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Determination of some toxic heavy metal accumulation in medicinal plants commonly used in Gondar area district, Northwestern Ethiopia

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ABSTRACT

In Ethiopia, up to 80% of the population uses traditional medicines. The common sources of these medicines are plants. However, medicinal plants are contaminated with environmental pollutants especially heavy metals, which pose a great health risks upon long term exposures. Hence, this work was done to determine the level of three most common and toxic metals (Arsenic, Cadmium and Lead) in ten medicinal plants commonly used in Gondar area district. The concentrations of selected heavy metals were determined in medicinal plant samples using Flame Atomic Absorption Spectrometer and Graphite Furnace Atomic Absorption Spectroscopy (for Cd and Pb) and Hydride Generation Atomic Absorption Spectrometer (for As) after they were digested following the already developed method. The accuracy of the analytical procedures was evaluated by performing spike recovery tests. The percentage recoveries were from 85 - 105%.The result showed that the concentrations of the metals detected in the samples range from 0.03 – 2.06, 0.001 – 6.75, and 0.002 – 35.97 mg of metal per kg of sample for As, Cd, and Pb, respectively. Over all, out of each ten samples analyzed two for Arsenic (20%), seven for cadmium (70%) and three for Lead (30%) were found to contain concentrations above maximum WHO permissible limit (1, 0.3 and 10 mg/kg, respectively) which showed possible risk of toxic effects of the studied metals in the medicinal plants used.

KEYWORDS: Arsenic, Cadmium, Lead, Heavy metals, Medicinal plants.

INTRODUCTION

Traditional medicine is used throughout the world as it is dependent on locally available plants, which are easily accessible, and capitalizes on traditional wisdom-repository of knowledge, simple to use and affordable. These medical systems are heavily dependent on various plant species and plant based products[1]. Medicinal plants are plants, either growing wild or cultivated, used for their medicinal purposes [2]. They are consumed worldwide for the treatment of several diseases and such plants are also an important source of raw material for pharmaceutical industries[3]. In Ethiopia, up to 80% of the population uses traditional medicine. The major reasons why medicinal plants are demanded in Ethiopia are due to culturally linked traditions, the trust the communities have in medicinal values of traditional medicine, relatively low cost in using them and difficult access to modern health facilities[4-5]. A common misconception is that medicinal plants are "pure and natural" and that this equates to "harmless". Based on their long history of use, users of traditional medicines deem them safe for human consumption. However, the absence of their regulation provides no such guarantee[6]. Plants are contaminated with environmental pollutants especially heavy metals, which pose a great health risks to all living organisms upon long term exposures[7]. Cultivation in soils containing high concentrations of heavy metals is one mechanism of contamination of herbal products. Farmland, which may have been used for generations to produce medicinal herbs, may be directly encroached upon by factories, roads and other high pollutant areas or contaminated by aerosolized particles of waste material[8]. Emissions from heavy traffic on roads contain lead (Pb), cadmium (Cd), zinc (Zn) and nickel (Ni), which are present in fuel as anti-knock agents contaminate the nearby areas. Rivers and streams may also become contaminated and lead to downstream dispersion of heavy metals [6, 8]. Fertilizers, herbicides or insecticides containing heavy metals may also be applied in some settings. Many agro-chemicals contain As, Cu, Fe, Mn and Zn. Some contaminants such as Cd and Pb enter the soil as impurities in fertilizers. Especially Phosphate fertilizers represent a potentially significant source of Cd to soils. Therefore, the continued uses of contaminated fertilizers over an extended period of time cause accumulation of these contaminants to high levels in the soil[6]. Regardless of the source, plants are ultimately grown in soils with high concentrations of heavy metals. Plants absorb a number of elements from this contaminated soil and accumulate them in their different parts. Heavy metals will get an access to human biological system as a result of using these contaminated medicinal plants to treat different ailments. Once being in human biological system, they will cause different health impacts[8-9]. With the ever-increasing use of herbal medicines worldwide and the rapid expansion of the global market for these products, the safety and quality of medicinal plant materials and finished herbal medicinal products have become a major concern for health authorities, pharmaceutical industries and the public[2]. Different researches concentrating on the heavy metal content of medicinal plants have been conducted in various parts of the world and most of them showed that the levels of some heavy metals in these plants is higher than the maximum recommended limits set by different international organizations[10]. National regulation and registration of herbal medicines varies from country to country. Maximum allowable limit recommended by WHO are 1, 0.3 and 10 mg/kg for As, Cd and Pb in medicinal plants, respectively[2]. The safety and toxicity information of herbal medicine or products are required prior to expanded clinical studies and to support the registration of the herbal products with the Drug Control Agency. The presence of toxic heavy metals such as Cd, As, and Pb in herbal products at high levels pose serious risk to public health. These heavy metals when consumed in considerable amount can result in damaged or reduced mental and central nervous function, damage to blood composition, lungs, kidneys, liver, and other vital organs[11]. Therefore, it is mandatory to assess toxic metal concentration in commonly used medicinal plants to take a corrective action. Though similar studies have been conducted in different parts of the world, there are very limited reports on heavy metal content of medicinal plants originated from Ethiopia. To the best of the investigators knowledge, no report on the heavy metal content of medicinal plants used in Gondar area district. It was with this notion this work was done to measure the level of these three most common and toxic metals (Arsenic, Cadmium and Lead) in ten selected medicinal plants commonly used in Gondar area district.

