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Environmental Risk Assessment of Heavy Metals in Selected Medicinal Herbs and Spices

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ABSTRACT

Spices and medicinal herbs is an important route for human exposure to toxic metals. This study determined total concentrations of heavy metals and risk assessment of ten types of herbs used in cooking, spices and medicinal plants available in local markets of Babylon province/Iraq. Concentrations of Cu, Ni, Zn, Pb, Co, As, Cd, Cr and Hg were estimated by ICP/Mass to identify toxic metals in the used portion of selected spices and medicinal plants. The average concentrations of these elements were within the limits allowed by the WHO. Culinary herbs and spices contained significantly Cu>Ni>Zn>Pb>Co>As>Cd>Cr>Hg. However, the concentrations of copper, zinc and nickel, respectively, were higher in all herbal samples. The study recorded the highest concentrations in the aerial parts of plants from the total elemental content of *Thymus vulgaris* leaves (109.4 $\mu g \cdot g^{-1}$). and barks of *Cinnamomum verum* was recorded (43 $\mu g \cdot g^{-1}$). Non-carcinogenic risks and estimated daily consumption of these herbs were assessed on the basis of the target hazard quotient (THQ) and Hazard Index (HI). THQ values for individual minerals were more than one indicating health risks for nickel (15.5) *Mentha verticillata* leaves, (12.3) for *Matricaria chamomilla*, other metals Cu, Co, Pb and Zn were recorded THQ>1which considered unsafe for human consumption. The mean Hazard Index (HI) for the nine metal elements is >1for all plants except *Zingiber officinale*, indicating that there are non-carcinogenic risks from these nine elements. this study provides a scientific basis to guide the safe consumption of certain culinary herbs and spices, it suggest potential health concerns for consumers of these products on a daily basis over a prolonged life span.

Keywords: heavy metals, medicinal herbs, risk assessment, human health.

INTRODUCTION

The culinary plant, herbs and spices have long been used for both food and medicinal purposes in different world civilizations in food preparation, the flavorings or nutritional supplements has been used also as pharmaceuticals for medicinal uses to treat many disease in traditional medicine or culinary additives. (Sarma, 2012), medicinal plant include herbs, herbal materials, herbal preparations and finished herbal products, either growing wild or cultivated, about 80% or more of the people in the world depend on herbal remedies in traditional medicine or primary health care. (WHO, 2004;WHO, 2007 Ekor, 2014). Although some widely used herbal medicines have promising potential, they are not monitored for potential adverse effects due to their frequent use, thus determining the safest and most effective treatments (WHO, 2002b). Heavy metals are naturally occurring elements of the Earth's crust and are usually present in all environmental system may be some their present with trace amounts, However, The possible sources of heavy metals in plan by polluted soil and water during cultivation and growth, fertilization, manufacturing process, anthropogenic activities can contribute to increasing of heavy metals concentration in the soil.

Generally, The level of heavy metals in plant varies primarily on a number of different parameters, including plant species, the frequency with which agrochemicals are used, the type of soil, pH, and soil organic matter (Tinker, 1981; Lubben, and Sauerbeck, 1991) One of the main ways that trace elements in the soil are cross to the food chain is through plant absorption. Therefore, heavy metals can have toxic effects on plants as well as human health through the food chain at high concentrations compared to what is physiologically necessary for vegetables (Waseam, et al., 2014).

Heavy metals have toxic risks of human and caused a lot of serious environmental damage around the world, major pathways of human exposure to heavy metals are direct ingestion of food, water and beverage. (Elekes et al. 2010; Ayodeji and Olorunsola, 2011; Sawut et al*.,* 2018). Many previous studies have shown that human consumption of herbs containing excessive levels of toxic metals can cause various cases of poisoning and health problems (Jarup, 2003; Mahan & Raymond, 2012).

