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ORIGINAL RESEARCH ARTICLE

Determination of Trace Elements Content of a Popular Traditional Herbal Medicine Used for Internal Hemorrhoids Treatment in Katsina State, Nigeria

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ABSTRACT
There has been an increase in the utilization of traditional herbal drugs for different medications and health care in Katsina state. This is possibly due to the growing need for complementary disease treatment, relative cheapness, availability, and wider distribution. Raw samples of a popularly used traditional herb were purchased from local markets, prepared, and analyzed using a flame atomic absorption spectrophotometer for Pb, Cr, As, Co, Ni, and Cd concentrations. The obtained concentrations were then summarized using descriptive statistics in SPSS Version 27. The results for the elements analyzed were Ni (0.83 ± 0.15), Cd (0.20 ± 0.01), Pb (21.5 ± 6.63), As (2.10 ± 0.70), Cr (2.36 ± 0.37), and Co (1.15 ± 0.22) ppm. Cr, Pb and As concentrations were greater than the WHO/FAO threshold limits for herbal medicines, while that of Cd was within the recommended threshold. Furthermore, the concentrations of these metals (As, Cr, and Pb) were greater than the WHO/FAO acceptable threshold in 50% of the herbal samples analyzed. Therefore, these metals can pose a threat to public health, especially considering the conventional way in which this herbal medicine is used. With deliberate improvement in hygiene during its preparation prior to consumption, the risk will be brought to a minimal. Future studies should perform a detailed chemical risk assessment, especially when we look at the increase in the number of diseases of unknown etiology in the region.

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heavy metals, World Health Organization, herbal medicine, health risk, herbal remedy toxicity

INTRODUCTION

There has been an increase in the utilization of traditional herbal drugs for different medications and health care in low and medium-income countries globally (WHO, 2011). This was ascribed to the inadequacy of modern medicines and the growing need for complementary disease treatment and prevention measures (Paulo et al., 2016). In the rural areas of Nigeria, the excessive use of herbal medicine is due to its relative availability, cheapness, and wider distribution and the belief that some diseases can only be treated using traditional means (Oni *et al.,* 2011; Njinga *et al.*, 2015 and Yahaya et al., 2019 and Anjorin et al., 2010). The World Health Organization and Food and Agricultural Organization, through their various publications, emphasized the need for quality control standards of medicinal plants and the need for health risk assessment to be carried out in order to protect people from unnecessary health implications (FAO/WHO: 2001, 2004, 2005, 2009 and 2011). Trace elements analysis of herbal medicines is therefore very necessary because medicinal plants can accumulate these trace metals, and when consumed by humans, it presents a detrimental effect (Imelouane et al., 2011; Brima, 2016 and 2017; Karahan et al., 2020; Werdemberg dos Santos et al. 2022 and Cigdem et al., 2023). Therefore, before using any traditional herb for medicinal purposes, it is very important to identify the trace element levels in order to prevent negative outcomes on human health. Suppose the concentration of some trace metals in the body is found to be greater than the physiological limits. In that case, several health complications may occur, such as neurological problems, cardiovascular dysfunction, disruption of the endocrine system, liver and kidney injury, hematological anomalies, and carcinogenic effects (Balali-Mood et al., 2021).

Literature reported varying levels of trace elements in medicinal plants and herbs due to location differences, the nature of the plants/herbs examined, and other parameters (Piero et al., 2012; Kim et al., 2016). A study determined the content of Ni, Zn, Mn, Cu, Cr, and Pb in samples of neem tree leaves (*Azadirachta indica*) obtained along the Katsina to Funtua highway of Katsina state