MATERIALS AND METHODS

INSTRUMENTS AND EQUIPMENTS

Graphite Furnace Atomic Absorption Spectrophotometer (novAA-400, Analytikajena), Flame Atomic Absorption Spectrometer (Spectra 20 plus, Varian) and Hydride Generation Atomic Absorption Spectrophotometer with appropriate hollow cathode lamps, analytical balance, What man filter paper, hotplate, plastic bags, flat bottom flasks, pipettes and volumetric flasks were used.

SAMPLES

Samples of ten medicinal plants commonly used by the people for their medicinal value were selected. Plants species name, code given, local name, parts used, their application and the route of administration for their traditional use are shown in Table 1.

Table1: List of medicinal plants analyzed for their heavy metal content and traditional use

Plant Species	**Code Given**	**Local Name (in Amharic)**	**Parts used**	**Application & route of administration**
Acalypha acrogyna	GUL	Gullo	Leaf	All types of cancer, orally
Adansonia digitata	BAM	Bamba	Bark	Cholera, bloating of stomach, orally
Brucea antidysentrica	ABA	Abalo	Leaf	Leprosy, scabies and other skin diseases, topically
Calpurnia aurea	DIG	Digta	Leaf	Ascariasis, gastric ulcer, oral
Carissa spinarum L	AGA	Agam	Leaf	Snake bite/external
Croton macrostachyus	BIS	Bsana	Bark	Gonorrhea, Abdominal colic, orally
Dodonaea angustifolia	KIT	Kitkita	Leaf	Jaundice, Malaria and Taeniasis, orally
Maytenus Ovatus	ATA	Atat	Leaf	All types of cancer, oral
Maytenus undata	CHE	Checho	Leaf	Eye infection, externally
Piliostigma thonningii	WAN	Wanza	Bark	Wound heal after male, female circumcision, externally

CHEMICALS AND SOLVENTS

Reference standards for As ($AsNO_3$), Cd ($CdNO_3$) and Pb ($PbNO_3$), distilled water, concentrated acids (like HNO_3, HCl), and H_2O_2 were used.

