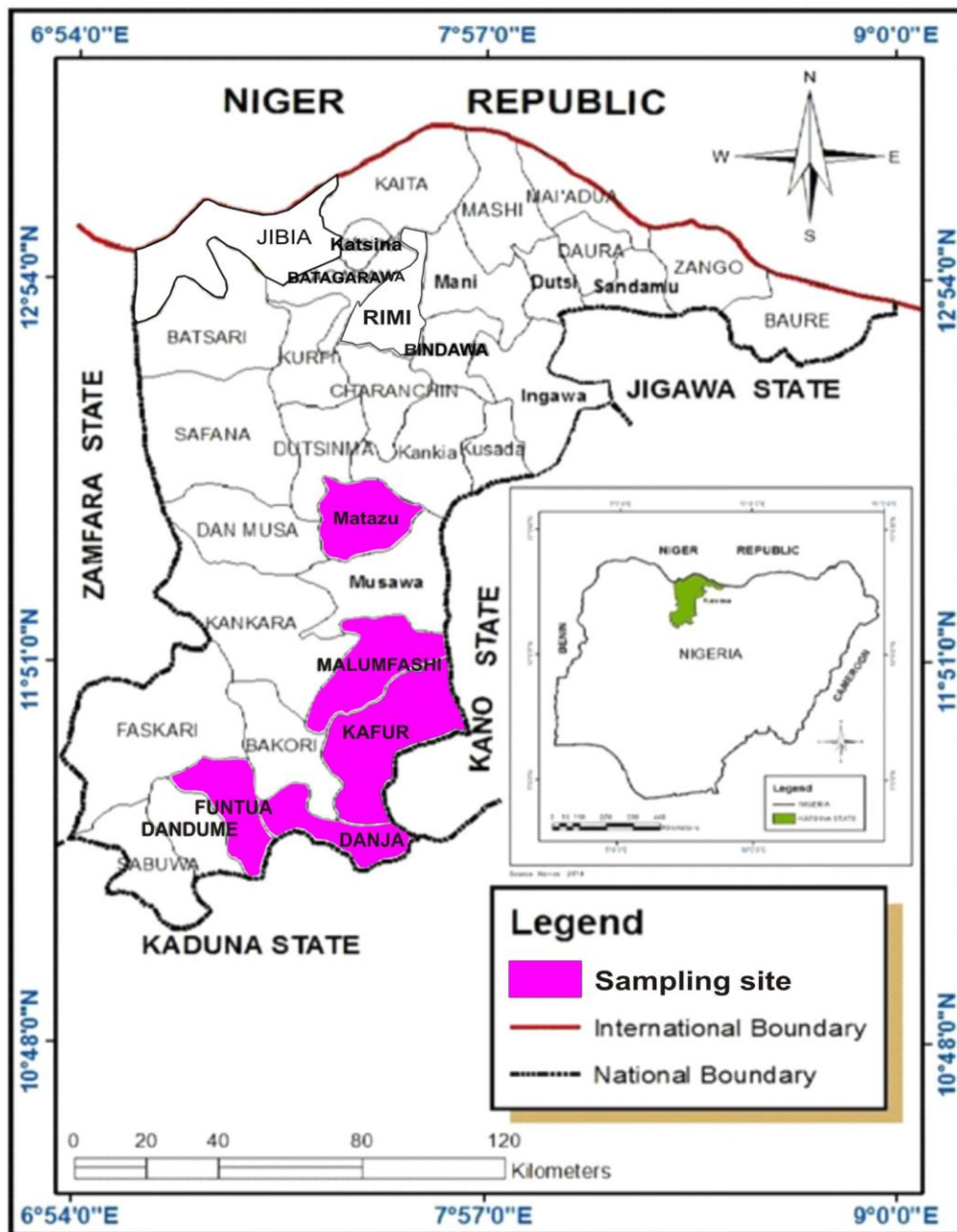
Fig. 1. Map of Katsina State showing the designated sampling sites

