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Assessing Heavy Metal Contamination Risk in Selected Vegetables Marketed in Dutsin-Ma, Katsina State, Nigeria

***ABUBAKAR, AA; ABDULLAHI, M; ALBASHIR, Y**

Department of Applied Chemistry, Federal University Dutsinma, Katsina State - Nigeria

**Corresponding Author Email: aaabubakar83@gmail.com*
Co-Authors Email: abchemist2020@yahoo.com; ayasir@fudutsinma.edu.ng

ABSTRACT: The study investigated the presence of heavy metals (Cd, Cu, Fe, Mn, and Pb) in cabbage, lettuce, onion, pepper, and tomato, as well as evaluated the potential human health risks/hazard that might be caused due to their consumption. Their concentrations were determined using atomic absorption spectrophotometer and subsequently transformed in evaluating their health risks via estimation using estimated daily intake (EDI), target hazard quotient (THQ), hazard index (HI), and target cancer risk (TCR). The concentrations of the trace metals detected were in the range of 0.02 ± 0.00 to 0.20 ± 0.01; 0.12 ± 0.00 to 0.43 ± 0.30; 0.60 ± 0.5 to 21.77 ± 0.02; 0.70 ± 0.02 to 3.01 ± 0.00; and 0.03 ± 0.00 to 0.67 ± 0.01 mg/kg, for Cd, Cu, Fe, Mn, and Pb respectively. Lettuce has the highest concentration of lead (0.67 mg/kg), whereas Pepper has the least (0.03 mg/kg). In all the samples analyzed, except for Pepper, the lead concentrations were greater than WHO/FAO recommended level. The EDI values of all metals in question were below the maximum tolerable daily intake (MTDI). The total target hazard quotient (TTHQ) was less than 1, in all the samples examined, hence posing no threat to human health. Also, there is no alarm for cancer as the TCR values for all samples were 4.93×10^{-8} for Pb which is less than the threshold value of USEPA (10^{-6}), indicating that the consumers' residents of these vegetables are not exposed to Pb. However, regular monitoring of Pb metal in vegetables should be carried out to avoid heavy metal toxicity associated with the consumption of those vegetables.

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Fresh vegetables have significant value and are the most common food in the human diet around the world. It contains vitamins, protein, carbohydrates, water, minerals, antioxidants, and antimetabolites (Gupta *et al.,* 2022). Though, it is a major source of metals, which contains both essential and toxic heavy metals over a wide range of concentrations (Arade and Ketema 2023). But, even the essential micronutrients become poisonous to humans as well as animals at concentrations higher than the recommended value resulting from accumulation in the tissues (Gang *et al.*, 2019). Both natural such as volcanic eruption, landslide, weathering, sea salt sprays, forest fire, and anthropogenic activities like mine tailings, industrial emission, dumping wastes, disposal of high metal wastes, from leaded gasoline and paints, application of fertilizer and animal manures, sewage sludge, pesticides, wastewater irrigation, coal combustion residues, run off of terrestrial systems, industrial and domestic effluents, spillage of petrochemicals, accidental leaks, atmospheric deposition have been considered for the release of trace metals into the environment (Gupta *et al.*,2022; Islam *et al.*, 2018). Heavy metal pollution is one of the most serious environmental and human health risks associated with industrial progress. A consequence of disastrous anthropogenic activities such as the discharge of hazardous materials poses a devastating threat to the safety of the environment and subsequently leads to severe cancer for human health worldwide (Fatima *et* *al*., 2020; Abdullahi *et al*., 2022). They are among the major contaminants in the food supply and may be considered the most important problem to our environment that can reduce both the productivity of plants and endanger the safety of plant products such as foods, (Sadi *et al*., 2021). This problem is even getting more attention all over the world, especially in developing countries. Heavy metals, in general, are non-biodegradable, have long biological half-lives, and have the potential for accumulation in the different body organs leading to undesirable side effects (Gupta *et al.*, 2022). The presence of heavy metals may have a negative influence on the quality of vegetables and human health. The processes of plant growth depend on the cycle of nutrients including trace elements, from soil to plant (Abdullahi *et al.*, 2022). Vegetables, especially leafy ones accumulate higher amounts of heavy metals because they absorb these metals in their leaves, hence consumption of vegetables contaminated with heavy metals over a long period of time can seriously diminish some of the essential nutrients in the body and can cause a reduction in immunological defenses, intrauterine growth retardation, impaired physical-social behavior, and disabilities associated with malnutrition (Sadi *et al*., 2021). Carcinogenic, mutagenic, or neurotoxic effects that may be chronic, sub-chronic, and acute have also been testified due to metal poisoning (Geoge *et al.*, 2023*;* Kadir *et al.*, 2008). Some employees were tested having kidney problems (Rai *et al*., 2019). The detrimental effects of heavy metals during pregnancy and fetal development have been widely documented. Heavy metals have the potential to damage the reproductive system of females by causing damage to the ovary, hormone production, and its release (Sankhla *et al.*, 2019). The presence of lead in the body of the host has been linked to preterm birthing, stillbirths, lower birth weightiness, spontaneous abortions, as well as hypertension, whereas, Cd exposure is linked to low birth weight. It has been reported that Pb was detected significantly in cabbage and tomato samples at higher levels above FAO/WHO standard permissible limit, (Bayissa and Gebeyehu 2021; Sadi *et al*., 2023). The consumption of leafy vegetables in Dutsin-Ma town is common. The people living in these areas consume substantial amounts of leafy vegetables. The objectives of this research is to evaluate the concentration and potential health risk of heavy metals in selected vegetables marketed in Dutsin-Ma, Katsina State, Nigeria.