SAMPLE PREPARATION AND DIGESTION

Plant parts with medicinal value were thoroughly washed with distilled water and rinsed with de-ionized water. They were cut into small pieces and dried at 50 °C for approximately 72 h. Once dried, the individual plant part was ground into a fine powder (< 0.5 mm) using pistil and mortar, and stored in plastic bag until analysis. Plant samples were digested according to the method described by Street *et al* [12]. To 0.5 gm of homogenized powdered plant material, 10 ml HNO_3-HCl-H_2O_2 (8:1:1, v/v/v) were added in a borosilicate flask. The mixture was heated at 120 ^oC over 3 h on a hotplate. After digestion was completed, the clear and colorless solution was transferred to a 50 ml volumetric flask. Each digestion tube was rinsed with distilled water to collect any possible residue, and added to the volumetric flask which made up to volume with distilled water. The dilute samples were then stored in 100 ml plastic bottles (high density polyethylene) until analysis. Each plant sample was digested and analyzed in triplicate to confirm precision of the result. The blank solution was prepared by taking a mixture of 8 ml HNO_3, 1 ml HCl and 1 ml H_2O_2 and treating similarly as that of the sample. Standard solutions were prepared from 1000 mg/L stock solutions for each metal. From these stock solutions, different concentration standards were prepared for each metal using the reagents and solvents used for sample preparation to minimize the matrix effect. Then absorbance (in terms of peak area) of Pb and Cd were determined using Flame Atomic Absorption Spectrometer (FAAS) for most samples and Graphite Furnace Atomic Absorption Spectroscopy (GFAAS) for samples that couldn't be detected by FAAS. Absorbance of As was detected using Hydride Generation Atomic Absorption Spectrometer where as Cd and Pb was detected using FAAS and GFAAS. The calibration line was obtained by plotting absorbance versus concentration. The elemental concentrations of the samples were determined from its absorbance/peak area using the linear regression equation of the calibration line. Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. The samples were carefully collected and stored in plastic bags. To minimize the risk of contamination, all glass wares were immersed in 10% HNO_3 for 24 h, washed with distilled water before use. The control solutions containing known concentration of the target elements were analyzed in between the analysis of samples to verify the accuracy of the instrument. A recovery test was carried out by spiking pre-analyzed samples. Each of the target elements was spiked at 50% of the initial concentrations of the respective element in the original sample. The spiked samples were treated as that of the respective samples and then reanalyzed. The spike recovery values of the elements were calculated as:

$$\%\ \text{recovery} = \frac{\text{amount in the spiked sample} - \text{amount in the unspiked sample}}{\text{added amount}} \times 100$$

Accepted recoveries ranged from 85% to 105%, and batches with recoveries less than 85% were reanalyzed. Excel 2007 was used for summarizing the raw.

RESULTS AND DISCUSSIONS

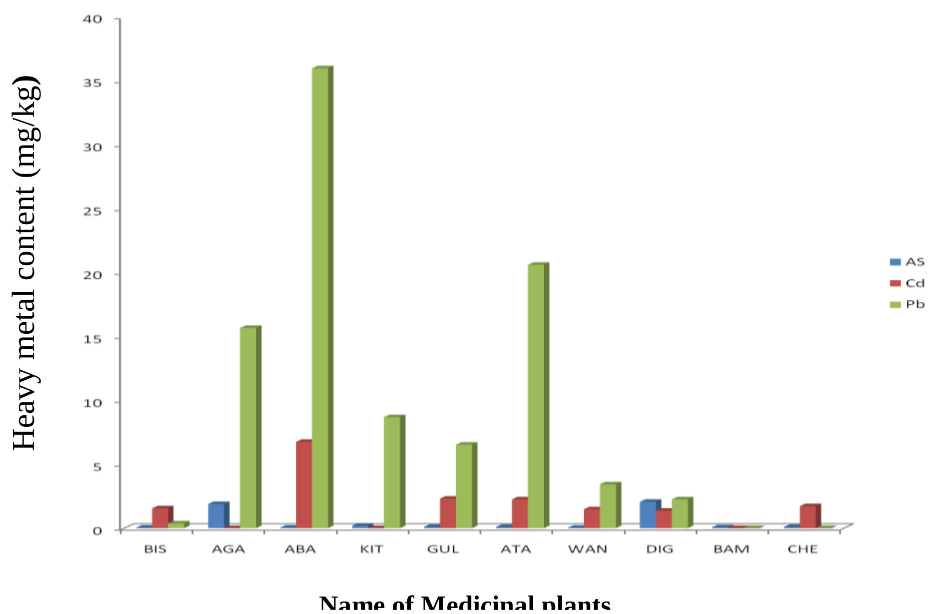
In this study, levels of As, Pb and Cd in the plant samples were analyzed and the result is shown in Table 2. All the 10 analyzed samples were found to contain detectable levels of all metals investigated.

