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Determination of Cadmium and Trace Elements in Some Spices Cultivated in Turkey

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In recent years, there has been a growing interest in monitoring trace element and cadmium contents of spices and herbs. Some spices widely cultivated and consumed in Turkey were monitored as to their trace element and heavy metal contents including cadmium, iron, copper, manganese and zinc by ICP-AES. Samples of cumin (*Cuminum cyminum* L.), linseed (*Linum usitatissimum* L.), aniseed (*Pimpinella anisum* L.), fenugreek (*Trigonella foenum-graceum* L.), coriander (*Coriandrum sativum* L.), fennel (*Foeniculum vulgare* Mill.), poppy (*Papaver somniferum* L.) and tarragon (*Artemisia dracanculus* L.) were subjected to chemical analysis. Cadmium was not detectable in any samples of poppy seed. Linseed contained the highest amount of cadmium (128 μg kg^{-1}) in all plant samples while fenugreek contained the least (7 μg kg^{-1}). The level of copper in the samples varied from 6.0 to 17 mg kg^{-1} with the highest value was in tarragon. Content of iron ranged from 29 mg kg^{-1} in poppy seed to 129 mg kg^{-1} in cumin. The tarragon sample occurred as the richest (42 mg kg^{-1}) in manganese content whereby the lowest manganese level was detected in fenugreek (8 mg kg^{-1}). Zinc concentrations of the plant samples ranged between 11 and 28 mg kg^{-1} with linseed containing the highest. The results of the present study revealed that trace metal contents of some selected spices commonly cultivated in Turkey were within the low range.

Key Words: Heavy metals, Micronutrients, Spices, Toxicity.

INTRODUCTION

Trace and heavy metal contamination of soils and its subsequent food chain transfer has long been considered a critical issue due to its potential adverse effect on biota and human health[1-3]. Although heavy metals occur naturally at low concentrations in soils, they are of considerable environmental concern because of their widespread occurrence, cumulative behaviour and toxicity[4].

Several factors contribute the input of heavy metals to agricultural soils including phosphorous fertilizers, manure and atmospheric deposition from town wastes, metal production and fossil combustion[5,6]. Increased trace metal concentration in soil is a severe problem and it becomes a threat to human health because metal ions may enter the food chain, when the soil is contaminated by these metals. Numerous factors affect heavy metals behaviour in soils and their transfer and availability to plants[7,8]. The level of trace metals in plants is conditional, the content being affected by geochemical characteristics of the soil and by the ability of plants to selectively accumulate some of these elements[9,10]. Reactions of plant species greatly differ in response to trace metal toxicity. This variation occurs not only among plant species but also within a given species[11,12].

Some of the trace elements including iron, copper, manganese and zinc are considered essential micronutrients to living beings. However, the benefits of micronutrients may be completely reversed if present at high levels[13,14]. Of the heavy metals, cadmium is perhaps the one that has attracted the most attention due to its relative high mobility in the soil-plant system and potential toxicity to biota at low concentrations[15,16]. Cadmium is not an essential element for human nutrition. On the contrary, higher cadmium intake may be resulted in serious illnesses in lungs, liver and kidneys[17,18].

Worldwide growing concerns about spices and herbs have become well evident in the past few years. Different parts of these plants have been used in various ways such as spice, herbal tea and traditional medicine, *etc.* in particularly local communities in some countries such as China, India, Brazil and the Middle East[19-22]. The place of spices in the diet can not be underestimated from the nutritional point of view. It was stated earlier[23] that spices are good sources of many nutrients, were indispensable in the daily diet of Indians. However, the safety of their use has recently been questioned due to the presence of heavy metals[20,24,25].

As in most countries, the use of spices has recently increased in Turkey. Particularly in the southeastern region of Turkey, the people traditionally consume significant quantities of spices. Although there is some information on the trace element and heavy metal contents of spices and herbal plants grown and consumed in Turkey, the data available so far is not adequate or complete. The objective of the present study was to determine the concentrations of selected trace elements and cadmium in some spices widely cultivated and traditionally consumed in Turkey. Although there is some information on the trace heavy metal contents of spices and herbal plants grown and consumed in Turkey, the data available so far is not adequate or complete. The objective of the present study was to determine the concentrations of selected trace heavy metals in some spices widely cultivated and traditionally consumed in Turkey.

