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ELEMENTAL ANALYSIS OF UNREGISTERED ANTIMALARIAL HERBAL MEDICINES SOLD IN SAMARU, ZARIA, KADUNA STATE

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ABSTRACT

The use of herbal medicines has been in the increase in many developing and developed nations and for centuries, Nigeria has a long history of relying on traditional herbal remedies, leveraging the natural healing properties of plants to address various health concerns and promote well-being. The listing of these herbal remedies by regulatory bodies has further encouraged the use of herbal remedies. The safety of these herbal remedies is not guaranteed. This study is aimed at estimating the concentration of heavy toxic metals (Cadmium, iron, manganese, lead and zinc) in unregistered Nigerian herbal products frequently used in the treatment of malaria. Ten brands of antimalarial herbal products were collected, processed using acid digestion method (nitric acid and hydrochloric acid) and then analyzed using flame atomic absorption spectrometer. The concentrations of Lead (17.74 – 27.33 mg/kg) was above the permissible limits in all the samples analyzed. The contents of Manganese, Iron and Zinc on the other hand were within the permissible limits for all the samples analyzed. The content of Cadmium was within the permissible limit for one of the samples (0.04 mg/kg) and above the permissible limit (0.39 – 0.81 mg/kg) for the other samples. In conclusion, unregistered Nigerian herbal products used to treat malaria contain high levels of toxic heavy metals, specifically Lead and Cadmium, which exceed permissible limits.

Keywords: Heavy metals, acid digestion, herbal products, antimalarial

INTRODUCTION

Heavy metals are defined as metallic elements that have a relatively high density compared to water (Fergusson, 2013). With the assumption that heaviness and toxicity are interrelated, heavy metals also include metalloids, such as arsenic, that are able to induce toxicity at low level of exposure (Duffus, 2002). In recent years, there has been an increasing ecological and global public health concern associated with environmental contamination by these metals. The widespread adoption of heavy metals in various industries, agriculture, household products, and technological applications has led to a staggering surge in human exposure, as their use has grown exponentially, posing significant health risks (He *et al.,* 2005; Kawamura *et al.*, 2012).

Some of the heavy metals are having so much of biological importance in trace amounts particularly the elements that are present in the 4th period in the modern periodic table. The biological importance of these metals is enzyme functioning (vanadium and manganese), hormone functioning, production (selenium), cellular growth (nickel), and metabolic growth (arsenic). But these metals are required for the human in trace amounts only if their amount in the body increases, they cause adverse effects on human health. Overall, the heavy metal should be considered as having high density and also biological importance in trace amounts. (Emslay, 2011).

The widespread use of herbal products to treat malaria is driven by their affordability and accessibility, despite the need for further research into their safety and toxicity. While some herbs have shown promise as effective antimalarial, the lack of regulatory oversight of unregistered products puts consumers at risk of adverse reactions, ineffective treatments, and other potential harm. In Nigeria, the National Agency for Food and Drug Administration and Control (NAFDAC) regulates both food and drugs under the NAFDAC (2004) ACT.

The aim of this study is to carry out elemental analysis on ten unregistered antimalarial herbal products sampled from herbal medicine shops in Samaru, Sabon Gari Local Government Area, Zaria, Kaduna State.

MATERIALS AND METHODS

Sample Collection

The herbal products were sampled from herbal medicines shops in Samaru, Sabon Gari local government area, Kaduna State, Nigeria on 12th April, 2021. The brand name, manufacturing and expiry dates were recorded. Selection of the products was based on the popularity of the products among the general public.

Sample Preparation

The method of Ang and Lee (2005) was used for sample digestion. A 0.5g of the powdered sample was taken in a beaker; 10ml of nitric acid (67%) was added and kept at room temperature for 24 hours in a fume cupboard. 4 ml of Hydrochloric acid (35%) was added to the sample and concentrated on a hot plate at 60°C until a suspension of approximately 1ml was left in the beaker. The residue was cooled, diluted with deionized water up to 50ml and filtered through filter paper. Sufficient deionized water was added to make the volume up to 100ml and was kept in a sample bottle. The levels of toxic heavy metals were analyzed using Atomic Absorption Spectroscopy (AAS) and expressed in units of milligrams per kilogram (mg/kg), allowing for accurate quantification of their presence.