2.4 Heavy Metals Determination

A Gallenkamp hotbox oven (CHF097XX 2.5) was used to dry 5 g of each of the powdered sample at 80°C for 2 hours and then blended using an electric blender, which was followed by ashing 0.5 g of the sample in an electric muffle furnace (Thermolyne FB131DM Fisher Scientific) at 550°C for 24 hours. The ash was diluted with a mixed ratio of 3:1 concentrated hydrochloric acid (HCl) and concentrated nitric acid (HNO$_3$) and allowed to stand for a while. To make up to 100 ml mark, 50 ml of distilled water was added to the diluents. Using standard methods [method 986.15] [12], the levels of heavy (Pb, Zn, Ni, Cd, Cr, Mn and Fe) metals in the soybeans samples were determined using AA210RAP BUCK Atomic Absorption Spectrometer flame emission spectrometer filter GLA-4B Graphite furnace (East Norwalk USA)) and the metal concentrations expressed in mg/kg.

Table 1. Background information of the atomic absorption spectrophotometer used in the study

Metal	Detection limit mg/l	Sensitivity check mg/l	Wavelength Nm	Linear range	Slit Nm
Fe	0.050	2.5	248.3	5.0	0.2
Pb	0.080	10.0	283.3	20	0.7
Mn	0.030	1.25	279.5	2.50	0.7
Zn	0.005	0.5	213.9	2.50	0.7
Cd	0.010	0.75	228.9	2.0	0.7
Cr	0.040	2.0	357.9	2.0	0.7

2.5 Heavy Metal Health Risk Assessment

2.5.1 Daily intake of metals (DIM)

The ingestion of heavy metals in the samples depicted as daily intake of metals (DIM) was calculated using the following equation:

$$DIM = \frac{Cmeta \; x \; Cfactor \; x \; Dintakel}{Bweight} \qquad \text{eqn. (1).}$$

With C metal standing for heavy metal concentration sample, C factor representing the conversion factor (CF) which was taken as 0.085 used in converting the soybean samples to their dry weight, D intake representing the daily intake of the sample taken from literature as 0.527 kg person$^{-1 \; d-1}$ [13], and B weight representing the average body weight which is also taken from the literature as 60 kg [14] for adults and 24 kg [15] for children. The same values were used to evaluate the HRI.

2.5.2 Non-cancer risks

The target hazard quotient (THQ), which gives an idea of the non-carcinogenic risks to the consumer population from intake of heavy metals were determined using the following equation [16].

$$THQ = CDI/RfD \qquad \text{eqn. (2).}$$

CDI represent the chronic daily heavy metal intake expressed in mg/kg/day and RfD represents the oral reference dose (mg/kg/day) which is quantification the maximum permissible risk to the consumer from daily exposure throughout an individual life span [17]. Individual reference doses taken from literature were used (Pb = 0.6, Cd = 0.5, Zn = 0.3, Fe = 0.7, Ni = 0.4, Mn = 0.014, Cr = 0.3) [18,19]. In conjunction with the THQ this research also uses the chronic hazard index (HI) that evaluate the potential risk to the population from exposure from more than

one heavy metal, which is the summation of all the hazard quotients (THQ) for each heavy metal for a particular exposure pathway [20], which is computed using the formula below:

$$HI = THQ_1 + THQ_2 + \cdots + THQ_n \qquad \text{eqn. (3).}$$

Where the subscripts 1, 2 .... n represent each heavy metal in the sample.

It is taken that the severity of the effect is equal to the total metal exposures and that organs affected by the exposure have similar working mechanism [21]. HI less than 1 infer that the consumer population is safe, while HI above 1 raise the level of concern to the consumer population [22].

2.5.3 Cancer risks

The risks of cancer to the consumer population from intake of the soybeans samples in the study were evaluated with the use of Incremental Lifetime Cancer Risk (ILCR) [23].

$$ILCR = CDI \; x \; CSF \qquad \text{eqn. (4).}$$

With CDI representing the chronic daily intake of individual carcinogenic heavy metal from a lifelong ingestion of the sample expressed in mg/kg, BW/day and CSF representing specific cancer factors for each heavy metal in the sample comparable to the individual weight [16]. Adapted from literature, the cancer slopes for Pb = 0.0085 mg/kg/day [24], Cd = 0.38 mg/kg/day [25] where used in this study.