MATERIALS AND METHODS

Study Area: Dutsin-Ma town is on a latitude of 12.4545^0N and a longitude of 7.4977^0E with an altitude of 543.267m. The market is located in Dutsin-Ma Town. It is the administrative headquarters of the Dutsin-Ma Local Government Area Katsina State, Nigeria. (Nura 2019; Abdullahi *et al.*, 2022).

Fig 1: Map of Katsina State, Nigeria showing Dutsin-Ma Local Government Area.
Source: Geography Department Umaru Musa Yar'adua University, Katsina.

Fig 2: Map of Dutsin-Ma Town Showing Wednesday Market Dutsin-Ma where the Samples were collected.

Reagents and Chemicals: All the chemicals and reagents were of analytical grade and were purchased from Sigma Aldrich or Merck (Germany)

Materials: Perkin-Elmer Pinnacle 900 H Atomic Absorption Spectrophotometer (AAS) was used for this analysis. Certified Atomic Absorption Spectroscopic standard stock solutions (1000 mg/L) of Cd, Cu, Fe, Mn, and Pb were prepared using, Cadmium (II) chloride ($CdCl_2$), Copper (II) chloride dihydrate ($CuCl_2.2H_2O$), Iron (II) chloride hexahydrate ($FeCl_2.6H_2O$), Manganese (II) chloride tetrahydrate ($MnCl_2.4H_2O$), and Lead (II) chloride ($PbCl_2$). Working standard solutions of 2, 4, 6, 8, and 10 mg/L were prepared by appropriate dilutions of the stock solution. Deionized water was used in the preparation of all the solutions.

Collection of Sample: Fresh vegetable samples were collected at different locations from the market as the primary sales ground. Samples were collected randomly (Three samples for each vegetable) to estimate the total heavy metal content (Cd, Cu Fe, Mn, and Pb) in the samples.

Preparation and Treatment of the Samples: About 200 g of each of the five edible portions of vegetable samples were used for analysis while damaged or rotten samples were removed. The samples were stored in polythene bags until analysis under refrigerated conditions (<10°C). The samples were thoroughly washed and then dried using the oven-dry method at 105 °C for 48 h to determine the moisture content (Sadi *et al*., 2021). Dried samples were powdered in a manual grinder and were used for heavy metal analysis.