Table 2: Heavy metal content of medicinal plants in mg per Kg of sample in dry weight

Sample code	**Heavy metal content (mg/kg ± SD)**		
	AS	**Cd**	**Pb**
BIS	0.037± 0.002	1.55±0.01	0.37±0.002
AGA	1.89±0.006	0.01±0.002	15.66±0.013
ABA	0.04±0.001	6.75±0.03	35.97±0.011
KIT	0.19±0.004	0.01±0.001	8.68±0.014
GUL	0.10±0.002	2.31±0.007	6.53±0.013
ATA	0.09±0.001	2.25±0.004	20.60±0.018
WAN	0.03±0.001	1.48±0.009	3.43±0.011
DIG	2.06±0.008	1.37±0.003	2.25±0.012
BAM	0.06±0.003	0.001±0.0003	0.002±0.0001
CHE	0.09±0.002	1.72±0.004	0.002±0.0001
MPL*	1	0.3	10

*MPL – Maximum permissible limit

Regarding the relative heavy metal content of the samples, lead (Pb) was found to be detected in higher concentration than arsenic (AS) and cadmium (Cd) in most of the samples (Fig. 1).



Medicinal Plants

Figure 1: Heavy metal content of selected medicinal plants

In this study, the concentration of Arsenic ranged from 0.03 to 2.06 mg/ kg of samples. The minimum was detected in WAN, whereas, the maximum was detected in DIG (Fig. 2). From the total medicinal plants analyzed, most of them (80%) contain arsenic in the level which is far below the maximum permissible limit specified by WHO (1 mg/kg). This imply that they are safe to be used at least with respect to As. However, two of the samples namely DIG and AGA contain arsenic in the level above the permissible limit, which may cause health hazard.

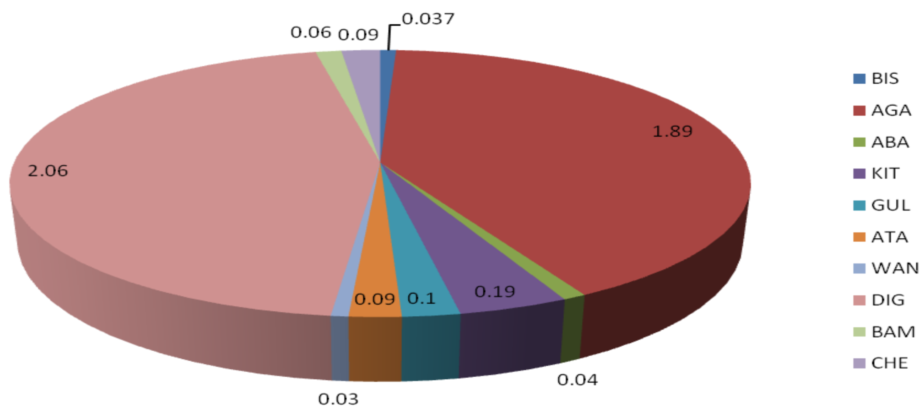


Figure 2: Arsenic (As) content of medicinal plants in mg/ kg of samples

The level of Cadmium in the analyzed medicinal plants ranged from 0.001 to 6.75 mg/kg of samples. The maximum and minimum content of Cd was detected in ABA and BAM, respectively (Fig. 3). Most of the medicinal plants (70%) analyzed contained Cd very far above the maximum WHO permissible limit (0.3mg/kg) showing there is significant contamination by cadmium. The most probable cause of this Cd contamination is extensive fertilizer use in the area. Fertilizers, especially phosphate based ones like DAP obtained from natural phosphate rock contain significant amount of Cd as impurity. Once Cd dispersed in the environment, it can persist in soils and sediments for decades.

