ASSESSMENT OF HEAVY METALS CONCENTRATIONS IN HERBAL MEDICINE USED FOR TREATMENT OF TYPHOID AND BACTERIAL INFECTIONS IN KADUNA STATE, NIGERIA

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Abstract: Herbal medicine is the oldest health care system based on holistic management and treatment of diseases. This study was aimed at assessing the heavy metals concentrations in herbal medicines used for the treatment of some diseases in Kaduna State. Ten (10) sample of the medicine were randomly collected, air dried, pulverized and subjected to digestion using concentrated HNO_3, and HCl at 1:3. The concentration of the heavy metals were analyzed using Atomic Absorption Spectrophotometer (AAS). The results range of the analyzed elements were; Cu (0.007 – 0.003 µg/g), Fe (0.0532 – 0.445 µg/g), Co (0.017 – 0.020 µg/g), Ni (0.001-0.170 µg/g), Zn (0.0059 – 0.161 µg/g), Cd (0.025 – 0.113 µg/g), Pb (0.055 – 0.055 – 0.098 µg/g). The estimated daily intake of the metals in the herbal medicines were calculated as; Cu (0.011 – 0.110 µg/day), Fe (0.3568 – 2.0585 µg/day), Co (0.011 – 0.0284 µg/day), Ni (0.002 - 0.0491 µg/day), Zn (0.0019 – 0.0372 µg/day), Cd (0.00056 – 0.0396 µg/day), Pb (0.2569 – 0.6159µg/day). The concentration of the metals was within the permissible limit of World Health Organization (WHO). The study concludes that consumption of these herbal medicine has no adverse effect on health challenges problem related to heavy metals toxicity considering the concentrations of the heavy metals in the samples. It is recommended that Herbalists should engage in hygiene practice right from planting, harvesting, processing/preparation and packaging of herbs.

Key words: Bacterial infection, Typhoid, Heavy metals, Herbal medicine,

INTRODUCTION

Traditional herbal medicines are the oldest health care system used in managing and curing diseases based on believe, theories and experience. According to the world health organization about two third of the world population depends on indigenous herbal medicines for primary health care due to it availability, accessibility and affordability (Shakya, 2016; Rania *et al.,* 2015, Saganuwan, 2010; WHO, 2008). Medicinal plants are the integral backbone of traditional system of medicine and their use date back before the written of human history. The use of herbal medicines is growing rapidly in Nigeria and the practice have been prevalent among the locals for many years and have impacted significantly on the lives of majority of Nigerian by providing holistic healing, community welfare and wealth creation (Omoseyindemi, 2003). In Nigeria people have been suffering from various infectious diseases. Typhoid and Urinary tract infection are common health concern and are the most frequent bacterial infection responsible for the majority of admissions into hospitals and with an increasing incidence of death associated to them (Lee, 2008, W.H.O., 2018).

Despite the therapeutic action of herbal medicines, contaminants such as heavy metals cannot be ignored dues to their adverse effect on human health. The heavy metal toxicity in traditional medicines may occur due to contaminated soil in which the plant grows, contaminated irrigated water, storage and/or manufacturing processes (Mihreteab *et al.,* 2020). The negative effect of consumption of herbal medicines with

such contaminants may lead to severe cases of life-threatening condition such as hepatic injury, neurological and gastrointestinal disorder and even death (Dghaim *et al.*, 2015, Stewart, 1998, Desmet *et al.*, 1997). Due to high dependence of people on traditional herbal medicines, there is need to explore the level of heavy metals as a result of their potential threat to human health as shown by several researches conducted (Harris *et al.*, 2011, Helal Uddin *et al.*, 2013, Mahupawar, 2015, Maiga *et al.*, 2005). This research was aimed to determine the level of heavy metals in tradition medicines sold in Kaduna State in order to ensure the safety of the general public.

MATERIALS AND METHODS

Sample Collection and Pretreatment

A total of ten (10) samples acclaimed by herbalists to treat typhoid and microbial infection were selected and purchased from herbal shops and vendors within and around markets in Zaria, Kaduna central and Kafanchan. The samples selected are the most common sold to patients with typhoid and infection cases. The crude forms of herbal samples were milled with an electrical grinder to convert them into powder form. The powdered form was then kept in a moisture free, air tight bottles prior to analysis.