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revealed that the levels of these metals were within the WHO/FDA permissible levels of trace elements in herbal plants (Lawal et al., 2011). Plant mix of *Anogeissus leiocarpus, Prosopis africana, Boswellia odorata, and Guiera senagalensis* in herbal remedies sold in Katsina were examined by measuring the activity concentration of the natural radionuclides (^{238}U, ^{232}Th, and ^{40}K), the results indicated that these herbal remedies are not safe for consumption (Okunola et al., 2020). In Kano state, a recent study of the levels of some essential (Copper, Cadmium, Cobalt, Iron, Manganese, Nickel, and zinc) and toxic (Cd and Pb) metals content of herbal preparations sold in the Kurmi market revealed that 100% of the samples analyzed contained Cadmium above WHO permissible limits (0.3µg/g) specified for herbal medicine and 33% of the samples contained lead (Pb) above WHO permissible limits (10µg/g) specified for herbal medicine (Samali et al., 2017). In Saudi Arabia, an analysis of macronutrients, micronutrients, and toxic trace metals in popular herbal plants and spices revealed some of the concentrations of the analyzed elements were within the safety limits in most of the investigated samples, and some were not (Ibrahim, 2014; Maghrabi, 2014 and Seddigi et al., 2016).

The trace elements associated with the intake of herbal preparations have been linked to most forms of leukemia and with cancer of many organs, such as the bone, lung, breast, and thyroid, in the long term (NJinga et al., 2015). Data on studies of toxicity in herbal medicines in Katsina State is extremely scarce. Though there are a few studies on the trace elements content of some predominant herbs used in Katsina State, the available studies only considered less than 1% of the medicinal herbs available in this region. It is, therefore imperative to determine the trace elements content in a popular traditional herb consumed in Katsina and other states in Northwestern Nigeria for internal hemorrhoid treatment. This will help in creating a database containing elemental compositions of medicinal and aromatic plants for standardization purposes by the relevant national and international organizations such as the National Agency for Food, Drug Administration and Control (NAFDAC), World Health Organization (WHO), food and agricultural organization (FAO) and US environmental protection agency (US EPA).

METHODOLOGY

Samples were purchased directly from the local vendors (in their ground form as it is used when purchased) in the major markets in the state capital. The samples were transferred to a sample collection bag and moved to the laboratory for preparation and analysis. The samples were kept at an ambient temperature in the laboratory and were allowed to dry for 1 week prior to analysis. Each of the samples was then further ground using mortar and pestle and sieved using a 20 mm mesh sieve before wet digestion. An electronic weighing balance was used to weigh 1g of each sample and put it in an empty, clean beaker. 10 mL of HNO_3, 2 mL of 60% $HClO_4$, and 5 mL of H_2SO_4 were added to each sample in the beaker and mixed up with the sample using a glass rod to aid digestion. The samples were then heated on a hot plate for about 1 hour at 100 °C for complete digestion and allowed to be digested up to dryness. It was then allowed to cool at room temperature, filtered into a standard 60mL sample bottle, and made up to the mark with distilled water. The trace elements analysis was then carried out using an atomic absorption spectrophotometer (AAS). Different concentrations of standard solutions were run on the instrument to obtain the calibration curves for each metal using measured absorbances and the corresponding concentrations. The instrument was set to zero by reading a reagent blank. Each of the prepared samples was aspirated into the instrument and read three (3) times. The average value of the concentration was taken for each metal in each sample. The results of the analysis were validated by digesting and analyzing standard reference materials (Lichens coded IAEA-336) following the same procedure. The values obtained in the standard materials in this work were compared with the certified values and were found to be within 10% uncertainty. Distilled and Deionized water were used throughout the experiment and all reagents used were of analytical grades. Table 1 provides the spectrophotometric parameters used in the analysis, and Table 2 provides the calibration equations determined from the calibration curve.

Statistical analysis

The concentrations of these elements in the samples were summarized using descriptive statistics available in the Statistical Package for Social Scientists (SPSS) version 27.

Table 1: AAS parameters used in the trace elements analysis

Element	Lamp current (mA)	Wavelength (nm)	Slit width (nm)	Fuel gas flow rate (L/min)	Flame type	Burner height (mm)
Ni	25	232.0	0.2	1.6	Air C_2H_2	7
Cr	100-600	357.9	0.5	2.1	Air C_2H_2	9
Co	12-400	240.7	0.2	1.6	Air C_2H_2	7
Pb	20	283.3	1.0	2.0	Air C_2H_2	7
Cd	25-242	228.8	1.0	1.8	Air C_2H_2	7
As	20-25	283.3	1.0	2.0	Air C_2H_2	7

