Heavy Metals in Egyptian Spices and Medicinal Plants and the Effect of Processing on Their Levels

A. A. K. Abou-Arab* and M. A. Abou Donia

Food and Dairy Technology Department, National Research Center, Dokki, Cairo, Egypt

To determine the contamination of Egyptian spices and medicinal plants with heavy metals, a total of 303 samples, which represent 20 different types of spices and medicinal plants that were collected from areas of exportation in Egypt, were analyzed for heavy metals. Some of them have different growing seasons, and each has its own agricultural practices and several shipments. The results revealed that heavy metal contents in spice and medicinal plants depend on the plant species. The maximum levels of heavy metals in the analyzed samples were 14.4, 2.44, 33.75, 2.85, 0.10, 68.80, 343.0, 11.40, and 1046.25 μg/g for Pb, Cd, Cr, Ni, Sn, Zn, Mn, Cu, and Fe, respectively. Cobalt was not detected in any of the various samples under investigation. The levels of heavy metals determined in the analyzed samples were found to exceed the maximum allowable levels of Zentrale Erfassungs und Bewertungsstelle für Umweltchemikalien. The investigated medicinal plants were also processed by two different methods to determine the behavior of their metal contents during processing. It has been found that boiling the plant in water leads to the extraction of higher amounts of the metal from the plant than immersing it in the hot water. The achieved results were tabulated.

Keywords: *Egyptian; spices; medicinal; plants; processing; levels*

INTRODUCTION

During the past decades, spice and medicinal plants gained a more important role in agronomy production, pharmacy, and exportation because of their increased use as a raw material for the pharmaceutical industry and pharmaceutical preparations and in the everyday life of the general population. In recent years the cultivation of medicinal and aromatic plants has been achieved with increasing interest in Egypt. The interest in our country for these plants is much greater because of the possibility of exportation. From plant nutrition studies, it is known that plants require a certain amount of trace elements, that they respond differently to an enhanced or lowered trace element supply, and that, in some cases, agricultural products may be contaminated with toxic heavy metals (Krug, 1986).

There are two major reasons (De Smet, 1992) to monitor levels of toxic metals in medicinal plants. The first reason, contamination of the general environment with toxic metals, has increased (Ali, 1983). The sources of this environmental pollution are quite varied, ranging from industrial and traffic emissions to the use of purification mud and agricultural expedients, such as cadmium-containing dung, organic mercury fungicides, and the insecticide lead arsenate (Schilcher, 1983; Gosselin et al., 1984; Schilcher et al., 1987). The second reason, exotic herbal remedies, particularly those of Asian origin, have been repeatedly reported to contain toxic levels of heavy metals and/or arsenic.

Several investigators have performed several studies on the residual levels of toxic metals in medicinal herbs (Schilcher, 1982; Ali, 1983, 1987; Peters and Schilcher, 1986; Schilcher et al., 1987). Most studies on residual levels of toxic metals in medicinal herbs have focused on lead, cadmium, and mercury (Schilcher, 1985; Ali, 1987; Schilcher et al., 1987).

In the experience of Peters and Schilcher (1986) and Schilcher et al. (1987), medicinal plants occurring wild show more anomalous values than cultivated herbs, in particular with respect to lead levels. The reason is, of course, that drugs grown wild are more difficult to control for all the potential ways of environmental pollution. As was to be expected, the research group also demonstrated that the levels of lead and cadmium in the same crude herb may vary considerably with plant part and habital.

The aim of this study is to investigate the heavy metal contents in spices and medicinal plants from shipments for exportation from Egypt to ensure the quality and also to detect the effect of extracting the investigated plants in hot water on the transfer of their metal contents into the used water.

MATERIALS AND METHODS

Materials. *Sample Collection.* A total of 303 samples, which represent 20 different types of spices and medicinal plants that have different growing seasons and each with its own agricultural practices, were collected from different sources of exportation in Egypt. Eleven types of them belong to the leafy group, 6 to the fruity group, and 3 to the flowery group. The collected samples were kept in plastic bags until their analysis. The various collected samples (item, scientific name, number) are presented in Table (1).

Standards. Standard solutions of heavy metals, that is, lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni), cobalt (Co), tin (Sn), zinc (Zn), manganese (Mn), copper (Cu), and iron (Fe), were provided by Merck (Darmstadt, Germany). The standards were prepared from the individual 1000 mg/L standard (Merck), 100 mL of nine combined standards were prepared in 0.1 N HNO_3. Working standards were prepared from the previous stock solutions.

