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**Research Article** 

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## Quantitative Determination of Heavy Metals in Some Commonly Consumed Herbal Medicines in Kano State, Nigeria

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Abstract Evaluation of heavy metals in twelve commonly consumed herbal medicines/preparations in Kano State, Nigeria was carried out. The samples comprised of five unregistered powdered medicines, namely, *Zuwo*, (ZW); *Rai Dorai*, (RD); *Miyar Tsanya*, (MTS); *Bagaruwar Makka*, (BM); and *Madobiya*, (M); five unregistered liquid herbal medicinal concussions for pile (MB), yellow fever (MS), typhoid (MT), stomach pain (MC), sexually transmitted diseases (STDs); and two registered herbal medicines; Alif Powder (AP) and Champion Leaf (CL). The heavy metals evaluation was carried out using Atomic Absorption Spectroscopy (AAS) and the result revealed the concentrations (ppm) ranges of the heavy metals as follows: Cadmium (0.0045 – 0.1601), Chromium (0.0418 – 0.2092), Cobalt (0.0038 – 0.0760), Copper (0.0547 – 0.2465), Iron (0.1197 – 0.3592), Manganese (0.0123 – 1.4462), Nickel (0.0073 – 0.0960), Lead (0.185 – 0.0927) and Zinc (0.0244 – 0.2444). Comparing the results obtained in this work with the standards of the World Health Organization (WHO), the Food and Agricultural Organization (FAO) and permissible limits of other countries, the concentrations of heavy metals in the herbal medicine/preparations are within the allowed permissible limits range in herbal medicines and their use could be safe.

Keywords Herbal medicines, Registered, Unregistered, Kano State,

### Introduction

Metals are a major category of globally-distributed pollutants and are natural elements that have been extracted from earth and harnessed for human industry and products for millennia. (An exception to metals being "natural" is polonium). Heavy metal is a term used to define metallic elements with atomic weight higher than 40.0 (the atomic mass of Ca) [1]. Metals are notable for their wide environmental dispersion from such activity; their tendency to accumulate in selected tissues of human body; and their overall potential to be toxic even at relatively minor levels of exposure.

Some metals, such as Copper and Iron, are essential to life and play an irreplaceable role in, the function of critical enzyme systems. Other metals are Xenobiotic, *i.e.*, they have no useful role in human physiology (and most other living organisms) and, even worse, as in the case of lead and mercury may be toxic even at trace level of exposure. Metals that are essential have the potential to be toxic at very high level of exposure. One reflection of the importance of metals relative to other potential hazard is their ranking by the United State Agency for Toxic Substances and Disease Registry (ATDSR), which lists all hazards present in toxic waste site according to their prevalence and the severity of their toxicity The first, second, third, and sixth hazards on the list are heavy metals: lead, mercury, arsenic and cadmium respectively.

Exposure to metals can occur through a variety of routes. Metals may be inhaled as dust or fume (tiny particulate matter, such as the lead oxide particles produced by the combustion of leaded gasoline). Some metals can be vaporized (*e.g.* mercury vapour in the manufacture of fluorescent lamps) and inhaled. Metals may also be ingested involuntarily through food and drink. The amount that is actually absorbed from the digestive tract can vary widely, depending on the chemical form of the metal and the age and the nutritional status of the individual. Once a metal is absorbed, it distributes in tissues and organs. Excretion typically occurs primarily through the kidney and digestive tract, but metals tend to persist in some storage sites, like the liver, bone,

kidneys, for years or decades. Pollution of soil with heavy metals from polluted irrigation water, atmospheric dust, automobile and industrial exhausts, pesticides and fertilizers play important roles in heavy metal accumulation of medicinal plants/products [2].

#### Metals in herbal medicinal products

The reasons for the presence of metals in Herbal Medicinal Products (HMPs) are varied. Plants may accumulate heavy metals from the environment during growth on contaminated soil or by deliberate addition [3-4]. Whatever the route by which heavy metals are introduced in HMPs, if the level of these metals is high [5-6], poisonings may occur [7-8].

The toxicity of metals most commonly involves the brain and the kidney, but other manifestations occur, and some metals, such as arsenic, are clearly capable of causing cancer [9].