Flame Atomic Absorption Spectroscopy

Elemental analysis was carried out using a flame atomic absorption spectrophotometer (280FS AA). The aqueous sample was aspirated in the flame atomizer by the nebulizer to measure the analyte concentration at milligrams per kilogram (mg/kg) concentration level with good precision. This process was carried out for all the 10 samples.

RESULTS

Collected Samples of Antimalarial Herbal Medicines

The collected herbal antimalarial samples from herbal medicine shops in Samaru Zaria with their Manufacturing dates and expiry dates are shown in Table 1 below.

Heavy Metal Concentration of the Collected Samples Antimalarial Herbal Medicines

The heavy metal concentration of the collected antimalarial herbal medicines is shown in Table 2.

DISCUSSION

The safety and quality of medicinal herbal products have become a major concern for health authorities, pharmaceutical industries and the general public (WHO, 2007). The practice of herbal treatment is well established in Nigeria (Abubakar, et al., 2015). The products have substantial share in drug market. Apart from affordability and availability, the advocates of herbal products believe that they are safe and harmless because of their natural origin without any scientific evaluation.

Table 1: Manufacturing and Expiry Dates of Sampled Antimalarial Herbal Products used in Samaru, Sabon Gari Local Government Area, Kaduna State

S/NO	Sample	Manu-Date	Exp. Date
1.	A	Nil	2022
2.	B	Nil	2023
3.	C	2018	2023
4.	D	08/08/20	31/12/23
5.	E	Nil	2022
6.	F	2018	2022
7.	G	2019	2023
8.	H	Nil	2022
9.	I	30/11/23	30/11/23
10.	J	Nil	12/2023

Table 2: Heavy Metal Concentration (mg/kg) of Sampled Antimalarial Herbal Products used in Samaru, Sabon Gari Local Government Area, Kaduna State

Sample	**Concentration (mg/kg)**				
	Zinc	Lead	Manganese	Cadmium	Iron
A	1.44 ±0.0012	21.22 ±0.0026*	15.28 ±0.0003	0.04 ±0.0010	65.03 ±0.0066
B	3.93 ±0.0025	19.14 ±0.0026*	30.21 ±0.0008	0.39±0.0031*	51.23 ±0.0043
C	2.62 ±0.0014	17.74 ±0.0041*	12.01 ±0.0010	0.88 ±0.0010*	46.28 ±0.0050
D	2.16 ±0.0057	26.26 ±0.0034*	24.72 ±0.0007	0.81 ±0.0034*	22.21 ±0.0015
E	3.10 ±0.0012	26.50 ±0.0026*	28.08 ±0.0003	0.60 ±0.0010*	65.59 ±0.0066
F	1.44 ±0.0025	26.14 ±0.0026*	17.12 ±0.0008	0.45 ±0.0031*	69.64 ±0.0043
G	0.19 ±0.0014	22.37 ±0.0041*	6.43 ±0.0010	0.63 ±0.0010*	57.21 ±0.0050
H	1.46 ±0.0057	27.33 ±0.0034*	10.71 ±0.0007	0.79 ±0.0034*	45.74 ±0.0015
I	1.28 ±0.0012	23.04 ±0.0026*	8.45 ±0.0003	0.76 ±0.0010*	73.06 ±0.0066
J	1.58 ±0.0025	27.23 0±.0026*	21.99 ±0.0008	0.52 ±0.0031*	56.99 ±0.0043

Concentration expressed as mean± standard deviation. World Health Organization (WHO)/ Food and Agricultural Organization (FAO) Permissible limit: Zn: 50mg/kg, Pb: 10mg/kg, Cd: 0.3mg/kg, *Exceeded permissible limit.

Similarly, the potential serious side effects of synthetic drugs also diverted attention towards natural products. Regrettably, a large number of herbal products on the market contain hidden or unlisted ingredients, and sometimes dangerous amounts of toxic heavy metals, which can have harmful effects on those who use them. It has been noticed that the exposure to heavy metals like cadmium are toxic to human health even in traces (Marcus and Grollman, 2002).

