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Potential Health Risk of Heavy Metal Concentration in Selected Medicinal Plants in Jos Metropolis

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Abstract

Plants are the primary pathway for the transfer of heavy metals from polluted soil to humans. The purpose of this study is to ascertain the concentrations of copper, iron, lead, cadmium, and manganese in some medicinal plants (lemon grass, bitter leaves, mint leaves, and moringa leaves) in Jos metropolis using Atomic Absorption Spectrophotometer (AAS) and the possible health risk by assessing the estimated daily intake, the hazard quotient, hazard index and the carcinogenic risk based on the United States Environmental Protection Agency (USEPA) model for a period of six months. Non-carcinogenic risk assessment of ingesting medicinal plants for children and adults was also established. The results were compared with the WHO permissible limits for heavy metals in medicinal plants. The findings demonstrated that all the heavy metals were detected except Cadmium in the two locations. The concentration of Pb and Fe in both locations exceeded the WHO permissible limit. The concentration of Cu in both locations in lemon grass was within the WHO permissible limit and Mn was consistently within the WHO permissible limit in the two locations. The Estimated Daily Intake (EDI) values for adults were higher than those for children in all the samples. The Hazard Quotient (HQ) values for Pb was below 1. Fe, Mn and Cu were above 1 in all the samples for adults and children except for lemon grass which recorded less than 1in both adult and children for Fe and Cu. Hazard Index (HI) values were greater than 1. Calculated cancer risks associated with Pb for both adults and children was 10^{-3}. From the result of this findings, medicinal plants should be taken with moderation as frequent and prolong usage may be injurious to health.

Keywords: *AAS, Concentration, Health, Heavy metal, Medicinal plants, Risk, WHO.*

Introduction In most countries, the use of herbal treatments has increased in recent decades due to the widespread belief that "natural" equates to "harmless" (Zigau, Aliyu, Abubakar, Bello, Yakasai, and Sanusi, 2024; Mafulul, Joel, and Gushit, 2024). It may not necessarily be true, especially when the plant components utilized in the preparation of the herbal remedies are polluted. However, the safety of herbal products has grown to be a significant public health concern as their market share and popularity have expanded globally (WHO, 2019). The sources of heavy metal contamination in the environment are generally from anthropogenic activities, including mining, smelting procedures, chemical industry, traffic, agriculture, industrial effluents, etc (Nedjimi, 2021). Heavy metals can accumulate in the food chains and present toxic effects to humans and other living beings, even at very low concentrations (Omotehinse and Ako, 2019). Some heavy metals such as copper (Cu), iron (Fe) manganese (Mn), nickel (Ni), and zinc (Zn) are required by plants in trace levels and are necessary for life, excessive consumption of these metals can have disastrous consequences, such as toxic and cancer causing effects (Magelsir, 2016). They are also linked to the development of a number of acute and chronic illnesses, including hypertension, atherosclerotic disease, cardiovascular disease, renal failure, infertility, hyperglycemia, diabetes, inflammation, autoimmune diseases, prostate dysfunction, osteomalacia, and osteoporosis disease (Martins, Carneiro, Grotto, Adeyemi, and Barbosa, 2018; Moreno, Navas Acien, Escolar, Nathan, Newman, 2018).

