

## Artículo de investigación

## Determination of the concentration of heavy metals in medicinal plants and assessment of the risk to health

Determinación de la concentración de metales pesados en plantas medicinales y evaluación del riesgo para la salud

Determinação da concentração de metais pesados em plantas medicinais e avaliação do risco para a saúde

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### Abstract

The present study was conducted to determine the concentration of heavy metals in some selected medicinal plants in the Shiraz. Heavy metals such as Cd, Pb, Cu, Zn and Fe were investigated in *Solanum alatum*, *Peganum harmala*, *Achillea*, *Eryngium*, *Fumaria officinalis*, *Artemisia* by using Atomic Absorption Spectrometry. Plants showed different metal concentration in the range of: 0.0116-0.079 mg/kg for Cd, 0.024- 0.306 mg/kg for Pb, 3.8-9.87 mg/kg for Cu, 11.55- 34.19 mg/kg for Zn and 17.94- 187.24 mg/kg for Fe. Health risk assessment showed that consumers are not in danger as far as these metals are concerned.

The findings of this study indicated that although most of the sampling plants were contaminated, the estimated daily intake of each metal (EDI) showed that except, Cu in *Achillea*, *Artemisia* in children, other samples have EDI below the Acceptable Daily Intakes (ADI) recommended by the Institute of Standards and Industrial Research of Iran and FAO/WHO.

**Keywords:** Heavy metals, Medicinal Plants, Atomic Absorption Spectrophotometer, Health Risk.

### Resumen

El presente estudio se realizó para determinar la concentración de metales pesados en algunas plantas medicinales seleccionadas en el Shiraz. Metales pesados como Cd, Pb, Cu, Zn y Fe se investigaron en *Solanum alatum*, *Peganum harmala*, *Achillea*, *Eryngium*, *Fumaria officinalis*, *Artemisia* mediante el uso de espectrometría de absorción atómica. Las plantas mostraron una concentración de metal diferente en el rango de: 0.0116-0.079 mg / kg para Cd, 0.024- 0.306 mg / kg para Pb, 3.8- 9.87 mg / kg para Cu, 11.55-34.19 mg / kg para Zn y 17.94- 187.24 mg / kg de Fe. La evaluación de riesgos para la salud mostró que los consumidores no están en peligro en lo que respecta a estos metales.

Los hallazgos de este estudio indicaron que aunque la mayoría de las plantas de muestreo estaban contaminadas, la ingesta diaria estimada de cada metal (EDI) mostró que, a excepción de Cu en *Achillea*, *Artemisia* en niños, otras muestras tienen EDI por debajo de las ingestas diarias aceptables (IDA) recomendado por el Instituto de Normas e Investigación Industrial de Irán y FAO / OMS.

**Palabras claves:** Metales pesados, Plantas medicinales, Espectrofotómetro de absorción atómica, Riesgo para la salud.

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## Resumo

O presente estudo foi conduzido para determinar a concentração de metais pesados em algumas plantas medicinais selecionadas no Shiraz. Metais pesados como Cd, Pb, Cu, Zn e Fe foram investigados em *Solanum alatum*, *Peganum harmala*, *Achillea*, *Eryngium*, *Fumaria officinalis*, *Artemisia* por espectrometria de absorção atômica. As plantas apresentaram diferentes concentrações de metais na faixa de: 0,0116-0,079 mg / kg para Cd, 0,024-0,306 mg / kg para Pb, 3,8-9,87 mg / kg para Cu, 11,55-34,19 mg / kg para Zn e 17,94-187,24 mg / kg para Fe. A avaliação dos riscos para a saúde mostrou que os consumidores não estão em perigo no que diz respeito a esses metais.

Os resultados deste estudo indicaram que, embora a maioria das plantas de amostragem estivessem contaminadas, a ingestão diária estimada de cada metal (EDI) mostrou que exceto Cu em *Achillea*, *Artemisia* em crianças, outras amostras têm EDI abaixo das Ingestões Diárias Aceitáveis (ADI) recomendado pelo Instituto de Padrões e Pesquisa Industrial do Irã e FAO / OMS.

