

## Determination of Contaminants in Some Iranian Popular Herbal Medicines

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### Abstract

**Introduction:** Medicinal plants have been used therapeutically all around the world, being an important aspect of various traditional medicine systems. However, the use of phytotherapy according to the efficacy and safety criteria is not sufficient to guarantee the quality of both the herbal medicine and its use.

**Objective :** The aim of this study was to determine the level of Cd, Pb, Ni, Cu, Hg and pesticide residues in some commonly used herbal medicines and herbal plants to assess the relative safety of these products based on world standard limits and potential health risk to local inhabitants.

**Materials & Methods:** 6 different medicinal herbal products and 6 herbal plants were purchased from Tehran's market. Certain weight of each samples were digested with nitric acid by wet digestion method for determining of toxic metals. Atomic absorption spectrometry (AA) was used for the determination of the concentration of Pb, Cd, Ni, Cu and an automated continuous – flow hydride vapor generation system was used for Hg and pesticide residues were analyzed by gas-chromatography – mass spectrometry after extraction performed using solid – phase micro extraction technique.

**Results:** Results were compared with the permissible limits (PI), acceptable daily intake (ADI) and provisional maximum tolerable daily intake (PMTDI) as set by world health organization (WHO), Food and Drug administration (FDA). Mercury, Lead and Cadmium were highly present in the majority of samples. In 100% of medicinal plant samples which brought randomly from the markets, the amounts of Diazinon, Fenitrothion, Malathion was higher than the allowed limits when compared to the MRL scale.

**Conclusion:** By a comparison between acceptable global standards and the level of Hg, Cd and Pb on investigated herbal medicine and medicinal plants, our results showed that the majority of medicinal plants samples had higher level of these heavy metals. Due to lack of research data and technical limitations at present, more research is needed in order to establish the scientific criteria for herbal medicines.

**Keywords:** Herbal medicine; Contamination; Heavy metals; Phytotherapy

### Introduction

Physical evidence of the use of herbal remedies has been found from some 60,000 years ago in a burial site of a Neanderthal man uncovered in 1960 in a cave in northern Iraq [1].

Medicinal plants have played a key role in world health. They are distributed worldwide, but they are most abundant in tropical countries. It is estimated that about 25% of all modern medicines are directly or indirectly derived from higher plants [2,3]. A vast number of plants have medicinal properties; in fact, many pharmaceutical drugs were originally derived from plants. Herbal materials and medicinal plants are also often used as food, functional food, nutritional or dietary supplements in many countries such as Iran.

Due to poverty and limited access to modern medicine, about four billion people, 80% of the world's population, are living in developing countries use herbal medicine as their source of primary health care [4-6]. Natural products in medicine constitute a vast array of "raw materials", making clear definitions important. Quality criteria are based on clear scientific definitions of raw material. The term "herbal drugs" denotes plants or plant parts that have been converted into phytopharmaceuticals by means of simple process involving harvesting, drying, and storage [7]. The quality of a plant product is determined by the prevailing conditions during growth, and accepted Good agricultural Practices (GAP) can control this. These include seed selection, growth conditions, use of fertilizers, harvesting, drying and

storage. In fact, GAP procedures are, and will be, an internal part of quality control. Apart from these criteria, factors such as the method of extraction, contamination with microorganisms, heavy metals, and pesticides can alter the quality, safety and efficacy of herbal drugs. Using cultivated plants under controlled conditions instead of those collected from the wild can minimize most of these factors [8,9].

By far the majority of potentially hazardous contaminants and residues are found in the herbs and herbal materials.

The objectives of this research was to determine the level of Cd, Pb, Ni, Cu, Hg in some commonly used herbal plants. In general determining undesired chemical contaminants and impurities in some commonly used Iranian herbal medicines by considering their dosages, quantities and frequency of their uses. The other matter of concern in this research is toxicological assessment of the trace heavy metals and comparing this data by the highest nationally recommended,

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authorized or registered uses and FAO/WHO evaluated safety of them and the joint FAO/WHO Expert Committee on Food Additives (JECFA) for contaminants in herbal medicines and in foods.

## Methods

A total of 8 commonly used medicinal plants were collected from Tehran's markets (Table 1). The samples were washed with deionized water and allowed to dry in oven for 72 hours at a temperature of 65°C. The samples were then ground and sieved through 0.5 mm sieve. The powdered samples then subjected to the acid digestion using nitric acid and perchloric acid [10].

