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Assessment of Heavy Metal Content in Herbal Medicines Used in Pakistan by Atomic Absorption Spectroscopy

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Abstract

Herbal medicines are being regarded with great favor and are appealing for the benefit of the people all over the world. World Health Organization gave importance to the maintenance of standards of herbal medicines specially to test the presence of heavy metals in them. In this research project, branded herbal medicines have been analyzed for the assessment of various heavy metals. The samples were collected from local market of Faisalabad, Punjab, Pakistan. These medicines were prepared by popular herbal companies of Pakistan and the products are famous among the public. The quantity of the medicine analyzed, were in the form of capsules, pallets, syrups, and pastes (majoons). The medicines were manufactured for analysis by wet digestion method treating them with nitric acid and perchloric acid. The prepared samples were analyzed by atomic absorption spectrometer (AAS) using graphite tube atomizer and equipped with an injection system of MHS-10. The results exhibited that most of the samples crossed the permissible levels of heavy metals such as cadmium (95 %), lead (95), chromium (90 %) and nickel (90 %). However, the quantity of the other metals (Cu, Zn and Fe) were not enough to be harmful for human ingestion in several samples. Few herbal medicines like Ambreena tonic, Hab moqal khas, Ireefal zamani, Zarjam, Majoon sang danamurgh, Allergex and Akseer e jigar demonstrated harmful quantities of almost all the heavy metals analyzed. The results indicated that there is a great requirement to establish a suitable system of maintenance of quality that can elaborate the public for the validation of herbal medicines and make sure the implementation of old laws and standard procedures for public safety.

Keywords: AAS; Heavy metals; Herbal; Majoons; Toxic

Introduction

The use of herbs and plants for medicinal purposes and the study of their use is called Herbalism or Herbology. Herbs are the basic source of herbal medicines but there are many herbs that are thought to have adverse effects on human health. [1]. Many herbal plants have been used for herbal medicines since ancient times such as *Aloe vera*, Marshmallow, Great burdock, Pot marigold, Gota kola, Chinese Yam, Siberian ginseng and Tea tree etc. Herbal products are used for various ailments as in Pakistan many branded herbal medicines are used by the people such as Anab is used for blood purifier, Arq-mako is anti-inflamatory, Dinar used for liver disorder, Saduri and Toot siah is used to treat cough etc. [2].

There are many advantages and disadvantages of herbal medicine. Before treating yourself with herbs or seeking advice from a qualified herbalist or naturopath, consider the pros and cons of herbal treatments and remedies [3].

Advantages: There are many advantages of herbal products such as their cost-effectiveness as compared to allotropic medicines, these have very little side effects, these have very easy access to a common person, these can be brought without a prescription etc.

Disadvantages: Herbal treatment is time consuming and requires patience, herbs contain various elements which they absorb through roots from soil, it may be noticeable that body chemistry

of patients could not matches with the ingredients of the herbal products so it may cause allergy [4].

In spite of the advantages and benefits of the herbal medicines there are many reports that shows that herbal medicines have been contaminated with heavy metals. As the metals have low excretion rate when excreted into the kidney which have damaging effect on humans, even at very low concentrations. Heavy metals have a tendency to accumulate in the food chain, however, an increase in their uptake can become toxic. The main intention of current research is to investigate the content of heavy metals in herbal medicines due to these reasons. Other aim of this study is the inherent capacity for coming into being the risks of heavy metals present in herbal medicines based on the World Health Organization (WHO) standard limits.

Materials and Methods

Materials

Herbal products were used in this research to assess the heavy metal concentration for the presence of cadmium, mercury, lead, arsenic, cuprum and zinc from their solutions prepared in distilled water. The quantity of herbal medicine analyzed were in the form of capsules, pallets, syrups, and pastes (majoons) frequently used in Punjab, Pakistan. The medicines were manufactured for analysis by wet digestion method treating them with nitric acid and perchloric acid.

