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Health Risk Assessment of Heavy Metal Accumulation in Medicinal Plants Available in the Shiraz Consumer Market

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ABSTRACT

Introduction: In recent years, the use of medicinal herbs has grown significantly due to their perceived safety compared to synthetic drugs. However, these plants are capable of accumulating toxic heavy metals, which may enter the food chain and pose potential health risks. This study aimed to assess the levels of zinc (Zn), lead (Pb), and cadmium (Cd) in commonly consumed medicinal plants—*Matricaria chamomilla*, *Hyssopus officinalis, Thymus vulgaris,* and *Origanum majorana*—sold in Shiraz.

Methods: Three samples from each plant species were collected, acid-digested, and analyzed using atomic absorption spectrophotometry in triplicate. Statistical analyses were performed using SPSS software.

Results: The mean concentration of Zn ranged from 10.49 ± 0.51 mg/kg in *Thymus vulgaris* to 27.73 ± 0.86 mg/kg in *Matricaria chamomilla*. For Pb, values ranged from 8.42 ± 0.43 mg/kg in *Origanum majorana* to 12.23 ± 0.45 mg/kg in *Matricaria chamomilla*. Cd levels ranged from 2.93 ± 0.19 mg/kg in *Matricaria chamomilla* to 23.2 ± 0.24 mg/kg in *Thymus vulgaris*.

Conclusion: Health index (HI) values for all samples were below 1, suggesting no immediate health threat. However, compared to World Health Organization (WHO) standards, Zn levels were within acceptable limits in all samples, while Cd exceeded the permissible threshold in every sample. Additionally, Pb concentrations in *Thymus vulgaris* and *Matricaria chamomilla* surpassed WHO limits. Based on these findings, cautious and regulated consumption of these herbs is recommended to mitigate potential health risks.

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Introduction

One of the significant environmental issues today is soil contamination with heavy metals (Alloway, 2013). The sources of heavy metals in soil can be natural or result from human activities such as atmospheric deposition due to air pollution, the use of pesticides and chemical fertilizers, the release of waste into the environment, or the application of sewage sludge on agricultural land (Kabata-Pendias, 2010). Heavy metals can accumulate in living organisms and enter the food chain, leading to both acute and chronic toxicity upon entering the body (Sobhanardakani et al., 2014; Ahmed et al., 2011). Metals such as zinc, lead, and cadmium can cause effects such as nausea, vomiting, diarrhea, and decreased secretion of certain enzymes within body tissues, particularly the liver (Sobhanardakani et al., 2014). They may also result in damage to the brain and nervous system, which can lead to reduced cognitive abilities in children and, in severe cases, trigger psychological disorders in humans (Pierroti and Ghasemzadeh, 2012). Cadmium is classified as a carcinogen; its accumulation in living organisms can lead to kidney and liver failure, increased blood pressure, and hinder the transfer of zinc and copper to the fetus (Oliver, 2007).

Numerous studies have reported on the contamination of medicinal plants with heavy metals and their high capacity for the absorption and transfer of these metals to usable parts (Baye et al., 2010; Karimi et al., 2015; Dawoud, 2025). Currently, the preference for using medicinal plants has increased due to their perceived lack of adverse effects compared to synthetic drugs (Khalifezadeh, 2014). Contamination of medicinal plants with heavy metals can occur during cultivation through contaminated water, soil, and air (Denholm, 2010). Agricultural soils are considered one of the main pathways for transferring heavy metals into the human food chain (Tóth et al., 2016).

Today, there has been limited attention paid to the quality and safety of medicinal plants, but as these plants are recognized as safer alternatives to synthetic drugs, their environmental safety and quality have become major concerns (Ekor, 2014). Although the use of medicinal plants is on the rise, it is crucial to consider that the cultivation of these plants in areas contaminated with heavy metals poses a risk for the introduction of these metals into the bodies of humans and animals. Therefore, the World Health Organization has recommended that medicinal plants be monitored for the presence of pesticides, heavy metals, and bacterial and fungal contamination (Jena et al., 2012).

The objective of this study is to assess the health risks associated with the accumulation of heavy metals such as zinc, lead, and cadmium in broadleaf thyme, chamomile, hyssop, and marjoram consumed in Shiraz.

