UJMR, Conference Special Issue Vol. 9 No. 3. June, 2024, pp.207 - 213

E-ISSN: 2814 – 1822; P-ISSN: 2616 – 0668

https://doi.org/10.47430/ujmr.2493.025

Received: 18th February, 2024 **Accepted:** 17th June, 2024

Determination of Heavy Metal Concentrations on some Selected Herbal Medicinal Preparations Marketed in Kano State, Nigeria

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Abstract

Herbs are extensively consumed in Nigeria, for their medicinal properties and availability. This study aimed at determining the concentration of heavy metals residues in some selected traditional medicinal herbs consumed in Kano State, Nigeria. This is to assess their relative safety and potential health risks to local inhabitants based on the World Health Organization standard limit. A total of ten (10) powdered samples of medicinal preparations were purchased from local markets in Kano metropolis and analyzed for the presence of lead, chromium, cadmium, and mercury contents. The samples underwent digestion, and heavy metal concentrations were determined using Atomic Absorption Spectrometry (AAS). The metals found to be present varied in different concentrations in the herbal samples. The presence of heavy metal ranges as follows: 0.6-6.5mg/kg for chromium, 3.10-22.10mg/kg for lead, 0.08-0.60mg/kg for cadmium, and 0.09-0.30mg/kg for mercury. However, mercury content was not detected in some samples. The study's findings conclude that the lead (Pb) values were found to be the highest in concentration, exceeding the W.H.O limits in sample J, which is used as a remedy for cough. This indicates that the sample is highly contaminated compared to others. Mercury was not detected in four of the samples, and when present, it was below the limits set by the World Health Organization, making all the samples free from mercury contamination.

Key words: Traditional medicine, heavy metals, permissible limits and contamination.

INTRODUCTION

Herbal remedies are utilized as supplementary treatments for a variety of illnesses and injuries, all derived from plant materials. These remedies are prepared in forms such as infusions, decoctions, tinctures, and water baths using leaves, roots, tree bark, fruits, soil, and animal products (Sulayman *et al.*, 2015). It is estimated that over 80% of individuals in Africa rely on complementary and alternative medicine to address conditions like malaria, diabetes, ulcers, cough, asthma, and other ailments (Muwanga, 2017). A significant portion of Nigerians (80%) depend on herbal remedies, which are increasingly accessible in the local markets (Sanda, 2015). These traditional medicines are subject to various market environments with high levels of pollution due to the continuous demand for natural remedies, exposing them to multiple risks (Rausan *et al.*, 2019).

Due to the current rate of environmental pollution, reports of heavy metal contamination in our soils, cities, wetlands, and rivers have been made (Daniel *et al.*, 2016). Using medicinal herbs to heal diseases is perhaps the oldest known method of treating ailments; as a result, phytotherapy is now a part of all conventional medical systems and is often the main form of treatment in low- and middle-income countries (Sulayman *et al.*, 2015). The popular notion that "natural" equals "harmless" has led to the rise in the usage of herbal remedies in affluent nations in recent decades. However, as their popularity and global market have increased, the security

of herbal products has become a major public health problem (WHO, 2019). Low-quality herbal goods might cause negative reactions due to regulation and unrestricted distribution channels, especially online sales. The absence of hazardous metal pollution, the ensuing health risks, and the impact on the ecology continue to be major concerns. While certain heavy metals are beneficial to human health in tiny amounts, their presence above specific thresholds can be harmful and result in acute or chronic poisoning (Sanda, 2015). Factors such as geography, soil characteristics, and the presence of toxins in soil, water, and air, along with other variables like transit and storage conditions, can significantly affect the quality of these herbal treatments (Saad *et al.*, 2020).

In recent years, there has been much discussion about the detrimental effects of trace metals on the environment and human health. The primary pathway through which heavy metals are transferred from contaminated soil to humans is through plants.