ILCR value in a particular sample is representative possibility of the consumer lifetime health risks from exposure to heavy metal carcinogens [26]. The range 10^{-6} to 10^{-4} is considered safe for the consumer population (17). The CDI was computed by the use of the below equation [23].

CDI = (EDI x EFr x EDtot)/AT…………….eqn. (5)

In which the EDI is the estimated daily intake of metal from intake of the samples; EFr represents the frequency of exposure (365 days/year); EDtot is the length of exposure which is taken as the average life time of 60 years for Nigerians; AT represent the duration of exposure for non-carcinogenic effects (EFr × EDtot), and 60 life years for carcinogenic effect [16]. The Human exposure to more than one carcinogenic heavy metal through food intake may result in cumulative cancer risk is the summation of the individual heavy metal increment risks and it is computed as below [23].

$$\sum 1n = ILCR_1 + ILCR_2 + \cdots + ILCR_n \ldots\ldots\ldots..eqn.~(6).$$

With the subscripts 1, 2 …n, representing each carcinogenic heavy metal.

3. RESULTS

The current work evaluated the presence of heavy metals in soybeans, one of the major legumes that are consumed by the population of Katsina State, Nigeria. As portrayed in Table 2, the mean concentration (mg/kg) in decreasing order of concentrations to be Zn (range: 1.204-1.432), Pb (range: 0.658-0.998), Fe (range: 0.563-0.687), Cr (range: 0.128-0.151) and Cd (range: 0.041-0.046), with the concentrations of Mn and Ni below the level of detection. The heavy metal Pb mean value in the sample was above the permissible values for Pb in legumes, while the remaining heavy metals evaluated exhibited mean values that were within the permissible range.

The results for the calculated daily intake of the heavy metals from ingestion of the soya bean samples were displayed in Figs. 1 and 2. Compared to the daily intake limit for heavy metals in legumes as set by the USEPA [27], all the calculated metal intakes were within the permissible limit. The sequential order for the daily metal intake of heavy metals from the designated sampling locations is represented as: Kafur (Zn>Pb>Fe>Cd); Malumfashi (Zn>Pb>Fe> Cr>Cd); Dabai/Danja (Zn>Pb>Fe> Cr>Cd); Funtua (Zn>Pb>Fe>Cr>Cd); Matazu (Zn>Pb> Fe>Cr>Cd). The sequential order being the same in both the children and adult's consumer population.
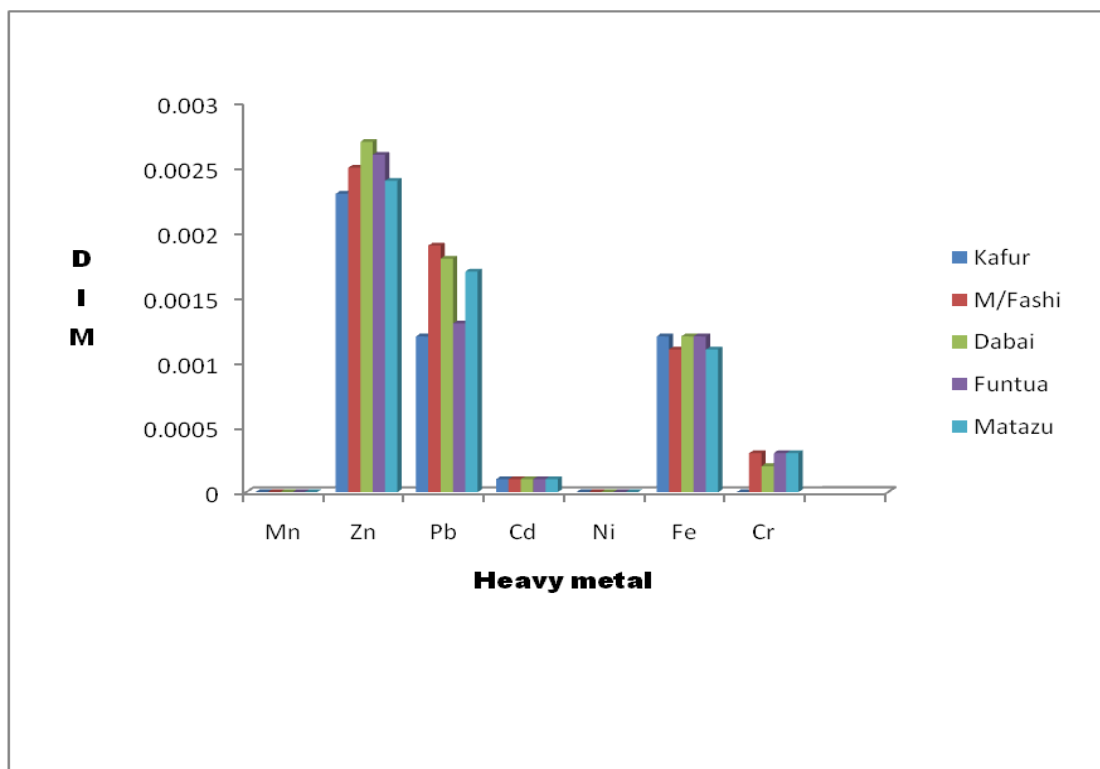