**Palavras-chave:** Metais pesados, Plantas Medicinais, Espectrofotômetro de Absorção Atômica, Risco à Saúde.

## Introduction

Due to rapid industrialization, heavy metals have been heavily transmitted to the environment, causing major global concern. The word heavy metals refers to metals and quasimetals with an atomic number greater than 20 and a density greater than 5 grams per cubic meter (Alloway, 2010). These metals include antimony, bismuth, cadmium, cesium, cobalt, chromium, gallium, gold, iron, lead, manganese, mercury, nickel, platinum, silver, uranium, tin, vanadium and zinc (Forte, Petrucci & Bocca, 2008), which have a positive and negative role important in human life (Oktem & et al., 2005; Colak, Soyak & Turkoglu, 2005). The major problem with heavy metals is that these inorganic pollutants are not decomposable to organic pollutants. This fact has made heavy metals one of the most dangerous pollutants of the environment (Vodyanitskii, 2016). Metals such as cadmium and lead are cumulative poisons that cause environmental damage and are reported to be highly toxic. Cadmium-containing foods can have long-term adverse effects on the kidneys, lungs, cardiovascular system, chromosome and bone (Hongxing & Yu-Kui, 2011). Lead can have effects on the blood, intestine, stomach, kidney, gland secretion, reproductive organs, central nervous system and neuromuscular effects (Baudouin & et al., 2002; WHO, 1997). According to reports, children are much easier to absorb lead in their gastrointestinal tract than adults, and on the other hand, the nervous system of children is growing rather than the destructive effects of this metal. Lead-induced poisoning in children causes neurological damage, and these injuries lead to decreased IQ, short-term memory loss, learning disabilities,

and coordination disorders among members (Yang & et al., 2004). Iron, copper and zinc are essential metals for humans because they play an important role in biological systems, but these heavy metals, if they are too high, can have toxic effects, it is an essential element for many enzymes. Therefore, it plays an important role in a wide range of physiological activities. Iron use processes, free radicals removal, connective tissue bone growth, melanin production, and many more. However, over-consumption of copper causes dermatitis, gastrointestinal tract, abdominal pain, nausea, diarrhea, vomiting, and liver damage (Ullah & et al., 2012; Martin & Griswold, 2009). It is an essential element for growth, blood clotting, thyroid function and protein synthesis, and DNA synthesis. There is little information about zinc poisoning; however, excessive consumption of zinc outside the toxicity of the immune system has levels of lipoprotein (Fosmire, 1990). Iron has several key functions in the human body: oxygen supply, energy production and safety. Excessive iron intake causes dizziness, nausea, vomiting, diarrhea, joint pain, shock and liver damage (Martin & Griswold, 2009). In the last decade, the acceptance and popularity of medicinal plants has increased in developed and developing countries. According to the World Health Organization, about 80 percent of the world's population uses plants and other traditional drugs to treat, given that the use of medicinal plants grown in contaminated areas or inappropriate processing can be one of the ways to enter pollutants. The World Health Organization (WHO) recommended heavy metals, pesticides, and bacterial and fungal contamination (WHO,

1997), including heavy metals to humans and animals. So far, studies have been done to verify the concentration of heavy metals in medicinal plants in Iran and other countries as well as to determine the health risks of these products; A Survey on the Investigation of Lead and Cadmium Elements in Eleven Iranian Herbal Medicines (Mousavi & et al., 2014), a study to investigate the contamination of Brazilian herbs with cadmium, mercury and lead (Caldas & Machado, 2004), to investigate the elements of mercury, cadmium, arsenic, palladium In some Indian herbs (Ahmad, Khan & Yadav, 2013), check the amount of heavy metals found in 8 types of medicinal plants in Pakistan (Rehman & et al., 2013). Iran has about 80 crops of medicinal plants with an area of 180 thousand

hectares and production of more than 200 thousand tons, and the export volume in 1396 is more than 400 million dollars. Iran's medicinal plants are often from car plants (natural) and places usually unknown of nature are collected, and because of the possibility of harvesting from contaminated sites, there is a potential danger to the health and safety of consumers. On the other hand, the use of medicinal plants is increasing, and given that the most important route is the transfer of metals Heavy to the human food chain and biological cycles, this study aims to investigate the concentration of age metals the cadmium, lead, copper, zinc and iron were performed in 6 types of medicinal plants in the city.

### Materials and Methods

Samples of herbs were arranged in the spring from the slopes of the mountains of Fars

province and transferred to the lab in polyethylene bags. Examples are given in Table I.