Table 2: AAS calibration equations used in the trace elements analysis

Element	Calibration equation	R^2 value
Ni	$Abs = 0.262131conc + 0$	0.9953
Cr	$Abs = 0.152958conc + 0$	0.9997
Co	$Abs = 0.167098conc + 0$	0.9966
Pb	$Abs = 0.0959407conc + 0$	0.9980
Cd	$Abs = 1.13367conc + 0$	0.9994
As	$Abs = 0.4354conc + 0$	0.9976

RESULTS AND DISCUSSION

The concentrations of the trace elements Ni, Cd, Pb, As, Cr, and Co in the collected samples were analyzed quantitatively using atomic absorption spectroscopy due to their ability for single-element analysis, and the summary statistics were presented in Table 3. The results in the form of average ± standard error for the elements analyzed were: Ni (0.83 ± 0.15), Cd (0.20 ± 0.01), Pb (21.5 ± 6.63), As (2.10 ± 0.70), Cr (2.36 ± 0.37) and Co (1.15 ± 0.22). The concentration of the trace elements for most of the samples analyzed was such that Pb > Cr > As > Co > Ni > Cd. Lead has the highest concentration, while Cadmium has the lowest concentration, which is consistent with the reports from other research on food, medicinal plants, and herbs. This is due to the fact that herbal drugs in different geographic regions absorb toxic elements in similar ratios. The variation in concentration among different toxic metals is normal because these elements have different relative abundance, plants differ in the way they accumulate and absorb toxic elements, and the nature of the element and its form determine the extent to which it will be absorbed in addition to the physicochemical properties of the soil on which that plant grow (Cindrić et al. 2013; Begaa and Messaoudi 2019; Zinicovscaia et al. 2020). Lead is a ubiquitous element; it originates from natural and anthropogenic sources and is easily deposited onto plants, herbs, and food samples due to its availability in vehicular emissions and other anthropogenic activities.

The Agency for Toxic Substances and Disease Registry of the United States has identified Nickel (Ni), Cadmium (Cd), Lead (Pb), Arsenic (As), Chromium (Cr), and Cobalt (Co) among the elements that can pose carcinogenic and non-carcinogenic health risks depending on their concentration. For this reason, and to protect the public from health consequences associated with the intake of these elements, the Food and Agricultural Organization (FAO) and the World Health Organization (WHO) have stipulated the maximum recommended levels of Pb, Cr, Cd, and As in food/medicinal plants and herbs. Currently, there is no WHO/FAO recommended threshold for Ni and Co in the food and herbs, but that of Pb, Cr, Cd, and As were 10, 2.3, 0.3, and 1.0 mg/kg, respectively (WHO/FAO 2001, 2005, 2011). The average concentration and some of the individual sample concentrations for the Pb, As, and Cr were greater than the WHO/FAO thresholds, while that of Cd was within the recommended threshold, as can be observed in Figures 1, 2, 3, and 4.

Table 3: Statistics of the trace metals concentrations

Statistics	Trace metals concentrations					
	Ni	Cd	Pb	As	Cr	Co
Mean	.833	.200	21.565	2.096	2.355	1.150
Std. Error of Mean	.151	.017	6.633	.703	.3700	.219
Median	.861	.208	9.685	.916	2.247	1.320
Std. Deviation	.4770	.052	20.977	2.223	1.170	.694
Skewness	-.198	-.802	.980	1.197	.114	-.552
Std. Error of Skewness	.687	.687	.687	.687	.687	.687
Kurtosis	-1.720	-.121	-1.070	-.187	-2.126	-1.384
Std. Error of Kurtosis	1.334	1.334	1.334	1.334	1.334	1.334
Minimum	.085	.097	1.34	.398	1.050	.129
Maximum	1.412	.253	55.46	6.314	3.824	1.997