Fig. 2. Daily Intake of Heavy Metal in Children from Consumption of Cultivated Soybeans from Katsina State

Table 2. Heavy Metal Concentration (mg/kg) in Cultivated Soybean Samples from Katsina State

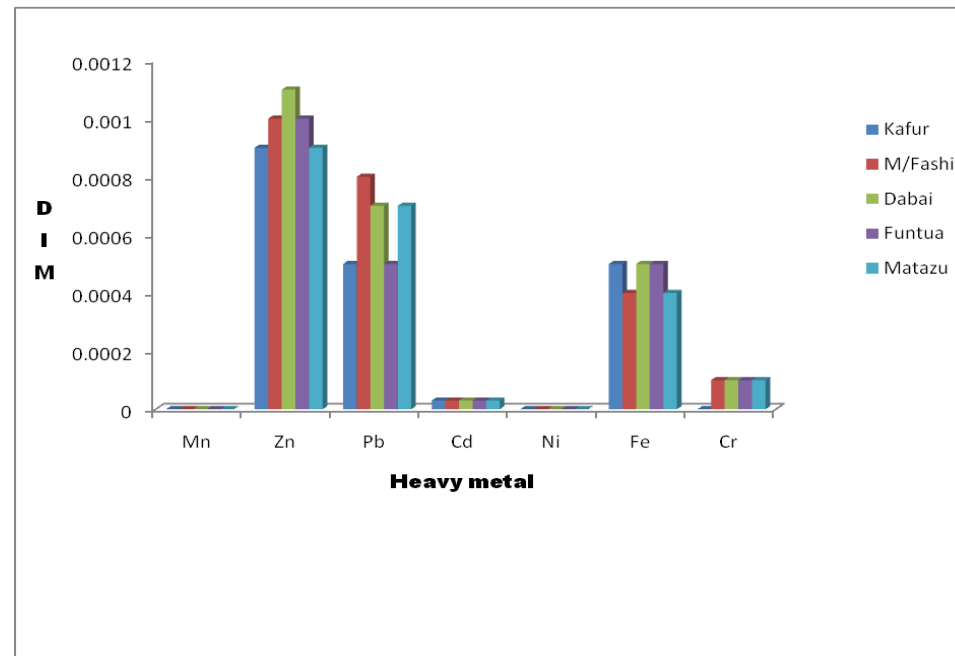| Location			Heavy metal				
	Mn	Zn	Pb	Cd	Ni	Fe	Cr
Kafur	BDL	1.204 ± 0.0130	0.658± 0.0002	0.045± 0.0003	BDL	0.687± 0.0010	BDL
Malumfashi	BDL	1.347± 0.0002	0.998± 0.0003	0.046± 0.0001	BDL	0.563± 0.0002	0.15± 0.0006
Dabai/Danja	BDL	1.432± 0.0002	0.926± 0.0008	0.041± 0.0003	BDL	0.648± 0.0004	0.128± 0.0003
Funtua	BDL	1.381± 0.0004	0.673± 0.0003	0.046± 0.0003	BDL	0.652± 0.0002	0.147± 0.0002
Matazu	BDL	1.257± 0.0008	0.913± 0.0003	0.042± 0.0006	BDL	0.574± 0.0004	0.132± 0.0007

