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Original Research

Using Stripping Voltammetry to Determine Heavy Metals in Cooking Spices Used in Iraq

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Abstract

Monitoring heavy metals in herbs and spices is increasingly being reported from different parts of the world. In Iraq and the Middle East, there is limited information on the levels of heavy metals in these additives, although several spices are widely used in daily diets. In the present study the concentrations of Cu, Zn, Fe, Mn, Cr, Ni, Co, Cd, Pb, and Hg in 32 kinds of natural spices traditionally consumed in Iraq were determined by stripping voltammetry. The highest concentration found (in mg kg^{-1} dry weight) was that of iron (32.44-1,147) followed by manganese (6.42–285.8), zinc (5.45-129.3), and copper (2.58-30.71). The essential elements of chromium, nickel, and cobalt were found at comparatively lower concentrations of less than 5.900 mg kg^{-1}, whereas the levels of the toxic elements Cd, Pb, and Hg were even lower (0.010-4.159 mg kg^{-1}). Strong elemental correlations (≥0.500) exist between the transition metals Cu, Zn, Fe, Ni, Co, and Cr, reflecting their chemical nature and physiological functions. The average daily contribution of Cu, Fe, Mn, and Cr from spices to the Iraqi requirements was estimated to be 21-61% of the internationally recommended standards, while the levels of the toxic elements Cd, Pb, and Hg were low and were found to pose no threat to consumers.

Keywords: spices, heavy metals, daily intake, voltammetry, Iraq

Introduction

Spices can be defined as vegetable products used for flavoring, seasoning, and imparting aroma in foods [1]. In addition, many spices are widely regarded as having medicinal properties such as antioxidant and antimicrobial potential, and hence play a role in the preparation of a number of medicines [1-2].

Among several components, spices may contain trace heavy metals in a wide concentration range. Heavy metals like Cu, Zn, Fe, Mn, Cr, Co, and Ni are required in trace quantities for the proper functioning of enzyme systems, hemoglobin formation, and vitamin synthesis, whereas metals like Cd, Pb, and Hg are non-essential metals that can be toxic even in trace amounts. The essential metals can also have harmful effects when their intakes exceed the recommended quantities [3].

Concentrations of heavy metals in spices may be influenced by geochemical characteristics of the soil in which the plants are cultivated, and environmental pollution levels. Also, industrial processing, packaging, transportation, and storage conditions can play a significant

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role in elevating the contaminant levels of heavy metals, which may affect the quality and safety of spices.

Generally, the addition of spices that may be contaminated with heavy metals to foods can lead to serious consequences for human health, because most heavy metals are not biodegradable and have the potential for accumulation in different human body organs. For this reason, monitoring heavy metals in herbs and spices has recently been an important topic in many articles and is one of the most important aspects of food quality assurance [4-9].

In Iraq, there is limited information on the levels of heavy metals in these additives, although several spices are widely used in daily diets. Thus, the primary objective of the present study was to determine the concentration of seven essential elements (Cu, Zn, Fe, Mn, Cr, Ni, and Co) and three toxic elements (Cd, Pb, and Hg) in the most frequently consumed spices in Iraq, and to assess the total daily intake of these elements compared to the internationally permissible values. The stripping voltammetry technique was used in this work due to its low-cost instrumentation and its well-known advantages of sensitivity, selectivity, and multi-element analysis capability. It is also superior to other trace analysis techniques as it works in the presence of high salt concentrations [10]. Monitoring the levels of heavy metal toxicity in spices would help to establish the health impact of consuming these spices and provide relevant data on spices throughout the country.

Materials and Methods

Sampling and Classification

The concentrations of Cu, Zn, Fe, Mn, Cr, Ni, Co, Cd, Pb, and Hg were measured in a total of 160 samples of 32 different spices that represent the most widely used taste enhancers in Iraq. The samples were chosen from various shops and supermarkets in Babil (100 km south of Baghdad), so as to have as representative samples as possible [11]. Most of the natural spices used in food preparation are either from Iraq or imported from Turkey, Iran, Pakistan, India, China, and south Asia. The spices analyzed were the bark, buds, flowers, leaves, fruit, seeds, rhizomes, or roots of a plant without any commercial milling to avoid the introduction of trace elements into the seasonings due to wear and tear of the machinery. The spices were divided according to the used part of the plant and their scientific and local names [1] (Table 1). The samples were kept in polyethylene bags in dry cardboard in a cool place prior to analysis.

Reagents

Deionized water (Milli Q Millipore 18.2 MΩ cm^{-1} resistivity) was used to prepare the solutions. All the reagents used were of analytical purity (Merck, Darmstadt, Germany). The working solutions were prepared immediately before analysis from the basic solution with 1,000 mg L^{-1} concentration for all metals. The glassware and polyethylene containers used for analysis were washed with tap water, then soaked overnight in 6 M HNO_3 solution and rinsed several times with ultra-pure water to remove any residual metals.

Sample Preparation

All the collected spices (except tamarind and saffron) were washed thoroughly with running tap water and followed by washing with deionized water to remove the dust particles and possible parasites. Tamarind and saffron lose a significant amount of their weight during washing processes. Thus, instead of washing they were wiped thoroughly with tissues. All the samples were then oven-dried at 80ºC for 24 hours to remove inherent water molecules and were kept for further analyses.

Dry, wet, and microwave digestions are the most common methods of bringing the analytes from organic matrix into solution in order to analyze. Based on the experience with these methods and to minimize reagent contamination, the following recipe was adopted. One gram of dry sample was introduced into a muffle furnace and the temperature increased very slowly to 550ºC. The white ash obtained after 12 hours was dissolved in 10 mL of 0.25% nitric acid. When necessary, more acid was added and the mixture was heated slowly until a clear solution was obtained. The resulting solutions were diluted to 50.00 mL and analyzed immediately.

Apparatus and Quantification

The digested solutions were analyzed by anodic, cathodic, and adsorptive stripping voltammetric using a Metrohm 797 VA Trace Analyzer. The pH of the samples was measured using a pH-meter (Model 720, WTW). Quantification of metal concentrations in the samples was carried out using the standard addition method (this is the preferred method as the sensitivity of the analysis may vary between samples of different ionic strengths). The best fit line through data pairs was calculated by linear-squares regression analysis. The concentration of each element in the sample was equal to the quotient of the intercept and the regression coefficient. Instrument control, data acquisition, and voltammetric data analysis were accomplished using Wizaard software (Application Bulletin Metrohm, Method No. 114/1e) with certain modifications (Table 2).