Materials and Methods

In this study, four medicinal plant species were utilized: broadleaf thyme (*Zataria multiflora*), chamomile (*Matricaria chamomilla*), hyssop (*Hyssopus officinalis*), and marjoram (*Origanum majorana*), which are widely consumed among the people of Shiraz. Three samples of each species were purchased from popular herbal shops located in the city. After collection, the samples were kept at room temperature, ground, and then mixed in a 3:1 volumetric ratio with concentrated nitric acid and perchloric acid. This mixture was placed in a water bath at 80 degrees Celsius for two hours until the sample became clear. The resulting solution was filtered using Whatman filter paper.

Subsequently, the main solution (stock) was prepared, along with standard solutions of zinc, lead, and cadmium salts, for calibrating the Shimadzu AA680 atomic absorption spectrometer. The concentrations of the elements in the samples were determined with three repetitions. The average values were calculated to assess the acceptable daily intake (ADI) and to compute the health index for each element using equations.

To estimate the average annual consumption of each food item by individuals (F), a questionnaire was distributed, determining the annual consumption rates as follows: 35 grams for chamomile, 40 grams for broadleaf thyme, 30 grams for hyssop, and 25 grams for marjoram**.**

The equation for estimating the daily acceptable intake is as follows**:**

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

Where **:**

C = Average concentration of each element in the studied food item (mg/kg)

D = Number of days in a year (365)

F = Average annual consumption of the food item by each individual

W = Average body weight (70 kg for adults and 15 kg for children)

The health index (HI) is calculated using the following equation:

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

Where:
EADI= Estimated average daily intake of each element (mg/kg/day)
ADI = Acceptable daily intake of each element (mg/kg/day)

To assess the normal distribution of the data, the Kolmogorov-Smirnov test was employed. For comparing the mean concentrations of elements against the World Health Organization (WHO, 2005) guidelines, a one-sample t-test was conducted. Furthermore, to compare the mean concentrations of heavy metals between samples, one-way analysis of variance (ANOVA) was performed using the least significant difference (LSD) method. The Pearson correlation test was used to examine the correlation between the average concentrations of the accumulated elements in the samples.

Results

The concentrations of heavy metals—zinc, lead, and cadmium—in the samples of the medicinal plants broadleaf thyme, chamomile, hyssop, and marjoram, along with the results obtained from the health index (HI) associated with the consumption of these medicinal plants, are presented in Tables 1 and 2, respectively.

Table 1. Concentration of cadmium, lead, and zinc in evaluated medicinal plant samples in this study

Medicinal Plants	Cadmium (mg/kg)	Lead (mg/kg)	Zinc (mg/kg)
Broad-leaved Thyme	2.23 ± 0.24^{a}	11.21 ± 0.29b	10.49 ± 0.51^{c}
Chamomile	2.93 ± 0.19^{a}	12.23 ± 0.45^{a}	27.73 ± 0.86^{a}
Hyssop	2.38 ± 0.30^{a}	11.63 ± 0.80^{ab}	12.34 ± 1.12^{c}
Marjoram	2.75 ± 0.69^{a}	8.42 ± 0.43^{c}	21.65 ± 1.41^{b}

Data are presented as mean ± standard deviation; Broad-leaved Thyme (*Zataria multiflora*), Chamomile (*Matricaria chamomilla*), Hyssop (*Hyssopus officinalis*), and Marjoram (*Origanum majorana*); different superscript lowercase letters (a, b, c) within each column indicate statistically significant differences between the means of medicinal plants for each heavy metal ($p < 0.05$) based on one-way ANOVA followed by Tukey's post hoc test.

The results of the comparison of the average effects of the herbal treatment on zinc accumulation indicated that chamomile and broadleaf thyme had a significant difference in the concentration of this metal compared to other plants ($P<0.05$), while no significant difference was observed in the case of hyssop ($P>0.05$). For lead, chamomile showed no significant difference in the accumulation of this metal compared to hyssop and broadleaf thyme; however, marjoram exhibited a significant difference in lead accumulation compared to the other plants at a 5% significance level.
Regarding cadmium, no significant differences in the accumulation of this metal were observed among the studied plants ($P>0.05$). Additionally, based on the results obtained in Table 2, the health index for these plants was less than one, indicating that controlled consumption of these plants does not pose adverse effects to consumers.

Table 2. Estimated Average Daily Intake (EADI), Health Index (HI), and Acceptable Daily Intake (ADI) of zinc, lead, and cadmium (mg/kg body weight/day) through the consumption of medicinal plants.