Fig. 3. Daily Intake of Heavy Metal (DIM) in Adults from Consumption of Cultivated Soybeans from Katsina State

Tables 3 and 4 represent the THQ and HRIs in the children and adult population from consumption of the soybean samples. It is observable from the displayed results that the THQ value is below 1, with the heavy metal Zn of the sample from Dabai/Danja with the highest THQ (0.0036 in adult; 0.0084 in children) and the heavy metal Cd in the Dabai/Danja sample (0.00006 in adult; 0.00015 in children) having the lowest THQ. Similarly the HRIs were below 1 in both the children and adult population, with the sample from Dabai/Danja (0.0073 in adult; 0.0139 in children) manifesting the highest HRI and the sample from Kafur (0.0046 in adult; 0.0115 in children) having the lowest HRI.

The consumer is believed to be safe when HI < 1 and in a degree of concern when 1 < HI < 5. THQ is taken as either above 1 (>1) or below 1 (<1), where THQ >1 indicate a degree of concern to human health.

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The risk of cancer from consumption of the samples for the carcinogenic heavy metals Cd, and Pb are displayed on Tables 5 and 6. From the results in the Soybeans samples in adults, the ILCR for Cd has reached the safety threshold risk limit ($>10^{-4}$) in all the studied samples, while the ILCR for Pb lies within the moderate risk limit ($>10^{-3}$). In children population, the calculated cancer risk for Pb and Cd was within the moderate risk limit ($>10^{-3}$). The sequential order of risk for developing cancer from likely ingestion of the samples is: Malumfashi> Dabai/Danja > Matazu> Funtua > Kafur.

The soybeans samples have a calculated cumulative cancer risk ($\sum$ILCR) that has reached the moderate risk limit ($>10^{-3}$). The Soybeans sample from Malumfashi has the greatest possibility of cancer risks to the Katsina State population (ILCR 5.2×10^{-3} in adults, 1.3×10^{-2} in children) and the studied soybean from Kafur has the least possibility of cancer risk (ILCR 3.5×10^{-3} in adults, 8.9×10^{-3} in children). The observed values implies that there is a possibility of 52 cancer manifestations in 10,000 of the adult population and 13 cancer manifestations in 1000 of the children population, that may likely arose from the consumption of the sample from Malumfashi. While consumption of the sample

from Kafur would likely result in a possibility of 35 cancer manifestations in adults and 89 cancer manifestations in children for every 10,000 people that are exposed.

The limit of safety for cancer risk is less than about 1 incidence in 1,000,000 lifetime exposure (ILCR < 10^{-6}) and threshold risk limit (ILCR > 10^{-4}) for incidence of cancer is above 1 in 10,000 exposure where remedial steps are desirable, and moderate risk level (ILCR > 10^{-3}) is above 1 in 1,000, where public health safety consideration is of paramount importance.

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4. DISCUSSION

Lead was seen in all the samples at a higher concentration than the 0.01mg/kg, 0.02 mg/kg and 0.05 mg/kg safety limits set by the WHO/FAO, EU and USEPA respectively, raises a level of concern. The recorded Pb concentrations in the study were low when compared to the reported Pb concentration range for soybeans samples from Iran [28] and the reported concentrations for soybean samples from Northeast China [29]. These values are also lower than the WHO safe limit for Pb in Cereals reported in literature [30]. But the results are higher when compared to results reported for the concentration of Pb from Kano State Nigeria, the result of a study conducted in Southeast Nigeria, Anhui province in China and in Zhejiang also in China [31- 34]. A possible explanation for the difference may be due to disparities in anthropogenic contribution to heavy metal pollution in the various sites were the studies were conducted, or contamination during production or handling process, and from industrial or vehicular exhaust [35].