Lead and cadmium have been shown to be at unsafe levels in 88 type of plants, for example, the lead and cadmium were detected in 21 and 44 respectively types of plant (Sarma et al., 2012). However, Mousavi et al*.*, (2014) detected the presence of both metals (Pb and Cd) above the maximum allowable daily in different herbal products in Iran, in contrast, Kohzadi et al., (2018); It was reported that the average concentration of As, Cd, Cr, Fe, Mn, Ni, Pb and Zn within the limits allowed by the WHO. According to WHO, the highest acceptable concentrations of lead, zinc and cadmium group are 10, 0.2 and 0.3 μg/g respectively in all parts of the plant(WHO, 1998). On the other hand, Ozyigit et al., (2022) found that some selected medicinal plants have contain higher concentrations from 14 heavy metals were detected from the places near factories, mining areas and farming land in Turkey. Medicinal herbs and spices are widely used in Iraq, but they have not been adequately evaluated for their content of toxic substances and elements, few studies indicated the content of some medicinal herbs and tea leaves to some heavy metals concentrations higher than the acceptable limits (Raouf et al., 2014; Jawad, 2016; Wali, 2022).

This study aimed to detect some heavy metals in ten types of different herbs and medicinal plants that are commonly used as food flavorings or as a traditional remedy in the Middle East, also to estimate the health risks posed by heavy metals by calculating the estimated daily intake. (EDI), Non-Carcinogenic Risk Assessment.

MATERIALS AND METHODS

Samples collection and preparation

The samples of herbs and medicinal plants categorized according part used, which included of selected parts of plants commonly used in cocking and phytotherapy Table 1 (Rhizome and roots, Flowers, seeds and bark). The samples were purchased from the city center shops in Babylone province, Iraq. These dried herbs were imported from different sources of the world (locally of Iraq, India, Iran and Turkey) were selected for the toxic metal analysis.

The samples were dried at 70 °C for 24 h, then crushed and sieved (mesh = 100 µ00 to obtain a fine powder then labeled and kept in polyethylene bags until digestion and analysis.

Digestion and chemical analysis of plant samples

Analytical-grade reagents and acids (Nitric acid, perchloric acid, deionized water, and H_2O_2) were purchased from Merck KgaA in Darmstadt, Germany. Prior to usage, the digesting vessel glassware and other equipment were properly cleaned with deionized water after being extensively scrubbed with 10% HNO_3.

All reagents and solvents, including sulfuric acid and H_2O_2, were of the analytical reagent grade (Merck). Each sample and standard solution were analyzed in triplicates to assure data accuracy, and the average values were taken as the final concentrations of the elements.

The chemical analysis of herbs powder were done by using the digestion method with Nitric acid and Perchloric acid, 0.2 g of samples were placed in a flask (100 ml), five ml from each nitric acid and Perchloric acid was sequentially added. The mixture was heated on a hot plate with slowly increasing heating to 300 °C in the muffle furnace until the content became colorless or gray of herbal ash residue. The digested material was diluted by double distilled water and filtered through a filter paper (Whatman 0.45µ). Deionized water was used to dilute the final mixture before it filtered by a Millipore filter (Whatman 0.45), then the final filtrate was collected to 25 ml. Finally, samples were kept in plastic vessels at 4 °C until analysis (Arumugam et al., 2012; Mahmood, et al., 2013).

The toxic metals contents were measured using Inductively coupled plasma mass spectrometry ICP-MS (Agilent 7500a). The operation conditions for (ICP-MS) were: The excitation power of the plasma (Radio Frequency RF) was 1200 Watt; pump speed: 30 $L \cdot min^{-1}$, the flow rates for cooling gas, carrier gas, and plasma gas were 1.0, 1.1, and 1 $L \cdot min^{-1}$, respectively,, sampling depth: 15 mm. The concentrations of each toxic metals in the plant samples were calculated by preparing a calibration curve by means of a series of dilutions of the studied elements (Wang and Hansen, 2004).

Risk assessment of toxic metals

Daily exposure to heavy metals that causes human health risks to consumers was measured by estimated daily intake (EDI), target hazard quotient (THQ), and hazard index (HI) (Zhang et al., 2018).