Procedure for Ashing: Powdered samples (3 g each) with three replicates for each vegetable were accurately weighed and placed in a porcelain crucible and two drops of concentrated nitric acid were added to the solid as an ashing aid. The dry ashing process was carried out in a muffle furnace by the stepwise increase of temperature up to 550°C and then left to ash at this temperature for 6 h (Sadi *et al.*, 2021).

Statistical Analysis: In the analysis of the data, IBM SPSS Statistics Software Version 23 was used and the results were expressed as Mean ± Standard deviation (SD). Parametric tests of one-way analysis of variance (ANOVA), a confidence level of 95%, and a significance level of 0.01 were considered in comparing the average concentration of the metals in the vegetable samples.

Health risk assessment: The long-term effects of these heavy metals due to the consumption of vegetables were evaluated by calculating the Estimated Daily Intake of metals (EDI), Hazard index (HI), Target Hazard Quotient (THQ), and Total Target Hazard Quotient (*T*THQ). The carcinogenic and non-carcinogenic health risks were also calculated using the relationship below: AT = Average exposure time for non-carcinogenic effects (ED × 365 days/year); RfDo = Oral reference dose (mg/kg/day). The RfDos are 0.001, 0.04, 0.7, 0.14, and 0.0035 mg kg^{-1} day^{-1} for Cd, Cu, Fe, Mn, and Pb, respectively. To determine the appropriate THQ, it is assumed that all lead ions are inorganic. If the value of THQ is less than the unity, the exposed population is unlikely to experience obvious adverse effects. If THQ is greater than unity there is a potential health risk. The target carcinogenic risk (TCR) factor (lifetime cancer risk), Oral slope factor from the Integrated Risk Information System USEPA database was 8.5×10^{-3} (mg/kg/day)$^{-1}$ for Pb, (Islam *et al*., 2018; Asrade and Ketema 2023).

Table 1: Description of Factors Involved in Health risk Assessment.

Risk exposure factors	Symbols	Values	Units
Vegetable Ingestion Rate	FIR	0.17	Kg/person/day
The concentration of heavy metals	CM		mg/kg/fresh weight
Average body weight	BW	60	Kg for adult
Exposure frequency	EFr	365	Days/year
Exposure duration	ED	70	Years
Average time, carcinogens	AT	25, 550days	Years
Oral Slope factor	SF		(mg/Kg/day)$^{-1}$

$$\text{EDI} = \frac{FIR \times CM}{BW} \dots\dots\dots\dots\dots\dots (1)$$

$$\text{THQ} = \frac{EFr \times ED \times FIR \times CM}{RfD_{\circ} \times BW \times AT} \times 10^{-3} \dots\dots\dots (2)$$

TTHQ ($_{\text{individual vegetable}}$) = THQ $_{\text{metal (1)}}$ + THQ $_{\text{metal (2)}}$ + THQ $_{\text{metal (3)}}$ + ...+ THQ $_{\text{metal (n)}}$ … (3)

HI = TTHQ $_{\text{vegetable (1)}}$ + TTHQ $_{\text{vegetable (2)}}$ + TTHQ $_{\text{vegetable (3)}}$ +.....+TTHQ $_{\text{vegetable (n)}}$ ……….. (4)

$$\text{TR} = \frac{EFr \times ED \times FIR \times CM \times SF}{BW \times AT} \times 10^{-3} \dots\dots\dots.. (5)$$

TCR = CR $_{\text{vegetable (1)}}$ + CR $_{\text{vegetable (2)}}$ + CR $_{\text{vegetable (3)}}$ +.......... +CR $_{\text{vegetable (n)}}$ ……………… (6)

RESULTS AND DISCUSSION

Table 2: shows the percentage of moisture in different samples of vegetables in the range of 80.22 % to 86.78 %. The water content of the fresh samples was in the order Tomato > Onion > Lettuce > Cabbage> Pepper. This shows that, in all the samples analyzed, tomato has the highest amount of water (86.78%) while pepper has the least (80.22%). Table 3 shows the mean concentrations of the metals that were in the range of 0.02 to 0.20, 0.12 to 0.43, 0.60 to 21.77, 0.70 to 3.01, and 0.03 to 0.67 mg/kg for Cd, Cu, Fe, Mn, and Pb respectively.