Equipments and Instruments

Atomic Absorption Spectrometer (JIEBO, AA320N), Electrical oven (memert Celsius 2005), Hot plate (Lab Tech), Electrical balance (G.C 2102), Grinder (Frith pulverize He.2), De-ionizer, Whatman filter paper, Fume Hood.

Chemicals

Nitric acid (HNO_3), Perchloric acid ($HClO_4$), Hydrochloric acid (HCl), Hydrogen peroxide (H_2O_2), Chromic acid H_2CrO_4, Cadmium Nitrate $Cd(NO_3)_2$, Zinc granules Zn, Ferrous Ammonium Sulphate $(NH_4)_2.Fe(SO_4)_2.6H_2O$, Manganese chloride $MnCl_2.2H_2O$, De-ionized water. All reagents were analytical grade and contain very low concentration of trace metals.

Sampling of Herbal Medicines

The branded herbal medicines were purchased from Jhang Bazar, a local market in the district of Punjab, Pakistan. The samples were mostly syrups and tablets as shown in Table 1. Only those samples were collected for the study which were famous among the public and extensively used as over the counter (OTC) products. OTCs are those medicinal products which can be used without the prescription of physician or pharmacist. The formulations collected were mostly manufactured by Qurshi, Hamdard manufacturer and some other well-known herbal industries.

The list of the twenty sampled herbal medicines is given below in the table.

No. of obs	Herbal Medicine	Batch Number	Brand name/ Company	Usage	Quantity
1	Ahmareen	171214	Qarshi	Liver tonic	10 mL BD
2	Akseer e Jigar	210116	Qarshi	Liver disorders	10 mL TDS
3	Allergex	111215	Qarshi	Allergies	5 mL BDS
4	Ambreena tonic	131015	Qarshi	Digestive, appetite stimulant	10-15 mL BD
5	Bukhareen	251017	Qarshi	Malaria, fever	2 tab TDS
6	Corezcol	251215	Qarshi	Whooping cough	5-10 mL TDS
7	Deedani	241012	Qarshi	Worms killer	5g OD
8	Epitize	1904	Himont	Eppetite stimulant,	15ml TDS
9	Garlicare	1	Hamdard	Heart disorders	1 tab TDS
10	Hab Moqil khas	251114	Qarshi	Acidity, stomach burns	2 tab TDS
11	Habis	300316	Qarshi	Blood coagulation	5-10 mL TDS
12	Itreefal zamani	L12-019	Hamdard	Constipation and headache	5 mL OD

13	Jawahar mohara khas	241114	Qarshi	Heart, brain and nerves disorders	1 cap BDS
14	Khameera Abresham	8	Ajmal	Protects vital organs	5 mL BDS
15	Majoon sang dana murgh	311215	Qarshi	Strengthen digestive system	5 mL TDS
16	Majoon suranjan	290216	Qarshi	Goat, relieve ankle and toe pain	5-10 mL BDS
17	Taryaq e nazla	261142	Qarshi	Acute catarrh, sore throat, cough	5-10 mL BDS
18	Zanjabeen	11114	Qarshi	Hyperacidity, nausea, vomiting	2 cap BDS
19	Zarjam	170415	Qarshi	Sexual debility	1 cap TDS
20	Zubex	30216	Qarshi	Anti-diabetic	1 tab TDS
Key: tab = tablet, OD = once daily, BD= twice daily, TDS= thrice daily					

Table 1: Basic information about the herbal medicines.

Results

The level of concentration of heavy metals in the herbal medicines are given in Table 2. All the samples except Itreefal zamani (4.3 ppm) exceeded the permissible limit for lead (5 ppm). Cr was analyzed in 90 % of the samples to have crossed the admissible amount (30-35 ppm) whereas Ni concentration exceeded the permissible limit (1.5 ppm) in 18 samples except Allergex and Epitize. In the case of cadmium, 19 products (i.e., 95 %) go above the admissible range (0.5 ppm). Mn, Zn and Fe were discovered within the allowed range in all the samples.