In the food chain, heavy metals have a tendency to accumulate. Even at very low concentrations, they can have harmful effects on humans due to their low renal excretion rates (Abdullahi, 2019). Lead, arsenic, cadmium, and mercury are among the heavy metals known for their negative side effects. Even in extremely low concentrations, these metals can be hazardous and potentially lead to mutations. The overconsumption of dietary heavy metals has been linked to various health issues, including weakened immune systems, impaired neurological and psychosocial functions, fetal malformations, cardiac malfunctions, gastrointestinal cancer, and more (Singh *et al.*, 2021). This study aims to determine the level of heavy metal contamination in some commonly consumed herbs to evaluate their relative safety and potential health risks based on the World Health Organization (WHO) standard limits.

MATERIALS AND METHODS

Study area

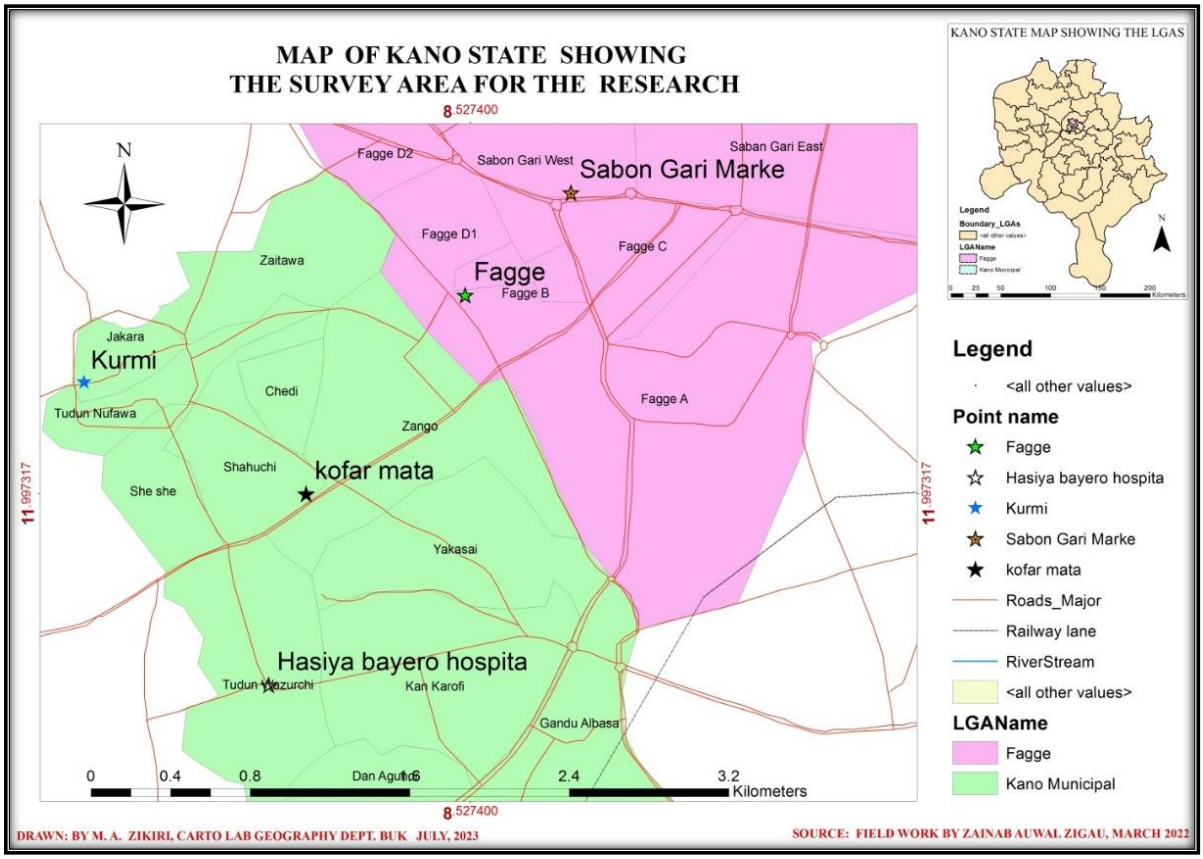

Figure 1: A Google map of Kano State showing the survey areas (markets) where samples were collected for the research.