Validation of Analytical Methods

All spice samples were analyzed under strict analytical control with the use of regular check standards and the analysis of a certified reference material. The calibration curves for all the elements were built on 6 different concentrations. The curves were linear over the concentration range from the limit of detection of the corresponding element up to 300 µg L^{-1}, with a correlation

Table 1. Spice samples with their common and scientific names.

No.	Common name	Botanical name	Family	Part analyzed
1	Mint	Mentha	Lamiaceae	Leaf
2	Thyme	Thymus vulgaris	Lamiaceae	Leaf
3	Bay Laurel	Laurus nobilis	Lauraceas	Leaf
4	Basil	Ocimum basilicum	Lamiaceae	Leaf
5	Celeriac	Apium graveolens	Apiaceae	Leaf
6	Betony	Stachys officinalis	Lamiaceae	Leaf
7	Parsley	Petroselinum crispum	Lamiaceae	Leaf
8	Black seed	Nigella Sativa	Ranunculaceae	Seed
9	Fenugreek	Trigonella foenumgraecum	Fabaceae	Seed
10	Fennel	Foeniculum vulgare	Apiaceae	Seed
11	Coriander	Coriandrum sativum	Apiaceae	Seed
12	Cumin	Cuminum cyminum	Apiaceae	Seed
13	Cardamom	Elettaria cardamomum	Zingiberaceae	Seed
14	Mustard	Brassica junceae	Brassicaceae	Seed
15	Sesame	Sesamum indicum	Pedaliaceae	Seed
16	Long pepper	Piper longum	Piperaceae	Fruit
17	Green pepper	Piper nigrum	Piperaceae	Fruit
18	Black grainy pepper	Piper nigrum	Piperaceae	Fruit
19	Cubeb	Piper cubeba	Piperaceae	Fruit
20	Omani limes	Citrus aurantifolia	Rutaceae	Fruit
21	Sumac fruit	Rhus Coriaria	Anacardiaceae	Fruit
22	Chili pepper	Capsicum annuum	Solanaceae	Fruit
23	Aniseed	Pimpinella anisum	Apiaceae	Fruit
24	Tamarind	Tamarindus indica	Cesalpiniaceae	Fruit pulp
25	Garlic	Allium sativum	Liliaceae	Bulb
26	Onion	Allium cepa	Liliaceae	Bulb
27	Cloves	Syzygium aromaticum	Myrtaceae	Flower bud
28	Nutmeg	Myristica fragrans	Myristicaceae	Kernel
29	Ginger	Zingiber officinale	Zingiberaceae	Rhizomes (root)
30	Turmeric	Curcuma longa	Zingiberaceae	Rhizomes (stem)
31	Cinnamon	Cinnamonum cassia	Lauraceae	Bark
32	Saffron	Crocus sativus	Iridaceae	Stigma

coefficient between 0.9975-0.9990 for the 10 elements. The limits of detection and quantification were calculated (from these curves) with three- and 10 times the standard deviation of the blank solution divided by the slope of the analytical curve, respectively [12]. The values of LOD were in the range of 0.4-8.0 (μg L^{-1}) and the LOQ were 1.3-26 (μg L^{-1}) (Table 3).

Precision was obtained as percent coefficient of variation (CV%) from the relative standard deviation of 10 repeated determinations of one sample [6]. The percent coefficients of variation (CV%) obtained for all trace elements were below 5.80%, ranging from 2.50 to 5.80% (Table 3).

The reliability of the method has been checked by the certified standard reference material: spinach leaves (SRM 1570a) from NIST, Gaithersburg, Maryland, USA. The reference material was analyzed immediately after preparation following the same procedure described

Table 2. Optimal conditions for detecting metal ions in digested spices using stripping voltammetry.

	Cd, Pb, Cu, Zn	Fe	Mn	Co, Ni	Cr	Hg
Technique	ASV/HMDE	ADSV/HMDE	ASV/HMDE	ADSV/HMDE	ADSV/HMDE	ASV / RDE
Volume of sample*+ supporting electrolyte	10 mL diluted sample + 1mL KCl-sodium acetate solution	10 mL diluted sample + 100µL catechol (1M) +1mL phosphate buffer	10 mL diluted sample +50 µL NH_3 solution (25%) +2.5 mL borate buffer+10 µL Zn solution (100 mg/L)	10 mL diluted sample + 0.5 mL NH_4Cl buffer +0.1 mL DMG	20 mL sample + 1 mL Supporting electrolyte	10 mL sample + 1 mL Supporting electrolyte
pH	4.6	7.0	10.5	9.5	6.2	< 1
Drop size	4	7	4	4	9	------
Stirring/min	2000	2000	2000	2000	2000	2000
Purging time/s	300	300	300	300	300	------
Deposition potential/V	-1.15	-0.300	-1.650	-0.7	-1.0	0.37
Deposition time/s	30-120	60	90	90	20	90
Equilibrium time/s	10	5.0	5	10.0	20	10
Start potential/V	-1.15	-0.20	-1.620	-0.8	-1.0	0.40
End potential	0.05	-0.50	-1.250	-1.25	-1.4	0.70
Pulse amplitude	0.05	0.050	0.075	0.05	0.05	0.05
Pulse time/s	0.04	0.04	0.04	0.04	0.04	0.04
Voltage step/V	0.006	0.004	0.004	0.004	0.006	0.002
Voltage step time/s	0.1	0.4	0.5	0.3	0.1	0.1
Sweep rate/V/s	0.060	0.010	0.008	0.013	0.06	0.02
Peak position/V	Zn(-0.980) Cd(-0.560) Pb (-0.380) Cu (-0.100)	-0.380	-1.440	Ni (-0.97) Co (-1.13)	-1.2	0.60