Table 1: Profile and Botanical Information of the Traditional Herbal Medicine Used in this Study

Sample	code	Composition	Therapeutic treatment
1	ZARM1	*Anageisus leiocarpus "marke", Guiera senrgalensis "sabara", Cassia occidentalis "raidore"*	Malaria and typhoid fever
2	ZARI1	*Albizia chevalion "katsari", Anageisus leiocarpus "marke", Piliostigma reticulatum "kalgo"*	Rheumatisms, UTI, STD
3	ZARM2	*Azadirachta indica "dogon yaro", Cochlospernum planchoni "rawaya", Boswella dalzilli "ararrabi", Cassia singuena "runfu"*	Malaria and typhoid fever
4	ZAR2	*Anageisus leiocarpus "marke", Piliostigma reticulatum "kalgo", Cassia singuena "runfu", Securidaca longe pedunculate "sanya"*	Rheumatisms, UTI, STD
5	KADM1	*Cochlospernum planchoni "rawaya", Cassia singuena "runfu", Anageisus leiocarpus "marke", Detarium microcarpum "taura"*	Malaria and typhoid fever
6	KADI1	*Senna itelia "filasko", Stiga hermontheca "wuta-wuta"*	Rheumatisms, UTI, STD
7	KADM2	*Cochlospernum planchoni "rawaya", Erythrina senegalensis "minjirya", Ximenia americana "tsada"*	Malaria and typhoid fever
8	KAFM1	*Cassia singuena "runfu", Anageisus leiocarpus "marke", Cassia occidentalis "raidore"*	Malaria and typhoid fever
9	KAFI1	*Moring oleifera "zogale", Cajanus cajan "adduwa", Stachytarpheta angustifolia "wutsinyan kadangare"*	Rheumatisms, UTI, STD
10	KAFI2	*Ziziphus mauritiana "magarya", Urelytum giganteum "jeemaa", Stzygium guineense "malmo"*	Rheumatisms, UTI, STD

Sample Digestion

About 1 g of each sample was accurately weighed and transferred to a 50.0 cm^3 beaker. 20 cm^3 of aqua regia solution (a mixture of HNO_3 (70%) and HCl (37%) in (1:3) v/v) ratio) was added to a beaker, and the sample solution was heated until all the samples

dissolved completely and a clear and colourless solution was obtained. After digestion was complete, the sample solution was allowed to cool at room temperature, and 30 cm^3 of distilled water was added. Then, the solution was filtered using Whatman filter paper and diluted to 50.0 cm^3 with distilled water.

Heavy metal Analysis

The concentration of the elements, iron, zinc, copper, nickel, cobalt, cadmium and lead were determined at University Central Laboratory of Umaru Musa University, Katsina, Nigeria using atomic absorption spectrophotometer (Agilent 55AA) as described by the Association of Official Analytical Chemists (AOAC, 2000). All the determinations were done in triplicates and the concentration of heavy metals was calculated according to the formula below.

Concentration in dry weight (mg/g) = $\frac{conc.of\ metal(mg/L) \times V(L)}{weight\ of\ the\ sample(g)}$

Where V is the final volume (50 ml) of the digested sample solution

Approximate Daily dosage determination of the metals

The approximate daily intake of metal (µg/day) in an analysed herbal medicine

= amount of metal in 1g of traditional medicines (µg) x the average daily dosage used traditionally

The average daily dosage used was estimated by the herbalist and it ranges from 4.4g to 8.g

Table 2.0 results of heavy metals analysis in the herbal samples in µg/g

s/n	Code	Cu	Fe	Co	Ni	Zn	Cd	Pb
1	ZARM1	0.0024 ±0.0003	0.2416±0.0007	0.0030 ± 0.0004	0.0035 ± 0.001	0.0033 ± 0.0001	0.0013± 0.0006	0.0575 ± 0.0011
2	ZARI1	0.0042± 0.0007	0.445±0.0059	0.0085 ± 0.0008	0.0001 ± 0.0019	0.0080 ± 0.0019	0.0015 ± 0.0003	0.0555 ± 0.0016
3	ZARM2	0.0018± 0.0006	0.053±0.0012	0.0003 ± 0.0005	0.0006 ± 0.0005	0.0022 ± 0.0009	0.0014 ± 0.0003	0.0560 ± 0.0012
4	ZARI2	0.0072± 0.0006	0.2225± 0.0028	0.0021 ± 0.0006	0.0125 ± 0.0007	0.0028 ± 0.0001	0.0057 ± 0.0005	0.0715 ± 0.0008
5	KADM1	0.0026±0.0005	0.1289± 0.0665	0.0035 ± 0.0010	0.0025 ± 0.0016	0.0025 ± 0.0001	0.0018 ± 0.0005	0.0850 ± 0.0008
6	KADI1	0.0029± 0.0002	0.1120± 0.0566	0.0021 ± 0.0005	0.0017 ± 0.0002	0.0041 ± 0.0014	0.0020 ± 0.0008	0.0820 ± 0.0018
7	KADM2	0.0024± 0.0006	0.0888± 0.0436	0.0047 ± 0.0004	0.0006 ± 0.0007	0.0021 ± 0.0006	0.0019 ± 0.0003	0.0855 ± 0.0007
8	KAFM1	0.0024± 0.0005	0.08 ± 0.0338	0.0039 ± 0.0002	0.0085 ± 0.0007	0.0027 ± 0.0016	0.0020 ± 0.0006	0.0975 ± 0.0011
9	KAFI1	0.0037 ± 0.000	0.108± 0.0493	0.0038 ± 0.0008	0.0029 ± 0.0010	0.0013 ± 0.0007	0.0022 ± 0.0003	0.0970 ± 0.0005
10	KAFI2	0.0033± 0.0001	0.2667 ± 0.0030	0.0050 ± 0.0003	0.0061 ± 0.0003	0.0003 ± 0.0008	0.0885 ± 0.0013	0.0885 ± 0.0014
Range		0.007 – 0.002	0.445 – 0.0532	0.020 – 0.017	0.170 – 0.001	0.161 – 0.0059	0.113 – 0.025	0.098 – 0.055
Mean		0.00329	0.172	0.00396	0.0039	0.00293	0.01083	0.0776
WHO PL		20	15	n.a	1.63	100	0.3	10