Studies conducted in Zhejiang China that evaluate heavy metals in Romaine lettuce and cabbage [34] and in Katsina State, Nigeria that evaluate heavy metals in unprocessed and processed bean samples [36] and for locust beans from Odo-Ori market Iwo, Nigeria [37] all reported Cd concentrations similar to the results of the present study. But the reported Cd in the

Table 3. Heavy Metal Target Hazard Quotient and Health Risk Index in Adults from Consumption of Cultivated Soybeans from Katsina State

Location	Target Hazard Quotient							Health Risk Index (HRIs)
	Heavy metal							
	Mn	Zn	Pb	Cd	Ni	Fe	Cr	
Kafur	BDL	0.0030	0.0008	0.0001	BDL	0.0007	BDL	0.0046
Malumfashi	BDL	0.0034	0.0012	0.0001	BDL	0.0006	0.0004	0.0056
Dabai/Danja	BDL	0.0036	0.0012	0.0001	BDL	0.0007	0.0019	0.0073
Funtua	BDL	0.0034	0.0008	0.0001	BDL	0.0007	0.0004	0,0057
Matazu	BDL	0.0031	0.0011	0.0001	BDL	0.0006	0.0003	0.0053

Table 4. Heavy Metal Target Hazard Quotient and Health Risk Index in Children from Consumption of Cultivated Soybeans from Katsina State

Location	Target Hazard Quotient (THQ)							Health Risk Index (HRIs)
	Heavy metal							
	Mn	Zn	Pb	Cd	Ni	Fe	Cr	
Kafur	BDL	0.0075	0.0021	0.0002	BDL	0.0018	BDL	0.0115
Malumfashi	BDL	0.0075	0.0031	0.0002	BDL	0.0015	0.0010	0.0132
Dabai/Danja	BDL	0.0084	0.0029	0.0002	BDL	0.0017	0.0010	0.0139
Funtua	BDL	0.0089	0.0021	0.0002	BDL	0.0017	0.0009	0,0138
Matazu	BDL	0.0086	0.0028	0.0002	BDL	0.0015	0.0008	0.0119

Table 5. Incremental Life Time Cancer Risk in Adults from Consumption of Cultivated Soybeans from Katsina State

Location	ILCR		∑ILCR
	Pb	Cd	
Kafur	3.0952E-03	5.0400E-04	3.5992E-03
Malumfashi	4.6941E-03	5.1450E-04	5.2086E-03
Dabai/Danja	4.3533E-03	4.5900E-04	4.8123E-03
Funtua	3.1689E-03	5.1450E-04	3.6834E-03
Matazu	4.3004E-03	4.7100E-04	4.7714E-03

Table 6. Incremental Life Time Cancer Risk in Children from Consumption of Cultivated Soybeans from Katsina State

Location	ILCR		∑ILCR
	Pb	Cd	
Kafur	7.7372E-03	1.2599E-03	8.9971E-03
Malumfashi	1.1735E-02	1.2879E-03	1.3023E-02
Dabai/Danja	1.0889E-02	1.1479E-03	1.2036E-02
Funtua	7.9136E-03	1.2879E-03	9.2015E-03
Matazu	1.0736E-02	1.1759E-03	1.1912E-02

present study concentrations were above the concentrations of Cd reported for wheat flours from Calabar, Nigeria [38]. Likewise, the reported Cd values in the present study were lower than Cd values reported for soybean samples, cereals, and cereal products [29-31, 33; 39]. These differences could be due to differences in the concentration of the metal in the soils where these various food produce were grown.

The Fe concentrations in this study were above the concentrations recorded in wheat flours from Calabar, Nigeria [38], but still fall within the FAO/WHO 40.7 mg/kg Fe permissible limit in legumes [40]. The result is similar to the results reported for market sold beans from Katsina, Nigeria [36]. But the concentrations are lower to values reported in a study in eastern Nigeria [33] and that recorded by Zahir et al. [41] in a study conducted in Pakistan and the results for the study conducted by Li et al. [29].

"The heavy metal Zn values obtain in this study are similar to values of Zn in food reported in some studies" [31, 42], but are higher than the range (0.04 to 0.19 mg/kg) reported by Edem et al. in wheat flours [38]. But the values far below the range reported by Ahmed and Mohammed [30] and the values reported in a study conducted by Sulyman et al. [43].These values also falls below the WHO permissible limit for Zn as reported by Umar et al. [44].

