

Original Article

A Quantitative Investigation on Some Toxic and Non-toxic Metals in Popular Medicinal Herbs in Iranian Market

Fazel Shamsa^a*, Shamsali Reza Zadeh^b, Hashim Shamsa^a and Khosro Abdi^a

^aDepartment of Pharmaceutical Chemistry, Faculty of Pharmacy, Tehran University of Medical Sciences, Tehran, Islamic Republic of Iran. ^bMedicinal Herbs Research Center, Jihad Institute, Tehran, IR Iran.

Abstract

This study was performed to explore the presence of heavy metals in some popular herbal medicines of the Iranian market. Primarily, this study was planned to do the task by the USP method. In the second step, those plants which contained more than 10 ppm of total heavy metals were analyzed by atomic absorption method. In this study, 43 popular herbal medicines were investigated according to the limit test procedure in the USP. It was found that 27 of these plants contained less than 10 ppm heavy metals compared to a standard lead solution, while 16 contained more than 10 ppm. The presence of Pb (<2.5), Cd (<0.25), Co (<1) and Ni (<1.5) could be considered safe.

Keywords: Popular herbal medicines; Toxic metals; Non-toxic metals; Iranian market; Quantitative analysis.

Introduction

During recent decades, there has been an increasing attention using herbal products for combating diseases. One of the reasons that people prefer herbal medicines over modern chemicals is their low price, while another reason is their safety compared to the chemical drugs, ignoring the fact that there are chemical materials in plants as their active ingredients. Because the plants are directly in contact with air, water and soil, the constituents of these sources might contaminate the plants. In addition to toxic elements such as mercury, arsenic, lead, nickel and cadmium which might be present in some plants and threaten the consumer health, especially the children and elderly, useful elements such as calcium, magnesium, zinc,

manganese and iron are also usually present in plants, which helps the good health. Many countries have already evaluated their popular herbal medicines with regard to toxic heavy metals (1-5).

Therefore, a study was undertaken to evaluate the herbal medicines in Iranian market with regard to the toxic and nontoxic metals, according to USP, by using atomic absorption spectrometry.

Experimental

Materials

Herbal medicines

The popular plants used in this study were purchased from herbal medicines stores in Tehran and were diagnosed by a botanist.

Chemicals

Sulfuric acid, nitric acid, hydrogen peroxide,

* Corresponding author:

E-mail: shamsa@sina.tums.ac.ir

Table 1. Common and scientific names of the popular Iranian herbal medicines under study with less than 10 ppm of total heavy metals based on USP method.

	Common name	Scientific name	AM*	AP*
1	Saatar	<i>Zataria multiflora</i> Boiss	+	+
2	Blond plantain	<i>Plantago Ovata</i> Forsk.	+	+
3	German madwort	<i>Dracocephalum moldavica</i>	+	-
4	Cinnamon	<i>Cinnamum zeylanicum</i> Nees	+	+
5	Santolin yellow	<i>Achillea santolina</i> L.	+	+
6	Maiden hair	<i>Adianthum capillus-veneris</i> L.	+	-
7	Common mallow	<i>Malva silvestris</i> L.	+	-
8	Will fumitory	<i>Fumaria parviflora</i> Lam	+	-
9	Licorice	<i>Glycyrrhiza glabra</i> L.	+	+
10	Fenugreek seeds	<i>Trigonella foenum graecum</i> L.	+	+
11	Fennel	<i>Foeniculum vulgare</i> Mill	+	+
12	Sebestan plums	<i>Cordia myxa</i> L.	+	-
13	Madwort and pepper weed	<i>Alyssum homolocarpum</i> Fisch C.A. Mey Boiss	+	-
14	Corn crest	<i>Zea mays</i> L.	+	+
15	Caster beans	<i>Ricinus communis</i> L.	+	+
16	Needle root	<i>Urtica dioica</i> L.	+	+
17	Chicory	<i>Cichorium intybus</i> L.	+	-
18	Common purslane seeds	<i>Portulaca oleracea</i> L.	+	-
19	Senna leaves	<i>Cassia angustifolia</i> Vahl.	+	-
20	Betony	<i>Stachys lavandulifolia</i> Vahl.	+	-
21	Camel's thorn mama	<i>Alhagi persarum</i> Boiss & Buhse	+	-
22	Olive leaves	<i>Olea europaea</i> L.	+	+
23	Coriander fruits	<i>Coriandrum sativum</i>	+	+
24	Black cumin	<i>Nigella sativa</i> L.	+	-
25	Damask rose	<i>Rosa damascene</i> Mill	+	+
26	Hawthorn	<i>Crataegus microphylla</i> C. Koch	+	+
27	Mint	<i>Mentha piperita</i> L.	+	+

AM*=Available in Iranian market, AP*=Available in Iranian pharmaceutical products.

thioacetamide, glycerin, acetic acid, ammonium acetate, and lead nitrate were bought from the Iranian agent of Merck Co.