Methods. *Preparation of the Plant Extract.* Medicinal plants, that is, spearmint, caraway, anise, chamomile, karkade, and tea, were extracted in hot water by two different

10.1021/jf990508p CCC: $19.00
Published on Web 05/06/2000

Table 1. Spices and Medicinal Plant Samples Collected from Several Selected Shipments in Egypt

item	no. of collected samples[a]	scientific name
leafy		
geranium	20	*Pelargonium graveolens* L.
basil	20	*Ocimum basilicum* L.
marjoram	20	*Marjorana hortensis* L.
peppermint	20	*Mentha piperita* L.
spearmint	20	*Mentha viridis*
Jew's mallow	10	*Corchorus olitorius*
dill	10	*Anethum graveolens* L.
celery	10	*Apium graveolens* Mill.
parsley	10	*Petroselinum sativum* Hoffm.
cumin	10	*Cuminum cyminum* L.
tea	6	*Thea sinensis* Linn.
fruity		
caraway	15	*Carum carvi* L.
anise	15	*Pimpinella anisum* L.
fennel	15	*Foeniculum vulgare* L.
coriander	15	*Coriander sativum* L.
dill	5	*Anethum graveolens* L.
black pepper	5	*Piper nigrum*
flowery		
chamomile	66	*Matrcaria chamomila* L.
karkade	6	*Rosella jamica*
saffron	5	*Crocus sativua* Linn.

[a] Each sample consists of three subsamples that were collected from at least three different boxes/places throughout the lot. Each subsample consists of 3−4 kg.

methods as commonly used in homes. In the first method, 2 g of the dry plant was left to boil in 100 mL of deionized water for 5 min in a glass beaker. In the second method, 2 g of the dry sample was immersed in 100 mL of hot deionized water for 5 min (tea method). The liquid extracts in the two previous procedures were filtered separately and were prepared for analysis.

Determination of Heavy Metal Contents in the Plant. A dry ashing method (Gorsuch, 1970; Giron, 1973) was used for the destruction of organic matter to determine all of the heavy metals. The weights of the samples used were between 0.5 and 1.0 g. The crucibles were partially covered and then were left to dry in an oven at 100−105 °C overnight (∼16 h). Magnesium nitrate, as an ashing aid, was added to the dried sample at an approximately 1:1 ratio. The samples were charred on a hot plate (∼2 h) and then ashed in a muffle furnace at 470 ± 5 °C for 16 h. After ashing, the ash was extracted with 20% (v/v) redistilled nitric acid and then completed to a definite volume with 1% nitric acid (v/v) for analysis. A Perkin-Elmer model 2380 atomic absorption spectrophotometer with flame atomization (air−acetylene) 60−20, equipped with a 10 cm burner and a deuterium lamp for background correction, was used for the flame atomic absorption measurement of the metals. Maximum absorbance was obtained by adjusting the cathode lamps at specific slit width and definite wavelengths as recommended by the method as follows: 0.7, 228.8 nm (Cd); 0.7, 357.9 nm (Cr); 0.2, 240.7 nm (Co); 0.7, 324.8 nm (Cu); 0.2, 248.3 nm (Fe); 0.2, 279.5 nm (Mn); 0.2, 232.0 nm (Ni); 0.7, 283.3 nm (Pb); 0.7, 286.3 nm (Sn); and 0.7, 213.9 nm (Zn), respectively. The recovery of this method was carried out as follows:

Portions of dry plant material were ground and weighed (0.5−1.0 g) and were spiked with 0.5 mL of the nine combined standards to get a concentration for each metal of 0.1, 1, and 10 μg/g of dry plant. The spiked samples were placed in a dehydrator at 80 °C until dry. These samples were analyzed according to the method of Gorsuch (1970) and Giron (1973), a dry ashing method, to get the recovery of this method. The recovery of the method ranged between 95 and 97% for all of the studied heavy metals. All of the obtained results were corrected according to the recovery percent.

The detection limits of the method, as expressed in micrograms per gram of dry plant, for the studied metals were 0.05 for Pb, Cd, and Cr, 0.1 for Ni, and 0.01 for Sn, Zn, Mn, Cu, and Fe.

Determination of Heavy Metal Contents in the Liquid Extract. The obtained filtrates were acidified by 1% nitric acid (v/v) for analysis by atomic absorption spectrophotometer at the same conditions that were mentioned before (AOAC, 1995).

RESULTS AND DISCUSSION

Levels of Heavy Metals in Some Egyptian Spices and Medicinal Plants. Heavy metal contents in spices and medicinal plants depend on climatic factors, plant species, air pollution, and other environmental factors (Sovljanski et al., 1989). The levels of heavy metal contents in the analyzed samples of spices and medicinal plants are listed in Tables 2−4.