EXPERIMENTAL

Plant materials from eight species were obtained from spice wholesalers and local spice shops in southeastern Turkey (Adana, Mersin, Kilis, Gaziantep and Hatay provinces), one of the most important trade centers for medicinal and spice plants[22], in October 2005. For chemical analysis, 25 different samples of 100 g from each eight plant species (a total of 200 plant samples) were derived from five local spice shops and wholesalers in each province. The samples in the plastic bags were kept at room temperature until to be analyzed. Some characteristics of eight plant species subjected to this study are given in Table-1.

TABLE-1
SOME CHARACTERISTICS OF EIGHT SELECTED SPICES

Plant scientific names	Common name	Turkish name	Parts used
Artemisia dracanculus L.	Tarragon	Tarhin	Leaf
Coriandrum sativum L.	Coriander	Kisnis	Fruit
Cuminum cyminum L.	Cumin	Kimyon	Fruit
Foeniculum vulgare Mill.	Fennel	Rezene	Fruit
Linum usitatissimum L.	Lineseed	Keten	Seed
Papaver somniferum L.	Poppy	Hashas	Seed
Pimpinella anisum L.	Aniseed	Anason	Fruit
Trigonella foenum-graceum L.	Fenugreek	Çemen	Seed

All the plant samples were cleaned and washed with deionized water and air dried. Afterwards, the samples were dried at 70 °C for 48 h in an oven and ground for chemical analysis. The ground materials were stored in polyethylene bottles in room temperature. For chemical analysis, 0.2 g ground samples were put into burning cup and 5 mL HNO_3 65 % (Merck, Darmastadt, Germany) and 2 mL H_2O_2 30 %, (Merck, Darmastadt, Germany) were added. The samples were incinerated in a HP-500 CEM MARS 5 microwave (crop. Mathews NC, USA) at 200 °C and cooled at room temperature for 45 min. The extracts were passed through a Whatmann filter paper no. 42 and the filtrates were collected by high-deionized water in a 20 mL polyethylene bottles and kept at 4°C in laboratory for ICP-AES analysis. Each sample was analyzed in triplicate.

Distilled-deionized water was used for all analytical works. All the polyethylene bottles and glassware were carefully leached with 2-4 % HCl, then, they were passed through deionized water for 3 times. As analytical reagent grade chemicals, Merck standards (R1 and R2 groups) were used. Standard solutions of Cd, Cu, Fe, Mn and Zn were prepared in 1 % HNO_3 immediately before the analysis by serial dilution of 1000 mg L^{-1} stock solution stored in polyethylene bottles. Peach Leaves (Standard Reference Material, 1547) and Corn Bran (Standard Reference Material, 8433) were used as reference materials[26].

Scanning ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometer; JY-138 Ultrace) instrument with high-resolution nitrogen purged with 1 m monochromator was used in the present study. This system was equipped with a 40.68 MHz generator with a demountable standard torch, including a sheath gas flow and a 1 m focal length of monochromator. A Meinhard C type nebulizer with a Scott type spray chamber was used for sample nebulization. The ICP-AES system was operated at 1.1 kW forward powers with a coolant flow of 12 L min^{-1} and a sample uptake rate of 3 mL min^{-1} with a peristaltic pump. The sheath gas flow wasn't used throughout this work. The ICP-AES was used to determine Cu, Fe, Mn and Zn in the extracts. The wave lengths and detection limits of the method were 214.438 nm/0.025 mg kg^{-1}, 324.754 nm/0.054 mg kg^{-1}, 259.940 nm/0.062 mg kg^{-1}, 257.610 nm/0.014 mg kg^{-1} and 213.856/0.018 mg kg^{-1} for Cd, Cu, Fe, Mn and Zn, respectively. For determining Cd concentration in the extracts, ICAP-OES (Inductively Coupled Argon Plasma-Optical Emission Spectrometer) U-5000AT+ Ultrasonic Nebulizer (214,438 nm/0.1 μg kg^{-1}) was also used.