Table I. Hillside herbs transferred to the laboratory

Indications	Family	scientific name	Plant name
Housing, anti-rheumatism, fever, cough and asthma	Potato	Solanum alatum	Routarab
Sleepy, Mucus, Milk maker, Relaxing	Espanol	Peganum harmala	Espanol
Elimination of gastrointestinal, tonic, augmentative, anticonvulsant	Chicory	Achillea	Yarrow
Kidney stones and bladder, diarrhea, bloating of the stomach and intestines	Apiaceae	Eryngium	Eryngium
Killer of fever, appetizer, tonic and laxative	Fumaria	Fumaria officinalis	Fumaria
Improvement of the function of stomach, Killer of fever, disinfectant	Chicory	Artemisia	Artemisia

Measurements of heavy metals were performed according to standard AOAC 991.11 method (AOAC). Weigh 5 g of each sample on a digital scale and burn it in a crucible and burn on heat to remove smoke from burning them. The samples were then placed in a 450 ° C electric furnace for 4 to 5 hours, resulting in a white ash

showing the loss of organic matter. (Organic matter in plants affects atomic absorption). After cooling the samples in a desiccator, 30 ml nitric concentrated nitrile was added to each crucible, and after complete dissolution, by the Watton filter paper 42 was straightened and then dissolved in 2% nitric acid and distilled into

volume and the dilution factor was recorded. Finally, the concentrations of heavy lead, cadmium, copper, zinc and iron elements in the medicinal plants were determined using the atomic absorption device (American Analyst 700

model) found in Shiraz Food and Drug Lab. The machine parameters for reading the levels of cadmium, lead, zinc, copper and iron were in accordance with Table 2.

Table 2. Device parameters for determination of cadmium, lead, copper, zinc and iron content in simples

		Cadmium	Lead	Zinc	Copper	Iron
Wave length	nm	228.8	217	324.8	213.9	248.3
Slit width	nm	0.7	0.7	0.7	0.7	0.2
Hallow-cathod lamp current	mA	4	10	15	15	30
Atomization temprature	°C	1800	1650	2300	2300	2300

In order to determine the concentration of the elements from the solution of cadmium, lead, copper, zinc and iron, standards were prepared with 6 concentrations and a standard curve was drawn for each one and the criterion was to determine the final concentration of the elements.

Estimated Average Daily Intakes (EADI) Estimates were estimated by the following equation (Apau, & et al., 2014).

$$EDI = \frac{C \times F}{W \times D}$$

In this relation:

K = Mean concentration of each element in the foodstuff studied in mg / kg

D = Number of days of the year (365)

F = Average annual consumption of food by each person;

W = the average body weight was 70 and 15 kg, respectively, for adults and children

And the health risk index can also be derived from the ratio of the average daily absorbance of each element to the acceptable absorption of that element.

$$HI = \frac{EADI}{ADI}$$

If a Health Index (HI) is found to be less than one, the consumer of the food has adverse health effects in the safe area, if more than one, the probability of adverse effects increases with increasing HI levels. In this research, a completely randomized design was used to

analyze the data. Comparison of the meanings obtained from three replications was performed using Duncan's multi-domain test with 5% error probability. Also, SPSS software (version 24) was used to analyze the data.

## Results and Discussion

Food contamination with heavy metals may be created during harvesting, transportation, storage, process and preparation. The absorption of heavy metals from contaminated lands by plants is one of the most important ways of entering these elements into the food chain (Salehipour & et al., 2015). The results of this study showed that in the medicinal plants the cadmium content varied from 0.06 mg/kg to 0.0793 mg/kg. Maximum and minimum cadmium concentrations were related to Chahar and Rutbark respectively. Figure 2 shows that the highest amount of lead in Achilles was 306 mg / kg, which showed a significant difference with the rest of the samples. Artemisia was ranked second in terms of lead absorption. The lowest levels of lead in the routardbatch sample were 0.0246 mg/kg. The statistical test showed that there was a significant difference between the samples ( $P < 0.05$ ). As shown in Fig. 3, among the selected samples, the highest amount of arsenic copper was 9.87 mg/kg and the lowest was 3.8 mg/kg in Esfand. In this sample and the rest of the samples, there was a significant difference ( $P < 0.05$ ). The results for copper indicated that the

highest copper content was found in the two herbs of Achillea and Artemisia, which did not have a significant difference. The amount of copper in Shahrverd, Esfand and Rutherbak plants was lower than other plants and there was no significant difference between them.