#### Experimental

#### Sample collection

The sampling was conducted in three phases (three months) to ensure representation of samples. All the results obtained are averages of the three samples collected at the three sampling sites.

A total of twelve herbal medicines/preparations meant for oral administration were purchased directly from local herbal markets in Kano state. The samples collected were categorized into three groups with their codes as follows:

- a. Five powdered unregistered herbal medicines intended for oral administration
  - i. Bagaruwar makka (BM) used as remedy for typhoid.
  - ii. Madobiya (M) used as blood purifier
  - iii. Zuwo (ZW) used as a remedy for diarrhea.
  - iv. Rai Dorai (RD) used for stomach pain
  - v. *Miyar Tsanya* (MTS) used as remedy for delivery pains.

b. Five liquid unregistered herbal medicines/preparation intended for oral administration.

- i. Maganin Basir (MB) used as remedy for pile and as energizer.
- ii. Maganin Shawara (MS) used for immune booster and as remedy for body weakness.
- iii. Maganin Typhoid (MT) used as a remedy for Typhoid
- iv. Maganin Ciwon Ciki (MC) used against stomach pain
- v. Maganin Sanyi (STDs) used for sexually transmitted diseases.
- c. Two registered herbal medicines in powder form intended for oral administration
  - i. *Alif Powder* (**AP**) used for immune booster, energizer, bacterial infections and pile.
  - ii. *Champion Leaf* (CL) used for sexually transmitted diseases and toilet infections.

#### **Pre treatment and Digestion**

Twelve herbal medicine/preparations in both powder and liquid dosage forms were purchased and prepared for Atomic Absorption Spectroscopy (AAS) analysis. 1.0 g each of the powdered samples was weighed and transferred into a 250 cm<sup>3</sup> beaker. Some 10 cm<sup>3</sup> of concentrated nitric acid (HNO<sub>3</sub>) was added and allowed to stand overnight in a fume hood. The solution was then heated carefully in a water bath at 60 °C until the emission of red nitrous oxide fume ceased to evolve. The container was allowed to cool at room temperature. Exactly 4 cm<sup>3</sup> of 70 % perchloric acid (HClO<sub>4</sub>) was added to the sample and heat on a hot plate at 60 °C until almost dried. The residue was cooled and transferred into a 50 cm<sup>3</sup> volumetric flask, diluted with deionized water and filtered through Whatman filter paper no. 42. The solution was made to mark with distilled water and kept in a plastic sample bottle until analysed by AAS.

For liquid samples, 100 cm<sup>3</sup> of the herbal liquid samples were transferred into to a beaker. Exactly 5 cm<sup>3</sup> of conc. HNO<sup>3</sup> was added, heated to a small boil and evaporated on a hot plate to about 2 cm<sup>3</sup>. Heating and addition of conc. HNO<sup>3</sup> was continued until digestion was completed by appearance of a clear solution. The content was transferred quantitatively to a 100 cm<sup>3</sup> volumetric flask and diluted to mark with deionized water. The diluted digest was analysed for heavy metals using Atomic Absorption Spectrometry (AAS) (Model: VPG 210) [10].