Cadmium is one of the most toxic natural elements. Chronic exposure to cadmium through environment or using contaminated food causes kidney and lung failure and also affect bones and stomach (Shukla et al., 2007). The toxic impact of cadmium on human health is universal, affecting both adults and children similarly, with no age-related differences in its harmful effects (Saeed *et al.,* 2010). However, studies have shown that younger animals absorb more cadmium than adults (ATSDR, 2008). The permissible limit set by WHO is 0.3 mg/kg

in medicinal herbs (WHO, 2007). The range of cadmium concentrations in this work is between 0.04 – 0.88 mg/kg. Sample A with a concentration of 0.04mg/kg of cadmium was within the permissible limit while the concentration for the other 9 herbal products was above the permissible limit. However, due cadmium cumulative effect as reported by Jabeen *et al.,* (2010), the low-level content of cadmium does not make sample A safe for consumption. Consuming even a moderate amount of cadmium can lead to sudden and severe poisoning, causing rapid damage to the liver and kidneys, and potentially resulting in long-term health consequences

The concentration range of manganese in the tested samples is between 6.43 - 30.21 mg/kg. The WHO permissible limit for manganese in medicinal herbs have not yet been set. In humans, manganese toxicity represents a serious health hazard, resulting in severe pathologies of the central nervous system. Exposure to excessive manganese can result in severe neurological damage, leading to a crippling and permanent disorder akin to Parkinson's disease in its most severe form, and a range of troubling symptoms such as hyperirritability, violent outbursts, hallucinations, and coordination problems in its milder form (Keen *et al.,* 2005). Manganese is an essential mineral that has various beneficial effects on human health, including, supporting bone health and preventing osteoporosis, regulating metabolism and managing diabetes, reducing inflammation, boosting vitamin absorption and cognitive function; and improving digestion and overall health (Keen *et al.,* 2005).

The concentration range of zinc in the tested samples is between 0.19 - 3.93mg/kg. According to WHO/FAO guidelines, the acceptable limit for zinc content in herbal medicines is 50 mg/kg, providing a standardized benchmark for manufacturers and regulatory agencies to ensure the safety and efficacy of these products (WHO, 2007). Based on the set standard, the concentration of the tested herbal medicine/preparation is within the permissible limit. Zinc is an essential element required for normal body growth, proper thyroid function, blood clotting and DNA synthesis. Though there is little information about its toxicity, consumption of zinc beyond the permissible limit may result in toxic effect on the immune system and reduced copper level in the body (Waheed *et al.,* 2013).

The concentration range of lead in the tested samples is between 17.74 - 27.23 mg/kg. The FAO/WHO, set the limit for lead in medicinal herbs as 10 mg/kg (WHO, 2007). Based on the set standard, the concentration of lead in all the tested herbal antimalarial is above the permissible limit. Lead is an extremely toxic substance that can cause both acute and chronic poisoning, resulting in severe and widespread harm to various body systems, including the kidneys, liver, digestive system, brain, and central nervous system, leading to damage, disorders, and potentially irreversible consequences (Khan *et al.,* 2008). Young children are particularly vulnerable to the toxic effects of lead and can suffer profound and permanent adverse health effects, particularly affecting the development of the brain and nervous system. Exposure of pregnant women to high levels of lead can cause miscarriage, stillbirth, premature birth and low birth weight (UNEP, 2019).

The concentration range of iron in the tested samples is between 22.21 - 73.06 mg/kg. Individuals demonstrate signs of GI toxicity after ingestion of more than 20 mg/kg. Moderate intoxication occurs when ingestion of elemental iron exceeds 40 mg/kg. Ingestions exceeding 60 mg/kg can

cause severe toxicity and may be lethal. (Madiwale *et al.,* 2006). The WHO limit for iron in herbal medicine has not been set or established yet. Iron poisoning is a leading cause of toxic ingestions and a major threat to children's lives, ranking among the most common and deadliest childhood poisonings, highlighting the need for vigilant prevention and prompt treatment. (Madiwale *et al.,* 2006). Iron toxicity can be classified as corrosive or cellular. Ingested iron can have an extremely corrosive effect on the gastrointestinal (GI) mucosa, which can manifest as nausea, vomiting, abdominal pain, hematemesis, and diarrhea; patients may become hypovolemic because of significant fluid and blood loss (Baranwal *et al.,* 2003). Iron is essential for human health, playing a crucial role in oxygen transport and energy metabolism, brain function and cognitive development, immune system function, healthy pregnancy and fetal development, physical performance and endurance; and red blood cell production, preventing anemia. Iron deficiency can lead to fatigue, weakness, and impaired cognitive and physical function (Saeed et al 2010).

CONCLUSION

In conclusion, unregistered Nigerian herbal products used to treat malaria contain high levels of toxic heavy metals, specifically Lead and Cadmium, which exceed permissible limits. hence their unsupervised use could have cumulative effect, which in turn may lead to their toxicity.

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