The results of Figure 4 indicate that zinc levels ranged from 11.55 mg/kg to 19.19 mg/kg/g. Maximum and minimum zinc concentrations were related to Artemisia and Rutherbak,

respectively. Figure 5 shows the results of the iron concentration. The results indicate that there is a significant difference between the samples ( $P < 0.05$ ). The highest concentrations of iron were found in Achilles (11.5 mg/kg) and Artemisia (7.57 mg/kg). The lowest iron concentrations were related to rutardab (4.25 mg/kg).

(Acceptable Daily Intakes; ADI) was calculated as follows (Apau, & et al., 2014).

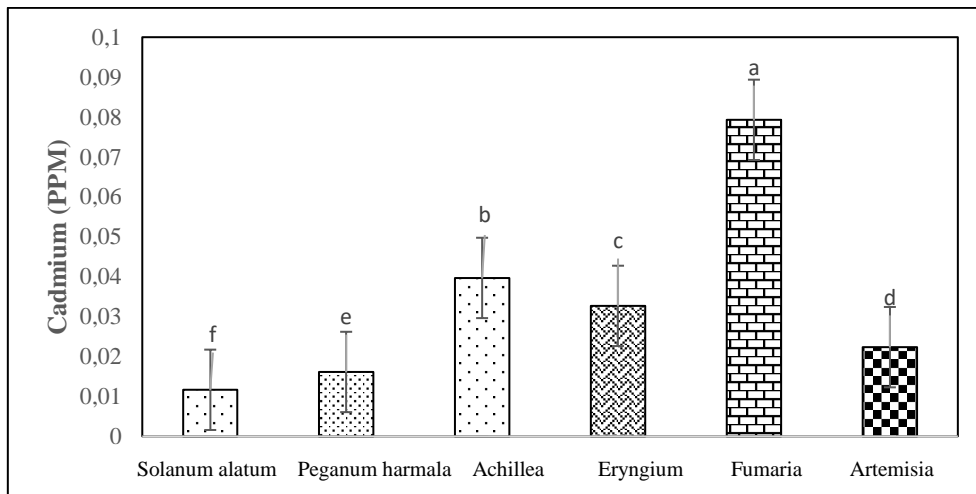


Figure 1. Cadmiun heavy metal concentration in medicinal plants sample

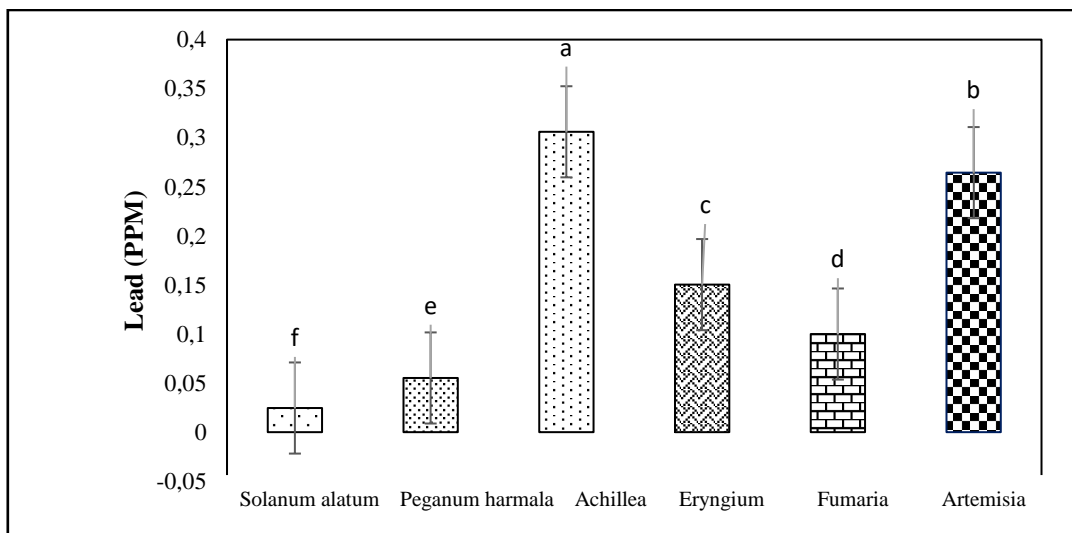


Figure 2. Heavy metal concentration of lead in the sample of herbs

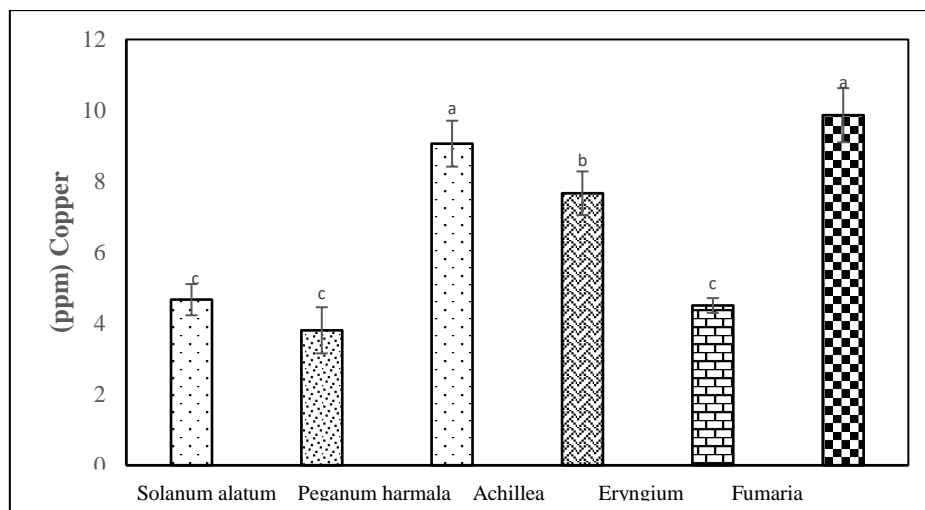


Figure 3. Heavy Metals Concentration in the Medicinal herbs sample

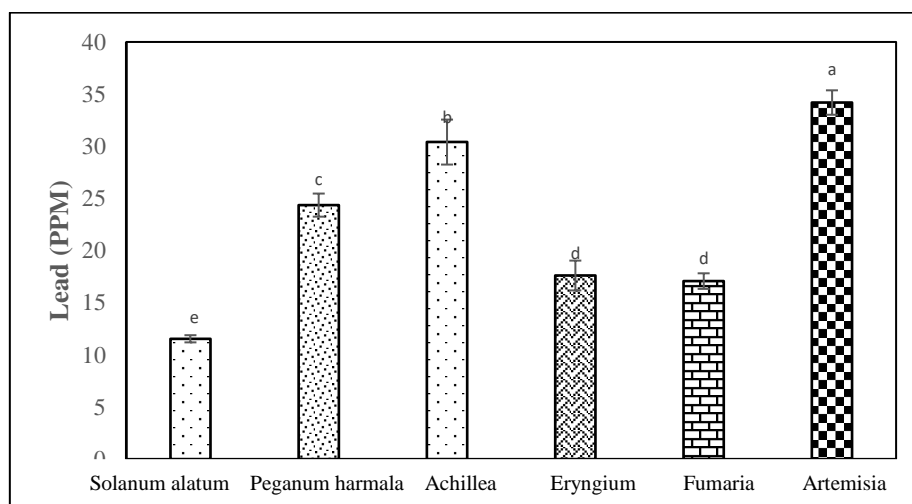


Figure 4. Concentration of heavy metal zinc in the sample of medicinal herbs

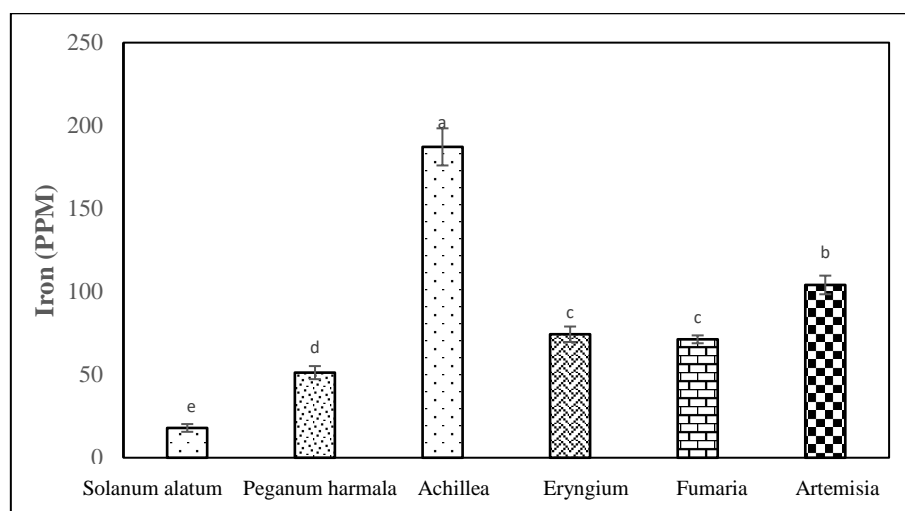


Figure 5. Heavy metal concentration in the sample of medicinal plants