Estimated Daily Intake of Heavy Metals

The estimated average daily intake (EDI) depends on both the metal concentration in plants and the dose of consumption of the individual herbs. The EDIs of Cu, Ni, Zn, Pb, Co, As, Cd, Cr and Hg were calculated according to the mean concentration of each elements in each plant and the consumption rate. The ADI for human exposure of heavy metals was calculated using Eq. 1, which recommended by (Zheng et al., 2020).

$$ADI = \frac{C \times IR \times ABS \times EF \times ED}{BW \times AT} \quad (1)$$

where: ADI – the estimated average daily intake ($mg \cdot kg^{-1}$ from BW/d);
C – the calculated metal concentration ($mg \cdot L^{-1}$ or $mg \cdot kg^{-1}$);
IR – the daily ingestion rate per capita, is 0.5 $g \cdot d^{-1}$ (Luo et al., 2021);
ABS – the absorption coefficient (=1), (Zheng et al., 2020);
EF – the exposure frequency was set 90 days per year (Luo et al., 2021);
ED – is the exposure days over a lifetime (20 year);
BW – the body weight, the average adult body weights were considered to be 60 kg (Sohrabi et al., 2015);
AT – is the average lifetime (day) (AT = 60 years·365 days per year), d.

Health hazard estimation

The human health risk assessment from exposure to toxic heavy metals was estimated by the target hazard quotient (THQ) (Cao et al., 2010; Zhang et al., 2018), which indicates to the average ADI induced by exposure to concentrations close to or higher than the reference dose (RfD) for a single toxicant prescribed by (Integrated Risk Information System, 2011; Zheng et al.,2020). Non-carcinogenic risk was assessed by calculating target hazard quotient (THQ) and Hazard Index (HI) (Wang et al., 1998).

$$\text{THQ} = \text{ADI}/\text{Reference dose RfD} \quad (2)$$

Table 1. Selected herbal and medicinal plant collected from public markets categorized according parts used

No.	Common name	Scientific name	Family
Parts used: Rhizome and roots			
1.	licorice	*Glycyrrhiza glabra L.*	(Fabaceae)
2.	Ginger	*Zingiber officinale*	Zingiberaceae
3.	Turmeric	*Curcurma longa*	Zingiberaceae
Parts used: Aerial parts and bark			
4.	Thyme	*Thymus vulgaris*	Lamiaceae
5.	Mentha	*Mentha verticillata*	
6.	Cinnamon	*Cinnamomum verum*	Lauraceae
Parts used: Flowers			
7.	Rosell	*Hibiscus sabdariffa*	Malvacae
8.	Chamomile	*Matricaria chamomilla*	Asteraceae
Parts used: Seeds			
9.	Cumin	*Cuminum cyminum*	Apiaceae
10.	Black seed	*L. Nigella sativa*	Ranunculaceae

where: RfD – the ingestion references dose for adults (mg per kg per bw per day).
Ingestion reference dose = (0.0035) for Pb, (0.00083) for Cd and (0.02) for Ni, 0.0003 for As, 0.0003 for Hg and 0.00057 for Cu, Cr = 0.0083, Cu = 0.133, Zn = 0.006 $mg \cdot kg^{-1}$ $bw \cdot d^{-1}$ (Li et al., 2019; Zheng et al., 2020).

Hazard index (HI) – the total hazard index (HI) was applied to calculate the total value of non-carcinogenic risks causing by various heavy metal pollution. HI index is calculated from the equation below:

$$HI = \sum_{n=9}^{m} \text{THQi} \quad (3)$$

where: *HI* – is the total hazard index;
THQi – is the summation of THQ of heavy metal i.

When the target hazard quotients less than 1, indicated that there is no potential adverse effects of non-carcinogenic risk of consumers (USEPA, 1986; Zhang et al., 2018; Meng et al., 2022).

Statistical analysis

The random design was done to analyze current data by determining the significant differences LSD values between metals concentration at P value = 0.05 to compare the results obtained for various samples. SPSS.Version.16 statistical analysis software and Microsoft Excel software were both used in statistical analysis.

RESULTS AND DISCUSSION

Concentrations of toxic elements in medicinal herbs and spices

Heavy metals concentrations of Zn, Pb, Ni, Hg, Cu, Cr, Co, Cd and As ($\mu g \cdot g^{-1}$) were detected in the commonly consumed herbal and culinary samples in Iraq, according to the part used of the plant:

Rhizome and roots; the results indicated that the highest concentration of Cu 12.62, 8.41 and 6.9 $\mu g \cdot g^{-1}$ was recorded for the three plants *Glycyrrhiza glabra L., Curcurma longa* and *Zingiber officinale* respectively, then arsenic also recorded 0.67, 0.3 and 0.24 $\mu g \cdot g^{-1}$ for same plant respectively, while the cumulative total concentration of the nine elements was 26.1 $\mu g/g^{-1}$ in the turmeric plant *Curcurma longa* Figure 1.