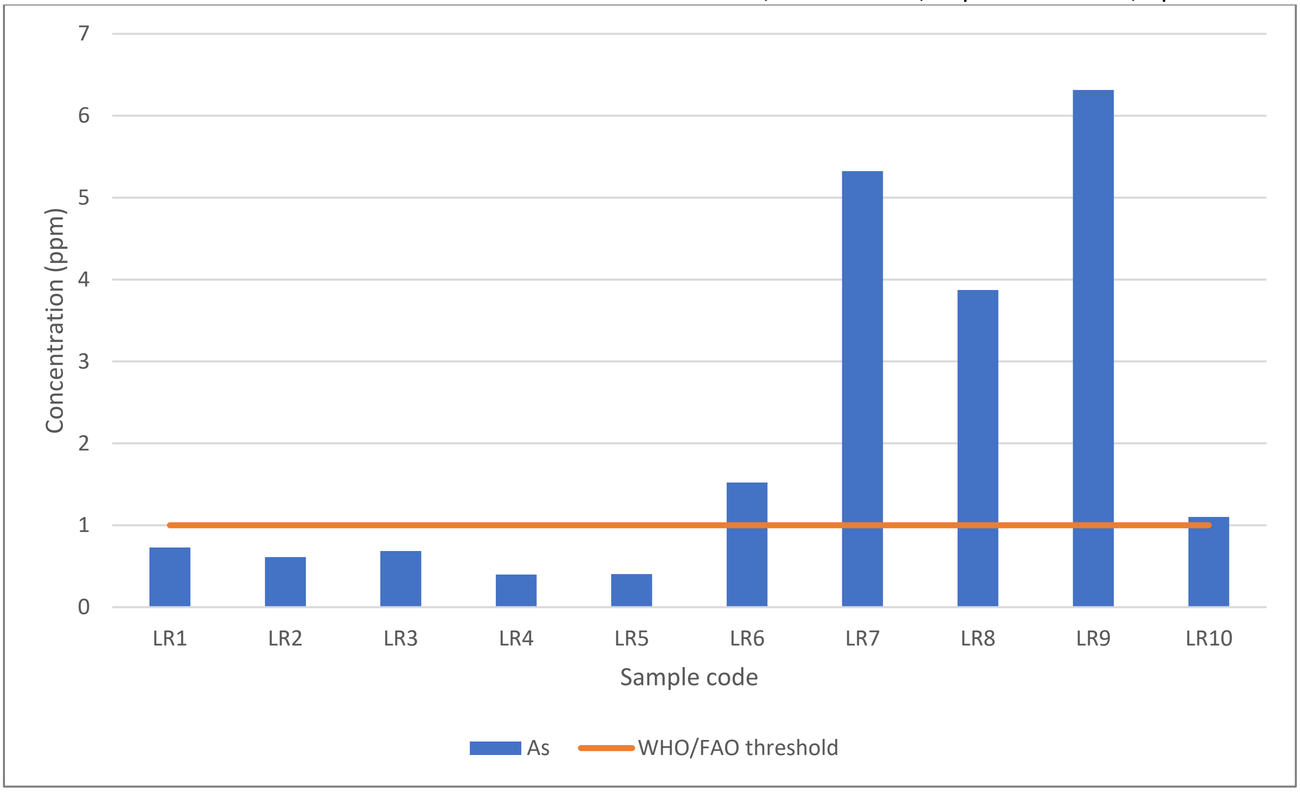

Figure 1: Concentrations of As and the WHO-recommended threshold

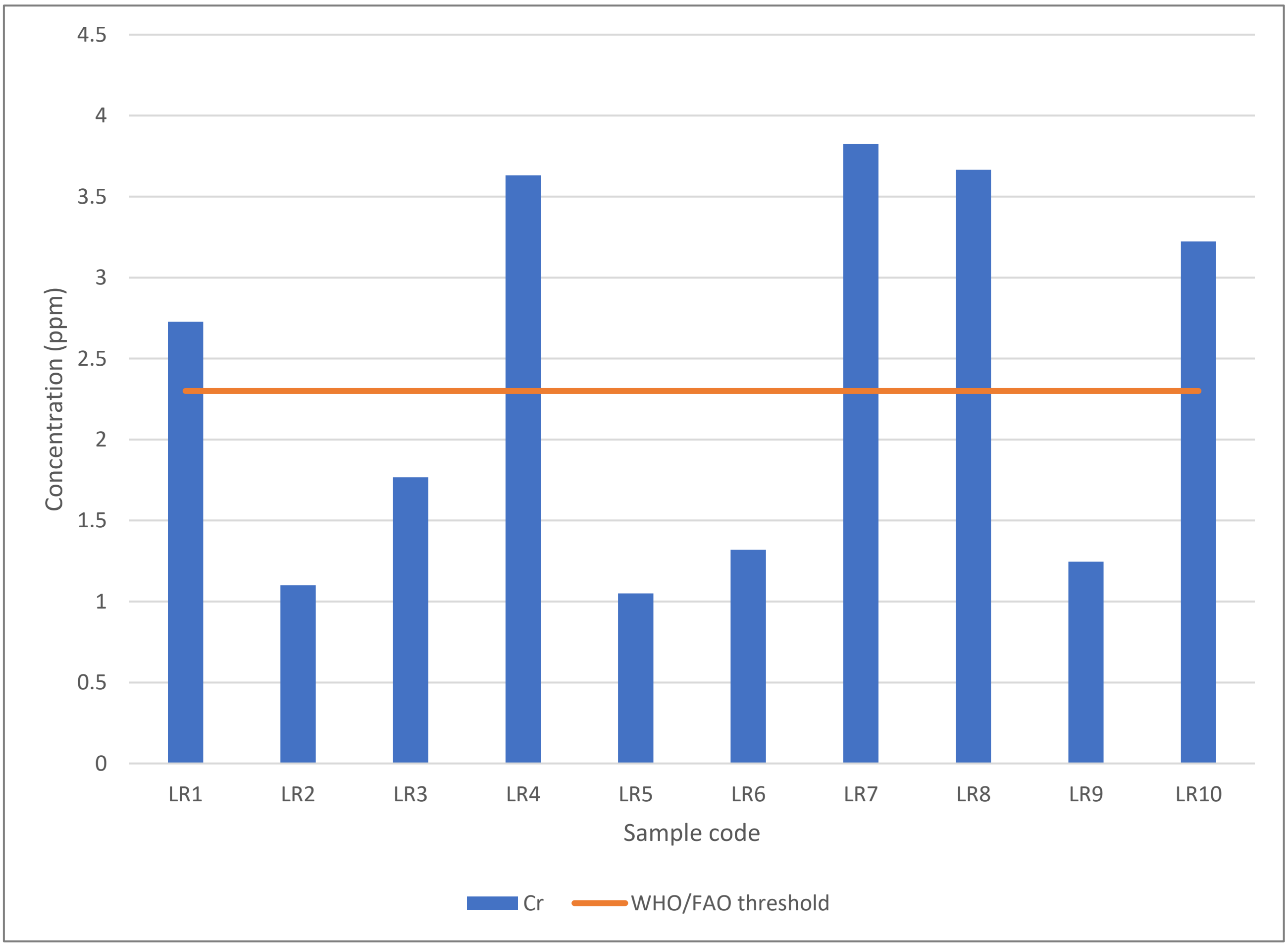

Figure 2: Concentrations of Cr and the WHO-recommended threshold

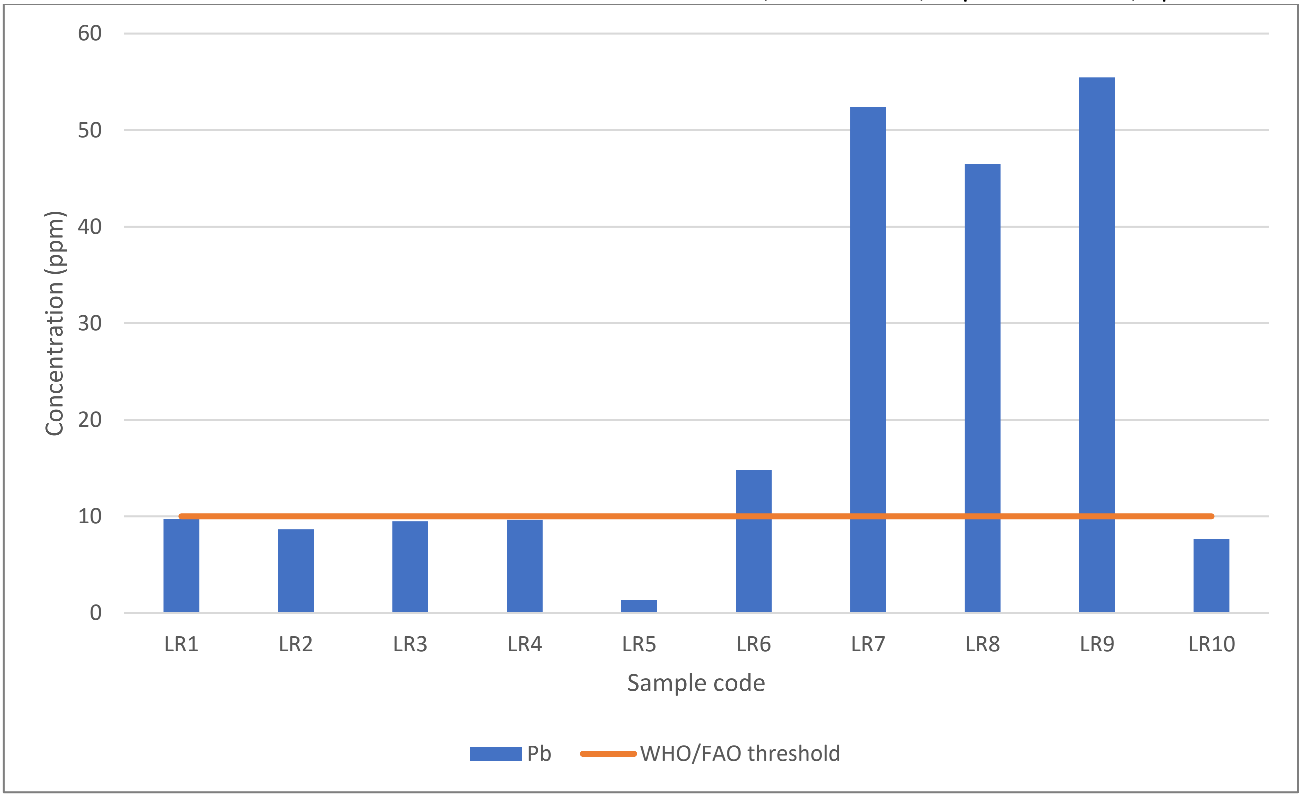

Figure 3: Concentrations of Pb and the WHO-recommended threshold

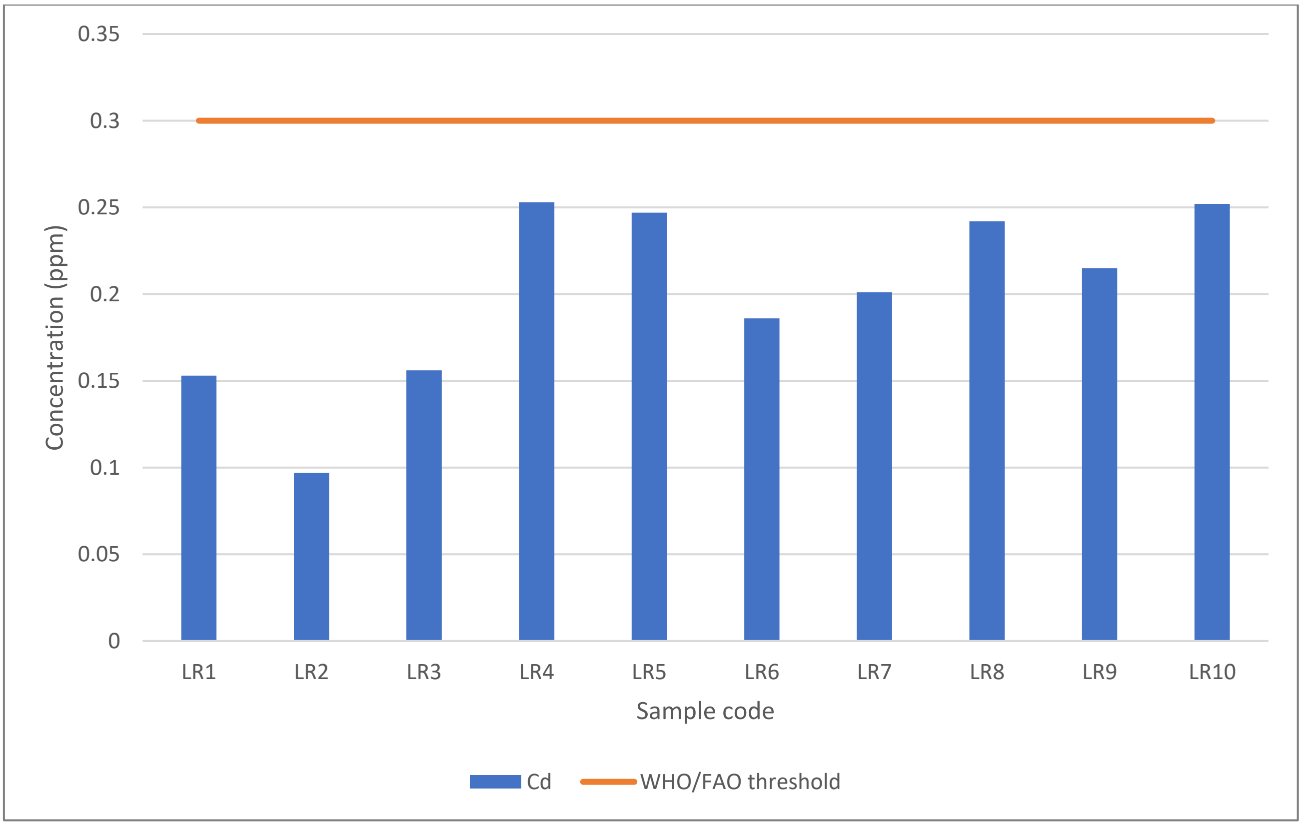

Figure 4: Concentrations of Cd and the WHO-recommended threshold

Though some of these elements were detected in concentrations greater than the WHO/FAO acceptable threshold, they may only constitute a serious health risk when used too frequently and when hygiene is neglected as it is in the present practice (not rinsing the herb before use). Discussion with the users of this traditional medicine pointed out that the mode of usage of this herb is that it is consumed when it forms a colloidal solution by directly pouring it in water, pouring it in cattle milk, or