Methods

Solution

Acetate buffer (pH=3.5), thioacetamide TS, thioacetamide-basic glycerin TS, and glycerin basic were prepared according to USP.

Standard lead solution

Lead nitrate (160 mg) was dissolved in double distilled water (ddW) and nitric acid (6N, 1 ml) and was made to volume (100 ml). One ml of

this solution was diluted to 100 ml with ddW to contain 10 µg/ml of Pb.

General screening method

The 3rd method of USP was applied for monitoring the total heavy metal content in herbal medicines under study. In this method, three different solutions should be prepared, i.e., a standard, a monitor and herbal sample solutions.

Standard solution

A mixture of sulfuric acid (8 ml) and nitric acid (10 ml) was transferred to a Kjeldal flask

Table 2. Popular Iranian medicinal herbs which contain more than the allowed limit* (10 ppm) of heavy metals based on USP method.

	Common name	Scientific name	Total heavy metals, ppm
1	Staechus	<i>Nepeta menthioides</i> Boiss & Buhse	20
2	Wonderberry	<i>Solanum nigrum</i> L.	20
3	Flixweed seeds	<i>Descurainia Sophia schur</i> L.	30
4	Celery seeds	<i>Apium graveolens</i> L. var. duke	25
5	Small caltrops	<i>Tribulus terrestris</i> L.	12
6	Savory seeds	<i>Satureja hortensis</i>	20
7	Indian valerian	<i>Nordostachys jatamansi</i> DC.	15
8	St. John's wort	<i>Hypericum perforatum</i> L.	15
9	Violet flower	<i>Viola odorata</i> L.	30
10	Saf flower	<i>Carthamus tinctorius</i> L.	30
11	Sesame seeds	<i>Sesamum indicum</i> L.	25
12	Sage	<i>Salvia officinalis</i> L.	20
13	Anise	<i>Pimpinella anisum</i> L.	15
14	Wild caraway	<i>Bunium persicum</i> (Boiss) B. Fedisch	20
15	Wild chamomile	<i>Tripleurospermum disciforme</i> (C.A. Mey) Schutz-Bip	15
16	Coriander fruits	<i>Coriandrum sativum</i> L.	20

*Allowed limit in this study is defined as the amount of heavy metals equivalent to the USP limit for acetaminophen tablet (10 ppm), which is equivalent to 0.001%.

(100 ml). The mixture was heated until the appearance of white vapors. Double distilled water (10 ml) was added cautiously and boiled until a white vapor appeared. After cooling, ddW (5 ml) was added to reach a volume of 2-3 ml, mixed and boiled. After cooling, ddW (5 ml) and lead standard solution (2 ml, 20 µg Pb²⁺) were added and mixed. For color comparison the mixture was transferred to a 50 ml test tube (TT), the flask was washed, transferred to the TT and the volume is made to 25 ml.

Preparation of the herbal product under study for heavy metal limit test

Herbal sample (2.0 g) was transferred to a dry Kjeldal flask, moistened by a mixture of sulfuric and nitric acids (8:10 v/v) and heated gently until the reaction started. This process was repeated until 18 ml of the acid mixture was consumed. The solution was boiled gently until the appearance of a deep color, and then cooled. Nitric acid (2 ml) was added, and re-heated until the solution became deeply colored. Heating and adding nitric acid were repeated until the solution was no more deeply colored. After cooling, ddW (5 ml) was added and boiled until the appearance of white vapors.

The volume was reduced to 2-3 ml and then cooled. If the solution was colored yellow, it should have been decolorized using hydrogen peroxide. The solution was transferred to a 3rd TT, the flask was rinsed by ddW and transferred to the same TT and the volume was made to 25 ml.

Monitor solution

The solution was prepared and processed similar to the preparation method of the herbal product under study and was then cooled and diluted with ddW. Then, the standard lead solution (2 ml, 20 µg) was added and mixed. The mixture was transferred to the 2nd comparison TT, the flask was washed and transferred to the same TT, and made to 25 ml by ddW.

Heavy metal levels of the herbal products under study

The pH values of the monitor, standard and herbal solutions were adjusted to 3-4 using dilute ammonia solution. The volumes were made to 40 ml by ddW, acetate buffer (2 ml, pH=3.5) and thioacetamide glycerin base TS solution (2 ml) were added, to each TT, the volumes were made to 50 ml by ddW, and left for

Table 3. The percentage of ash after ignition of the aqueous extracts of the samples under study (2 g original sample) at 400-500 °C.