Though studies now show that exposure of heavy metals through general dietary consumption contribute negatively to human health (Wei and Cen, 2020). However, an increase in their intake above certain permissible limit can become toxic (Henry, Ogenyi, Henry, and Dogun, 2019). Heavy metals have toxic risks in humans and cause a lot of serious environmental damage around the world. Major pathways of human exposure to heavy metals are direct ingestion of food, water and beverage (Al-keriawy, Nehaba and Alwan, 2023; Ayodeji and Olorunsola, 2011). Health risk assessment of heavy metals in contaminated vegetables and plants is being carried out in developed countries; however, little is being done in developing countries (Kohzadi, Shahmoradi, Loqmani and Malek, 2018). The health risks associated with heavy metal contamination has led to the establishment of quality and safety standards for herbal remedies that stipulate the maximum allowable concentrations for heavy metals in herbal remedies (USEPA, 2011). The World Health Organization (WHO) and various regulatory agencies and institutes have introduced guidelines for the permissible limits of heavy metals in so-called consumable herbal plants (Mafulul et al., 2024). Although there are concerns about some of these standards not being based on scientific research on herbal medicines, they provide a reference value based on which a plant or plant parts can beused as (or as part of) traditional remedy. Trends are toward increased use of traditional and complementary medicine due to various cultural reasons, and there are growing concerns about the use of herbal plants because of the lack of adequate scientific research and absence of control by corresponding organizations (Zigauetal., 2024). The purpose of this study is to ascertain the concentrations of copper, iron, lead, cadmium, and manganese in a few commonly used herbs (lemon grass, bitter leaves, mint leaves, and moringa leaves) sold in Jos metropolis and the possible health risk by assessing the estimated daily intake, the hazard quotient, hazard index and the carcinogenic risk based on the United States Environmental Protection Agency (USEPA) model.

Materials and Methods: Collection of Medicinal Plant Samples: Leaf samples of selected herbal plants (lemon grass, bitter leaves, mint leaves, and moringa leaves) were sourced commercially from herb sellers in Jos metropolis. Each sample was purchased separately from different selling points in the month of April through May, 2023. The medicinal plant sample was packed in sterilised polyethylene bags each and transported to the laboratory where it was taken to the herbarium in Federal College of Forestry, Jos for identification. Exactly 20 g of each leaf sample was thoroughly washed with distilled water to remove dust particles and subsequently oven-dried to constant weight at 40 °C. Each dried sample was ground to powder using a pestle and mortar and sieved through a 2 mm sieve. The powdered samples were kept in clean polyethylene bags at room temperature for further analysis.

Digestion of The Samples: Plant samples were digested according to VG 101 analytical package method and protocol described by Bureau Veritas Mineral Laboratories (former ACME Analytical Laboratories), Vancouver, Canada (Cecconi, Incerti, Capozzi, Adamo, Bargagli, Benesperi, Candotto, Favero-Longo, Giordano, Puntillo, Ravera, Spagnuolo and Tretiach, 2019). 1.0 g of each previously pulverized plant sample was cold leached with 2 ml nitric acid (HNO_3) and subsequently digested in a hot water bath for 1 hour. After cooling to room temperature, 6 ml of a modified Aqua Regia solution of equal parts 2:2:2 of concentrated HCl, HNO_3, and distilled H_2O was added to each sample and allowed to leach on a heating block of hot water bath at 95 °C for 1 hour. Samples were later made up to 20 ml mark with dilute HCl and filtered (Cecconi *et al*., 2019). Atomic Absorption Spectrophotometer (AA-6800) was used determine heavy metals concentration in the herbal plant samples. Duplicate determinations were made.

Human Health Risk Assessment: Human health risk assessment was used to estimate the health effects that might result from exposure to noncarcinogenic and carcinogenic chemicals (Zheng, Zhang, Li, Sun, and Zhong, 2020., USEPA, 2015., Kamunda, Mathuthu, and Madhuku, 2016). Human health risk upon ingestion of contaminated medicinal plants was calculated based on the model developed by USEPA, and the values used for specific variables were adapted for Nigerian population statistics (Khan, Malik and Muhammad, 2013, USEPA, 2011).