Aerial parts and bark; The leaves of thyme *(Thymus vulgaris*) and mint *Mentha verticillata* recorded the highest concentration 2197.3 and 127.6 $\mu g \cdot g^{-1}$ of copper and nickel respectively. Moreover, they also contained the highest total concentration 2220.94 and 159.57 $\mu g \cdot g^{-1}$ of all the studied elements and plants, while Zn elements was recorded high concentration 5.21 $\mu g \cdot g^{-1}$ in the bark of Cinnamon (*Cinnamomum verum*) which was accumulated by 80.4 as the third element that recorded the highest accumulation of total heavy elements as shown in Figure 2.

Flowers; In the flowers of the hibiscus plant, the highest concentration of heavy metals was 18.8; 8.9 and 2.2 $\mu g \cdot g^{-1}$ for Zn, Cu and Pb

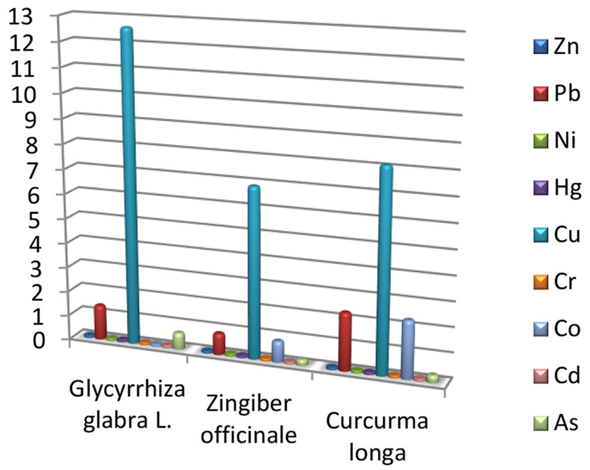

Figure 1. Mean concentration of heavy metals ($\mu g \cdot g^{-1}$) at Rhizome and roots

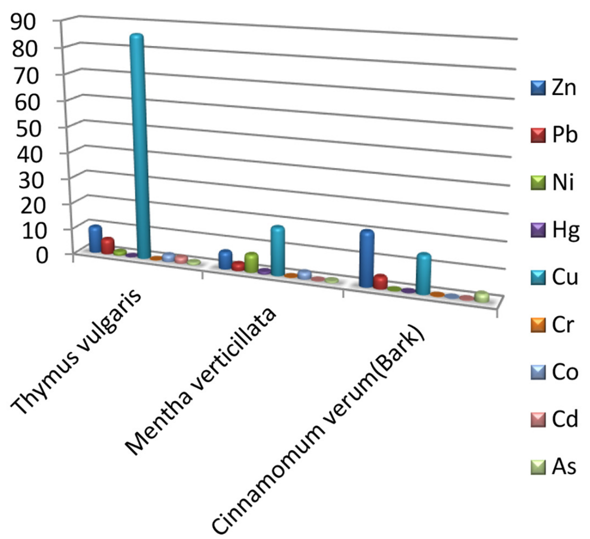

Figure 2. Mean concentration of heavy metals ($\mu g \cdot g^{-1}$) at Aerial parts and bark

respectively, while they were recorded 15.2 6.5, and 5.4 μg·g^{-1} in the Chamomile flowers for Cu, Zn and Ni respectively Figure 3. Both Cu and Zn as nutrient elements and have a vital role in metabolism reactions as activators of enzymatic systems in plant. The current study found that the accumulation of heavy metals was higher in the leaves, then flowers and seeds compared to other parts of the plants, Several authors reported that some heavy metals accumulated in leaves and flowers (Milošević and Milošević, 2014; Hladun et al., 2015; Dhiman et al., 2017). The Cadmium in *Thymus vulgaris* was recorded high value 2.3 μg·g^{-1}, which is higher than the permissible limits 0.3 μg/g (WHO, 2007).

