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HEAVY METALS PROFILES AND POTENTIAL RISK IN HERBAL REMEDIES PRODUCED IN NORTH WESTERN NIGERIA

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ABSTRACT

Herbal remedies are widely used for cure of array of ill health conditions. However, they are less regulated and hardly undergo pharmacological screening, despite the fact that a lot of reports showed toxicities arising from their use. The aim of this study is the evaluation of Co, Cu and Ni concentrations in some herbal remedies formulations prepared in North West Nigeria and enumerate the potential risk using hazard quotients and hazard index. Samples were dried, digested and the solutions aspirated into atomic absorption spectrometer according to standard methods. The result showed that Co, Cu and Ni concentrations occur in the range of 12.00–34.60, 6.3–35.0 and 52.6–631.5 mg/kg, respectively. 100% and 37% of sample showed that they exceeded the World Health Organization permissible limits for Ni and Cu, respectively. The values of hazard quotient showed that 32.04 and 93.11% of samples for adult and children respectively are at risk. Also, the hazard index values showed that 31.03% and 93.10% of the remedies pose potential risk for adult and children, respectively. Therefore, the herbal remedies are considered unsafe for consumption and necessitate appropriate action by regulatory bodies.

Key words: Herbal remedies, Cobalt, Copper, Nickel, Concentration, Hazard quotient

INTRODUCTION

The use of herbal remedies for cure of various ailments is ubiquitous among nations for a long time, every community utilizing one or more of its advantages of being accessible, affordable, good efficacy and safe application relative to conventional medicine. The World Health Organization (WHO) stated that users of herbal remedies globally exceed that of conventional orthodox medicine by two to three times¹. The herbs are important in the orthodox medicine as the active phytochemicals of herbs serves as template in the manufacture of conventional medicine. It is estimated that about 30% of the drugs in the modern pharmacopeias were derived from plants and many others, which are synthetic analogues, were built on prototype compounds isolated from plants²⁻⁴.

Herbal remedies are lacking regulations governing preparation and administration, contamination in the course of cultivation, preparation, transportation or storage by pesticides, radioactives, microbes and heavy metals⁵. These contaminants result in deleterious health effects of the consumers. A typical example is a child who suffered from renal failure symptoms when he was treated with remedies containing high concentration of Fe, Mn, and to a lesser extent As and Pb⁶. Other reports indicated metals poisoning causing morbidity and death in children^{7,8}. Numerous studies were conducted on heavy metals content in herbal remedies^{9,10}.

This study was aimed at determination of concentrations of Co, Cu and Ni in the herbal remedies and the potential risk

posed by the metals via computation of hazard quotient and hazard index in both children and adult populations.

MATERIALS AND METHODS

All glass and porcelain wares were scrupulously cleaned with 10% HNO₃, rinsed several times with deionised water and air-dried. Reagents of analytical grades were used except otherwise stated.

Sampling

A total of 29 samples, 6 packaged and 23 unpackaged were purchased from wholesaler herbalists in Kurna Kano, Chiranci, Maigatari, Kawo Kaduna and Zaria towns of Kano, Katsina, Jigawa and Kaduna State, respectively. It should be noted that none of the samples is registered with the regulatory body, National Agency for Drugs Administration and Control (NAFDAC). Table 1 showed the samples and their cures as acclaimed by the herbalists.

Digestion and analysis

Samples were air dried and grinded to powder. 0.5g of a sample was weighed into a pre-weighed porcelain crucible and placed in a muffle furnace. Sample was heated gradually to 500°C and maintained at $\pm 10^\circ\text{C}$ in the furnace for four hours, then removed and cooled. 10ml of 6 M HCl was added to the ignited sample covered with watch glass and heated for 15min on a steam bath. Another 1ml HNO₃ was added and evaporated to dryness with further heating

for 1 hour to dehydrate silica and digest organic compounds completely. 5ml of 6M HNO₃ and 10ml of deionized water were added and the mixture heated on a steam bath to complete dissolution. The mixture was cooled and filtered through Whatman No. 1 filter paper into a 50ml volumetric flask and made up to the mark with deionized water¹¹.