<b>Table 1</b> : Result for concentrations of heavy metals (ppm) in the unregistered powdered herbal medicines.									
Cd	Cr	Со	Cu	Fe	Mn	Ni	Pb	Zn	
$0.0045 \pm 0.0$	$0.2092 \pm 0.0$	$0.0076 \pm 0.0$	0.1643±0.03	$0.1197 \pm 0.0$	$0.4752 \pm 0.0$	$0.0147 \pm 0.0$	$0.0556 \pm 0.0$	$0.2444 \pm 0.0$	
0.0091±0.0	$0.0418 \pm 0.0$	$0.0114 \pm 0.0$	0.1369±0.0	$0.2694 \pm 0.0$	$0.7438 \pm 0.0$	$0.0147 \pm 0.0$	$0.0371 \pm 0.0$	$0.11 \pm 0.0$	
$0.0045 \pm 0.0$	$0.0836 \pm 0.0$	$0.076 \pm 0.01$	$0.1643 \pm 0.0$	$0.1497 \pm 0.0$	$1.1157 \pm 0.0$	$0.0147 \pm 0.0$	$0.0278 \pm 0.0$	$0.1466 \pm 0.0$	
0.0091±0.0	$0.0418 \pm 0.0$	$0.0152 \pm 0.0$	0.1369±0.0	$0.1197 \pm 0.0$	$0.6611 \pm 0.0$	0.0221±0.0	$0.0927 \pm 0.0$	$0.1224\pm0.0$	
$0.0068 \pm 0.0$	0.0836±0.0	$0.0114 \pm 0.0$	0.2465±0.0	0.1497±0.0	1.4462±0.0	0.0221±0.0	$0.0278 \pm 0.0$	0.1222±0.0	
	Cd 0.0045±0.0 0.0091±0.0 0.0045±0.0 0.0091±0.0	Cd         Cr           0.0045±0.0         0.2092±0.0           0.0091±0.0         0.0418±0.0           0.0045±0.0         0.0836±0.0           0.0091±0.0         0.0418±0.0	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Cd         Cr         Co         Cu           0.0045±0.0         0.2092±0.0         0.0076±0.0         0.1643±0.03           0.0091±0.0         0.0418±0.0         0.0114±0.0         0.1369±0.0           0.0045±0.0         0.0836±0.0         0.076±0.01         0.1643±0.0           0.0091±0.0         0.0418±0.0         0.0152±0.0         0.1369±0.0	Cd         Cr         Co         Cu         Fe           0.0045±0.0         0.2092±0.0         0.0076±0.0         0.1643±0.03         0.1197±0.0           0.0091±0.0         0.0418±0.0         0.0114±0.0         0.1369±0.0         0.2694±0.0           0.0045±0.0         0.0836±0.0         0.076±0.01         0.1643±0.0         0.1497±0.0           0.0091±0.0         0.0418±0.0         0.0152±0.0         0.1369±0.0         0.1197±0.0	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	

Result

Key: RD= Rai Dorai; ZW= Zuwo; BM= Bagaruwar Makka; M= Madobiya; MTS= Miyar Tsanya.

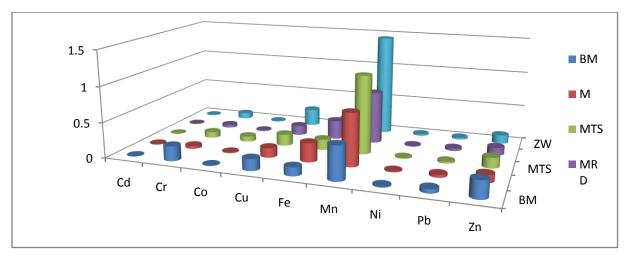


Figure 1: Graph showing bar representation of the concentrations of heavy metals in the unregistered Powdered herbal medicines.

**Table 2:** Result for concentrations of heavy metals (ppm) in the unregistered Liquid herbal medicines/preparations.

Sample	Cd	Cr	Со	Cu	Fe	Mn	Ni	Pb	Zn
MB	$0.0182 \pm 0.0$	$0.0836 \pm 0.0$	$0.0076 \pm 0.0$	$0.1369 \pm 0.0$	$0.2994 \pm 0.0$	$0.1239 \pm 0.0$	$0.0073 \pm 0.0$	$0.0649 \pm 0.0$	0.1222 ±0.0
MC	$0.0457 \pm 0.0$	$0.0836 \pm 0.0$	$0.019 \pm 0.0$	$0.0547 \pm 0.0$	$0.1197 \pm 0.0$	$0.0445 \pm 0.0$	$0.0369 \pm 0.0$	$0.0371 \pm 0.0$	$0.0244 \pm 0.0$
MS	$0.0137 \pm 0.0$	$0.2092 \pm 0.0$	$0.0076 \pm 0.0$	$0.0547 \pm 0.0$	$0.1197 \pm 0.0$	$0.0123 \pm 0.0$	$0.0591 \pm 0.0$	$0.0185 \pm 0.0$	$0.0855 \pm 0.0$
MT	$0.0114 \pm 0.0$	$0.1255 \pm 0.025$	$0.0076 \pm 0.0$	$0.0821 \pm 0.0$	$0.1497 \pm 0.0$	$0.0619 \pm 0.0$	$0.096 \pm 0.0$	$0.0556 \pm 0.0$	$0.1466 \pm 0.0$
STDs	$0.1601 \pm 0.04$	$0.1255 \pm 0.025$	$0.0114 \pm 0.0$	$0.0821 \pm 0.0$	$0.2694 \pm 0.0$	$0.0413 \pm 0.0$	$0.0295 \pm 0.0$	$0.0278 \pm 0.0$	$0.1466 \pm 0.0$
									10