* Sample: digested solution.
KCl-sodium acetate solution: 11.18 g KCl + 5 mL (30% NaOH) + 2.84 mL (100% CH_3COOH) filled up to 100 mL with ultrapure water. **Phosphate buffer**: 2.3996 g NaH_2PO_4 and 4.2594 g Na_2HPO_4 filled up to 100 mL with ultrapure water. **Borate buffer:** (3.81 g $Na_2B_4O_7$.10 H_2O in 50 mL water + 3 mL NaOH solution 30%) filled up to 100 mL with distilled water. **NH_4Cl buffer**: 22.5 mL NH_3 + 10.6 mL HCl filled up to 100 mL with high purity water. **DMG solution:** 0.304 g Dimethylglyoxim disodium salt in 10 mL ultrapure water. **Supporting electrolyte for Cr:** 1.96 g diethylenetriaminepentaacetic acid + 1.64 g sodium acetate + 21.3 g sodium nitrate +1.0 mL NaOH solution (30%) filled up to 100 ml with H_2O**.** **Supporting electrolyte for Hg:** 0.372 g Na_2EDTA and 0.146 g NaCl are dissolved in 40 mL ultrapure water and 5.56 mL H_2SO_4. After cooling down to room temperature they are made up to 50 mL with ultrapure water. **ADSV**: adsorptive stripping voltammetry; **HMDE**: hanging mercury drop electrode; **CSV**: cathodic stripping voltammetry; **ASV**: anodic stripping voltammetry; **RDEV:** Rotating disk electrode voltammetry.

for the other samples. The determined concentrations in reference materials were within the range 90.0-94.9% of the certified range (Table 4). The accuracy was also checked with recovery assays by spiking the samples at different selected concentrations prior to the digestion step [10]. The spiked samples were prepared by adding 1,000 µL of 1,000 mg L^{-1} Fe, 300 µL of 1,000 mg L^{-1} Mn, 150 µL of 1,000 mg L^{-1} Zn, 100 µL of 1,000 mg L^{-1} Cu, and 100 µL of 100 mg L^{-1} Cr, Ni, Cd, Pb, and Hg standard solutions to 1.0 g of each spice. The spiked and non-spiked samples were digested and analyzed in similar conditions. The results of the percentage recoveries for the studied elements (Table 3) were within the acceptable range (90-106%). Both analytical accuracy and precision were acceptable by the AOAC international guidelines [12], suggesting that this method can be used as routine analysis of heavy metals in vegetables and spices.

Statistical Analysis

The Data were analyzed using the Statistical Package for Social Sciences (SPSS, version 17.0). T-testing was employed when necessary to detect significant differences among means. Unless otherwise indicated, data are presented as mean ± SD. A probability level of $p < 0.05$ was considered statistically significant.

Table 3. Limits of detection (LOD), limits of quantification (LOQ), precision (CV%), and spike recovery (%) for the trace elements analyzed.

Trace Element	LOD (μg L^{-1})	LOQ (μg L^{-1})	Precision (CV%)	Spike Recovery (%)
Cu	4.0	13	2.95	105
Zn	4.0	13	2.50	93.1
Fe	8.0	26	4.72	109
Mn	5.0	16	3.44	101
Cr	7.5	25	5.20	90.6
Ni	1.0	3.4	4.18	92.8
Co	3.5	12	3.97	91.3
Cd	0.4	1.3	5.76	88.9
Pb	1.0	3.4	3.98	91.6
Hg	0.4	1.5	5.80	88.1

Table 4. Determination of Cu, Zn, Mn, Ni, Co, Cd, and Hg in certified reference material (NIST-1570a, spinach leaves).

Element	Accuracy		
	Certified value (mg kg^{-1})	Observed values (mg kg^{-1})*	Recovery (%)
Cu	12.2±0.6	11.3±0.7	92.6
Zn	83±3	78.8±4	94.9
Mn	75.9±1.9	70.9±3	93.4
Ni	2.14±0.10	1.98±0.14	92.5
Co	0.39±0.05	0.36±0.07	92.3
Cd	2.89±0.07	2.63±0.12	91.0
Hg	0.030±0.003	0.027±0.004	90.0

* Mean of five determinations obtained by the proposed procedure.

Results

All the spices tested in this study were found to contain detectable levels of heavy metals. The concentrations of Cu, Zn, Fe, Mn, Cr, Ni, Co, Cd, Pb, and Hg elements in 32 spices commonly consumed in Iraq are presented in Tables 5 and 6: Cu (2.58-30.71), Zn (5.45-129.3), Fe (32.44-1147), Mn (6.42-285.8), Cr (0.052-1.317), Ni (0.280-5.900), Co (0.020-0.754), Cd (0.011-1.389), Pb (0.250-4.159), and Hg (0.032-2.870) mg kg^{-1}. In general, the average Cu+Zn+Fe+Mn level of leafy spices (773 mg kg^{-1}) was higher than seeds (346 mg kg^{-1}) and fruits (229 mg kg^{-1}). The highest variability and concentration found was that of iron, followed by manganese, zinc, and copper. These results were compared with the ranges currently available in the literature (Table 7) [4, 5, 13-23].

The effect of washing on the level of trace elements in 23 spices (usually used without washing) was determined. The levels of Cu, Cd, Pb, and Hg in unwashed spices were found to be 36, 14, 7, and 11% higher than those in washed spices, respectively (Cu is presented in Fig. 1).

A linear regression correlation test was performed to investigate correlations between the metal contents in all tested spice samples. The values of correlation coefficients between metal concentrations are given in Table 8. The pairs of Fe-Ni (0.723), Cu-Zn (0.694), Fe-Co (0.673), Ni-Co (0.658), Cr-Co (0.586), and Cu-Fe (0.500) showed high and significant correlations at 95% confidence level, while the toxic metals Cd, Pb, and Hg show weak correlation with the rest of the elements.

To evaluate the average quantity of spices consumed by ordinary Iraqis, 30 undergraduate female students were requested to monitor the time required by their families to consume certain amounts of the studied spices. After six months the average quantity of spices consumed by ordinary Iraqis was found to be 18.3±5.4 g daily. The average daily intake of trace elements from spices was calculated by multiplying the mean concentration of each element by the average consumption rate of ordinary Iraqis. The results were compared with the internationally recommended standards [24] and both are presented in Table 9.

Discussion

Variability of Elemental Concentrations in Different Spices

As can be seen from Tables 5 and 6, there are large differences in elemental levels of different spices due to differences in genotypes and various ecosystems. The highest variability was found in iron, followed by manganese, zinc, and copper. This pattern is recorded in most of the published articles on herbs and spices (Table 7). Moreover, a remarkable variability of elemental concentrations was also observed in individual spices cultivated at different sites. This was reflected in the relatively high standard deviations and the remarkable differences between the mean and the median of the elemental contents (Tables 5-6). Similar results were reported by other studies for different spices [17, 19, 23, 25-26]. This variability is usually explained in terms of the effects of various ecosystems on the elemental levels at different regions. The major source originated from the difference in mineral uptake in plants as a function of mineral concentrations in soils [27]. The other probable reasons could be the influence of soil pH, cation exchange capacity, organic matter content, age of the plant, and that some sample sites may be exposed to intensive industrial activities [28]. To overcome these obstacles and to set adequate reference values for different elements (especially toxic ones), a great number of samples of the considered plant must be analyzed and the values fixed so that 90-98% of the samples remain below them.