Collection of Samples

A total of ten (10) finished powdered herbal preparations were purchased from five different sources in the markets of Kano State Metropolis. The herbal samples used in this study were obtained from shops and roadside sellers. Table 1 presents the composition of herbal formulations and their scientific names, the ailments treated by the herbal preparations, and the plant parts used in the study. Powdered samples were collected and stored in plastic bags for metal analysis.

Table 1:The composition of plants, ailments treated, and plant parts of powdered HMPs.

S/N	Samples	Ailments treated	Whole plant Composition	Plant Parts
1	A	Typhoid	*Vitex Doniana*	Stem
			Prospis Africana	Leaves
			Senna italic	Stem
2	B	Pile	*Detarium microcarpum*	Fruits
			Diospyrous misipiliformis	leaves
3	C	Ulcer	*Carica Papaya,*	Leaves
			Anogeissus leiocarpus	Stem
4	D	High BP	*Vitex Doniana,*	Stem
			Combretum micranthum	Leaves
5	E	Malaria	*Cryptolepis Sanguinolenta*	Stem
			Terminalia ivorensis	leaves
6	F	Diarrhea	*Acacia nilotica,*	Leaves
			Vitex doniana	leaves
			Parkia biglobosa	Stem
7	G	Pain reliever	*Cassia abida,*	Stem
			Ipomea asarifolia	Stem
8	H	Gonorrhea	*Curcuma longa*	Roots
			Amaranthus spinosus	Leaves
9	I	General infections	*Jatropha curcas*	Leaves
			Garcinia kola	Stem
10	J	Cough	*Anogeissus leiocarpus*	Stem
			Allium sativum	Fruits

Heavy Metal Analysis

The herbal samples were digested following the procedure outlined by Street *et al.*, (2020). A mixture of 10ml of nitric acid (HNO_3), hydrochloric acid (HCL), and H_2O_2 was added to 1g of each homogenized powdered plant material in a borosilicate flask. The mixture was heated to 120°C on a hotplate for three hours. After digestion, the colorless, transparent solution was transferred to a 50 ml volumetric flask. Each digestion tube was then rinsed thoroughly with distilled water to eliminate any potential residue before transferring it to a volumetric flask filled with distilled water.

Prior to analysis, the diluted samples were stored in 100 ml high-density polyethylene plastic bottles. Each herbal sample was analyzed three times to ensure the accuracy of the results. Standard solutions were prepared from the stock solutions to minimize the matrix effect. Concentration standards for each metal were prepared using the same reagents and solvents used in sample preparation. The absorbance of Pb, cu, Hg, and Cd was determined using an atomic absorption spectrometer (AAS) and measured in terms of peak area. Stringent quality assurance measures were implemented to validate the reliability of the data.

Samples were collected with care and stored in plastic bags. All glassware was rinsed with distilled water before use and soaked in 10% HNO_3 for a whole day to minimize contamination risks. To validate the accuracy of the instrument, control solutions containing known concentrations of the target elements were analyzed in between sample runs.

Statistical Analysis

Three sets of measurements were made for each heavy metal, and the results were reported as mean ± standard deviation (SD).

RESULT

The results of heavy metals analysis in the selected samples are presented in Table 2.

Results of the analysis of heavy metals in the ten (10) powdered herbal medicinal preparations (HMP's) revealed the presence of cadmium, chromium, lead, and mercury in varying quantities. The highest contamination of lead (Pb) was observed in sample J, with approximately 22.0 mg/kg, while the lowest contamination was found in sample C, with around 0.09 mg/kg of mercury. It was noted that the samples analyzed were free from mercury contamination.