Plant	Element	EADI (Children)	EADI (Adults)	HI (Children)	HI (Adults)	ADI (mg/kg bw/day)
Hyssop	Zinc	1.085×10^{-4}	3.097×10^{-5}	1.074×10^{-10}	8.715×10^{-9}	0.025
	Lead	1.075×10^{-4}	3.074×10^{-5}	1.500×10^{-8}	1.024×10^{-4}	0.3
	Cadmium	3.057×10^{-5}	6.067×10^{-6}	4.510×10^{-10}	9.310×10^{-9}	0.007
Thyme	Zinc	2.010×10^{-4}	4.050×10^{-5}	9.889×10^{-9}	9.917×10^{-9}	0.025
	Lead	2.024×10^{-4}	4.080×10^{-5}	4.770×10^{-8}	6.010×10^{-5}	0.3
	Cadmium	4.046×10^{-5}	6.069×10^{-6}	7.663×10^{-8}	6.613×10^{-8}	0.007
Marjoram	Zinc	1.071×10^{-4}	5.080×10^{-5}	8.310×10^{-8}	1.123×10^{-4}	0.025
	Lead	1.005×10^{-4}	2.026×10^{-5}	5.130×10^{-8}	7.500×10^{-5}	0.3
	Cadmium	4.043×10^{-5}	3.077×10^{-6}	1.449×10^{-8}	5.310×10^{-8}	0.007
Chamomile	Zinc	1.086×10^{-4}	1.004×10^{-5}	4.219×10^{-8}	6.241×10^{-8}	0.025
	Lead	1.014×10^{-4}	5.059×10^{-5}	1.370×10^{-8}	5.210×10^{-5}	0.3
	Cadmium	1.013×10^{-5}	1.010×10^{-6}	3.073×10^{-8}	7.015×10^{-8}	0.007

Zufa: *Satureja hortensis*; Thyme: *Thymus vulgaris*; Chamomile: *Matricaria chamomilla*; Marjoram: *Origanum majorana*.

The mean concentrations in the samples, according to WHO guidelines, are as follows: 0.3 mg/kg for cadmium, 10 mg/kg for lead, and 50 mg/kg for zinc (WHO, 2005). Based on the one-sample t-test, there is a significant difference in the zinc levels of hyssop, broad-leaved thyme, marjoram, and chamomile compared to the standard concentration at a significance level of 1% ($P<0.01$), with all samples showing lower zinc levels than the standard. For lead, there was no significant difference between the lead levels in Hyssop flowers and the standard concentration at a 5% significance level ($P<0.05$). However, broad-leaved thyme and chamomile exhibited higher lead levels than the standard sample at the 5% significance level. Conversely, broad-leaved thyme showed lower lead levels compared to the standard sample at the same significance level ($P<0.05$). A significant difference was observed in the cadmium levels of Hyssop, broad-leaved thyme, and chamomile compared to the standard at a significance level of 1%, whereas the difference for marjoram was significant at the 5% level, with all plants exhibiting higher cadmium levels than the standard sample. The Pearson correlation test indicated a significant correlation between zinc and cadmium levels in all samples, with a correlation coefficient of 0.673 and a significance level of less than 0.05 (Table 3).

Table 3. Results of the correlation analysis between zinc, lead, and cadmium in plant samples

Heavy metals	Correlation coefficient / Significance level	Zinc	Lead	Cadmium
Zinc	Correlation coefficient	1	-0.071	0.673
	Significance level	–	0.826	0.017
Lead	Correlation coefficient	-0.071	1	0.031
	Significance level	0.826	–	0.925
Cadmium	Correlation coefficient	0.673	0.031	1
	Significance level	0.017	0.925	–

Discussion

One of the significant drawbacks of industrialization in modern societies is the gradual destruction and degradation of the environment (Shahbazi et al., 2012). The increase in industrial activities has led to the production of numerous pollutants, including heavy metals (Khodakarami et al., 2012). Among the primary pathways through which these pollutants enter the soil is the use of chemical fertilizers (Street, 2012) by local farmers. Medicinal plants, which are widely consumed today, serve as accumulation sites for these contaminants. Factors influencing the accumulation of these pollutants include air pollution, plant species, and various environmental conditions (Rahimi et al., 2012).