Table 2: Percentage of moisture in the vegetable samples

Samples	Weight of Fresh Sample (g)	Weight of Dried Sample (g)	Weight of Water (g)	Percentage of Moisture (%)
Cabbage	200	33.15	166.85	83.4
Tomato	200	26.44	173.56	86.78
Lettuce	200	32.49	167.51	83.76
Onion	200	30.82	169.18	84.59
Pepper	200	39.56	160.44	80.22

Table 3: Mean Concentration in the Sample and Standard Deviation

		Mean conc. (mg/kg) ± STD				
Vegetables	Scientific Name	Cd	Cu	Fe	Mn	Pb
Cabbage	*Brassica oleracea*	0.03 ±2.33	0.12 ±0.00	0.60 ±0.5	2.90±0.00	0.36±0.02
Lettuce	*Lactuca sativa*	0.06 ±0.00	0.43 ±0.30	21.77±0.02	2.26±0.02	0.67±0.01
Onion	*Allium cepa*	0.02 ±0.00	0.40 ±0.00	7.00±0.02	3.0 1±0.00	0.40±0.01
Pepper	*Capsicum sp*	0.20 ±0.01	0.20 ±0.01	3.62±0.04	0.70±0.02	0.03±0.00
Tomato	*Lycopersicume sculentum*	0.10 ±0.02	0.33 ±0.00	6.66±0.10	1.67±0.00	0.59±0.07
FAO/WHO		0.30	4.0	42.5	50	0.03

Key: FAO/WHO Maximum permissible limits of the elements in vegetables (mg/kg) dry weight, WHO (2013).

The highest mean concentrations of Cd, Cu, Fe, Mn, and Pb were detected in pepper, lettuce, lettuce, onion, and lettuce respectively, while the lowest mean concentrations of Cd, Cu, Fe, Mn, and Pb were detected in onion, pepper, cabbage, pepper, and pepper. In addition, lettuce was also found to have the highest mean concentration of Fe (21.77 mg/kg) and cabbage has the lowest (0.60 mg/kg). The levels of Pb were in the range of 0.03 to 0.67 mg/kg in Cabbage, lettuce, onion, pepper, and tomato. Lettuce has the highest concentration of lead (0.67 mg/kg), whereas pepper has the least (0.03 mg/kg). In all the vegetables analyzed, except for pepper, the concentrations of lead were greater than WHO/FAO recommended levels of contaminants in vegetables (0.03mg/kg).

Table 4. Estimated Daily Intake (EDI) of trace elements (for the adult population) through consumption of Vegetables in Dutsin-Ma Town.

	EDI values for each heavy metal (mg·day−1 kg−1 body weight)					
Vegetables	Cd	Cu	Fe	Mn	Pb	Total intake
Cabbage	8.50E-5	3.40E-4	1.70E-3	8.20E-3	1.00E-3	1.13E-2
Lettuce	1.70E-4	1.20E-3	6.20E-2	6.40E-3	1.90E-3	7.17E-2
Onion	5.60E-5	1.10E-3	2.00E-2	8.50E-3	1.10E-3	3.08E-2
Pepper	5.70E-4	5.70E-4	1.00E-2	2.00E-3	8.50E-5	1.27E-2
Tomato	2.80E-4	9.40E-4	1.90E-2	4.60E-3	1.70E-3	2.65E-2
EDI from all vegetables	1.16E-3	4.15E-3	1.13E-1	2.97E-2	5.79E-3	
MTDI	2.10E-02	4.00E-2	7.0E-1	(2.0 to 5.0) E+0.0	2.1E-01	

MTDI = Maximum tolerable daily intake; EDI values for each heavy metal (mg·day^{-1} kg^{-1} body weight)

Human health risk assessment

Estimated daily intake of heavy metals (EDI): Table 4. Showed the result of the estimated daily intake of heavy metals in the vegetables analyzed. The EDI values of heavy metals through the ingestion of different vegetables revealed that the mean values of total EDI of individual heavy metals from the consumption of all analyzed vegetables were 1.16×10^{-3}, 4.15×10^{-3}, 1.13×10^{-1}, 2.97×10^{-2}, and 5.79×10^{-3} for Cd, Cu, Fe, Mn, and Pb respectively which were less than the maximum tolerable daily intake (MTDI) as shown in Table 4. A similar result was reported by (Islam *et al*., 2018).