Key n.a = not available

WHO PL = world health organization permissible limit

Table 3: daily intake of metal (µg/day) according to the herbalists

S/n	Code	Cu	Fe	Co	Ni	Zn	Cd	Pb
1	ZARM1	0.0110	1.0677	0.0130	0.0155	0.0145	0.0056	0.2576
2	ZARI1	0.0194	2.0585	0.0039	0.0002	0.0372	0.0069	0.2569
3	ZARM2	0.1100	0.3242	0.0015	0.0034	0.0132	0.0085	0.3416
4	ZARI2	0.0504	1.5572	0.0147	0.0088	0.0197	0.0396	0.5005
5	KADM1	0.0149	0.7538	0.0202	0.0146	0.0146	0.0105	0.4973
6	KADI1	0.0151	0.5922	0.0111	0.0089	0.0219	0.0108	0.4338
7	KADM2	0.0097	0.3568	0.0189	0.0024	0.0085	0.0076	0.3546
8	KAFM1	0.0139	0.4624	0.0225	0.0491	0.0156	0.0116	0.5636
9	KAFI1	0.0232	0.6851	0.0238	0.0184	0.0082	0.0137	0.6159
10	KAFI2	0.0189	1.5306	0.0284	0.0289	0.0019	0.0123	0.5080
	Mean	0.0177	0.939	0.0158	0.0150	0.0155	0.0109	0.4330
	RDOD	24.2	45	8	100	50	5	10

Key

RDOD = recommended daily oral dose

Copper (Cu)

The mean concentration of copper in all the analyzed herbal samples was 0.0033µg/g and the concentration ranges from 0.002 – 0.007 µg/g. The highest concentration of copper was found in sample $ZARI_2$ followed by KAFI1. The lowest concentration was recorded in sample ZARM2. All the 10 herbal samples analyzed were within the maximum concentrations of copper (40 µg/g) set by WHO (2007). A Similar result of low concentrations of copper was reported by Chionyedua *et al* (2015) on toxic heavy metals in herbal medicines sold in Lagos. Copper is an essential element for the human metabolism. It regulates various biological processes like redox reactions, energy production, connective tissue formation, iron metabolism, and synthesis of neurotransmitters. However, chronic exposure to a high concentration of copper causes irritation of nasal mucosa, vomiting, nausea, diarrhea, kidney, and liver damage (Ekeanyanwu *et al.,* 2013; Zamir *et al.,* 2019).

Iron (Fe)

The result of Iron concentrations in all the herbal samples ranged from 0.445 µg/g to 0.053 µg/g and mean concentration was. These concentrations were lower than WHO permissible limit of 20 µg/g. The result of the concentration of iron in this study is compared to the ranged concentration of iron (3.75 to 35.24 µg/g) reported by Samali *et al.,* 2011. The level of iron in herbal medicine can boost optimum hemoglobin supply which suppress severity of malaria condition by meeting the body requirement (Bashawri *et al.,* 2002). Iron is an essential micronutrient it plays a vital role in biochemical activities such as oxygen transport and electron transfer. Deficiency of iron content in human lead to gastrointestinal infection, nose

bleeding and myocardial infection (Maiga *et al.,* 200). At high level intake iron causes dizziness, nausea, vomiting, diarrhea and liver damage (Martin and Griswold 2009).