No. of obs	Medicine	Pb	Cd	Cr	Cu	Ni	Zn	Fe
1	Ahmareen	27.2 ± 0.01	6.2 ± 0.02	120.8 ± 0.01	5.1 ± 0.03	8.3 ± 0.00	7.6 ± 0.01	19.3 ± 0.02
2	Akseer e jigar	29.5± 0.02	8.2± 0.01	135.4± 0.00	3.6 ± 0.01	13.2 ± 0.02	7.3 ± 0.01	17.1 ± 0.02
3	Allergex	23.4±0.01	7.1±0.00	136.3±1.30	ND	ND	7.3±0.01	9.1 ± 0.01
4	Ambreena tonic	31.7± 0.01	9.3±0.01	141.2± 0.06	ND	17.9 ± 0.00	16.5± 0.03	20.3 ±0.04
5	Bukhareen	25.4± 0.01	9.5± 0.05	131.3± 0.07	2.9± 0.00	7.9 ± 0.02	7.4 ± 0.00	17.3 ± 0.01
6	Corezcol	29.2± 0.01	7.3± 0.03	121.7± 0.08	5.3±0.01	7.4±0.00	19.3± 0.01	20.5 ± 0.03
7	Deedani	31.0±0.04	7.9± 0.00	137.2±0.05	3.0±0.00	7.5 ± 0.01	9.7 ± 0.00	21.3 ±0.04
8	Epitize	21.9± 0.05	4.3±0.00	121.7± 0.06	3.5±0.01	ND	8.1 ± 0.00	25.3 ± 0.07
9	Garlicare	25.6± 0	5.1± 0.01	130.8± 0.02	ND	5.4 ± 0.00	7.9 ± 0.02	11.9 ±0.01
10	Hab Moqal khas	31.2± 0.03	6.9±0.03	81.5 ±0.02	15.0± 0.03	7.3 ± 0.00	24.5± 0.07	601.8± 1.09

11	Habis	21.6± 0.06	3.9± 0.00	121.3± 0.07	4.3± 0	6.7 ± 0.01	ND	9.6 ± 0.01
12	Itreefal Zamani	4.3± 0	83.2± 0.04	9.5± 0.02	6.4±0.00	43.4± 1	51.8± 0.9	29.8± 2.02
13	Jawahar mohara khas	30.2± 0.03	5.1± 0	129.8± 0.04	5.9± 0	7.6 ± 0.00	34.4± 0.41	550.4± 1.25
14	Khameera Abresham	21.7± 0.03	3.2± 0	124.6± 0.05	14.1± 0.01	51.3 ±0.01	15.1± 0.01	601.3± 1.01
15	Majoon Sangdana	23.9± 1.01	6.8± 0.05	140.3± 0.05	7.8± 0	8.9±0.00	9.2 ±0.02	151.5± 1
16	Majoon suranjan	27.2± 1.03	6.7± 0	127.7± 0	4.1 ±0.00	11.3 ± 0.09	13.4 ± 0.21	75.7± 0.70
17	Taryaq e nazla	21.1± 1.04	ND	121.7± 0	4.9±0	4.5 ±0.02	9.7 ± 0.05	ND
18	Zanjabeen	23.9± 1	7.1± 0.01	132.4± 0	ND	7.1± 0.09	5.2± 0.01	17.5 ± 0.11
19	Zarjam	27.1± 1.04	81.0± 0.03	9.7± 0.02	7.3± 0.11	21.4± 0.11	31.8± 0.02	401.2± 0.6
20	Zubex	23.9±0.05	5.9± 0.08	132±1.03	ND	9.4± 0.16	9.6 ± 0.01	23.8 ±0.01

Table 2: Concentration of heavy metals in herbal medicines sampled from Punjab, Pakistan.