Seeds; The concentrations of heavy metals in the seeds showed that the black cumin seeds *Nigella sativa* recorded the highest concentrations of all elements and the highest of them was for Cu, Zn and Pb25.14, 12.6 and 2.54 μg·g^{-1} respectively in comparison with cumin seeds that recorded 11.2, 9.92 and 1.24 μg·g^{-1} for same elements Figure 4. However, the study showed that the elements that recorded high concentrations in total concentration of all elements the order was as follows; Cu >Ni >Zn >Pb >Co >As >Cd >Cr >Hg. But the plant that contained the high total concentration of nine heavy elements was a thyme leaves *(Thymus vulgaris*) plant, then mint *Mentha verticillata* plant. On the other hand, all the herbal plants were contained Cu, Ni and Zn concentrations out the allowable limits prescribed by the WHO and FAO for edible plants is 3.00, 1.63 and 27.4 μg·g^{-1} respectively (FAO/WHO (1984), these elements are essential to the organism because they function as components of proteins and enzymes in the human body. however, they may also accumulated in some organs of the human body and causes chronic poisoning (Ur Rehman et al., 2018). These values in this study were agreement with the determined values in (Kohzadi et al., 2019).

Copper is an essential element of many enzymes, an crucial role in extensive range of processes including scavenging of free radicals, iron utilization, bone and connective tissues and numerous disorders. However, in elevation consumption of Cu can cause upper respiratory tract irritation, abdominal pain, dermatitis, vomiting, nausea, liver damage and diarrhea (FAO/WHO 1984; Dghaim et al., 2015).

The Nickel concentration exceeded the permissible limits in three types of plants are *Mentha* leaves (127.6 μg·g^{-1}), *chamomilla* flowers (5.4 μg·g^{-1}) and thyem leaves (1.66 μg·g^{-1}), note that the permissible limits are (1.63 μg·g^{-1}) according to (Jabeen et al., 2010). Maobe et al. stated Ni concentration exceeded in the all samples to 1.6 μg·g^{-1}, while Raouf et al. (2014) pointed out the Ni content exceeded the WHO established limit in three medicinal herbs in Iraq was ranged (8.8–10.25 μg·g^{-1}), while these limits were not exceeded in the study conducted by Kohzadi et al. (2019) in eight different types of medicinal plants in Iran, this indicates that nickel concentrations vary according to the type of plant and the part used. The Nickel affects the lungs and upper respiratory tract as a suspected carcinogen, and it causes skin allergic and dermatitis on prolong exposure.

In current study all the culinary herbs and seasoning were contained Zn levels within the

Table 2. Mean concentrations of heavy metals (μg/g) in the consumed herbal medicines and culinary sample

Herbs and medicinal plant		Heavy metals con. (μg·g^{-1})									Total
Parts used	Plant name	As	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn	
Rhizome and roots	*Glycyrrhiza glabra L.*	0.67	0.1	0.1	0.1	12.62	0.1	0.1	1.4	0.1	14.69
	Zingiber officinale	0.24	0.1	0.86	0.1	6.9	0.1	0.1	0.86	0.1	8.86
	Curcurma longa	0.30	0.1	2.33	0.1	8.14	0.1	0.1	2.33	0.1	13.1
Aerial parts and bark	*Thymus vulgaris*	1.03	2.32	2.63	0.1	85.7	0.1	1.44	5.77	10.2	109.4
	Mentha verticillata	0.68	0.1	2.63	0.1	18.42	1.01	6.8	2.63	6.6	38.57
	Cinnamomum verum (Bark)	2.96	0.26	0.1	0.1	14.55	0.1	0.1	4.5	20.7	43
Flowers	*Hibiscus sabdariffa*	0.68	0.2	1.2	0.1	8.9	0.1	0.1	2.2	18.8	31.98
	Matricaria chamomilla	0.47	0.1	1.08	2.2	15.2	0.1	5.4	2.16	6.5	33.01
Seeds	*Cuminum cyminum*	0.4	0.1	0.1	0.1	11.2	0.1	0.1	1.24	9.92	22.76
	Nigella sativa	0.43	0.12	0.1	0.1	25.14	0.1	0.1	2.54	12.6	40.83
Total		7.86	2.9	10.98	2.2	300.2	1.01	134.44	25.63	122.83	Total

Table 3. Estimated daily intake EDI (g/kg/bw/day) of nine heavy metals from consumption of medicinal herbs and spices