Estimated Daily Intake of Heavy Metals : To appraise the health risk associated with heavy metal contamination for the studied medicinal plants' samples, the estimated daily intake (EDI) (mg/kg/day) of heavy metals through ingestion of contaminated medicinal plants was undertaken based on a daily dose for each heavy metal using equation 1:

$$EDIplant\ Ingestion = \frac{CxIRxEDxEF}{BWxAT} \quad \text{-------------------------- 1}$$

Where C is the concentration of heavy metal in the medicinal plant sample, while the other terms in the equation are ingestion rate (IR), exposure duration (ED), exposure frequency (EF), body weight (BW), and average time (AT), respectively, the IR for average daily ingestion of medicinal plants for adults and children was estimated to be 0.02 kg person-1day-1 and 0.01 kg person-1 day-1 respectively (USEPA, 2011; Tschinkel *et al*; 2020); the ED for children and adults were estimated to be 6 and 30 years respectively (Kamunda *et al*., 2016; Tschinkel *et al*., 2020); the EF for adults and children was 350 days (USEPA, 2015., Kamunda *et al*., 2016]; the AT for non-carcinogenic risk for adults and children was taken as the product of 365 days and the respective ED of adults and children while the carcinogenic risk was taken as 365 days X 70 years of age for both adults and children (Tschinkel, Melo, Pereira, Silva, Arakaki, Lima, Fernandes, Leite, Melo, Melnikov, Espindola, De Souza, Nascimento, Júnior, Geronimo, Dos Reis, and Nascimento, 2020), and the BW was 15 and 70 Kg for children and adults respectively (Kamunda *et al*., 2016; Tschinkel *et al*., 2020).

Hazard Quotient (HQ): The non-carcinogenic health risk associated with heavy metals exposure through the ingestion of medicinal plants was evaluated for each heavy metal determined in this study using the hazard quotient (HQ) which was obtained by dividing the EDI of each heavy metal via the oral exposure route to its corresponding reference exposure dose (R_fD) (USEPA,1989; Kumar *et al*., 2019).

using equation 2:

$$HQ = \frac{EDI}{R_fD} \quad \text{--------------------------- 2}$$

The values of R_fD maximum permissible oral dose 0.001 for Cd, 0.040 for Cu, 0.02 for Mn, 0.0035 for Pb and 0.7 for Fe mg·kg-1·day-1 respectively for the heavy metals analysed were adopted from Integrated Risk Information System (Kamunda *et al*., 2020; USEPA, 2010). Where HQ is less than 1 it is concluded there is no obvious risk from the heavy metal over a lifetime exposure, however, a value greater than 1, it is considered the presence of heavy metal may produce an adverse effect over a lifetime exposure. The higher the HQ value, the higher the probability of experiencing long-term carcinogenic effects (Kamunda *et al*; 2016; USEPA, 2011; Song, Chen, Zheng, Xie, Li, and Gao, 2009).

Hazard Index: To evaluate the overall potential for non-carcinogenic risk to human health through more than one heavy metal in the medicinal plant samples, the hazard index (HI), the sum of the hazard quotients for all the individual heavy metals determined, was calculated for each medicinal plant (USEPA,1989; Song *et al*, 2009). This was done using equation 3.

$$HI = \sum HQPb + HQFe + HQCu + HQMn \text{ ------------------------ } 3$$

Where the result for HI is less than 1 it shows that, it there is no carcinogenic risk to human health whereas HI is more than 1 predict cancerous risks are likely to occur.

Carcinogenic Health Risk: Cancer health risk estimates represent the incremental likelihood that an individual will develop cancer as a result of specific exposure to a carcinogenic chemical over a lifetime (USEPA; 1989). The CR of each of the carcinogenic element (Pb) was calculated by multiplying the EDI values for the ingestion of medicinal plants by the cancer slope factor (CSF) for the PTE via the ingestion route using equation 4 USEPA; 1989; Alsafran, Usman, Rizwan, Ahmed, and Jabri, 2021; Masri, Lebrón, Lebrón, Logue, Valencia, Ruiz, Reyes, and Wu, 2021).

$$CR = EDI \times CSF \ldots\ldots\ldots\ldots\ldots\ldots 4$$

where EDI (mg/kg/day) = estimated daily intake averaged over 70 years and CSF = Cancer slope factor (mg/kg/day). The cancer slope factors used for CR calculation was 0.0085 (mg/kg/day) for Pb [20]. According to the New York State Department of Health (NYSDOH), CR values $\leq 10^{-6}$, indicate low cancer-causing risks, between 10^{-5} and 10^{-4}, indicate moderate cancer-causing risks, and between 10^{-3} and 10^{-1}indicate high cancer-causing risks (Ashraf *et al*; 2021; Alsafran *et al*; 2021).