RESULTS AND DISCUSSION

The mean values of cadmium, copper, iron, manganese and zinc concentrations in spices studied are presented in Table-2. The values of the trace metals, based on plants' dry weight, are the means of three replicates and given as mean ± SD. Analytical recovery of the method was checked by a parallel analysis of the two certified reference materials and acceptable recoveries (> 95 %) were obtained. The correlation coefficients of the calibration curves were in the range of 0.995-0.999.

Cadmium content of the selected spices was noticeably variable. Cadmium was not detectable in any samples of poppy seed (*Papaver somniferum* L.). The results revealed that linseed (*Linum usitatissimum* L.) contained the highest amount of cadmium (128 μg kg^{-1}) in all plant samples while fenugreek (*Trigonella foenum-graceum* L.) contained the least (7 μg kg^{-1}). Other plant samples relatively richer in cadmium concentration were cumin (*Cuminum cyminum* L.), aniseed (*Pimpinella anisum* L.) and fennel (*Foeniculum vulgare* Mill.).

Concentrations of copper in eight spices were the ranges of 6.0-17 mg kg^{-1}, the lowest in coriander (*Coriandrum sativum* L.) and the highest in tarragon (*Artemisia dracanculus* L.) samples. The content of iron in the samples varied between 129 mg kg^{-1} in cumin (*Cuminum cyminum* L.) and 29 mg kg^{-1} in poppy (*Papaver somniferum* L.).

TABLE-2
CADMIUM AND TRACE ELEMENT CONCENTRATIONS OF EIGHT SELECTED SPICES (n = 25 ± SD, DRY WEIGHT)

Plant sample	Cd	Cu	Fe	Mn	Zn
	(µg kg^{-1})	(mg kg^{-1})			
A. dracanculus L.	29 ± 2.9	17 ± 0.6	69 ± 1.6	42 ± 2.3	24 ± 2.1
C. sativum L.	16 ± 2.3	6 ± 0.8	44 ± 1.5	12 ± 4.3	12 ± 0.2
C. cyminum L.	77 ± 1.3	8 ± 0.3	129 ± 2.1	14 ± 0.8	22 ± 0.5
F. vulgare Mill.	42 ± 3.8	7 ± 0.5	63 ± 2.7	19 ± 1.4	11 ± 0.8
L. usitatissimum L.	128 ± 3.7	10 ± 0.9	38 ± 2.2	13 ± 0.9	28 ± 0.6
P. somniferum L.	<LOD	11 ± 0.4	29 ± 1.4	24 ± 1.7	25 ± 0.4
P. anisum L.	69 ± 1.0	8 ± 0.2	87 ± 1.8	18 ± 0.9	25 ± 0.4
T. foenum graceum	7 ± 1.6	9 ± 0.6	36 ± 3.6	8 ± 1.0	19 ± 0.9
Peach leaves (NIST-SRM 1547)	26	3.7	218	98	17.9
Recovery values (%)	99	95	96	97	98
Corn Bran (NIST-RM 8433)	12	2.47	14.8	2.55	18.6
Recovery values (%)	95	98	97	99	96

LOD: Limit of detection

In the case of manganese, the tarragon (*Artemisia dracanculus* L.) samples occurred as the richest (42 mg kg^{-1}) whereby the lowest level (8 mg kg^{-1}) was recorded in fenugreek (*Trigonella foenum-graceum* L.). Similarly to copper, variability in zinc content was in lower ranges. Zinc concentrations of the plant samples varied between 11 and 28 mg kg^{-1} with linseed (*Linum usitatissimum* L.) sample containing the highest and fennel (*Foeniculum vulgare* Mill.) sample having the lowest, respectively.

Cadmium is naturally present in soils at trace amounts. However, it is of special concern because of its potential toxicity to biota at low concentrations[1,8]. Cadmium in food chain is mostly derived from various sources of environmental contaminations. World Health Organization[27] declared that tolerable amount of cadmium in medicinal plants is 300 µg kg^{-1}.