Cobalt (Co)

Cobalt (Co) is an essential element required in small amount. Co plays a vital role in body metabolism and considered useful for humans as it is a part of vitamin B_{12} and also used for treatment of anemia but toxic in high concentration (Broding *et al.,* 2009, Ojeda *et al.,* 2012). The adverse effects of cobalt are vasodilatation, flushing in humans and animals as well. The mean concentration of cobalt in all the analyzed samples was 0.0039 µg/g and level ranged from 0.0003 - 0.0085 µg/g. Umar *et al* (2016) reported similar finding from different unregistered herbal preparation marketed in Kano State.

Zinc (Zn)

Zinc is the basic component of a large number of different enzymes and plays structural, regulatory and catalytic functions. It also has very important role in DNA synthesis, normal growth, brain development, bone formation, and wound healing. At high level, Zinc is neurotoxin (Adelekan and Abeunde, 2011) causing vomiting, dehydration, abdominal pain, nausea, dizziness, lack of muscular coordination, and renal failure (WHO, 1993). The mean concentration of zinc was 0.0029 µ/g and concentration ranged between 0.0059 to 0.161 µg/g. The highest concentration of zinc was recorded in ZARI1. The concentration of Zinc in all the analyzed samples fall within the permissible recommended limit of 100 mg/kg (WHO 2007).

Nickel (Ni)

The concentration of nickel in the herbal samples ranged between 0.001±0.00019 µg/g and 0.0125±0.0007 µg/g. The maximum concentration of nickel was found in $ZARI_2$. Concentration of nickel in all the analyzed samples were below the maximum permissible limit of WHO (1.62 µg/g). The concentration of nickel in this study is relatively lower compared to the study conducted on toxic heavy metals in some selected branded Lagos herbal medicines in the range of 0.73 µg/g to 54.0 µg/g (Chionyedua *et al.,* 2015). A study of heavy metals concentration in Kano herbal preparation gave the concentration of nickel above this study in the range of 9.94 – 15.63 µg/g (Samali *et al.,* 2017). Research has shown that ingestion of nickel beyond the tolerable limit for an extended period of time affect stomach, liver, kidney, immune system (ATSDR 2008).

Lead (Pb)

Lead is a non-essential heavy metal. High level accumulation of Pb in human body causes anemia, colic, headache, brain damage, and central nervous system disorder (Rehman, 2013). The herbal samples contained concentration of Pb in a range of 0.055–0.09 µg/g. the herbal sample obtained from Zaria had the least level (0.055-0.057µg/g), while herbal samples obtained from Kafanchan ($KAFM_1$, $KAFI_1$ & $KAFI_2$) have the highest level (0.089-0.098 µg/g) of Pb. Too much Consumption of these product may pose a health challenge if we consider the level of lead found on the samples. Samali *et al* (2017) reported 3.75 – 35.24 µg/g Pb in herbal preparations from Kano State. WHO's permissible limit of lead in herbal medicine is 10 µg/g (WHO 2007).

Cadmium (Cd)

Cadmium is also a nonessential element for humans, because it can easily cause toxic effects on humans at low concentrations. It is classified as a human carcinogen (Mihreteab *et al.,* 2020). The effects of exposure to long-term at lower levels may cause toxic effects on the kidney, the

respiratory and the skeletal systems (Jaishankar *et al.*, 2014). The mean concentration of Cadmium in all the herbal samples was 0.01083 µg/g and the concentration ranged from 0.001 µg/g to 0.088 µg/g. These values were low compared to what is reported by Chionyedua *et al* (2015) and Adepoju-bellow *et al* (2012). According to the WHO, the permissible limit of cadmium in herbal medicines is 0.3 µg/g.).The recommended value estimated for Cd is 0.025 mg/day (Lenntech, 1998-2009).

CONCLUSION

Traditional herbal medicines used for treatment of malaria, typhoid fever, bacterial and viral infections in Kaduna metropolitans were analyzed for heavy metals concentration using Atomic Absorption Spectrophotometer. The concentration of all the elements analyzed in the herbal samples are within the permissible limit of WHO. Therefore, Consumption of these herbal medicines is unlikely to cause health problem associated with heavy metals considering their concentrations in the samples.

RECOMMENDATIONS

Based on the results of this study, the following recommendations were made.

i. Herbalists should inject hygiene practice right from planting, harvesting, processing/ preparation and packaging of herbs.
ii. Constant monitoring of herbal medicines by regulatory agencies such as NAFDAC and also strict rule should be enforced on vendors selling herbal medicines that lacks standardization so as to ensure the safety of the consumers.
iii. Sensitization and awareness to herbal practitioners on safety measures and ways of minimizing contaminants.

AUTHOR CONTRIBUTION

Author 1: conceptualization, methodology, Sample Collection and Pretreatment, validation, funding acquisition formal analysis, investigation, resources, writing – review and editing, visualization, supervision, project administration, funding acquisition.
Author 2: conceptualization, data curation, writing – original draft preparation validation, formal analysis, investigation, resources.

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