Table 5. Concentrations* of Cu, Zn, Fe, Mn, Cr, Ni, and Co (mg kg^{-1}) in dry spices

Common name *(part investigated)*	Cu	Zn	Fe	Mn	Cr	Ni	Co
Mint (*Leaf*)	18.66±2.21	51.37±6.69	864.3±155.1	83.81±18.41	1.258±0.252	3.181±0.859	0.754±0.241
Thyme *(Leaf)*	3.49±1.40	25.20±3.50	356.8±82.33	12.48±3.42	0.386±0.093	1.351±0.297	0.269±0.081
Bay laurel (*Leaf*)	8.19±1.07	18.44±3.41	235.2±42.15	72.32±13.75	0.521±0.112	0.969±0.292	0.085±0.034
Basil (*Leaf*)	25.52±2.89	128.7±15.51	1147±275.2	139.2±36.42	0.887±0.231	2.071±0.518	0.412±0.074
Celeriac (*Leaf*)	16.48±3.71	74.53±11.91	400.4±112.1	52.81±15.31	0.574±0.166	2.562±0.717	0.228±0.078
Betony (*Leaf*)	20.66±3.75	74.40±19.81	663.2±139.1	50.86±10.24	1.096±0.339	5.900±1.416	0.607±0.152
Parsley(*Leaf*)	9.69±1.51	89.66±15.17	732.7±182.7	32.71±7.53	0.472±0.104	2.671±0.695	0.341±0.099
Black seed (*Seed*)	9.57±2.62	85.36±17.11	110.3±15.12	22.74±3.46	0.249±0.055	2.342±0.492	0.081±0.018
Fenugreek (*Seed*)	16.74±2.88	43.72±4.80	111.7±21.94	12.02±1.91	0.340±0.136	1.181±0.366	0.466±0.084
Fennel (*Seed*)	16.69±3.62	66.63±11.91	268.4±62.05	30.14±5.42	0.164±0.029	1.561±0.281	0.175±0.049
Coriander (*Seed*)	13.78±1.30	65.50±9.15	152.4±40.77	18.28±2.92	0.256±0.061	0.565±0.152	0.043±0.020
Cumin (*Seed*)	11.19±0.96	97.42±12.70	404.8±72.82	27.02±5.73	0.282±0.056	1.099±0.308	0.455±0.146
Cardamom (*Seed*)	5.32±0.42	55.27±9.38	92.23±17.66	201.1±28.32	0.281±0.092	1.219±0.195	0.145±0.044
Mustard (*Seed*)	3.15±1.23	61.91±9.28	331.2±49.12	22.62±4.31	0.052±0.022	1.780±0.463	0.084±0.037
Sesame (*Seed*)	30.71±4.87	109.3±12.00	257.7±43.87	14.64±1.62	0.488±0.110	1.452±0.261	0.100±0.054
Long pepper (*Fruit*)	11.72±0.65	26.72±4.28	72.43±19.21	10.12±2.14	1.317±0.342	2.471±0.692	0.495±0.149
Green pepper (*Fruit*)	6.39±0.81	35.06±7.67	79.52±16.83	14.32±3.64	0.745±0.216	0.388±0.124	0.120±0.031
Black pepper (*fruit*)	8.09±1.34	9.06±0.89	194.1±39.03	189.1±53.22	1.044±0.424	4.904±1.618	0.412±0.136
Cubeb (*Fruit*)	8.39±2.09	10.52±1.39	91.61±26.58	6.55±1.24	0.244±0.078	0.369±0.148	0.020±0.018
Omani lime (*Fruit*)	3.26±0.67	5.45±1.68	336.4±108.1	6.42±2.56	0.448±0.085	0.462±0.139	0.107±0.039
Sumac (*Fruit*)	4.93±0.49	19.64±2.81	43.85±6.31	7.23±1.52	0.337±0.115	0.939±0.253	0.063±0.044
Chili pepper (*Fruit*)	13.35±0.85	26.14±4.71	66.72±6.24	11.68±2.71	0.254±0.096	0.897±0.215	0.158±0.061
Aniseed (*Fruit*)	12.79±1.71	58.06±12.78	243.1±61.08	23.82±5.66	0.297±0.103	0.869±0.121	0.231±0.042
Tamarind (*Fruit pulp*)	4.35±1.24	13.52±4.11	383.8±133.8	10.72±4.14	0.144±0.036	0.437±0.131	0.339±0.119
Garlic (*Bulb*)	5.73±1.12	26.39±2.88	35.11±4.87	11.92±1.86	0.234±0.050	0.905±0.298	0.063±0.029
Onion (*Bulb*)	7.76±1.04	16.89±2.69	45.64±6.42	16.43±2.01	0.379±0.091	1.577±0.252	0.087±0.024
Cloves (*Flower bud*)	5.22±0.41	14.77±1.76	232.3±51.12	285.8±63.22	0.146±0.040	0.276±0.055	0.104±0.037
Nutmeg (*Kernel*)	6.76±1.61	19.31±3.10	42.72±8.11	18.55±2.42	0.081±0.049	1.532±0.260	0.023±0.019
Ginger (*Rhizomes*)	5.18±1.90	14.51±1.20	32.44±5.22	186.3±30.01	0.250±0.075	1.356±0.325	0.121±0.024
Tumeric (*Rhizomes*)	2.58±0.76	11.53±1.51	159.6±15.33	181.3±44.11	0.500±0.115	0.683±0.200	0.279±0.101
Cinnamon (*Bark*)	7.87±1.08	22.80±2.09	112.3±21.02	85.21±19.33	1.197±0.311	0.620±0.198	0.116±0.044
Saffron crocus (*Stigma*)	11.77±2.45	36.93±9.91	255.3±87.06	17.11±6.41	0.431±0.259	1.375±0.611	0.431±0.236
Average/mg kg^{-1}	**10.50±2.03**	**44.21±8.88**	**267.4±86.08**	**58.60±20.43**	**0.480±0.171**	**1.560±0.536**	**0.232±0.094**
Median/mg kg^{-1}	**8.29**	**30.91**	**213.2**	**22.68**	**0.340**	**1.285**	**0.151**
Range/mg kg^{-1}	**2.58-30.71**	**5.45-129.3**	**32.44-1147**	**6.42-285.8**	**0.052-1.317**	**0.280-5.900**	**0.020-0.754**

*Values are expressed as Mean±SD, number of samples analyzed for each spice = 5, SD = standard deviation

Table 6. Concentrations* of Cd, Pb, and Hg (mg kg^{-1}) in dry spices.