Key: **MC** = Maganin c/Ciki; **MT**= Maganin typhoid; **MC**= Maganin c/Ciki;**MB**= Maganin basir; **MS**= Maganin Shawara;

STDs = Remedy for STDs

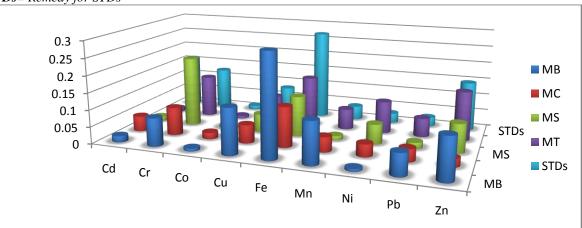


Figure 2: Showing bar representation of the concentrations of the heavy metals in the unregistered Liquid herbal medicine/preparations

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Table 3: Result for concentrations of heavy metals (ppm) in the registered herbal medicine/preparations.

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Sample	Cd	Cr	Со	Cu	Fe	Mn	Ni	Pb	Zn
AP	$0.0045 \pm 0.0$	$0.0836 \pm 0.0$	$0.0038 \pm 0.0$	0.1917±0.0	$0.1197 \pm 0.0$	$0.4132 \pm 0.0$	$0.0147 \pm 0.0$	$0.0556 \pm 0.0$	0.1711±0.0
CL	$0.0068 \pm 0.0$	$0.1673 \pm 0.0$	$0.076 \pm 0.01$	0.2191±0.03	$0.3592 \pm 0.0$	$0.6404 \pm 0.0$	$0.0221 \pm 0.0$	$0.0927 \pm 0.0$	$0.1833 \pm 0.0$
Ke	Key: CL= Champion leaf: AP= Alif Powder								

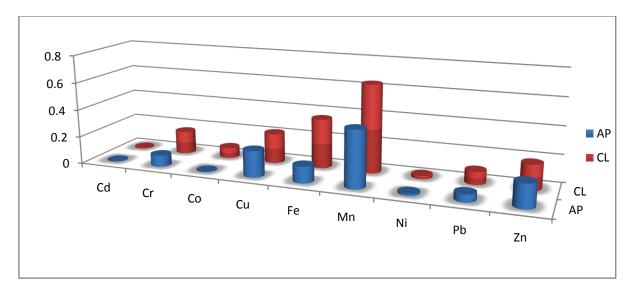


Figure 3: Showing Bar representation of the concentrations of the heavy metals in the registered herbal medicines

#### Discussion

The result for heavy metals determination in the various medicinal plants and product are presented in tables' 1, 2 and 3 for the 3 classes of herbal medicine/preparation. Table 1 shows the result of the heavy metals in unregistered powdered samples. The result is also presented in graphical form in figure 1. Table 2 shows the results of heavy metals concentrations in the five unregistered liquid samples while figure 2 presents the graphical form of the results. Table 3. and figure 3 presents the results and the graphical form of the concentrations of heavy metals in the solid registered samples respectively. The result presented shows the various samples along the first column while the metals determined are arranged on the first raw. Heavy metal concentrations pattern in all the samples roughly depicts: Mn > Zn > Cu > Fe > Pb > Co > Ni > Cd.

#### Cadmium

Among the five unregistered powdered herbal medicine presented (Table 1), samples M and RD have the highest concentration of cadmium (0.0091 ppm) while sample BM and MTS have the lowest concentrations of 0.0045 ppm each. For the unregistered liquid samples presented in table 2, the sample with highest concentration of cadmiumis STDs (0.1601ppm). While sample MT has the least (0.0114 ppm). The highest concentration of cadmium in the registered herbal medicine (table 3) was obtained in sample CL (0.0068 ppm), a local remedy for syphilis, toilet infections and sexually transmitted diseases.