The highest Pb concentration was found in Ambreena tonic (31.7 ppm) followed by Hab moqil khas (31.2 ppm), Deedani (31.00 ppm), Jawahar mohara khas (30.2 ppm) and Akseer e jigar (29.5 ppm). The concentration of Cd was 3.2 - 83.2 ppm with the highest concentration in Itreefal zamani (83.2 ppm), followed by Zarjam (81 ppm), Bukhareen (9.5 ppm), Ambreena tonic (9.3 ppm), Akseer e jigar (8.2 ppm), Deedani (7.9 ppm) and Majoon sang dana murgh (6.8 ppm). These values depict that all the samples crossed the upper limits of the admissible range for Cd.

The highest amount of Cr was found in Ambreena tonic (141.2 ppm) followed by Majoon sang dana murgh (140.3 ppm), Deedani (137.2 ppm), Allergex (136.3) and Akseer e jigar (135.4 ppm). The highest amount of Cu was discovered in Hab moqil khas (15 ppm) followed by Khameera abresham (14.1 ppm), Majoon sang dana murgh (7.8 ppm), Zarjam (7.3 ppm), Itreefal zamani (6.4 ppm), Jawahar mohara khas (5.9 ppm), and Ahmareen (5.1 ppm). Highest concentration of Iron was found in Hab moqil khas (601.8 ppm) and lowest concentration was found in Allergex (9.1 ppm).

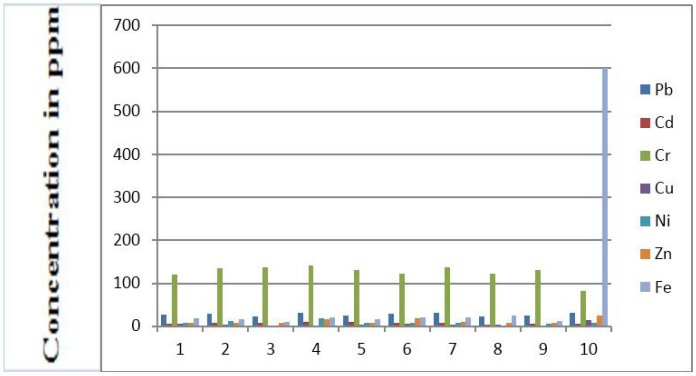

Figure 1: Heavy metal contents in first ten herbal medicines (as found on table. 2).

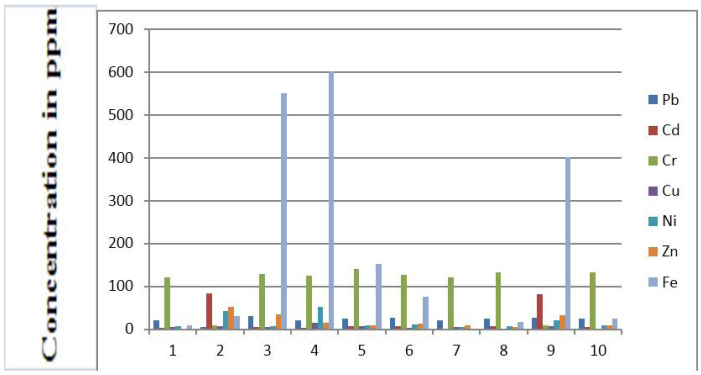

Figure 2: Heavy metal contents in last ten herbal medicines (as found on table. 2).

Discussion

The following advocated daily limits of some of the significant metals will help in the findings of this research study are given table 3.

Heavy Metal	Advocated daily limit	Reference
Cd	70 µg	(Obi et al., 2006) [5]
Cu	340-400 µg (child), 900 µg (adult)	(IOM, 2001; ATSDR, 2008) [6,7]
Cr	11-25 µg (child), 30 -35 µg (adult)	(IOM, 2001; ATSDR, 2008). [6,7]
Fe	8-10 mg	(IOM, 2001) [6]
Mn	8-11 mg	(IOM, 2001) [6]
Ni	35 µg	(Forstner & Wittmann, 1981) [8]
Pb	20-514 µg	(Obi et al., 2006) [5]
Zn	3-8 mg	(IOM, 2001) [6]

Table 3: Recommended daily limits for some metals.