Common name *(part investigated)*	Cd	Pb	Hg
Mint (*Leaf*)	0.043±0.006[b]	1.218±0.465	0.196±0.071
Thyme *(Leaf)*	0.015±0.007	0.862±0.138	0.365±0.146
Bay laurel (*Leaf*)	0.032±0.009	0.498±0.162	0.237±0.052
Basil (*Leaf*)	0.300±0.042	1.056±0.386	0.124±0.041
Celeriac (*Leaf*)	0.362±0.159	1.102±0.445	0.145±0.036
Betony (*Leaf*)	0.274±0.123	3.743±1.222	0.496±0.198
Parsley(*Leaf*)	0.243±0.009	3.069±0.888	0.217±0.082
Black seed (*Seed*)	0.073±0.027	0.258±0.016	0.351±0.072
Fenugreek (*Seed*)	0.043±0.027	0.276±0.096	0.082±0.046
Fennel (*Seed*)	0.016±0.006	0.316±0.151	0.082±0.064
Coriander (*Seed*)	0.039±0.021	0.584±0.059	2.870±1.320
Cumin (*Seed*)	0.157±0.032	0.992±0.367	0.494±0.148
Cardamom (*Seed*)	0.156±0.024	0.583±0.256	0.821±0.213
Mustard (*Seed*)	0.091±0.029	0.465±0.115	0.171±0.053
Sesame (*Seed*)	0.021±0.006	0.347±0.103	0.053±0.046
Long pepper (*Fruit*)	0.219±0.050	0.967±0.398	0.417±0.117
Green pepper (*Fruit*)	0.154±0.037	0.375±0.136	0.174±0.050
Black pepper (*fruit*)	0.093±0.050	0.317±0.157	0.311±0.087
Cubeb (*Fruit*)	0.015±0.002	3.353±1.243	0.182±0.069
Omani lime (*Fruit*)	0.022±0.003	0.432±0.078	0.662±0.179
Sumac (*Fruit*)	0.011±0.005	0.361±0.169	0.263±0.063
Chili pepper (*Fruit*)	0.022±0.004	0.550±0.168	0.032±0.030
Aniseed (*Fruit*)	0.070±0.008	0.269±0.111	0.240±0.067
Tamarind (*Fruit pulp*)	1.389±0.152	0.825±0.332	0.422±0.298
Garlic (*Bulb*)	0.067±0.051	0.250±0.083	0.282±0.104
Onion (*Bulb*)	0.025±0.021	0.480±0.209	0.156±0.052
Cloves (*Flower bud*)	0.123±0.004	0.541±0.210	0.128±0.031
Nutmeg (*Kernel*)	0.038±0.030	0.582±0.387	0.089±0.033
Ginger (*Rhizomes*)	0.121±0.029	0.812±0.323	0.177±0.030
Tumeric (*Rhizomes*)	0.227±0.130	0.387±0.101	0.076±0.021
Cinnamon (*Bark*)	0.874±0.177	4.159±0.943	1.055±0.359
Saffron crocus (*Stigma*)	0.216±0.161	2.007±0.851	1.445±0.882
Average/mg kg^{-1}	**0.171±0.070**	**0.968±0.481**	**0.400±0.306**
Median/mg kg^{-1}	**0.082**	**0.556**	**0.227**
Range/mg kg^{-1}	**0.011-1.389**	**0.250-4.159**	**0.032-2.870**

*Values are expressed as Mean±SD, number of samples analyzed for each spice =5, SD= standard deviation

Table 7. Comparison of average metal contents (mg kg^{-1} dry weight) of natural spices consumed in Iraq with other areas.

State/ No. of spices	Cu	Zn	Fe	Mn	Cr	Ni	Co	Cd	Pb	Hg	Ref.* No.
Iraq (32)	2.58-30.7	5.5-129	32.4-1147	6.4-286	0.052-1.317	0.280-5.900	0.020-0.754	0.010-1.281	0.250-4.159	0.032-2.87	Present work
Poland (13)	4.20-9.12	6.86-30.84	----	------	------	------	-------	0.01-0.14	0.25-1.45	-----	[13]
Turkey (12)	4.1-28.7	7.84-47.6	88.9-376.3	11.9-211.3	0.67-7.15	0.65-8.69	-----	0.10-0.14	0.47-1.89	-----	[14]
India (5)	0.195-2.581	BDL**-2.575	1.699-17.87	0.670-5.729	BDL-0.156	-----	-------	BDL-0.007	BDL-0.215	BDL	[15]
India (20)	nd-7.84	30.3-201	17.1-1920	nd-1060	nd-9.14	-----	0.008-0.740	----	-----	nd-1.970	[16]
Poland (15)	3.17-24.2	9.26-86	20.0-1280	5.50-802	-----	0.19-26.5	------	----	-----	----	[17]
Turkey (9)	2.78-12.17	16.5-45.7	53-409	17.6-164.7	0.44-3.14	0.32-8.05	0.06-0.52	<LOD-0.118	-----	----	[5]
Egypt (20)	nd-11.40	1.4-68.80	11.4-1046	23-343.0	3-33.75	nd-2.85	nd	nd-2.44	0.5-14.4	-----	[18]
Japan (18)	-------	6.7-51.2	25-1100	7.4-930	-------	1.3-2.9	0.02-0.87	-------	-------	-------	[19]
Turkey (11)	3.8-35.4	5.2-83.7	30.0-945.3	7.9-152.5	0.1-9.7	1.4-11.3	-------	0.2-2.7	0.1-2.8	-------	[20]
India (8)	3.83-27.53	31-115.77	80-1390	33.6-107.62	0-3.99	0-8.67	0-4.12	-------	0-8.68	-------	[4]
Serbia (4)	-------	-------	-------	-------	-------	-------	-------	-------	0.28-0.36	0.01-0.05	[21]
Pakistan (35)	BDL-293	BDL-498	129-9293	4.3-2840	-------	-------	-------	-------	-------	-------	[22]
Austria (9)	3.1-34.1	9.9-94.1	33.5-1398	14.2-175	-------	-------	-------	<0.01-0.98	<0.05-4.3	-------	[23]

Ref.* No.: Reference number; BDL**: Below detection limit; nd: not determined; LOD: Limit of detection

In a unique study, 12,000 samples originating from quality control analyses in various companies dealing with herbs and medicinal plants were used to set Cd, Pb, and Hg references [23].