Various studies have shown that the concentration of heavy metals in medicinal plants depends on their growth medium, plant species, dry conditions, storage, transport and processing. Increasing the use of chemical fertilizers, pesticides and pesticides to increase the yield of agricultural products on the one hand, and human activities such as mines, agriculture, production and disposal of waste at landfills are the common sources of heavy metals, and each of these can be A very high amount for some of the heavy metals found in these food samples. The World Health Organization has announced that maximum levels of cadmium and lead in herbs are 0.3 and 10 mg/kg respectively ( table 3). The amount of cadmium in the samples was less than standard. In some samples, it was lower than the maximum value, which is based on collecting the highest amount of lead in this plant. It seems that except for the Greek species that show the highest amount of lead collection, other plants have a case study by adopting Some mechanisms, such as the accumulation of heavy metals in the roots and the lack of entry into plant organs, stabilize the heavy element in the soil with the compounds secreted from the root, prevent the entry of heavy metals into the plant (Bizhani,

Asgharipor & SirousMehr, 2014). The permitted values for copper, zinc and iron are 40 and 60, respectively, by FAO/WHO (Vodyanitskii, 2016). Permitted iron in medicinal herbs has not yet been determined (Rania & et al., 2015). Among the similar studies, the results of which can be summarized as follows:

In July 2013, Kulhari et al reported that levels of lead and cadmium in some of the herbal medicines in the northwest of India have been set at a limit (Kulhari, & et al., 2013). Rehman et al., Research on Heavy Metals in Different Medicinal Plants in Pakistan. The results showed that the average concentration of iron, zinc, lead, cadmium, chromium, copper, manganese and nickel in mg / kg was 40.47, 10.11, 0.99, 0.1, 0.01, 15.06, 29.11 and 02.1, according to the obtained values, all elements except iron and manganese are within the permissible limits of the World Health Organization (Rehman & et al., 2013). In Poland, a study was conducted to investigate the amount of zinc and copper in 13 species of herbal medicine, and the results showed that in more than 60% of the samples, these elements were below the WHO target (Krejpcio, Krol & Sionkowski, 2007).

Table 3. Results of calculation of daily average intake and health risk of medicinal herbs based on the potential of the elements of cadmium, lead, zinc, Copper and iron