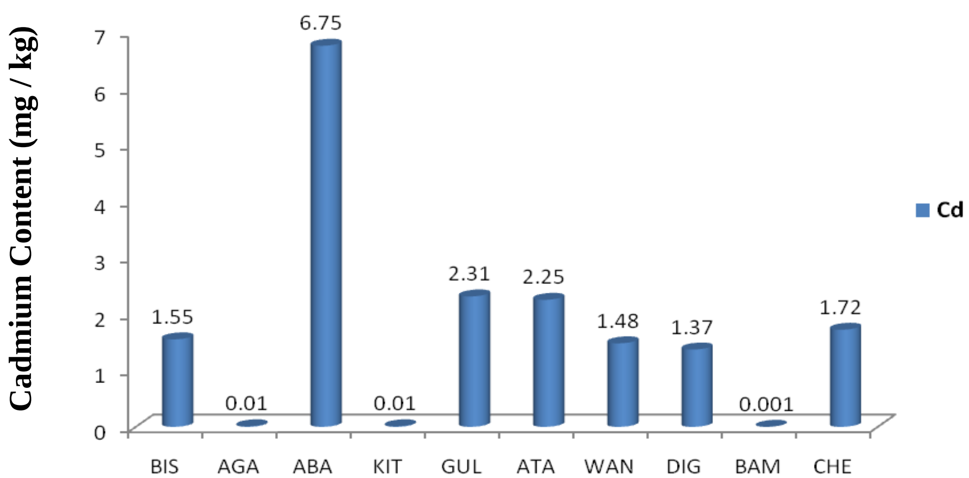
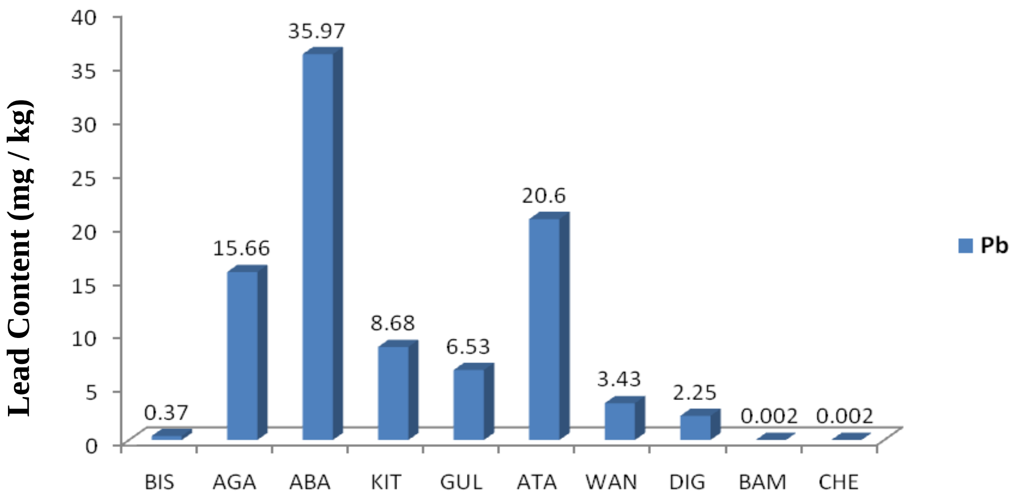


Figure 3: Cadmium content of medicinal plants in mg/ kg of samples

The concentration of lead (Pb) in the samples ranged from 0.002 to 35.97mg/ kg (Fig.4). The maximum was detected in ABA while the minimum was detected in both CHE and BAM. The minimum detected concentration of Pb in this study is below when compared to other research works of Ong *et al.*, in

Malaysia (5.59-63 µg/g)[13], Jeoma *et al* in Nigeria (1.55-10.47)[8] and Henok *et al* in Ethiopia (0.34 -98.19mg/kg) [14] while the maximum concentrations were higher than the work of the above researchers. Concentrations of Pb in ABA, AGA and ATA were observed to exceed the maximum permissible limit (10mg/kg) as specified by WHO (2005) showing obvious signs of environmental contamination. This environmental contamination by lead could be most probably from fuel exhausts as the study site is near to the heavy traffic road between Addis Ababa and Gondar. It cloud be also attributed by extensive chemical fertilizers used in the area.