The concentrations of cadmium obtained in this work is about four times lower than the one reported by Bushra (2011) who determined the occurrence of cadmium in some selected valuable herbal drugs (*G. glabra, O. bracteatum, V. odorata, F. vulgare, C. cyaminum, C. sativum,* and *Z. officinalis*) purchased from three different zones of Karachi city using AAS [11]. The level is also lower than the permissible limit set by WHO, China and Thailand as 0.3 ppm in medicinal herbs (FAO/WHO, 1984).The range of Cd concentrations in this work (0.0045–0.1601 ppm) were low which showed that the herbal formulations studied were safe for consumption. However, due to its cumulative effect as reported by Jabeen *et al*, (2010), the low level of the metal does not make what it contained to be safe for consumption [12].

The concentration of cadmium among the three groups of herbal medicines (solid unregistered, liquid unregistered and solid registered) is not significantly different at 95% confidence level (fig. 1).

#### Chromium

The concentration of chromium in the unregistered powdered herbal medicine/preparation is presented in table 1. Sample BM used as a remedy for typhoid has the highest level of chromium (0.2092 ppm) and samples M

and RD (remedies for blood purifier and stomach ache respectively) were the least with concentration of (0.0418 ppm). Table 2 showed the concentration of chromium in the unregistered liquid samples. Sample MS used as immune booster locally has the highest chromium concentration (0.2092 ppm) while samples MB and MC (used as energizer and stomach upset remedies respectively), have the least concentrations (0.0836 ppm). Table 3 above-showed samples CL to have the highest level of chromium (0.1673 ppm) and sample AP with the least concentration of (0.0836 ppm). The concentration of chromium among the tested samples is not significantly different at 95% confidence level (Fig. 2).

The medicinal herbs Dichrostachyscineria, Maeruaangolensis, Mimusopszeyheri, Albiziaanthelmintica, Plumbagozeylanica, Cambretumimberbe, Indegoferaflavicans, Clerodendrumternatum, Solanumpanduriforme, Capparistomentosa, TerminaliasericeaandMaytenus senegalensis used for the treatment of perceived HIV and AIDS – associated opportunistic infections by traditional healers in northern Botswana were assessed for chromium and other metals using AAS. Chromium was found to be the most abundant (0.15-1.27  $\mu$ g/g). The permissible limit for chromium in herbal medicinal plants/finished products have not been set by the World Health Organization [13]. However, 2.0 ppm was set by Canada as the permissible limit of chromium in raw medicinal plants. High intake of chromium is reported to have toxic effect, causing skin rash, kidney and liver damage, cancer of the lungs and nose irritations [14].

#### Cobalt

Table 1 showed the concentration level of cobalt in the unregistered powdered herbal medicine/preparations. The sample MTS (a remedy for pains during delivery) has the highest concentration of cobalt (0.2465 ppm). Among the unregistered liquid herbs (table 2), sample MC recorded the highest concentration of the heavy metal cobalt (0.019 ppm) while samples MB, MS and MT have the least (0.0114 ppm). Of the two registered herbal medicines tested, sample CL has the highest level of cobalt (0.076 ppm), while sample AP has the least concentration of the metal, (table 3)

Except for the sample MTS, the cobalt concentration obtained in this work is lower than the one reported by Jabeen *et al.* [12], in which seven herbal medicines used in Turkey had (0.14 ppm-0.48 ppm) concentration range of Cobalt. In a separate work, higher levels of cobalt ((0.967 ppm- 6.067 ppm) were obtained selected herbs used in the Kisii region, Southwest Kenya [15]. There is no significant difference in the concentration of cobalt among the solid unregistered, liquid unregistered and solid registered herbal medicine samples (fig. 3)

The toxic level of cobalt was assessed for efficacy and safety using Instrumental Neutron Analysis (INA). The result showed the tested herbs to contain high level of Cobalt [16]. For herbal plants and preparation, the WHO/FAO have not set any regulation limit for cobalt.