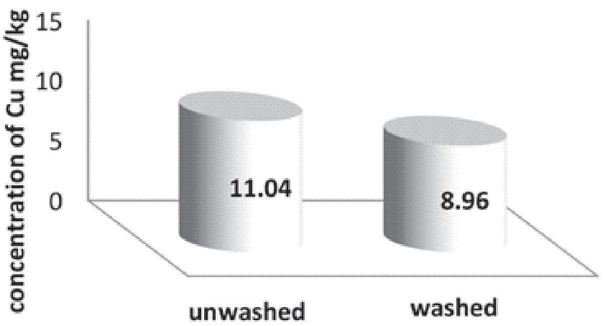

Fig. 1. Comparison of copper contents in 23 washed and unwashed spices consumed in Iraq.

Essential Elements

Iron

Iron is the most important metal and one of the major constituents of the lithosphere with an average content of about 5% of the Earth's crust. It is an essential micronutrient as it is required for adequate erythropoietic function, oxidative metabolism, and cellular immune responses [29]. Out of the 32 spice varieties, 21 contain Fe levels more than 100 mg kg^{-1}; among these spices were basil (1,147 mg kg^{-1}), followed by mint (864.3 mg kg^{-1}) and betony (663.2 mg kg^{-1}). These leafy spices attract special attention as a source of iron – especially for humans with low Fe intake and, consequently, for people with low hemoglobin levels. The Fe levels recorded in this study were close to the range reported in nine out of 11 published articles, but completely different from the other two (Table 7). These discrepancies between published data may be attributed to the influence of geochemical characteristics and

Table 8. Correlation matrix for the element concentrations in spices consumed in Iraq.

	Cu	Zn	Fe	Mn	Cr	Ni	Co	Cd	Pb	Hg
Cu	1.00									
Zn	**0.694**	1.00								
Fe	**0.500**	0.443	1.00							
Mn	-0.110	-0.097	0.186	1.00						
Cr	0.363	0.055	0.469	0.124	1.00					
Ni	0.475	0.313	**0.723**	0.009	0.461	1.00				
Co	0.423	0.252	**0.673**	0.050	**0.586**	**0.658**	1.00			
Cd	-0.155	-0.101	0.052	0.033	0.154	-0.129	0.120	1.00		
Pb	0.140	0.096	0.337	-0.045	0.396	0.246	0.290	0.349	1.00	
Hg	-0.038	0.035	-0.097	-0.096	0.024	-0.056	-0.076	0.159	0.183	1.00

environmental pollution of the soils and the processing steps (such as harvesting, transporting, drying, grinding, packing, fumigating, storing, and analytical procedures) that have the potential to introduce chemical contaminants into the spices [30-32].

Manganese

Manganese is known to be an essential nutrient for bone development as well as in the amino acid, cholesterol, and carbohydrate metabolisms. Mn also plays a critical role in the function of different enzymes, including arginase and Mn superoxide dismutase [33, 34]. On the world scale, the range of Mn average contents of soil [35] varies from 270 to 525 mg kg^{-1}. Referring to Table 5, manganese content of the analyzed spices ranges between 6.42 and 285.8 mg kg^{-1}. It was higher than 100 mg kg^{-1} in cloves (285.8), cardamom (201.1), ginger (186.3), curcumin (181.3), and basil (139.2) mg kg^{-1}. The concentration range of Mn in this work is within the values recently reported in 6 out of 9 spice studies (Table 7). In comparison to other plants, the Mn content was somewhat higher than the range recorded for a large number of vegetable samples [36].

Zinc

Zinc is essential for normal growth and reproduction. In addition, it is vital for the functionality of more than 300 enzymes, for the stabilization of DNA, and for gene

Table 9. Estimated daily intake of heavy metals (mg kg^{-1} DW) by Iraqi adults (19-50 years old and 60 kg body weight) from spices relative to the recommended standards.

M	Average metal content mg kg^{-1}	Quantity of spices consumed g day^{-1}	Estimated daily intake EDI^S mg day^{-1}	RDA or **AI** mg day^{-1} M	F	EAR mg day^{-1} M	F	TUL mg day^{-1} M & F	PTDI mg day^{-1}	% of Recommended Standard
Cu	10.50	18.3	0.192	0.9	0.9	0.7	0.7	10		EDI^S/RDA = 21%
Zn	44.21	18.3	0.809	11	8	9.4	6.8	40		EDI^S/RDA = 7-10%
Fe	267.4	18.3	4.89	8	18	6	8.1	45		EDI^S/RDA = 27-61%
Mn	58.60	18.3	1.07	**2.3**	**1.8**			11		EDI^S/AI = 46-59%
Cr	0.480	18.3	0.009	**0.035**	**0.025**					EDI^S/AI = 26-36%
Ni	1.560	18.3	0.029					1.0		EDI^S/TUL = 3%
Co	0.232	18.3	0.004							
Cd	0.171	18.3	0.003						0.050	EDI^S/PTDI = 6%
Pb	0.968	18.3	0.018						0.215*	EDI^S/PTDI = 8%
Hg	0.400	18.3	0.007						0.014	EDI^s/PTDI = 50%

*This reference health standard was recently withdrawn as they were considered to be no longer appropriate [24]. **M: Metal; RDA:** Recommended dietary allowance; **EAR**: Estimated Average Requirement; **AI:** is the mean intake; **(TUL):** Tolerable upper level; **PTDI**: Provisional tolerable daily intake mg/d.

expression [37]. The general values for the average total Zn contents in soils all over the world range between 60 and 89 mg kg^{-1}. Soluble forms of Zn are readily available to plants, and the uptake of Zn has been reported to be linear with metal concentration in the nutrient solution and in soils [35]. The levels of zinc in the current study vary considerably from 5.45 to 129.3 mg kg^{-1} (Table 5). It was more than 100 mg kg^{-1}only for basil (128.7 mg kg^{-1}) and sesame (109.3 mg kg^{-1}). Compared to other studies, the average zinc contents in this study are close to the range reported in 9 out of 12 articles on spices (Table 7) and to the range (21-92.5 mg kg^{-1}) recorded by Reddy et al. for leafy vegetables [38].