Table 5: THQ values of individual heavy metals through the consumption of different vegetables in the study area.

Vegetables	Target Hazard Quotient (THQ) values of each heavy metal for adults					
	Cd	Cu	Fe	Mn	Pb	∑THQ
Cabbage	2.55E-4	2.55E-5	7.28E-3	1.76E-4	8.74E-4	8.61E-3
Lettuce	5.10E-4	9.10E-5	2.64E-4	1.37E-4	1.63E-3	2.63E-3
Onion	1.70E-4	8.50E-5	8.5E-5	8.5E-3	9.72E-4	9.81E-3
Pepper	1.7E-3	4.25E-5	4.39E-5	1.83E-4	7.29E-5	2.04E-3
Tomato	8.5E-4	7.01E-5	8.08E-5	1.01E-4	1.43E-3	2.53E-3
∑TTHQ	3.49E-3	3.14E-4	7.75E-3	9.10E-3	4.98E-3	2.56E-2
RfDo mg/kg/day	0.001	0.04	7.0E-1	1.4E-1	3.5E-3	

Non-carcinogenic health risk assessment: Table 5. Showed the result of the target hazard quotient (THQ) of heavy metals in the vegetables analyzed. The estimated THQ for non-carcinogenic risk of heavy metals through five evaluated vegetable ingestion for adult inhabitants is presented in Table 5. The results showed that the THQ of all the metals in cabbage, lettuce, onion, pepper, and tomato was less than 1 indicating that if people consume these types of vegetables in their diet, they might not be at risk. The descending order of TTHQ for vegetable samples was in the order of Onion>lettuce>cabbage>pepper>tomato. Total THQ of individual metals from the consumption of all vegetables contributed 52.00 and 28.49, 18.88, 0.44, and 0.19%, Mn, Pb, Cd, Fe, and Cu respectively.

Table 6. Target carcinogenic risks (TCRs) assessment through vegetable consumption for Pb to the population in the study area.

Command name	**Pb**
Cabbage	8.67E-9 ± 4.82E-10
Lettuce	1.61E-8 ± 2.40E-10
Onion	9.63E-9 ± 2.40E-10
Pepper	7.23E-10 ± 0.00E-0
Tomato	1.42E-8 ± 1.69E-9
TCRs	4.93E-8 ± 2.65E-9

*Note: *Target cancer risks (Mean ± SD), **Total cancer risk (Mean)*

Carcinogenic health risk assessment: Table 6. Showed the result of the target carcinogenic risks (TCRs) assessment through the consumption of vegetables analyzed in the study area. The TCRs of Pb ranged from 7.23×10^{-10} in pepper to 1.42×10^{-8} in tomato. However, the total value of TCRs was 4.93×10^{-8} for Pb which is less than the acceptable risk limit (10^{-6}), indicating that the inhabitants consuming these vegetables are not exposed to Pb with a lifetime cancer risk. Therefore, based on the results of the present study, the potential health risk for the local inhabitants due to heavy metal exposure through the consumption of vegetables should be ignored.

Conclusions: The results revealed that the concentrations of Pb in all the vegetable samples were greater than the standard permissible limit, and all other heavy metals were within the safety tolerable limits for human consumption. The THQ and HI results showed that the consumption of vegetables may not result in adverse non-carcinogenic health risks to the consumers. The results also showed that Pb in vegetables might not have exerted lifetime carcinogenic health risks to consumers. Therefore, this study suggests the regular analysis and checking of the heavy metals present in vegetables, irrigated water, and foodstuff to avoid extreme accumulation in the food chain and hence get away from human health risks.

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