In the Soybeans samples, risk levels of Target Hazard Quotient (THQ < 1) were observed for all the evaluated heavy metals for both adults and children. Which is an Indication that intake of these heavy metals through consumption of the Soybeans will not pose a considerable non-cancer risk to the population. The THQs for the samples were in the decreasing order Zn>Pb>Fe>Cr>Cd, in both the samples respectively. The sequence of risks was the same for both adults and children although the children had higher THQ values in all cases compared to the adults. Further, the non-cancer risks for each sample were 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/Danja and lowest in the sample from Kafur. It suggests that the inhabitants of Katsina State might not be exposed to non-carcinogenic health risk through the intake of heavy metals from soya beans.

"The THQs of less than 1 reported for all the samples analyzed were lower than the results of THQ for cabbage and tomato" in a study conducted by Gebeyehu and Bayissa [45] in Mojo, Ethiopia that reported a THQ of more than 1and that of a study conducted by Yi et al. [46], that reported "a THQ of more than 1 in fish samples from upper Yangtze river, China, and in a study conducted in Enyigba, south eastern Nigeria for Pb in lemon grass and Mn in leafy vegetables" [47]. The value is also lower than the THQ values above 1 reported by Bhalkhair and Ashraf [13] in a study conducted in the western region of Saudi Arabia on Okra vegetable, "they

are also lower than the THQ values for cereals, green leafy vegetables, roots and tubers from Vadodara" [48] and the values reported by Mahfuza et al. [49] for vegetables and fruits from Bangladesh. But the results are similar to the THQ of less than 1 reported for Shrimp samples from Selangor, Malaysia [50] and in tea leaves from Puan County, Guizhou province China [51].

The samples evaluated in the study showed the risk level (HI < 1) in both adults and children. "The HI in the samples differs from the HI values reported for vegetables from Tamale metropolis, Ghana that showed that the hazard index (HI) for both adult and children exceeded 1" [52]. " The value is also lower than the HI values of more than 1 for cabbage and tomato from Mojo, Ethiopia" [45], the report of Obiora et al. [47] in a study conducted in Enyigba, south eastern Nigeria for Pb in lemon grass and Mn in leafy vegetables. The HRI values of above 1 reported by Bhalkhair and Ashraf (13) in a study conducted in the western region of Saudi Arabia on Okra vegetable, the values for cereals, green leafy vegetables, roots and tubers from Vadodara [48] and the values reported by Mahfuza et al. [49] for vegetables and fruits from Bangladesh are all higher than the reported HRI of the current study.. But the HRI values are similar to what was reported for *Clarias gariepinus* from Imo River, Nigeria [53] and the reported studies conducted in Katsina State, Nigeria on leafy and fruit Vegetables, and on cereals[54- 56].

"The range of ILCR and ∑ILCR from consumption of all the evaluated samples as highlighted above, which raises the level of health concern for the consumer population as they may contribute to the population cancer burden is similar" to what was reported by Gebeyehu and Bayissa [45], in vegetables from Mojo Ethiopia, the ILCR and ∑ILCR reported from consumption meat and sea food samples from Xiamen, China [57]. The ILCR and ∑ILCR in Vegetables from Pearl River Delta South China [58], in fruits and vegetables from Jamaica [59], in vegetables from a Pb/Zn smelter in Central China [60] in vegetables grown in Patuakhali province Bangladesh [61] and in fruit, root and leafy vegetables, and fruits in a study conducted in a sub urban industrial area of Bangladesh [47].are also in line with the result of the present study. But the results differ from the results for vegetables from some selected communities from ONELGA Rivers State, Nigeria that reported non carcinogenic cancer risks from the vegetable samples in the study [62].

5. CONCLUSION

This work contributes to the monitoring of heavy metal exposure in soybeans cultivated in Katsina State and the possible carcinogenic and non-carcinogenic risk to the population that may results from consumption of the samples. With the exception of the heavy metal Pb whose values were above their permissible values, the rest of the metals have concentrations that were within the permissible limit. The metal target hazard quotient (THQ) and the hazard index (Hi) for the heavy metals evaluated falls within the safety limit. The overall cancer risk to the adults based on pseudo-total metal concentrations exceeded the target value, mainly contributed by the heavy metal Pb. Zn is the primary heavy metal posing non cancer risks while Pb caused the greatest cancer risk. The study has revealed that low non-carcinogenic risks exist for the population on consumption of the samples but the cancer risk is a cause for public health concern.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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