However, the high concentrations of Fe, Mn, and Zn in spices compared to other metals may be attributed to the high availability of the metals (especially Fe) in the earth's crust, and the high mobility of their ions from soils to plants. Oxidizing conditions may greatly reduce the availability of Fe, Mn, and Zn, whereas reducing conditions may lead to the ready availability of these elements even up to the toxic range.

Copper

Copper in humans widely occurs in the form of organic complexes, many of which are metalloproteins. Cu plays a crucial role in cell physiology as a catalytic cofactor in the redox chemistry of enzymes, mitochondrial respiration, iron absorption, free radical scavenging, and elastin cross-linking [39]. Table 5 reveals that spices accumulate moderate levels of Cu (2.58-30.71 mg kg^{-1}) compared to Fe, Mn, and Zn – probably due to the moderate average concentration of Cu (1-16 mg kg^{-1}) in uncontaminated soils [35]. The other important factor that reduces the bioavailability of copper is the strong tendency of copper to bind organic materials to form stable coordination complexes [40]. Excluding the data given by Ansari et al. [22], all the surveyed studies gave copper levels within the range established in this study (Table 7). The same range of copper (1.0-19 mg kg^{-1} DW) was also reported by other workers for different varieties of fruits and vegetable [41].

Chromium

Chromium, nickel, and cobalt were found at comparatively lower concentrations of less than 5.900 mg kg^{-1} for all analyzed spices, reflecting both environmental and biological factors (Table 5). The contents of Cr in plants have recently received much attention due to the knowledge of its importance as an essential element in human metabolic processes and because of its carcinogenic effects. Chromium is known to regulate normal blood glucose levels, nucleic acid, and lipoprotein metabolism, and it also potentiates insulin action [42]. Most soils contain significant amounts of Cr, but its availability to plants is highly limited. The level of Cr obtained in the current study is 0.052-1.317 mg kg^{-1}, which is generally lower than the levels recently recorded in the literature (Table 7).

Nickel

Nickel is believed to play a role in physiological processes as a co-factor in the absorption of iron from the intestine [43]. The levels of Ni in the current study vary considerably from 0.280 to 5.900 mg kg^{-1} (Table 5). Five out of six surveyed studies gave an Ni range close to the present work, whereas the last one gave a higher range (Table 7). In contrast, Chao et al. [44] found little differences of Ni concentrations among leafy vegetables of celery, garlic, cole, and spinach, with the Ni average (2.445 mg kg^{-1}) slightly higher than that found in the current study (1.560 mg kg^{-1}).

Cobalt

The physiological role of cobalt in humans is based on its role in cobalamin (vitamin B12). Cobalamin acts as a cofactor for two enzymes in humans: methylmalonyl-CoA mutase and methionine synthase. Both enzymes are important for the normal formation of red blood cells and for nerve tissue health [45]. The worldwide mean value of Co in surface soils [35] is calculated as 10 mg kg^{-1}. Usually, higher levels of Co are found in heavy loamy soils and, sometimes, in organic soil. Co uptake by plants is a function of content of its mobile fractions and of the Co concentration in solution. Except for the study by Gupta et al. [4], the concentration range of Co in all the surveyed studies (Table 7) are in good agreement with the range (0.020 to 0.754 mg kg^{-1}) obtained in the present study (Table 5). On the other hand, the Co range of spices in this study was slightly higher than those reported by Erdogrul and Erbilir [46] for vegetables (0.0-0.020 mg kg^{-1}).

Toxic Elements

Cadmium

Cadmium is considered one of the most ecotoxic metals that exhibit highly adverse effects on soil biological activity, plant metabolism, and human and animal health. The principal toxic effect of cadmium is its toxicity to the kidney, although it has also been associated with lung damage and skeletal changes in occupationally exposed populations [47]. In uncontaminated soils its contents are highly governed by soil texture and range from 0.01 to 0.3 mg kg^{-1} in sandy soils and from 0.2 to 0.8 mg kg^{-1} in loamy soils [35]. Because Cd is readily available to plants from air and soil sources, its concentration rapidly increases in plants grown in polluted areas [48]. In the current study, the range of cadmium was 0.011-1.389 mg kg^{-1} (Table 6). The highest was in tamarind (1.389) followed by cinnamon (0.874 mg kg^{-1}). The rest of the samples had cadmium content of less than 0.350 mg kg^{-1}. Cadmium lies close to this range in most of the surveyed studies (Table 7). Compared to other foodstuffs, the Cd concentration range in this study was higher than the range (0.014-0.028 mg kg^{-1}) reported by

McBride et al. [49] for 195 vegetables. Overall, leafy green vegetables contained the highest Cd concentrations.

Lead

For centuries, lead toxicity has been one of the most significant causes of neurological morbidity from an environmental toxin. Lead has an effect on brain and intellectual development in young children, while long-term exposure in both children and adults can cause damage to the kidneys plus reproductive and immune systems, in addition to affecting the nervous system [47]. The overall mean value of total Pb for different soils is estimated as 27 mg kg^{-1}. Most of this Pb is unavailable to plant roots, most likely due to the high ability of Pb to form complexes with insoluble humic substances [50]. In the present work, Pb for spices ranges between 0.250 and 4.159 mg kg^{-1} (Table 6). Eight spices have lead content of more than 1 mg kg^{-1} (the highest were cinnamon at 4.159 and betony at 3.743 mg kg^{-1}), whereas the rest of the samples showed lower lead content of less than 1 mg kg^{-1}. These data are in good agreement with six out of eight spice studies recently published in the literature (Table 7), and comparable with Pb contents (0.27-3.8 mg kg^{-1}) recorded for different vegetables grown in uncontaminated areas [49].

Mercury

Mercury and its compounds (organic and inorganic) are highly toxic, the most common being methylmercury. Excessive exposure to mercury is associated with a wide spectrum of adverse health effects, including damage to the central nervous system and the kidney. The average content of Hg in soils of different groups all over the world range between 0.58 and 1.8 mg kg^{-1}, and the worldwide mean contents are estimated as 1.1 mg kg^{-1} [35]. Increasing soil Hg generally causes an increase in the Hg content of plants. In the present work, the concentration range of mercury in spices was (0.032-2.870 mg kg^{-1}). The highest was in coriander (2.870 mg kg^{-1}), followed by saffron crocus (1.445 mg kg^{-1}) and cinnamon (1.055 mg kg^{-1}) (Table 6). These levels are relatively higher than those reported for spices (Table 7) and selected vegetables (0.014-0.0301 mg kg^{-1}) [51].