Statistical Analysis: The results were analysed statistically using Microsoft Office Excel and the Statistical Package for Social Science (SPSS 23.0 for Windows, SPSS Inc., IL, USA). One-way analysis of variance (ANOVA) followed by Tukey-Kramer's Multiple Comparison was used to assess the variation in concentration of heavy metals in the herbal plants studied. Results were expressed as mean ± standard deviation and possibilities less than ($p < 0.05$) were considered to be statistically significant.

Results Heavy Metal Concentration

TABLE 1: Heavy metal Concentration (mg/kg) in Plant Samples (herbs) Sampled from Jos North L.G.A.

Plants/Elements	Moringa Leaf (mg/Kg)	Bitter Leaf (mg/Kg)	Mint Leaf (mg/Kg)	Lemon Grass (mg/Kg)	WHO Limits (mg/Kg)
Pb	44.11 ± 0.25^a	22.00 ± 0.04^b	20.03 ± 0.03^b	11.18 ± 0.23^c	10
Fe	550.00 ± 0.08^a	150.00 ± 0.86^c	342.00 ± 0.13^b	72 ± 0.20^d	20
Cu	23.00 ± 0.30^a	12.16^b	5.94 ± 0.83^c	1.02 ± 0.01^d	10
Mn	50.60 ± 0.12^b	44.23 ± 0.05^b	115.31 ± 0.31^a	18.35 ± 0.38^c	200
Cd	N.D	N.D	N.D	N.D	-

Means are expressed as mean±standard deviation of three replications
ND = Not Detected

Table 2: Heavy Metal Concentration (mg/kg) in Plant Samples (herbs) Sampled from Jos South L. G A.

Plants/Elements	Moringa Leaf (mg/Kg)	Bitter Leaf (mg/Kg)	Mint Leaf (mg/Kg)	Lemon Grass (mg/Kg)	WHO Limits (mg/Kg)
Pb	17.32 ± 0.01^b	13.72 ± 0.18^{bc}	77.31 ± 0.17^a	13.11 ± 0.02^{bc}	10
Fe	583 ± 0.02^a	122.00 ± 0.09^b	602.00 ± 0.18^a	53.91 ± 0.16^c	20
Cu	13.40 ± 0.04^a	13.02 ± 0.05^a	11.32 ± 0.01^a	1.12 ± 0.05^b	10

Mn	23.83±0.12[c]	84.25 ±0.07[b]	101.31±0.19[a]	19.31±0.11[cd]	200
Cd	N.D	N.D	N.D	N.D	-

Means are expressed as mean±standard deviation of three replications
ND = Not Detected

Health Risk Assessment Estimated Daily Intake (EDI) of Heavy Metals

Table 3: Estimated daily intake (EDI) values (mg/Kg/Day) of eight heavy metals through consumption of medicinal plants sold in Jos North L.G.A. via the ingestion route of exposure for adults and children

Plant/Element	Moringa Leaf		Bitter Leaf		Mint Leaf		Lemon Grass	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Pb	0.370	0.170	0.180	0.08	0.164	0.08	0.09	0.04
Fe	4.540	2.120	1.230	0.580	2.810	1.310	0.09	0.05
Cu	0.190	0.090	0.100	0.050	0.050	0.020	0.010	0.003
Mn	0.410	0.190	0.360	0.170	0.950	0.440	0.150	0.070

Table 4: Estimated daily intake (EDI) values (mg/Kg/Day) of eight heavy metals through consumption of medicinal plants sold in Jos South L.G.A. via the ingestion route of exposure for adults and children

Plant/Element	Moringa Leaf		Bitter Leaf		Mint Leaf		Lemon Grass	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Pb	0.142	0.066	0.112	0.052	0.635	0.296	0.107	0.050
Fe	4.791	2.236	1.002	0.467	4.950	2.309	0.443	0.206
Cu	0.110	0.051	0.107	0.049	0.093	0.043	0.009	0.004
Mn	0.196	0.091	0.692	0.323	0.832	0.388	0.158	0.074