Common name	Scientific name	Ash %
Staechus	<i>Nepeta menthoides</i> Boiss & Buhse	9.70
Wonderberry*	<i>Solanum nigrum</i> L.	10.75
Flixweed seeds*	<i>Descurainia sophia</i> schur. (L.)	12.00
Celery seeds	<i>Apium graveolens</i> L. var. duke	5.55
Small caltrops	<i>Tribulus terrestris</i> L.	14.70
Savory seeds	<i>Satureja hortensis</i>	17.50
Indian valerian	<i>Nordostachys jatamansi</i> DC.	3.00
St. John's wort	<i>Hypericum perforatum</i> L.	5.50
Violet flower	<i>Viola odorata</i> L.	8.25
Saf flower	<i>Carthamus tinctorius</i> L.	10.15

*2 g of these samples was directly ignited at 400-500°C after mixing with ddW (5 mL).

2 min. By watching of comparing the solutions against a white surface, the test solution color intensity should not exceed that of the standard and monitor solutions.

Sample preparation evaluation of toxic and non-toxic metals in herbal products

The herbal products under study (2-5 g) was moistened with 30 ml of ddW and boiled for 20 min. After filtration, the extraction was repeated twice, and the extracts were combined and concentrated 10 ml. The solution was transferred to a crucible and dried by gentle heating. The crucible was left for 2 h at 400-500°C in a furnace. After weighing the ash (Table 3), it was dissolved in hydrochloric acid 20 ml,

transferred to a 50-ml volumetric flask and made to volume by ddW. Individual metal content of this solution were analyzed by atomic absorption spectrometry.

Results and Discussion

For total heavy metal content in Iranian herbal medicines, the third USP method was successfully applied. Due to the presence of significant quantities of calcium, magnesium, sodium, and potassium, in the last step and before the addition of thioacetamide solution, the solutions should be filtered to make them transparent and clear. To these clear solutions, thioacetamide-glycerin basic TS solution was

Table 4. Estimation of the toxic and non-toxic metals in some Iranian herbal medicines. The numbers indicate µg/g or ppm.

Sample*	Fe	Zn	Cu	Mn	Mg	Ca	Pb	Cd	Co	Ni
1	20.4	11.8	ND	108	4060	75840	<2.5	<0.25	<1	<1.5
2	290.5	15.9	0.76	21.5	2150	16820	<2.5	<0.25	<1	<1.5
3	318.3	39.6	0.70	29.9	1848	24270	<2.5	<0.25	<1	<1.5
4	22.5	18.3	1.68	37.9	2546	32560	<2.5	<0.25	<1	<1.5
5	20.2	16.5	1.98	104.7	8428	70930	<2.5	<0.25	<1	<1.5
6	36.4	17.8	3.05	67.6	14918	78540	<2.5	<0.25	<1	<1.5
7	76.3	9.1	ND	80.8	1073	18660	<2.5	<0.25	<1	<1.5
8	16.0	28	0.43	34.5	1996	29960	<2.5	<0.25	<1	<1.5
9	44.3	39.2	ND	18.5	607.0	3270	<2.5	<0.25	<1	<1.5
10	30.8	38.5	0.03	13.5	465.8	6410	<2.5	<0.25	<1	<1.5

Sample*

1. Staechus; 2. Wonderberry; 3. Flixweed seeds; 4. Celery seeds 5. Small caltrops; 6. Savory seeds;
7. Indian valerian; 8. St.John's wort; 9. Violet flower; 10. Safflower

added and the turbidities of the solutions were compared. Those solutions with less turbidities than the standard and monitor solutions, were to contain less than 10 ppm of total heavy metals and (Table 1). Consumption of such products is considered safe. On the other hand, some of the herbal products showed more turbidities than the standard solution; therefore, they were considered to contain more than 10 ppm of total heavy metals (Table 2). These products were ignited, dissolved in hydrochloric acid, and analyzed by atomic absorption spectrometry. The results of this analysis are shown in Table 4. From this table, the followings could be concluded:

1. Toxic heavy metals including lead, cadmium, cobalt, and nickel are present in insignificant quantities.

2. Flixweed seeds contain the highest amounts of iron and zinc (Fe=318, Zn=39.6 ppm).

3. Calcium and magnesium are present in significant quantities in different herbal products. For example, Staechus contains 75 mg/g and 4 mg/g, Flixweed seeds contains 24 mg/g and 1.85 mg/g, Celery seeds contains 32 mg/g and 2.5 mg/g, Savory seeds contains 78.5 mg/g and 14.9 mg/g of Ca and Mg, respectively.

4. Copper contents were low and acceptable, while savory seeds and small caltrops contain the highest Cu contents, i.e. 3 and 2 ppm, respectively.

5. Some of the therapeutic activities of herbal products could be attributed to the presence of some metals such as iron (6), zinc (7), nickel (8, 9), manganese (10) and magnesium (11).

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