Herbs and medicinal plant	Metals								
	As	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn
Glycyrrhiza glabra L.	4x10^{-4}	7x10^{-5}	7x10^{-5}	1x10^{-5}	8x10^{-3}	7x10^{-5}	7x10^{-5}	1x10^{-3}	1x10^{-5}
Zingiber officinale	2x10^{-4}	1x10^{-5}	4x10^{-2}	1x10^{-5}	7x10-^{3}	7x10^{-5}	7x10^{-5}	6x10^{-4}	1x10^{-5}
Curcurma longa	2x10^{-4}	1x10^{-5}	1x10^{-3}	1x10^{-5}	5x10^{-3}	7x10^{-5}	7x10^{-5}	0.001	1x10^{-5}
Thymus vulgaris	7x10^{-4}	2x10^{-3}	2x10^{-3}	1x10^{-5}	0.1	7x10^{-5}	9x10^{-3}	4x10^{-3}	1x10^{-3}
Mentha verticillata	4x10^{-4}	1x10^{-5}	1x10^{-3}	1x10^{-5}	0.01	7x10^{-5}	0.08	0.002	5x10^{-3}
Cinnamomum verum	2x10^{-3}	2x10^{-4}	1x10^{-5}	1x10^{-5}	0.01	7x10^{-5}	1x10^{-5}	3x10^{-3}	0.014
Hibiscus sabdariffa	6x10^{-4}	7x10^{-5}	1x10^{-4}	1x10^{-5}	6x10^{-3}	7x10^{-5}	1x10^{-5}	2x10^{-3}	0.012
Matricaria chamomilla	3x10^{-4}	7x10^{-5}	1x10^{-4}	3x10-3	0.01	7x10^{-5}	0.002	1x10^{-3}	5x10^{-3}
Cuminum cyminum	3x10^{-4}	2x10^{-5}	1x10^{-5}	1x10^{-5}	7x10^{-3}	7x10^{-5}	1x10^{-5}	3x10^{-3}	0.06
Nigella sativa	3x10^{-4}	4x10^{-4}	1x10^{-5}	1x10^{-5}	0.07	7x10^{-5}	1x10^{-5}	0.03	0.01

acceptable limits (27.4 μg·g^{-1}) according to the WHO/FAO for consumable herbs except cinnamon bark recorded 58.21 μg·g^{-1}. Kulhari et al., (2013) indicated that the zinc levels in ten types of medicinal plants in diverse locations of north western India were (2.42–8.93) these values were higher than the values recorded in Maobe et al. (2012) amongst the local herbs used as phytomedicines in Southwest Kenya which recorded (0.989 to 1.833 μg·g^{-1}) and Kohzadi et al. (2019) in 8 different types of herbs sold in the Iran (Kurdistan region) markets, recorded (0.19–3.95 μg·g^{-1}) all these data were within the permissible limits of Zn levels. This difference may be due to the type of plant and the type of soil as well as the environmental pollution for irrigation water and soil by different sources also some properties of the metals such as mobility and chemical behavior. The highest allowable WHO level of Pb is 10 μg·g^{-1} in all parts of the plant (WHO, 2007). All plant samples showed low Pb concentrations compared to the acceptable limits set by WHO and FAO. In current study, the lead concentration was given in the range of 0.87–5.77 μg·g^{-1} these values were agreement with other studies Maobe et al. (2012); Kohzadi et al. (2019).

It is important to note that the total heavy metals calculated per one gram that an adult person eats per day may be caused healthy effects as they are toxic elements and their accumulated in the same tissues of the body. On the other hand, the *Thymus vulgaris Nigella sativa*, *Mentha verticillata*, *Matricaria chamomilla* and *Hibiscus sabdariffa* plant were hyperaccumulator of total heavy metals (Table 2), indicated that the differences may attributed to the uptake, distribution, accumulation and different geological background of cultivated area. The accumulation of metals in plants depending on several factors such as physicochemical properties of the soil, cultivation, bioavailability, chemical behavior