250 mg of air-dried of each homogeneously medicinal plant samples accurately weighed and 1.0 mL of the digestion mixture ( 2 parts by weight of nitric acid & 1 part by weight perchloric acid ) and heated slowly by an oven and then rise the temperature. Dissolved the remaining dry inorganic residue in 2.5 mL of nitric acid and used for the determination of heavy metals. Atomic absorption spectroscopy (AAS) was used for the determination of the concentration of Pb, Cd, Ni and Cu; standardized international protocols were followed for the preparation of material and analysis of heavy metals contents. Analytical grade reagents and distilled water were used throughout the experiment. All glassware and plastic containers used were washed with liquid soap, rinsed with water, soaked in 10% volume- volume nitric acid for 24hrs, cleaned thoroughly with distilled water and dried in such a manner to ensure that any contamination does not occur. An automated continuous flow hydride vapor generation system was used for mercury and all samples were tested as quickly as possible after collection.

## Results and Discussion

A total of 6 elements (Zn, Ni, Cd, Cr, Cu and Pb) were determined in the powdered Iranian herbal plants samples and all samples were analyzed three times. Each data present mean content ± SD of 60 samples. All results were calculated on a dry weight basis (µg/g dry weight).

The results of analysis are shown in table 2 and figures 1 & 2.

### Pb

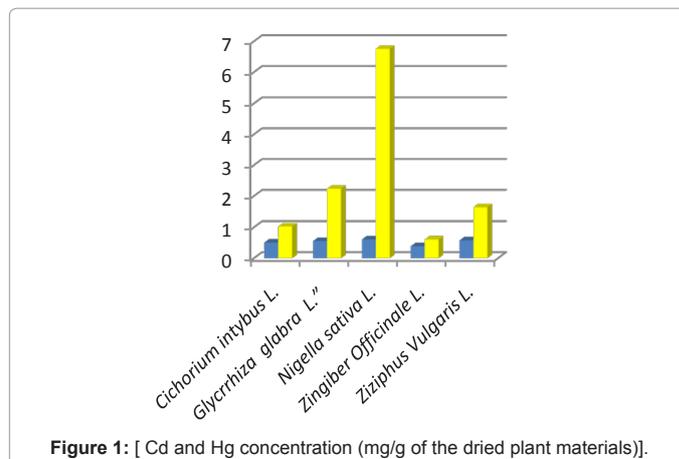
*Zingiber officinale* had the lowest Pb level and *Ziziphus vulgaris L.* the highest. The WHO maximum limit of Lead prescribed in herbal

Botanical name	Family	English name	Local name	Part used
<i>Cichorium intybus L.</i>	Compositae	Chicory	<i>Kasni</i>	Roots
<i>Cinnamomum L.</i>	Lauraceae	Cinnamon	<i>Darchin</i>	bark
<i>Glycyrrhiza glabra L.</i>	Leguminosae	Liquorice	<i>Shirin bayan</i>	Roots
<i>Nigella sativa L.</i>	Renunculaceae	Black Cumin	<i>Zirreh</i>	Seeds
<i>Zingiber Officinale L.</i>	Zingiberaceae	Ginger	<i>Zanjebil</i>	Fruits
<i>Ziziphus Vulgaris L.</i>	Rhamnaceae	Jujube fruit	<i>Annab</i>	Fruits

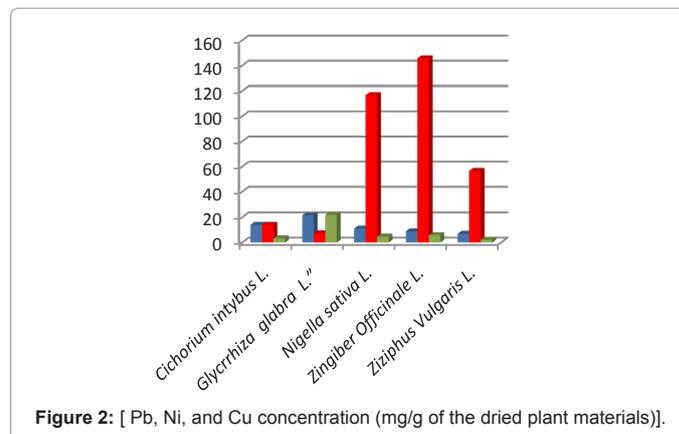
**Table 1:** Medicinal herbal plants analyzed in 2011 in Iran's market for heavy metals, local name, parts used