Table 2: Heavy metal concentrations (mean in mg/kg) of powdered herbal medicinal samples sold at some markets of Kano State.

Powdered samples	Chromium (*ch*) Mean± SD	Lead (*Pb*) Mean± SD	Cadmium (*Cd*) *Mean± SD*	Mercury (*Hg*) Mean± SD
A	1.2±0.15	3.60±0.55	0.08±0.01	ND
B	3.7±0.14	4.60±0.85	0.06±0.01	ND
C	0.6±0.15	3.10±0.12	0.03±0.00	0.09±0.01
D	3.5±0.07	7.20±0.25	0.40±0.26	0.21±0.02
E	3.1±0.45	10.2±0.31	0.52±0.02	0.10±0.16
F	2.1±0.12	14.1±0.20	0.21±0.01	ND
G	3.2±0.02	15.7±0.31	0.46±0.03	0.2±0.07
H	4.2±0.07	17.1±0.54	0.60±0.01	0.3±0.33
I	4.7±0.13	19.5±0.95	0.09±0.50	0.1±0.02
J	6.5±0.10	22.0±1.40	0.10±0.03	ND
WHO limits(mg/kg)	**	10.0	0.3	1.0

Key; ND not detected ** No acceptable limit by W.H.O

DISCUSSION

The continual advancements in industry, motorization, and the extensive application of fertilizers and pesticides have led to the persistent pollution of the air and soil with chemicals and heavy metals. The elevated levels of heavy metal contamination exceeding the limits established by the W.H.O indicate that the herbal samples are tainted and unsuitable for consumption.

According to Abdullahi (2019), cadmium is a non-essential element that is soluble in biological systems. It is known for its high toxicity and is comparable to other heavy metals in that it tends to bioaccumulate and interfere with the normal functioning of bodily organs.

The concentration of cadmium in the herbal samples varied from 0.03 to 0.6mg/kg. Six of the powdered samples labeled A, B, C, F, I, and J appeared to have concentrations lower than the limits set by the World Health Organization for many countries. However, four samples labeled D, E, G, and H recorded values of 0.4, 0.53, 0.46, and 0.6mg/kg respectively, exceeding the W.H.O limits. Consumption of these samples can pose serious health risks to consumers, making them unsafe for consumption. For instance, sample H, used as a remedy for gonorrhea, contained the highest value of about 0.6mg/kg, while the lowest cadmium value was detected in sample C, approximately 0.03mg/kg, used as a remedy for ulcer. The samples with high cadmium contamination are proven unsafe for consumption, while others are deemed safe. This study contradicts the findings of Muhammad, (2020), where cadmium levels in analyzed herbal formulations did not exceed the recommended limit. Cadmium has been detected in herbal medicines in various studies conducted worldwide (Umar *et al.*, 2016). In Nigeria, a study on cadmium content in herbal medicines reported high concentrations ranging from 16.438 to 29.796 mg/kg. Elevated cadmium levels have significant toxicological implications on human health, primarily targeting the kidneys. Cadmium accumulates in human kidneys over time and is excreted at a slow rate, leading to irreversible damage to the renal system (Li *et al.*, 2016). High cadmium levels also adversely affect the immune system, vascular system, and liver (Muhammad *et al.*, 2020).

The chromium levels in the powdered samples of this study ranged from 0.6 to 6.5mg/kg. Chromium is considered a micronutrient essential for fat, cholesterol, and glucose metabolism, with no specific intake limit set by the World Health Organization and other regulatory bodies. However, excessive chromium concentrations can be harmful and potentially carcinogenic (Chishti *et al.*, 2021).