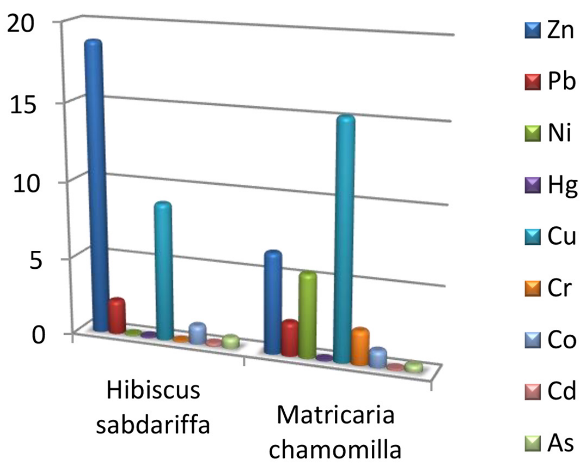

Figure 3. Mean concentration of heavy metals (μg·g^{-1}) at flowers

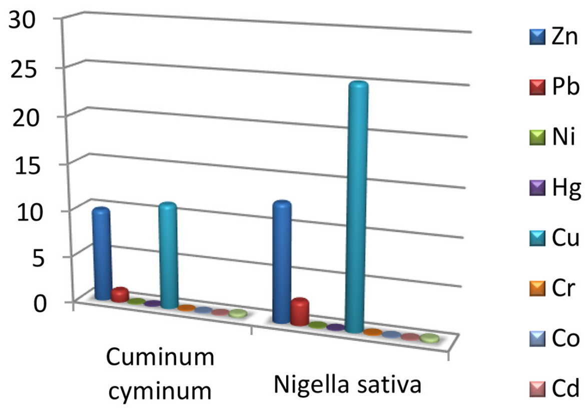

Figure 4. Mean concentration of heavy metals (μg·g^{-1}) at seeds

Table 4. Target hazard quotient (HQ) and Hazard Index (HI) of nine heavy metals from consumption of medicinal herbs and spices

Herbs and medicinal plant	Metals									Hazard index HI
	As	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn	
Glycyrrhiza glabra L.	0.15	0.08	0.12	0.01	0.29	0.12	0.23	0.27	0.01	1.29
Zingiber officinale	0.05	0.12	0.07	0.01	0.16	0.12	0.23	0.17	0.01	0.94
Curcurma longa	0.07	0.01	0.05	0.01	0.19	0.12	0.23	0.46	0.01	1.14
Thymus vulgaris	0.24	0.05	3.5	0.01	4.1	0.12	3.3	1.13	1.2	13.5
Mentha verticillata	0.16	0.12	3.2	0.01	0.42	0.12	15.5	0.51	0.75	20.78
Cinnamomum verum	0.59	0.59	0.12	0.01	0.33	0.12	0.23	0.88	2.3	9.52
Hibiscus sabdariffa	0.16	0.02	1.3	0.01	0.20	0.12	0.23	0.43	2.14	4.63
Matricaria chamomilla	0.11	0.01	1.3	0.17	0.34	0.12	12.3	0.42	0.8	15.54
Cuminum cyminum	0.09	0.08	0.12	0.01	0.25	0.12	0.23	0.24	1.1	2.28
Nigella sativa	0.10	0.12	0.12	0.01	0.57	0.12	0.23	0.49	1.4	3.20

of the heavy of metals in the soil (Sharma et al., 2018). One of the factors affecting the distribution of heavy metals between plant parts is the mobility of elements from soil to the aboveground plant parts and translocation index, Previous studies of many plant species showed that some metals accumulated in the highest concentrations in flowers compared to the other parts, then they had posed highest risk due to its high mobility within the plant. (Abou-Arab and Abou Donia, 2000; Hladun and Trumble, 2015; Kim et al., 2017). Singh and Garg (2006) find out the cinnamon was more content of Fe, Co, Cr, Na, K, P and Zn, while curry and turmeric leaves were detected high concentration of Se. Also seeds of mustard and Cumin were rich in Cu. Then, some elements were concentrated more in the leaves than other elements accumulated in the bark, roots, or rhizomes. Chromium, arsenic and mercury recorded the highest concentrations (2.2, 2.0 and 1.01 μg/g), which did not exceed the acceptable limits for all samples. The statistical analysis showed significant differences ($P<0.05$) between the content of herbal leaves for all the elements of thyme and mint plants, except for both zinc and arsenic, which recorded the highest concentration in the bark of the cinnamon plant.