The analyte solutions, a blank and calibration solutions of 0.000, 1.000, 1.500 and 2.000mg/l for Cobalt and 0.000, 0.400, 0.800 and 1.000mg/l for Copper, 0.000, 2.500, 5.000 and 7.500mg/l for Nickel were aspirated into AA500 pg instrument model atomic absorption spectrometer as recommended by manufacturer's settings. Detection limits were found to be 0.0012, 0.0009 and 0.0015 for Co, Cu and Ni respectively. In absence of reference materials for the herbal remedies, spiking experiment was conducted on some samples to validate the experimental protocol. Percentage recoveries on spiking revealed 93.50±0.0%, 92.35±4.50 and 78.95±0.00% for Co, Cu and Ni, respectively.

Hazard quotient and Hazard index

To assess the health risk of the individual metals at the determined concentration, hazard quotient HQ which is the ratio between exposure and the reference oral dose (RfD) was used. If the ratio is lower than one (1), there will be no obvious risk¹². HQ is computed and adapted as follows

$$HQ = (Dih) \times (C_{metal}) / RfD \times Bo^{13}$$

Where (Dih) is the daily intake of herbs (kg per day), in this study it is 3.0g as the herbalists usually prescribe an average of teaspoonful of formulation which is equivalent to 1.5g to be taken twice a day, (C_{metal}) is the concentration of metal in the herb (mg/kg), Bo is the human body mass (kg), RfD is the oral reference dose for the metal (mg/kg of body weight/ day). RfD is an estimate of a daily oral exposure for the human population, which does not cause deleterious effects during a lifetime.

Values of RfD for Ni is 0.02 mg/kg/day¹⁴, Cu is 0.04mg/kg/day¹⁵ and Co was estimated as 0.043 mg kg⁻¹ per day¹⁶. The average Bo was taken as 70 kg for adults¹⁷, and 19.25 kg for children 0-6 years old¹⁸.

The sum contribution of the metals to overall risk of the remedies was assessed by HI which has values as HQ.

$$HI = HQ_{Co} + HQ_{Cu} + HQ_{Ni}$$

RESULTS

Figure 1 shows the concentration of the heavy metals in the herb formulation samples. The heavy metals occur in the following ranges 12.00±0.05–34.60±0.04, 6.30±0.03–35.0±0.08 and 52.60±0.09–631.5±0.07mg/kg for Co, Cu and Ni respectively. This indicates that the metals occur in all the samples. Likewise, Ni has the highest concentration followed by Cu, then Co. It was observed that both the lowest and highest concentrations occur in different samples. Co and Cu concentrations showed significant positive correlation ($p < 0.05$) while Ni had significant negative correlation with Co ($p < 0.05$).

Hazard quotient/index

Table 2 presents the data on hazard quotient/index computation. Values of the hazard quotient indicate that Ni concentration has potential risk in some samples which contribute to the corresponding risk shown by the hazard index.

DISCUSSION

Copper range of 5.92–14.79 mg/kg in herbs was reported¹⁹ which are lower than the range in this study, while a relatively higher concentration of 17.6 to 57.3 mg/kg was reported in some medicinal plants of India²⁰. An almost equal concentration of 2.30 - 33.56 mg/kg Cu was recorded²¹. Cu concentration in 45% sample of this study exceeds the permissible limit of 20.0 mg/kg set by WHO, US Food and Drug Administration and China pharmacopoeia but may be said to be 100% below Singapore National limit of 150 mg/kg in herbal products²². High concentration of Cu may cause fever with flue like symptoms, hair and skin decolouration, dermatitis, irritation of the upper respiratory tract, metallic taste in the mouth and nausea²³.

Cobalt is an integral component of cyanocobalamin (vitamin B12), and as such is essential for red blood cell formation and the maintenance of nerve tissue in man²³. Co is the least occurring among the metals in this study. Its highest concentrations are associated with the packaged formulations, probably the formulations contain mineral constituents as sometimes are being added to herbs and contribute to metal content²⁴. Co range of 0.967- 6.067 mg/kg and 0.604-9.781 mg/kg were found, in which their highest concentrations are below the lowest of this study^{25,26}.

Nickel concentration in this study is greater than the highest concentration of 1.63 mg/kg of some Kenya medicinal plants²⁵ and FAO/WHO²⁰, 6.21±0.10 mg/kg was also reported by Baranowska²⁷. Even though Ni concentration is high, its toxicity in human is not a common occurrence because its absorption by the body is very low²⁰. It was stated that humans and animals absorb approximately 1-10 per cent of dietary nickel²⁸. The most common harmful health effect of nickel in humans is an allergic skin reaction in those who are sensitive to nickel²⁹.