pouring it in a local drink. Therefore, the major exposure route is ingestion and there is no concern with inhalation and dermal routes. From this observation, it can be seen that the major reason for detecting high levels of these elements is pouring it directly without washing. Some research indicated that as high as 50% of these toxic elements can be washed away if the plant or herb is rinsed with clean water (Brima et al., 2017). Therefore, based on conventional use, it can pose a threat to public health, but with improvement in hygiene during its preparation prior to consumption, the risk will be minimal, as confirmed by other research. Lastly, a health risk assessment should be carried out to ascertain the health impact or otherwise of the utilization of these herbs, as they are very cheap and looking at the socioeconomic realities, they cannot be ruled out, but behavioral change towards hygienic preparation and consumption of this herbs is highly necessary. One of the limitations of this study is that the concentrations of these toxic elements in the herbs should also be assessed after washing them to further evaluate the actual reduction in the concentrations of these toxic elements. In addition, the concentration of the major and minor elements also should be reported so that a comprehensive understanding of the total elemental composition will be obtained. The accuracy of the findings could also be improved if the herb's samples were collected directly by future researchers and processed in a clean laboratory environment, thereby reducing the contamination significantly and providing more reliable data on the content of trace metals in the medicinal herb.

CONCLUSION

The average concentrations of Cd (0.20 ± 0.01), Pb (21.5 ± 6.63), and As (2.10 ± 0.70) were greater than the WHO/FAO threshold limits for herbal medicines, while that of Cd was within the recommended threshold. Furthermore, the concentrations of As, Cr, and Pb were greater than the WHO/FAO acceptable threshold in 50% of the herbal samples analyzed. Therefore, based on the conventional use of this herbal medicine, it can pose a threat to public health, but with improvement in hygiene during its preparation prior to consumption, the risk will be minimal, as confirmed by other research. However, a detailed health risk assessment of these herbal medicines should be carried out to ascertain the health impact or otherwise of the utilization of these herbs. Looking at the socioeconomic realities, even though the concentration of As, Cr, and Pb indicates that they are not safe, these traditional herbal medicines cannot be ruled out but ensuring that it is rinsed prior to consumption is highly necessary.

CONFLICT OF INTEREST

None

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