According to the results of this study, the samples analyzed cannot be deemed contaminated. However, caution is warranted regarding the detrimental effects of long-term accumulation of these elements in the body due to prolonged consumption of these plants. Chamomile, known for its high capacity (Abou-Arab et al., 1999) to accumulate elements and transfer them to aerial parts (Parsafar et al., 2014), exhibited the highest mean concentration of zinc among the samples (Grejtovský et al., 2006), while the lowest concentration of zinc was found in broad-leaved thyme and hyssop (Kulhari et al., 2013). These findings align with research by Kulhari et al. (2013) and Grejtovský et al. (2006).

The highest mean concentration of lead was also observed in chamomile (Kulhari et al., 2013), consistent with the studies of Kulhari et al. (2013). Hyssop and thyme exhibited slightly higher concentrations than chamomile, potentially due to mechanisms related to the plants' ability to accumulate metals and their secretion of certain substances that prevent metal absorption (Bizhani et al., 2014). In contrast, marjoram showed the lowest lead concentration, likely because metal accumulation occurs predominantly in the root sections of marjoram (Burzynski et al., 1997), while its medicinal use primarily involves the leaf parts of the plant.

Cadmium, known to initially accumulate in the roots before being transferred to the leaves (Salt et al., 1995), also exhibited the highest concentration in chamomile (Turekian and Wedepohl, 1961) and the lowest in broad-leaved thyme. Due to its high mobility and dynamics, cadmium can enter plants more readily than other metals (Khatamipour et al., 2011; John et al. 2009), potentially leading to more significant impacts on plants and the higher food chain.

The results obtained from the t-test for comparing the mean concentrations of elements with global standards indicated that all samples had lower zinc levels than the global standard, a finding that corresponds with the results of Karimi et al. (2017). In relation to lead, broad-leaved thyme and chamomile contained higher amounts than the standard. A study on ten medicinal plant species in Brazil (Caldas and Machado, 2004) found that cadmium and lead concentrations exceeded permissible limits. Numerous similar studies from Poland (Les´niewicz et al., 2006), Egypt (Abou-Arab et al. 1999), Argentina (Gomez et al., 2007), and the USA (Khan et al., 2001) reported results that contradicted this study regarding lead absorption, likely due to climatic and environmental conditions affecting the uptake of these elements (Rahimi et al., 2012). All species demonstrated cadmium accumulation exceeding global standards, consistent with findings from De Pasquale et al. (1993). This may be attributed to cadmium's high mobility (Khatamipour et al., 2011; John et al., 2009), leading to greater concentrations compared to global standards. This research aligns with findings from Hashemi et al. (2014).

The observed correlation between zinc and cadmium in some studies could be attributed to their similar physical and chemical properties, leading to antagonistic interactions (Aravind et al., 2005). Some studies have indicated that the addition of zinc to soil can reduce the concentration of cadmium (Choudhary et al., 1995), while others have shown that adding zinc can increase cadmium uptake (Moraghan, 1993).

Based on the results of the average daily element uptake and the health index, values for all elements were below 1, indicating no risk for consumers of these medicinal plants. These findings correspond with studies by Karimi et al. (2017).

Conclusion

The results of this study aimed to assess the concentrations of heavy metals—zinc, lead, and cadmium in the medicinal plants chamomile, marjoram, broad-leaved thyme, and hyssop. The findings indicated that the average concentrations of lead and cadmium in chamomile and broad-leaved thyme exceeded the guidelines established by the World Health Organization (WHO). Although the health index suggests no negative impact on consumers, it is essential to consider the potential risks associated with consuming these plants without regard to their contaminants. Therefore, controlled consumption of these herbal remedies is advisable to mitigate risks, keeping in mind that individual sensitivity to pollutants may vary based on the amount consumed. Furthermore, due to a lack of comprehensive information regarding the growth and cultivation conditions of these plants, continuous monitoring of metal accumulation in medicinal plants is recommended to ensure safety and health.

Declarations

Conflict of interest

The authors have no competing interests to declare that are relevant to the content of this article.

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Consent for publications

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Authors' contributions

Conceptualization, data curation, investigation, methodology, and writing of the original draft: ZH and HRP
Formal analysis, software development, data visualization, interpretation of results, and assistance with manuscript preparation and editing: ARP
Funding acquisition, project administration, provision of resources, overall supervision, validation of findings, critical revision of the manuscript, and final approval: DR
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Ethics approval

This study was performed in line with the principles of the Declaration of Helsinki.

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