#### Copper

The concentration copper in twelve herbal medicine/preparations ranged from (0.2465 ppm to 0.0547 ppm). The highest concentration of copper among the unregistered powdered herbal medicines (table 1) was recorded in sample ZW (0.2465 ppm) a remedy for diarrhea. The least concentration was obtained in samples M and RD (0.1369 ppm). The highest level of copper in the liquid unregistered herbs (table 2) was recorded in sample MB (0.1369 ppm), while the least concentration was found in samples MC and MS (0.0547 ppm). The concentration of copper in the registered herbal medicine (table 3) was highest in sample CL (0.2191 ppm). Sample AP has a least value of copper concentration (0.1917 ppm).

The result of copper level in this work was well below that obtained by a similar analysis done by Jabeen *et al.* [12] on 50 medicinally important leafy materials grown in India with copper range of 17.6 ppm to 57.3 ppm. The copper content of some indigenous medicinal herb in Southwest Kenya was found to be in the range of (0.305 to 1.44 ppm) [15]. The concentration of copper among the herbal medicines (solid unregistered, liquid unregistered solid registered) is significantly different at 95% confidence level (Fig.3.7). The solid samples (registered and unregistered) have higher copper level than the liquid unregistered herbal medicines.

The regulatory limits of the WHO/FAO have not been established yet for copper in herbal medicines [17], but China and Singapore set the permissible limits of 20 ppm and 150 ppm respectively [13]. Copper is an essential component of many enzymes. It has both beneficial and toxic effects depending on its level of consumption. High intake of copper may result in hair and skin discoloration, irritation of the upper respiratory tract, and vomiting and liver damage among others [17-18].

#### Iron

The concentration of iron in the herbal medicine was in the range of (0.3592 ppm to 0.1197ppm). The highest concentration of iron in the unregistered powdered medicine (table 1) was found in samples M and RD (0.2694 ppm). Least concentration was found in sample BM (0.1197 ppm). Among the unregistered liquid herbal

medicines (table 2), the highest concentration of iron was detected in sample MB (0.2994 ppm) with samples MC and MS having the least (0.1197ppm). For the registered herbal medicine (table 3), the highest concentration of iron was found in sample CL (0.3592 ppm). There is no significant difference in the concentration of iron among samples (Fig. 3.8).

The results for iron obtained in this work are reported [12], of some selected medicinal herbs of Egypt as between 261ppm to 1239 ppm. The limit for iron in herbal medicine has not been set or established yet. Iron is an essential element that facilitates the oxidation of carbohydrates, protein and fat to control body weight.

#### Manganese

The highest level of manganese in the unregistered herbal powdered medicine is sample ZW (1.4462 ppm) (table 1) while sample BM has the least concentration (0.4752 ppm). In the liquid unregistered herbal medicines, (table 2), sample MB has the highest concentration (0.1239 ppm) while sample STDs has the least concentration (0.413 ppm). Among the registered herbal medicines (table 3), sample CL has the highest concentration (0.6404 ppm). Sample AP has the least concentration of the metal manganese (0.4132).

The concentration of manganese in this work is below the one obtained in some eight selected herbal medicines in Egypt with manganese range of (254 - 17.35 ppm) [12]. WHO permissible limit for manganese in medicinal herbs have not yet been set. There is significant difference in the concentration of manganese among the solid unregistered, liquid unregistered and solid registered samples at 95% confidence level (Fig. 3.9).

#### Nickel

Sample ZW (0.0221 ppm) was found with the highest concentration of nickel in the powdered unregistered herbal medicine tested (table 1). In the liquid unregistered herbal medicine (table 2), Sample MT (0.096 ppm) has the highest concentration of nickel. Of the registered herbal medicines (table 3), sample CL had the highest concentration of nickel (0.0221 ppm). Concentration of nickel among the herbal medicines tested is not significantly different (fig. 10). The result in this work is below that obtained by Muhammad *et al.*, (2011) [19], in some branded Pakistani herbal medicines (1.2 - 56.3 ppm) used in management of various ailments.