Estimated daily intake of elements

The estimated daily intakes (EDIs) of Cu, Ni, Zn, Pb, Co, As, Cd, Cr and Hg were calculated according to the concentration of each element in each herb and the daily intake rate for adults of 60 years of age with an average body weight of 60 kg. These data detected that the some herbs powder had lower values for daily metal consumption as shown in Table 3 the EDIs of selected trace metals were equivalent among many elements of 7×10^{-5} $\mu g\cdot g^{-1}$ $bw\cdot day^{-1}$ for several herbs, While it recorded o.1 $g\cdot kg^{-1}$ $bw\cdot day^{-1}$ for copper in *Thymus vulgaris* plant samples. The EDI values were higher than RfD of copper was detected (0.1, 0.01 and 0.7) for in *Thymus vulgaris*, *Mentha verticillata* and *Cinnamomum verum* plants and *Nigella sativa* respectively, also increased to 0.08, 0.03 and 0.01 for Ni, Pb and Zn in ment and *Nigella sativa* plant respectively, with ingestion references dose for these elements (0.02) for Ni, 0.00057 for Cu, (0.0035) for Pb and Zn=0.006 $mg\cdot kg^{-1}$ $bw\cdot d^{-1}$ (Li et al., 2019). These levels were below the WHO allowable daily dose of 240 g/day for adults weighing 68 kg (Garcia-Rico et al., 2007). The EDIs decreasing followed the order: Cu > Zn > Ni > Co > Cd>Cr >As >Co >Hg. The copper, Zink and Nickel contribution were the highest among the daily intakes of elements, the values of Non-cancer risk probability were 0.1, 0.01 and 0.7) which were more than the acceptable cancer risk of 10^{-6}~10^{-4}.

Statistical analysis showed that thyme, mint and cinnamon bark had the highest value of (EDIs) for Cu ($P<0.05$).

Health hazard estimation

Target hazard quotient and hazard index

Exposure to heavy metals daily through food and vegetables is a public health concern, the Target hazard quotients (THQ) and Hazard Index (HI) of nine heavy metals from consumption of culinary herbs and seasoning were calculated to assess Non-carcinogenic risks (Table 4). The maximum values of THQ was recorded more than one (15.5, 12.3, 3.3) for the Ni, (3.5, 3.2, 1.3) for Co, (2.14,2.3,1.41.2,1.1) for Zn, (4.1) for Cu and (1.13) for Pb in many samples of culinary herbs. Therefore, these values are higher than the permissible limits of FAO/WHO (≥1) according to the USEPA (Islam et al.,2014). In addition, Heavy metals (Cd, As, Cr, and Hg) have shown the lowest non-carcinogenic (HQ≤1) none pose more severe risks than five other heavy metals.

The value of Hazard Index (HI) which is the value of hazard resulting from the nine elements combined in one sample of herbs and seasoning were recorded values higher than one with except in *Zingiber officinale* was 0.94. Furthermore, if more than one herb is ingested, it is possible for the total HI value to be more than 1 and unsafe for consumers. The order of elements for THQ was Ni >Cu >Co >Zn >Pb >As >Cd >Cr >Hg, was observed for the average THQ values of each metal can be attributed to the anatomical and physiological differences of various herbs for accumulating metals, These results were consistent with the previous studies (Obi et al., 2006; Gao et al., 2021), they revealed that all the herbal samples contained elevated amounts of from Fe, Ni, Cd, Cu, Pb, Se and Zinc with Hazard quotient values higher than the standard value (HQ> 1) will cause dangerous health effects in humans when taken as recommended. Moreover (Lu et al., 2020) reported that the accumulation of heavy metals in some herbal remedies caused unacceptable health risks that exceeded permissible limits according to exposure and health risk assessments. Notably, this may be attributed to the fact that the heavy metal content of medicinal herbs has health risks for consumers.

CONCLUSIONS

In the current study, trace elements frequently accumulated in the aerial parts of the plant such as leaves, seeds and bark, and the high concentrations of heavy metals in the herbs consumed on the basis of daily consumption were high or within the permissible ranges thus posing health risks to the consumers. However, in Iraq and other countries, culinary, seasonings, herbal medicines and related products are introduced to the markets without any mandatory assessment of health safety or the content of these herbs from toxins, pesticide residues, and other contaminants. Many of these countries also lack effective oversight to regulate quality standards and manufacturing processes. However, medicinal plants must be obtained from trusted sellers or collected from uncontaminated areas. We hope that this study will be useful in providing information and advice to organizations and people who prepare natural herbal remedies.

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