Name of Medicinal Plants

Medicinal Plants

Figure 4: Lead content of medicinal plants in mg/ kg of samples

CONCLUSION

Over all, the present study showed that out of ten samples analyzed two for Arsenic (20%), seven for cadmium (70%) and three for lead (30%) of the analyzed samples were found to contain heavy metal concentrations above maximum WHO permissible limit (1, 0.3 and 10 mg/kg, respectively). This shows possible risk of toxic effects of heavy metals in the studied medicinal plants that are used in Gondar area district. Therefore, due attention should be given by both the users and responsible governmental bodies.

REFERENCES

[1]. Tesfaye A, Sebsebe D. Ethnobotanical study of medicinal plants in Kafficho people, Southwestern Ethiopia. Institute of Biodiversity Conservation, Ethiopia; 2009. P. 711-26.

[2]. World Health Organization. WHO guidelines for assessing quality of herbal medicines with reference to contaminants and residues. World Health Organization, Spain; 2007. P. 1-105.

[3]. Kirmani M, Mohiuddin S, Naz F., Naqvi I, Zahir E. Determination of some toxic and essential trace metals in some medicinal and edible plants of karachi city. *J Basic & Appl Sci* 2011; 7: 89-95.

[4]. Gidey Y. Assessment of indigenous knowledge of medicinal plants in Central Zone of Tigray, Northern Ethiopia. *African Journal of Plant Science* 2010; 4: 6-11.

[5]. Kebede D, Alemayehu A, Binyam G, Yunis M. A historical overview of traditional medicine practices and policy in Ethiopia. *Ethiop J Health Dev* 2006; 20:127-134.

[6]. Princewill-Ogbonna IL, Ogbonna PC. Heavy Metal Content in Soil and Medicinal Plants in High Traffic Urban Area. *Pakistan Journal of Nutrition* 2011;10: 618-24.

[7]. Hina B, Rizwani G H, Naseem S. Determination of toxic metals in some herbal drugs through atomic absorption spectroscopy. *Pak J Pharm Sci* 2011;24:353-58.

[8]. Justin D. Complementary Medicine and Heavy Metal Toxicity in Australia. *WebmedCentral Toxicology* 2010; 1(9):1-7.

[9]. Shehu A, Mullaj A, Harizaj F, Shehu J. Assessment of Heavy Metals Accumulation by Different Spontaneous Plant Species Grown along Lana River, Albania. Department of Chemistry, Faculty of Natural Sciences, University of Tirana, Tirana, Albania, 2011.

[10]. Saied S, Zahir E, Siddique A. Heavy metal levels in commonly used traditional medicinal plants. J Chem Soc Pak 2010; 32: 737-43.

[11]. Shafii K, Fazliana MS, Shamsiah AR. Preliminary Toxicology Evaluation and Heavy metal Determination of Selected Malaysian Medicinal Plants. *Health and the Environment Journal* 2011;2: 6-8.

[12]. Street RA. Heavy metals in South African medicinal plants. Research Centre for Plant Growth and Development, School of Biological and Conservation Sciences, University of KwaZulu-Natal, Pietermaritzburg, PhD dissertation paper, 2008.

[13]. Ong GH,Yap CK, Maziah M, Tan SG. Heavy metal accumulation in a medicinal plant *Centella asiatica* from Peninsular Malaysia. Journal of Biomedical Sciences 2011; 11:146-155.

[14]. Henok B, Ariaya H. Lead and Cadmium Accumulation in Medicinal Plants Collected from Environmentally Different Sites. *Bull Environ Contam Toxicol* 2010; 84:197–201.