Table 5: Hazard Quotient (HQ) Values of Heavy Metals In Jos North Medicinal Plants

Plant/Element	Moringa Leaf		Bitter Leaf		Mint Leaf		Lemon Grass	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Pb	0.360	0.169	0.179	0.079	0.163	0.079	0.089	0.039
Fe	6.485	3.028	1.757	0.828	4.014	1.871	0.120	0.071

Cu	4.750	2.250	2.500	1.250	1.250	0.500	0.250	0.075
Mn	20.500	9.500	18.000	8.500	47.500	22.00	7.500	3.500

Table 5: Table 5: Hazard Quotient (HQ) Values of Heavy Metals In Jos South Medicinal Plants

Plant/ Element	Moringa Leaf		Bitter Leaf		Mint Leaf		Lemon Grass	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Pb	0.141	0.066	0.112	0.051	0.632	0.294	0.106	0.049
Fe	6.844	3.194	1.431	0.667	7.071	3.298	0.632	0.012
Cu	2.750	1.275	2.675	1.225	2.325	1.075	0.225	0.100
Mn	9.800	4.550	34.600	16.150	41.600	19.400	7.900	3.700

Discussion: Heavy metals have been shown to be harmful, even at low concentrations, when ingested over a long time period, and their health effects may not be immediate but show up after many years due to their bioaccumulation tendency (Tabrez, Zughaibi and Javed, 2021). Hence, chronic intake of such heavy metals in the form of medicinal plants could potentially lead to their toxicity, which might affect the nervous system and cause neurological disorders. Based on the results of this study, all the samples in both locations were found to have heavy metals (Pb, Mn, Cu, and Mn) at different concentrations, indicating that the medicinal plants have the potential to pose health risk if consumed regularly. This is in tandem with previous work on heavy metal content in medicinal plants and showed that the heavy metals were also detected at varying concentration (Mafulul *et al*., 2024; Henry *et al*., 2019). In this study, the concentration of Pb, Fe and Cu were higher in both study sites than the reported WHO maximum permissible limits established for medicinal plants, just as previous studies have reported such high levels of essential metals in medicinal plants (Figas *et al*., 2021; Henry *et al*., 2019). Mn had concentrations that was within the WHO permissible limit for medicinal plants in both locations in this study. This was also observed in similar studies on medicinal plants but with higher concentrations (Mafulul *et al*., 2024; Dghaim *et al*., 2015; Ababneh, 2017).

Pb recorded the highest concentration value in moringa leaves and least in lemon grass in Jos North L. G.A while mint leaves recorded the highest in Jos North and South L.G.A. Moringa and mint leaves recorded the highest concentration of the heavy metals studied. This pattern was also observed in a similar study (Henry *et al.*, 2019). Pb interferes with a variety of metabolic processes, including calcium metabolism and protein reactions. Higher levels of lead exposure increase ROS levels while decreasing the levels of antioxidant (Naveed *et al*., 2022; Brochin *et al*., 2014). It was observed that, lemongrass has a low affinity for heavy metal uptake while mint plant showed a high affinity for heavy metal uptake. This may be due to several factors like soil type, type of activities in the area, the atmospheric condition of the area and the type of plant (Henry *et al*., 2019).