Numerous medicinal plants have the ability to accumulate heavy metals when grown under the natural conditions. Medicinally used *Seneciocoronatus (Thnb.) Harv. (Asteraceae)* is one of the nine Nickel (Ni) hyper accumulating plants in Africa [20]. Though toxic, Nickel absorption by the body is reported to be very low. The most common toxic effect of nickel is lung cancer, nickel itch especially on wet or moist skin and blockage of nasal cavities [21].

#### Lead

The concentration of lead in the tested herbal medicine/preparations ranged from (0.0927 to 0.0185 ppm). Samples M and RD were found to have the highest concentration (0.0927 ppm) in the unregistered powdered herbal medicine (table 1), the sample with the least concentration in that category were samples ZW and MTS (0.0278 PPM). Sample MB had the highest concentration of Pb (0.0649 ppm) among the unregistered liquid herbal medicines (table 2). While sample MS had the least concentration (0.0185 ppm). The sample with the highest level of Pb among the registered herbal medicines (table 3) was CL (0.092 ppm). The concentration of lead is not significantly different among the herbal medicines tested (Fig. 11).

Ten brands of herbal medicines sold in Ghanaian markets were selected for the analysis of trace element quality using AAS. The results were reported as mean concentration of the representative samples for each of the ten brands. The respective percentage incidence of the trace metals analysed was; Fe (20%), Zn (80%), Cu (20%), Cd (40%); Al (100%) and Pb (100%). The range of the mean concentrations measured for all the brands were; Fe (< 0.006-298mg/L), Zn (< 0.001-0.091 mg/L), Cu (< 0.003-0.009mg/L), Cd (< 0.002-0.003 mg/L), Al (0.278-0.533 mg/L) and Pb (0.0056-0.085 mg/L) [22].

The World Heath Organization, (WHO), Malaysia, China and Thailand set the limit for lead in medicinal herbs as 10 ppm. Lead is considered as one of the most toxic element, causing both acute and chronic poisoning with adverse effect on various body systems such as kidney, liver, renal, digestive, brain damage and disorder of the central nervous system [14]. Based on the set standard, the concentration of the tested herbal medicine/preparation is below the permissible limit.

#### Zinc

The highest concentration of zinc in the powdered unregistered herbal medicines (table 1) was found in sample BM (0.2444 ppm). Samples M and RD showed the least concentration of the heavy metal. In the unregistered liquid herbs (table 2), samples MT and STDs had the highest concentration of zinc (0.1466 ppm). Sample MC was found to have least concentration of zinc (0.0244 ppm). Among the registered herbal medicines (table 3),

sample CL had the highest level of zinc while sample AP had the least. There is no significant difference in the concentration of zinc among the tested herbal medicinal samples (Fig. 12).

The obtained result in this work is below that of Moses, *et al.*, 2012, who reported the concentration of zinc in some herbs within the range of (0.989 - 1.833 ppm). Though it was similar to the result of Umar, *et al.*, (2014) that reported the concentration of some herbal medicines commonly used in Kura local government of Kano State, Nigeria in the range of (0.0088 - 0.1 mg/Kg) [15,23].

The permissible limit for zinc in herbal medicine set by WHO/FAO is 50 mg/kg. 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 [24].

#### Conclusion

The contamination of herbal medicine/preparations by heavy metals is of great concern because of their toxic and cumulative tendencies. The overall analysis of the twelve herbal medicines/preparations commonly consumed in Kano State showed the presence of the heavy metals (Cd, Cr, Co, Cu, Fe, Mn, Ni, Pb, Zn) in various concentrations. Though the concentrations of the heavy metals in all the tested samples are below the international accepted permissible limit, their unsupervised use could have cumulative effect, which in turn may lead to their toxicity.

#### Recommendations

- ✓ Frequent analysis of metal content of herbal medicine/preparations
- ✓ Pharmacovigilance to improve quality and safety especially during collection, transporting, processing and packaging.
- ✓ Proper labeling should be made on containers indicating dosage, date of manufacture, date of expiry and appropriate storage condition.
- ✓ Sensitization of herbal medicine practitioners on safety and ways of reducing contaminations.
- ✓ Government and community based surveillance methods (in hospitals and point of sale) as tool for early detection of safety signals.

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