Hazard quotient

HQ values indicate that Cu and Co does not pose health threat to adult and children as their values are all below 1. This agrees with the concentration as greater percentage of the samples has Cu concentration below the WHO permissible limit. Also, all the samples have Cu below the Singapore limit of 150 mg/kg³⁰. The daily intake ranges 0.02-0.11mg/day but the safe highest level of intake for an extended period of time is 10 mg/day.³¹.

Table 1: List of Samples with Their Names, Treatment and Formulations

Sample no.	Local name	Treatments	Formulation*
1	Maganin malaria da shawara	Malaria, jaundice	<i>Anogeissus leiocarpus</i> (Marke), <i>Cassia singueana</i> (Runfu), <i>Ximenia americana</i> (Tsada)
2	Maganin sanyin mara	Waist weakness,	<i>Anogeissus leiocarpus</i> (Marke), <i>Cassia singueana</i> (Runfu),
3	Maganin basir	Pile	<i>Prosopis africana</i> (Kirya), <i>Detarium microcarpum</i> (Taura)
4	Danbashaana	Sexual stimulant	Undisclosed
5	Maganin basir	Pile	<i>Anogeissus leiocarpus</i> (Marke), <i>Mangifera indica</i> (mangwaro), <i>Diospyros mespiliformis</i> (Kanya), <i>Lannea microcarpa</i> (Faru)
6	Maganin kurajen baki	Mouth boils	<i>Acacia Senegal</i>
7	Maganin gyanbon ciki	Endo abdominal wound	Undisclosed
8	Maganin ciwon ciki	Stomach ache	<i>Lannea microcarpa</i> (Faru), <i>Piliostigma reticulatum</i> (Kalgo),
9	Maganin shawara	Jaundice	<i>Anogeissus leiocarpus</i> (Marke), <i>Guiera senegalensis</i> (Sabara),
10	Maganin majina	Moderate mucus	<i>Albizia chevalieri</i> (Katsari), <i>Senna italica</i> (Filasko)
11	Maganin basir da zafi	Haemorrhoids	<i>Prosopis africana</i> (Kirya), <i>Commiphora kerstingii</i> (Arárrábi)
12	Maganin shawara	Jaundice	<i>Cochlospermum tinctorium</i> (Rawaya)
13	Maganin kasala	Body weakness	<i>Combretum micranthum</i> (Geza)
14	Maganin shawara	Jaundice	<i>Anogeissus leiocarpus</i> (Marke), <i>Cochlospermum tinctorium</i> (Rawaya)
15	Maganin basir	Pile	<i>Commiphora kerstingii</i> (Arárrábi), <i>Ficus ovate</i> (Gamji), <i>Detarium microcarpum</i> (Taura)
16	Maganin rana	Sun burn	<i>Prosopis africana</i> (Kirya)
17	Maganin jinni	Haemorrhage	<i>Detarium microcarpum</i> (Taura)
18	Maganin basir	Haemorrhoids	<i>Cochlospermum tinctorium</i> (Rawaya), <i>Detarium microcarpum</i> (Taura), <i>Ficus ovate</i> (Gamji)
19	Maganin daji	Cancer	<i>Ficus ovate</i> (Gamji), <i>Detarium microcarpum</i> (Taura), <i>Cassia singueana</i> (Runfu), <i>Ficus sp</i> (Baure)
20	Dan kadafi	Sexual stimulant	Undisclosed
21	Maganin kaikai	Itches	<i>Tamarindus indica</i>
22	Maganin mura	Catarrh	<i>Anogeissus leiocarpus</i> (Marke)
23	Dan kadafi	Sexual stimulant	Undisclosed
24	Maganin basir	Haemorrhoids	Undisclosed
25	Sanamaki	Headache, spirit	“
26	Maganin gudawa	Diarrhoea	“
27	Maganin sihiri	Spirit	“
28	Zaiti	Pain, typhoid fever, yellow fever, worms, pile	“
29	Nauriyya	Ulcer, pile, jaundice, asthma, cold, typhoid	“

* Information about plants used in the formulation was given by the herbalists.