Fe is needed by the body and many biological systems. Fe plays an important role in sustaining aerobic life on earth but excessive consumption has been linked to accidental death in children aged below 6 years (Naveed *et al*., 2022). In this study Fe had the highest concentration of all the metals analyzed in the two study sites. The highest concentration of Fe was recorded in mint leaves in Jos South LGA and Fe in all the studied medicinal plants exceeded the WHO permissible limit for Fe in medicinal plants. Cu deficiency can cause extreme tiredness, lightened patches of skin, high levels of cholesterol in the blood, weak and brittle bones, loss of balance and coordination. Consumption of excess Cu can cause liver damage, abdominal pain, cramps, nausea, diarrhea and vomiting (Naveed et al; 2022). In this study, Cu concentration in the medicinal plants studied exceeded the WHO permissible limits except for lemon grass in both locations. This was also observed in similar work in literature with Cu having concentrations that exceeded WHO standard in medicinal plants (Mafulul *et al*., 2024; Figas, Tomaszewska-Sowa, Kobierski, Sawilska, and

Klimkowska, 2021;) In the environment, Cd is toxic to both plants and animals, with half life between 25-30 years, and can accumulate in the liver, kidney, and bones. Exposure occurs primarily via ingestion of contaminated food and water (Mafulul *et al*., 2024). In this study, Cd was not detected in all the sampled medicinal plants in the two study sites. The population's health risk within the study was assessed based on the United States Environmental Protection Agency (USEPA) model, where a noncarcinogenic risk assessment of ingesting medicinal plants for children and adults in the study was established based on HQ and HI values.

EDI values of all the heavy metals in the medicinal plants studied were less than one except for Fe which was above one. The EDI values for adults were generally higher than those for children in all the medicinal plants. This was also reported in a similar work where the EDI values were also less than one (Mafulul *et al*., 2024). The results showed that the HQ values for Pb was below one in all the medicinal plants studied. The results also showed HQ values were greater than one for Fe and Cu in all the medicinal plants for adults and children except for lemon grass which recorded less than one in both adult and children in the two study sites. Mn values were also greater than one for adults and children in all the medicinal plants studied in both sites. This implies that consumption of these medicinal plants regularly can pose health threat as HQ, less than 1, is concluded to pose no obvious risk from the heavy metal over a lifetime exposure, however, a value greater than 1, is considered that the presence of heavy metal may produce an adverse effect over a lifetime exposure. The higher the HQ value, the higher the probability of experiencing long-term carcinogenic effects (Kamunda *et al*., 2016; USEPA, 2011; Song *et al*., 2009). This study shows that HI values for the heavy metals were greater than 1, indicating a possible health risk associated with each heavy metal for children and adults through the consumption of the studied medicinal plants (Bhatti *et al*., 2020) The high HI values observed suggest longer use of the plant product might present possible noncarcinogenic health risks to the consumers even at lower concentrations. (Mafulul *et al*., 2024). The results of the calculated cancer risks associated with Pb through consumption of the medicinal plants investigation for both adults and children was 10^{-3}, indicating high carcinogenic risk. Similar studies previously done showed CR calculated values for Ni, Cd, Pb in medicinal plants for adults and children fall within the range of 10^{-5} to 10^{-4} (Mafulul *et al*., 2024).

Conclusion: This study assessed the level of the concentration of heavy metals in medicinal plants obtained in two sites in Jos metropolis and their associated health risk. The results of the heavy metals analysis were compared with the WHO permissible limits for heavy metals in medicinal plants. The findings demonstrated that copper, iron, lead and manganese were detected in the medicinal plant samples. Cadmium was not detected in all the plant samples in the two locations. The concentration of Pb and Fe in both locations exceeded the WHO permissible limit for heavy metal concentration in medicinal plant. The concentration of Cu in both locations in lemon grass was within the WHO permissible limit for heavy metals in plants and Mn was consistently within the WHO permissible limits in all the plant samples from the two study locations. The Estimated Daily Intake (EDI) values for adults were generally higher than those for children in all the medicinal plants studied. The Hazard Quotient (HQ) values for Pb is below 1 while Fe, Mn and Cu were above 1 in all the medicinal plants studied for adults and children except for lemon grass which recorded less than one in both adult and children in the two study sites for Fe and Cu. Hazard Index (HI) values for the heavy metals were greater than 1, suggesting that longer use of the plant product might present possible non-carcinogenic health risks. Calculated cancer risks associated with Pb through consumption of the medicinal plants investigated for both adults and children was 10^{-3}, indicating high carcinogenic risk.

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