Washed and Unwashed Spices

Spices may easily be contaminated by heavy metals during different processing steps such as drying on the ground and commercial milling. The t-test results indicated that there exist statistically significant differences at 95% confidence level in mean concentrations of these elements in washed and unwashed spices (Fig. 1). These increases were expected, since unwashed spices can accommodate uncontrolled amounts of contaminants. The possible effect of uncontrolled contamination was also observed in the case of saffron, which has been analyzed directly without washing. The estimated levels of Cd, Pb, and Hg for unwashed saffron in this work (0.216, 2.007, and 1.445 mg kg^{-1}, respectively) were much higher than those reported by Jia et al. [52] for Tibet saffron (0.024, 0.37, 0.032 mg/kg, respectively). These differences may support the assumption that unwashed saffron may be loaded with coloring agents that affect the accuracy of the results.

Elemental Correlations

A linear regression correlation test was carried out to determine inter-elemental correlations between all tested spice samples. Similar investigations have been carried out by others [14, 17]. Table 8 shows high positive correlations (≥0.500) between transition metals Fe-Ni, Cu-Zn, Fe-Co, Ni-Co, Cr-Co, and Cu-Fe. These correlation trends reveal that transition metals may form stable covalent complexes simultaneously with large molecules such as proteins, enzymes, and hormones according to their similar chemical characteristics, including oxidation state [53]. On the other side the toxic metals Cd, Pb, and Hg show weak correlation with other metals, indicating that the presence or absence of one metal does not affect the others.

Estimation of Dietary Exposure to Heavy Metals

The average quantity of spices consumed by ordinary Iraqis was found to be 18.3±5.4 g daily. This figure is lower than the level (25.0 g) reported by Singh and Garg for Indians [16]. Curcuma is a staple additive widely consumed in Iraq. The Dietary Reference Intakes (DRIs) were used to determine nutritional adequacy. The DRIs consist of four reference intakes: recommended daily allowances (RDA), tolerable upper intake level (TUL), estimated average requirement (EAR), and adequate intake (AI). An AI is given when the RDA cannot be set. Table 7 collects (in mg) the estimated daily intake of Iraqi consumers from Cu (0.192), Zn (0.809), Fe (4.89), Mn (1.07), Cr (0.009), Ni (0.029), and Co (0.004); together with the recommended dietary allowance (RDA) [54] for Cu (0.9), Zn (8-11), and Fe (8-18); the mean intake (AI) for Mn (1.8-2.3) and Cr (0.025-0.035); and the tolerable upper level (TUL) for Ni (1.0). None of the Dietary Reference Intakes (DRIs) were determined for Co due to lack of data of adverse effects and concern with regard to lack of ability to handle excess amounts. However, although spices are consumed in small quantities, Table 9 reveals that Cu, Zn, Fe, Mn, Cr, and Ni of these additives contribute significantly (21, 7-10, 27-61, 46-59, 26-36, and 3%, respectively) to the recommended dietary allowance. On the other hand, the average amounts of Cu, Zn, Fe, Mn, and Ni consumed daily by Iraqi adults from spices (Table 9) were still far below the specified tolerable upper level (TUL) of these metals (10, 40, 45, 11, and 1.0 mg kg^{-1}, respectively). This means that any adverse health effects from the consumption of these elements cannot be expected under normal conditions.

Moreover, living organisms have evolved transport mechanisms for the active uptake, enabling cells to regulate their intercellular concentrations. Accordingly, essential elements practically do not produce toxic effects on human and animals due to this homeostatic mechanism. Meanwhile, toxic elements can compete with the essential ones for protein-binding sites and this is the underlying cause of the toxicity effects of many of them.

The average concentrations of toxic elements in spices were Cd (0.171), Pb (0.968), and Hg (0.440) mg kg^{-1} (Table 6). Using the average quantity of spices consumed by ordinary Iraqis (18.3 gram daily), these quantities contribute 3, 18, and 7 µg daily of cadmium, lead, and mercury, respectively (Table 9). The estimated dietary exposures for toxic elements were usually compared with either the provisional tolerable weekly intake (PTWI), or provisional tolerable monthly intake (PTMI). In the present work these references were recalculated in terms of the provisional tolerable daily intake (PTDI).

The FAO/WHO Joint Expert Committee on Food Additives (JECFA) has recommended a provisional maximum tolerable daily intake (PTDI) of Cd and Pb from all sources (food, air and water) [24] of 50 and 215-µg, respectively (assuming an average Iraqi weight of 60 kg). Based on these references, the average daily intake of Cd and Pb by Iraqi consumers from spices alone are 6 and 8% of the PTDI values, respectively, which would not cause any adverse health concerns to consumers.

With respect to Hg, the average quantity of spices consumed by ordinary Iraqis contributes 7 µg daily (Table 9). Both JECFA and the Agency for Toxic Substances and Disease Registry (ASTDR) address mercury specific to its more toxic organic form of methyl mercury. The JECFA provisional maximum tolerable daily intake (PTDI) for a 60-kg adult is 13.7 µg day^{-1}, while the ASTDR minimal risk level (MRL) for chronic oral consumption is 18 µg day^{-1} for the same adult [55-56]. If these two directives are applied to spices, the daily intake of Hg by Iraqi consumers from spices alone (50% of PTDI and 39% of MRL) would not cause any adverse health concerns to consumers.

In summary, Iraqi spices are good sources of essential metals and safe for human consumption. However, if other Pb, Cd, and Hg sources are included and if spices are unwashed and contaminated during processing, the daily intake may exceed the recommended levels. Serious consequences may appear due to the accumulation of toxic elements through years of frequent use of these spices. Periodic surveillance of these spices is therefore advisable in order to be able to guard consumers against health risks associated with these heavy metals, which is the major aim of this work.

Conclusions

The concentrations of trace elements were found to be largely dependent upon the type of spice analyzed and different factors that play an important role in ecosystem functioning. The spices analyzed were rich sources of at least one or more nutritionally essential trace elements (Cu, Zn, Fe, Mn, Cr, Ni, Co). This obviously leads to the conclusion that spices not only add flavor to food, but also enhance its nutritive value by providing several essential elements in bioavailable form. On the other hand, the contribution of the toxic elements Cd, Pb, and Hg to the overall intake from the analyzed spices was very low and could not pose any threat to the consuming population. However, if other Pb, Cd, and Hg sources are included and if spices consumed are unwashed and contaminated during processing, the daily intake may exceed the recommended levels. Keeping a close watch on these contaminants is recommended in order to guide consumers against the imminent health risks associated with these heavy metals.

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