ADI	HI(Adult)	HI(Childre)	EDI(Adult)	EDI (Children)	Mean concentration (mg/kg)	Element
						Routarabek
$1.0 \times 10^{-3}$	$1.135 \times 10^{-2}$	$5.29 \times 10^{-2}$	$1.135 \times 10^{-5}$	$5.29 \times 10^{-5}$	0.0116	Cadmium
$3.6 \times 10^{-3}$	$0.65 \times 10^{-2}$	$3.027 \times 10^{-2}$	$2.34 \times 10^{-5}$	$0.109 \times 10^{-3}$	0.024	Lead
0.3	0.583	0.175	0.0113	0.0527	11.55	Zinc
0.04	0.112	0.532	0.0045	0.0213	4.67	Copper
0.93	0.0188	0.088	0.0175	0.0819	17.94	Iron
						Espand
$1.0 \times 10^{-3}$	$1.575 \times 10^{-2}$	$7.351 \times 10^{-2}$	$1.575 \times 10^{-5}$	$7.351 \times 10^{-5}$	0.0116	Cadmium
$3.6 \times 10^{-3}$	$1.5 \times 10^{-2}$	0.07	$5.401 \times 10^{-5}$	$0.252 \times 10^{-3}$	0.0552	Lead
0.3	0.079	0.37	0.0238	0.1112	24.36	Zinc
0.04	0.092	0.432	0.00371	0.0173	3.8	Copper
0.93	0.053	0.251	0.0501	0.2340	51.26	Iron
						Yarrow
$1.0 \times 10^{-3}$	$3.874 \times 10^{-2}$	0.1808	$3.874 \times 10^{-5}$	$0.1808 \times 10^{-3}$	0.0396	Cadmium
$3.6 \times 10^{-3}$	0.083	0.388	$0.2994 \times 10^{-3}$	$1.397 \times 10^{-3}$	0.306	Lead
0.3	0.099	0.462	0.0297	0.1388	30.4	Zinc
0.04	0.22	1.032	0.0088	0.0413	9.06	Copper
0.93	0.196	0.918	0.1831	0.8547	187.22	Iron
						Boughanagh
$1.0 \times 10^{-3}$	$3.189 \times 10^{-2}$	$14.8 \times 10^{-2}$	$3.189 \times 10^{-5}$	$14.8 \times 10^{-5}$	0.326	Cadmium
$3.6 \times 10^{-3}$	0.0408	0.19	$0.147 \times 10^{-3}$	$0.686 \times 10^{-3}$	0.1503	Lead



0.3	0.573	0.268	0.0172	0.0804	17.61	Zinc
0.04	0.187	0.875	0.0075	0.035	7.67	Copper
0.93	0.078	0.364	0.0727	0.339	74.37	Iron
						Fumaria
$1.0 \times 10^{-3}$	$7.759 \times 10^{-2}$	$0.362 \times 10^{-2}$	$7.759 \times 10^{-5}$	$0.362 \times 10^{-3}$	0.0793	Cadmium
$3.6 \times 10^{-3}$	$2.715 \times 10^{-2}$	0.126	$9.774 \times 10^{-5}$	$0.456 \times 10^{-3}$	0.999	Lead
0.3	0.055	0.259	0.0167	0.0779	17.08	Zinc
0.04	0.11	0.512	0.044	0.0205	4.5	Copper
0.93	0.073	0.349	0.0697	0.325	71.32	Iron
						Artemisia
$1.0 \times 10^{-3}$	$2.299 \times 10^{-2}$	0.107	$2.299 \times 10^{-5}$	$0.107 \times 10^{-3}$	0.0235	Cadmium
$3.6 \times 10^{-3}$	0.071	0.334	$0.258 \times 10^{-3}$	$1.205 \times 10^{-3}$	0.264	Lead
0.3	0.111	0.52	0.0334	0.156	34.19	Zinc
0.04	0.24	1.125	0.0096	0.045	9.87	Copper
0.93	0.109	0.494	0.1018	0.475	104.06	Iron

Identifying any heavy and toxic metal in contaminated medicinal plants and highlighting it in controlling the health of consumers is necessary. Also, the results of daily absorption and Risk and Health Indicators indicate that the highest daily absorption of heavy metals resulting from the use of medicinal plants is related to iron, zinc and copper. This suggests that adult and pediatric consumers of medicinal plants have consumed a high proportion of the studied metals. In the meanwhile, the highest absorption of metals was done by the children. On the other hand, the risk and health index of less than one for Cd, Pb, Zn and Fe indicates a relative lack of risk for drug users. In addition, it is clear that children are more at risk than adults. The contamination of medicinal plants with heavy metals can lead to mutagenic and carcinogenic diseases in the human body. Also, the Risk and Health Indicators in children for copper in yarrow and Artemisia plants were more than 1, which indicates the potential hazard of the plant in these plants. The results of this study, which were conducted in Taiwan in order to determine the health risk index of arsenic, lead, cadmium and chromium in teaspoon tea in Taiwan, in all samples smaller than one and in HI showed that the safe area is in good health (Shen & Chen, 2008). The results of this study aimed at investigating the concentration of heavy metals in cadmium, lead, copper, zinc and iron in some medicinal plants used in Shirazan city. The average concentration of heavy metals examined was less than the permissible level determined by the WHO. The study of health risk index except for copper in Achilles and Artemisia was less than one. Because these products may be used by patients who are susceptible to toxic elements, it is advisable to avoid the use of medicinal herbs to avoid exposure to small amounts of toxic metals.

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