	Pb	Cd	Cu	Ni	Hg
<i>Cinnamomum L.</i>	14.22±0.23	1.03±0.05	14.2±3.3	3.49±0.06	0.51±0.03
<i>Cichorium intybus L.</i>	21.66±0.22	2.24±0.06	7.51±0.07	22.11±0.10	0.56±0.06
<i>Glycyrrhiza glabra L.</i>	11.23±0.16	6.76±0.25	117±25	4.87±0.16	0.61±0.04
<i>Nigella sativa L.</i>	9.06±0.03	0.61±0.03	146±45	6.03±0.10	0.39±0.04
<i>Zingiber Officinale L.</i>	7.06±0.21	1.64±0.11	56.8±3.4	2.37±0.61	0.58±0.05
<i>Ziziphus Vulgaris L.</i>	38.21±0.06	8.74±0.04	8.00±0.07	22.96±0.12	0.69±0.06

**Table 2:** Heavy metal in medicinal plants determined in 2011. [ metal concentration ( µg/g of the dried plant materials)]



**Figure 1:** [ Cd and Hg concentration (mg/g of the dried plant materials)].



**Figure 2:** [ Pb, Ni, and Cu concentration (mg/g of the dried plant materials)].

medicines and products is 10 mg/kg while the dietary intake limit for Pb is 3 mg/week [11]. The finding for Lead residue in medicinal herbal plants is much higher than acceptable global standards. Lead is known to cause neurological disorders, anemia, kidney damage, miscarriage, lower sperm count and hepatotoxicity in higher concentration [12].

### Cd

Cd concentration varies between 0.61- 8.74 mg/kg with *Ziziphus Vulgaris*, had the highest Cd level and *Nigella sativa* had the lowest. WHO [11] prescribed limit for Cd contents in medicinal plant is 0.3 mg/kg and the maximum acceptable concentration for food stuff is around 1 ppm [13]. Cd intoxication can lead to kidney, bone and pulmonary damages [14].

### Cu

The concentration of Cu varied from 7.51 to 146 mg/kg, *Nigella sativa L.* contains the highest level of Cu and *Cichorium intybus L.* contains the lowest. There is no permissible limit prescribed in local food law or by WHO, There is no permissible limit prescribed in local food law or by WHO, but WHO (1996) has recommended the lower limit of the acceptable range of Cu as 20 µg/mg body weight per day [15,16], however, national limits in Sangapore for herbal medicines and products is 150 ppm.

### Ni

The highest level of Ni occurred in *Ziziphus vulgaris L.* and the lowest in *Zingiber Officinale*. The Ni concentration varied from 2.37 to

22.96 mg/kg. Except for *Ziziphus vulgaris L.* and *Cichorium intybus* the results of present study shows Ni contents well within the permissible limits of 8 mg/kg. Although Ni is required in minute quantity for body as it is mostly present in the pancreas and hence plays an important role in the production of insulin. EPA has recommended daily intake of Ni should be less than 1 mg beyond which is toxic [17].

## Hg

The Hg concentration varied from 0.39 to 0.69 mg/kg, *Ziziphus vulgaris L.* had the highest Hg concentration and *Nigella sativa L.* the lowest. Most samples had Hg contents more than the permissible limits of national limits in herbal medicines and products. For the Ph. Eur. monograph Herbal drugs, the mercury limit was drafted as 0.1 mg/kg [18]. Permissible limit set by FAO/WHO (1984) in edible plants was 0.02 mg/kg [19].

## Conclusion

One of the major environmental pollution in the developing countries like Iran is the heavy metal pollution and the pollution from the use of excessive insecticides, pesticides and fertilizers in the agriculture fields. By a comparison between acceptable global standards and the level of Hg, Cd and Pb on investigated herbal medicine and medicinal plants, our results showed that the majority of medicinal plants samples had higher level of these heavy metals. Due to lack of research data and technical limitations at present, more research is needed in order to establish the scientific criteria for herbal medicines.

The problem is rather more serious in Iran and the other developing countries. Because medicinal plants neither controlled nor probably regulated by quality assurance parameters.

The results suggest that medicinal plants used for human consumption or for preparation of herbal products and standardized extracts should be collected from an unpolluted natural habitat.

As heavy metal toxicity through contamination of preparation continues to be recognized risk, voluntary programs to provide community education regarding the potential risk of herbal preparations should be

Supported by the availability free heavy metal testing services. These testing services would most hopefully be provided through existing general practice and pathology testing services, and would contribute to health protection.

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