Table 2: Values of Hazard Quotient and Hazard Index of the Heavy Metals for the Herbs in Both Adult and Children

Sample	Hazard Quotient						Hazard Index	
	Ni		Cu		Co		Adult	Child
	Adult	Child	Adult	Child	Adult	Child		
1	0.39	1.43	0.017	0.062	0.019	0.071	0.426	1.563
2	0.56	2.04	0.013	0.049	0.019	0.071	0.592	2.16
3	0.56	2.05	0.007	0.025	0.027	0.098	0.594	2.173
4	0.62	2.26	0.017	0.062	0.012	0.043	0.649	2.365
5	0.79	2.87	0.010	0.037	0.016	0.057	0.816	2.964
6	0.85	3.08	0.017	0.062	0.019	0.071	0.886	3.213
7	0.85	3.08	0.007	0.025	0.023	0.084	0.88	3.189
8	0.90	3.28	0.007	0.025	0.016	0.057	0.923	3.362
9	0.79	2.87	0.007	0.025	0.019	0.071	0.816	2.966
10	0.85	3.08	0.017	0.062	0.016	0.057	0.883	3.199
11	0.79	2.87	0.015	0.056	0.016	0.057	0.821	2.983
12	1.01	3.69	0.020	0.074	0.019	0.071	1.049	3.835
13	1.07	3.90	0.022	0.080	0.016	0.057	1.108	4.037
14	1.13	4.10	0.020	0.074	0.019	0.071	1.169	4.245
15	1.24	4.51	0.024	0.087	0.019	0.071	1.283	4.668
16	1.30	4.72	0.027	0.099	0.012	0.043	1.339	4.862
17	1.35	4.92	0.026	0.093	0.016	0.057	1.392	5.07
18	1.30	4.72	0.038	0.136	0.019	0.071	1.357	4.927
19	0.11	0.41	0.022	0.080	0.016	0.057	0.148	0.547
20	0.11	0.41	0.026	0.093	0.016	0.057	0.152	0.56
21	0.39	1.43	0.029	0.105	0.019	0.071	0.438	1.606
22	0.56	2.05	0.017	0.062	0.027	0.098	0.604	2.21
23	0.62	2.26	0.019	0.068	0.023	0.084	0.662	2.412
24	0.68	2.46	0.015	0.056	0.031	0.112	0.726	2.628
25	0.73±	2.67	0.036	0.130	0.027	0.098	0.793	2.898
26	0.79±	2.87	0.017	0.062	0.027	0.098	0.834	3.03
27	0.85±	3.08	0.034	0.124	0.027	0.098	0.911	3.302
28	1.01±	3.69	0.019	0.068	0.034	0.125	1.063	3.883
29	1.07±	3.90	0.022	0.080	0.023	0.084	1.115	4.064
Average	0.80±0.32	2.92±1.17	0.02±0.008	0.07±0.03	0.020±0.006	0.074±0.020	0.842±0.323	3.066±1.174
% Risky (>1)	32.04	93.11	0	0	0	0	31.03	93.10

Figure 1: The concentration of Co, Cu and Ni in the samples

The hazard quotient of this study showed a maximum Co daily intake of 0.10 mg/day in samples 24 and 26. This value is lower than the average of 0.12 mg/day Co daily intake among United Kingdom population in 1994^{32, 33}. However, it is greater than the estimated 0.005–0.040 mg/day Co intake in the United States³⁴ and 0.029 mg/day³⁵ for France. These reports showed that the contribution to daily intake of cobalt due to the herbs alone may be significant to the total intake.

In case of Ni, 32.04% and 93.11% of the samples exhibits some cautions on their use by adult and children respectively, particularly as it is a potent haematotoxic, immunotoxic, neurotoxic, genotoxic, reproductive toxic, pulmonary toxic, nephrotoxic, hepatotoxic and carcinogenic agent²⁹.

Hazard index

It is the sum contribution of individual metals to the hazard caused by a particular sample. The Ni high percentages in the herb contribute to the high hazard index values. As such 31.03% and 93.10% of the remedies may be risky for use by adult and children respectively.

The Cu and Co low contribution to the HI contrast with the result of³⁶ whereby Cu has mean HQ of 4.48, translating to 72% of HI.

CONCLUSION

The study indicates that the herbal samples contain Cu and Co at concentration that may be deemed to be safe as the results of hazard quotient revealed. But a metal of concern is the Ni which has concentrations that are very high compared to other studies and WHO permissible limit. Even though its solubility is low, there are reported health hazards associated with the metal. Therefore, possible sources of nickel should be investigated.

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