Lead

Lead is one of the most toxic among toxic heavy metals. It enters the human body in various ways including inhalation, drinking water, ingestion of food polluted with lead and absorption by skin. It accumulates in the body has no known useful biological function [9]. It has adverse effects on various body systems such as the reproductive, renal, digestive, cardiovascular and immunological systems [10]. The concentration of lead in the tested products was in the range of 4.3 - 31.7 ppm. The recommended limit for finished herbal products is 5 ppm while for crude herbal material it is 10 ppm [11]. Since our study was conducted on finished herbal products, we consider the 5 ppm as the applicable limit. Thus, only one product out of the twenty samples assessed had lead concentration below the recommended limit. It is clear, therefore, that these products are potentially toxic to man. Incidentally, all these products are very popular among the people and adult and children chronically use some of them such as Hab e moqal khas and Majoon suranjan. There is a large body of data on the adverse effect of lead in herbal formulations human health [12].

Cadmium

Cadmium is a toxic metal of occupational and environmental concern. Cadmium exposure leads to a variety of adverse effects due to its extremely long biological half-life. The rate of cadmium transfer from soil to plants is very high and the metal is considered to have carcinogenic effects on liver, pancreas and stomach [13]. Its recommended concentration in plant is 3 ppm while for finished plant products, it is 0.5 ppm [11]. The recommended consumption of cadmium is 70 µg/day [5]. Based on this, 19 products out of 20 exceeded the permissible limit. The highest Cd consumption are: Itreefal zamani (83.2 ppm), followed by Zarjam (81 ppm), Bukhareen (9.5 ppm) and Ambreena tonic (9.3 ppm).

Chromium

Chromium is exists in two ionic forms, trivalent and hexavalent, the latter being toxic and is correlated to human carcinogenesis and acute toxicity of aquatic organisms [14]; on the other hand, its reduced form (trivalent) is an essential element for animals. The concentration of chromium was in the range of 9.5 - 141.2 ppm. There is no recommended limit for chromium in herbal finished products. The recommended limit of chromium is 11 - 25 µg/day for children and 30 - 35 µg/day for adults [6,7]. Highest consumption was observed for Ambreena tonic (141.2 ppm). Only two products had chromium concentration below the recommended limit. Such a high concentrations could be toxic due to the chronic use of some of the tested products.

Copper

Copper has both beneficial and toxic effects depending on its level of consumption. Monitoring of copper limit is essential as it is beneficial in low concentrations but exhibit various toxic effects above the safety limit. In this study, copper was found in the products in the concentration range of 2.9 - 15 ppm. The recommended consumption of copper is 900 and 340 - 400 µg/day for adult and for child, respectively [6,7]. Thus, all the tested products were safe with regard to copper content.

Nickel

Nickel is abundant in nature. Possible exposure sources of nickel are food, drinking water, absorption by contact and inhalation in nickel-polluted area [15]. Ingestion of large amounts of nickel affects stomach, liver, kidneys, immune system and reproduction in rats and mice [15]. The most common toxic effect is dermatitis

while lung cancer has also been reported in some studies [15]. Nickel was found in a concentration range of 4.5 - 51.3 ppm. The highest concentration was found in Khameera Abresham (51.3 ppm), followed by Itreefal zamani (43.4 ppm) and Ambreena tonic (17.9 ppm). The recommended consumption limit of nickel is 35 µg/day [8]. Thus, almost all the tested samples exceeded the daily allowed limit for nickel. The contents of Mn, Zn and Fe in the tested products were within their permissible limits, they did not exceed the daily consumption limits of these metals.

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