The levels of cadmium compared well with the levels in spices form other parts of the world. The highest cadmium concentration found in the present work was in linseed (128 µg kg^{-1}) followed by cumin (77 µg kg^{-1}) and aniseed (69 µg kg^{-1}), which were lower than those reported in other parts of the world. Cadmium concentrations of medicinal plants studied in Italy and Egypt were in ranges of 10-750 and 50-300 μg kg^{-1}, respectively[19,28]. Levels of cadmium in medicinal herbs in Brazil[20] ranged from < 0.2 to 0.74 μg g^{-1}. Basgel and Erdemoglu[29] found cadmium content at higher levels (1.2-440 μg kg^{-1}) in some herbal teas consumed in Turkey. The cadmium range of some spices and herbal plants from western Anatolia was reported as 0.1-2.8 μg g^{-1} by Divrikli *et al.*[18]. The values of cadmium obtained in the present study were also much lower than the limit of 300 μg kg^{-1} recommended for herbs and spice plants by World Health Organization[27].

Although they are required in very low quantities, certain trace heavy elements including iron, copper, zinc and manganese are the essential micronutrients with one or more structural or functional roles for living organisms[18,30]. Of all the micronutrients, iron is required by plants in the largest amounts. Copper, manganese and zinc play important roles in many enzyme systems[31,32]. However, in excess amounts, these elements may also become toxic[14,21,23]. The recommended dietary allowances (RDA) for iron, copper, manganese and zinc are 18, 2, 5 and 15 mg d^{-1} $person^{-1}$, respectively[33].

Comparing with the previous investigations on the trace metals Cu, Fe, Mn and Zn in herb and spice plants from other parts of the world, reported levels for these elements in the present study were generally in lower ranges. The levels of Cu, Fe, Mn and Zn were also lower than the levels given above by Food and Nutrition Board of the National Academy of Sciences-United States[33].

Pradeep *et al.*[23] reported concentration ranges of iron, manganese and zinc for coriander from India as 25.09, 3.09 and 3.96 mg 100 g^{-1}, respectively. Chizzola *et al.*[24] stated that contents of micronutrient such as Cu, Fe, Mn and Zn were generally within the usual range in herbs, spices and medicinal plants from Austria. The ranges for iron, copper, manganese and zinc in coriander, cumin, fenugreek and anise have been reported by Gupta *et al.*[14] as 0.11-0.35 g kg^{-1}, 7.72-13.23 mg kg^{-1}, 26.39-82.99 mg kg^{-1} and 27.48-50.20 mg kg^{-1}, respectively. Basgel and Erdemoglu[29] reported that the average levels of Fe, Cu, Mn and Zn were in the ranges of 228-810, 3.92-35.8, 23-244 and 21.9-47.2 mg kg^{-1}, respectively, in major herbs consumed as tea for medicinal purposes in Turkey. In spices and herbal plants from western Anatolia, Turkey[18], the concentrations of copper, iron, manganese and zinc were found in the ranges of 3.8-35.4, 30.0-945.3, 7.9-152.5 and 5.2-83.7 μg g^{-1}, respectively.

Significant differences might be expected in cadmium and trace element contents of herbs and spices due to the great heterogeneity in the species studied, used plant parts and growing regions. A number of studies have indicated that the plants collected from rural areas or grown in less industrialized regions had lower contents of heavy metals than those growing in industrialized regions[10,19,28,34]. As in the case of this study, since spices are produced on a rather small scale or plant materials are collected form nature in less industrialized areas of Turkey[35], lower levels of heavy metal contamination may be expected.

Conclusion

The present study revealed that, contents of heavy metal and trace elements of some selected spices commonly cultivated and consumed in Turkey, particularly for cadmium, were far below the recommended critical levels. Therefore, it was concluded that these levels may not constitute a metal toxicity concern over safety of spices consumption. From the nutritional point of view, these findings would also be worthwhile in determining the value and contribution of spices as good sources of many nutrients in the diets of the people consuming large amounts of spices.

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