The chromium levels found in this study were higher than those reported by Umar *et al.*, (2016) in a similar study conducted in Kano state. Environmental sources of chromium release include stainless steel, welding, and tannery industries (Daniel *et al.*, 2016). Prolonged exposure to high chromium levels can lead to skin rashes, skin cancer, and nasal irritation (Goulart *et al.*, 2015). The presence of chromium in the samples contradicted the findings of Yusuf *et al.* (2020), who reported its absence in certain anti-malarial herbal preparations from the same area. This discrepancy may be attributed to variations in chemical composition and soil content of the collection sites. Cadmium accumulation can result in liver and kidney damage, alterations in reproductive hormones, and renal injury (Akinloye *et al.*, 2016).

In this study, the concentrations of lead in the samples analyzed ranged from 3mg/kg to 22mg/kg. The maximum concentrations of lead in the samples were 22.0, 19.5, 17.1, 15.7, 14.1, and 10.2mg/kg. These values exceed the permissible daily intake set by the WHO (2019) of 10mg/kg, posing a toxic risk to human health when consumed. From Table 2 below, the highest concentration of lead at 22.0 mg/kg is found in a remedy for cough. The highest concentration was observed in powdered sample J, also used as a remedy for cough. The levels of lead reported in this study were higher than those in a study by Odoh and Ajiboye (2019), possibly due to variations in chemical components and the geographic regions where herbal items were gathered (Alaribe *et al.*, 2018).

The concentrations of lead in this study varied considerably among the samples analyzed other than the study conducted by Felagha and Ogbolosingha 2018, where three herbal medicines of malaria are having the same concentrations compared to other medicines. This variance most likely stems from the various sources these raw materials used to make the various herbal medications came from. It has been documented that lead can bioaccumulate in human tissues and cause spontaneous abortions. Patients who take herbal remedies with increased lead content over an extended period of time may be at risk of Chronic lead toxicity and should be closely watched for any signs of lead poisoning. Low birth weight in females and delayed neuron development are two more potential side effects (Daniel *et al.*, 2016).

The levels of mercury were not detected in samples A, B, F, and J, but it was found in a few samples in minute quantities that did not exceed the WHO limits (1.0mg/kg), making these samples free of mercury toxicity. The detected mercury values in this study ranged from 0.09 to 0.3mg/kg. This finding contradicts the study by Rausan *et al.*, 2019, where higher levels of mercury were found in an antidiabetic herbal preparation, exceeding the WHO limits and rendering the preparation unsafe for consumption. Alpana and Arun, 2015, conducted a similar study on *Viola* odorata and *Zingiber officinalis* collected from the Southern and Eastern parts of Pakistan, revealing mercury levels below the permissible limits set by the WHO regardless of the collection sites. Individuals who use herbal medicines with high mercury content are at risk of developing organ toxicity, such as hepatotoxicity, renal failure, and damage to the central nervous system (Bala *et al.*, 2018). Environments exposed to mining activities or certain industrial processes may contain mercury. While mercury was previously used in medicine, safer pharmaceutical drugs like chlormerodrin have now replaced its use (Chen *et al.*, 2020).

CONCLUSION

The findings of this study revealed that extracts from certain herbal preparations sold at major markets in Kano State, Nigeria showed varying levels of toxicity. The results indicate that lead (Pb) concentrations were notably high, surpassing the W.H.O limits in sample J, which was used as a remedy for cough, indicating significant contamination compared to other samples. Mercury was undetectable in four samples, and when present, it was within the World Health Organization's safety limits, certifying all samples as free from mercury contamination. Minimal to no cadmium and chromium contamination was observed in the herbal samples.

It is recommended that the Kano State Government regulate, monitor, and assess the use, safety, quality, manufacturing, handling, distribution, and importation of herbal products through the National Agency for Food and Drug Administration and Control (NAFDAC) and other regulatory bodies in Nigeria.

ACKNOWLEDGEMENT

The authors are grateful to tertiary education trust fund (TETfund) Abuja, Nigeria for funding

this project. We also acknowledge the technical assistance of the Technologists in the Central

Laboratory, Centre for Dryland Agriculture, Bayero University Kano, Nigeria.

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