Data in Table 2 demonstrate the contents of heavy metal in the analyzed samples of spices and medicinal plants (leafy group). The highest mean levels of Sn and Mn were detected in tea samples, which were recorded as 0.1 and 343 μg/g, respectively. However, marjoram contained the highest mean levels of Pb (14.4 μg/g) and Cr (25.25 μg/g). On the other hand, the highest mean levels of Ni (2.85 μg/g) and Zn (35.5 μg/g) were detected in basil samples. The results proved also that celery, parsley, and spearmint contained the highest mean levels of Cd (2.44 μg/g), Cu (11 μg/g), and Fe (1046 μg/g) in this order. It could be observed from the data that the lowest mean levels of Pb, Cd, Ni, Sn, and Mn were detected in Jew's mallow, which recorded as 1.14, 1.06, 0.61, 0.01, and 22.4 μg/g, respectively. Also, the lowest mean levels of Zn (8 μg/g), Cu (1.8 μg/g), and Fe (145 μg/g) were detected in tea. The results revealed that celery contained the lowest mean levels of Cr (2.47 μg/g). Concerning geranium, peppermint, dill, and cumin samples, data indicated that the mean levels of the metals were moderate. Cobalt was not detected in the different leafy samples under investigation.

With regard to fruity group samples in Table 3, data indicated that Pb, Ni, and Sn were detected at the highest mean levels, that is, 6.4, 1.3, and 0.09 μg/g, respectively, in caraway samples. In contrast, the lowest mean level of Pb was found in black pepper (1.1 μg/g), and the lowest mean levels of Ni and Sn were reported in coriander, which were recorded as 0.71 and 0.05 μg/g, in the same order. On the other hand, the highest mean levels of Cd (2.4 μg/g) and Cu (11.4 μg/g) were found in anise. In contrast, the lowest of both (Cd and Cu) were detected in black pepper (1.16 μg/g) and coriander (2.78 μg/g). The results indicated also that coriander, fennel, black pepper, and dill recorded the highest mean levels of Cr (33.75 μg/g), Zn (68.8 μg/g), Mn (118.5 μg/g), and Fe (289 μg/g), respectively. However, the lowest mean levels of Cr and Fe were detected in black pepper, which were recorded as 11.19 μg/g and 26.96 μg/g, respectively. Data in Table 3 reveal also that the lowest mean levels of Zn (26.16 μg/g) and Mn (23 μg/g) were detected in caraway samples. It could be observed from the data that Co was not detectable in all of the analyzed samples. In addition, Ni and Sn were not detectable in anise, fennel, dill, and black pepper.

Data in Table 4 summarize the contents of heavy metals in flowery group samples. The obtained results indicated that chamomile samples recorded the highest mean levels of Pb (6.19 μg/g), Cr (21.6 μg/g), Ni (2.78 μg/g), Sn (0.08 μg/g), Zn (24.7 μg/g), and Fe (825 μg/g). However, karkade contained the highest mean levels of Cd (1.34 μg/g) and Mn (289 μg/g). The highest mean

Table 2. Distribution Levels (Micrograms per Gram ± SD) of Heavy Metal Contents[a] in Egyptian Spices and Medicinal Plant Samples (Leafy Group)

metal	geranium, 20	basil, 20	marjoram, 20	peppermint, 20	spearmint, 20	Jew's mallow, 10	dill, 10	celery, 10	parsley, 10	cumin, 10	tea, 6
Pb	11.4 ± 4.6	4.45 ± 1.48	14.4 ± 4.5	5.1 ± 1.9	5.8 ± 1.6	1.14 ± 0.88	1.49 ± 0.94	1.13 ± 0.22	2.59 ± 0.98	1.7 ± 0.6	3.45 ± 1.9
Cd	1.06 ± 0.24	1.26 ± 0.64	2.05 ± 1.09	1.35 ± 0.66	1.9 ± 0.9	1.06 ± 0.41	1.90 ± 0.64	2.44 ± 0.94	1.95 ± 0.48	1.68 ± 0.2	*b*
Cr	16.2 ± 3.8	8.75 ± 2.44	25.25 ± 6.49	12.88 ± 4.19	19.75 ± 3.66	3 ± 0.99	9.25 ± 1.49	2.47 ± 0.96	9 ± 1	21.88 ± 3.2	9.75 ± 0.9
Ni	1.1 ± 0.6	2.85 ± 1.85	2.15 ± 0.99	0.96 ± 0.14	1.45 ± 0.49	0.61 ± 0.26	1.59 ± 0.69	0.64 ± 0.22	1.93 ± 0.64	0.74 ± 0.20	1.9 ± 0.3
Sn	0.03 ± 0.01	0.02 ± 0.006	0.04 ± 0.01	0.05 ± 0.02	0.06 ± 0.02	0.01 ± 0.006	0.06 ± 0.02	0.02 ± 0.01	0.03 ± 0.01	0.07 ± 0.02	0.1 ± 0.06
Zn	12.1 ± 6.1	35.5 ± 4.1	10.59 ± 2.49	11.14 ± 3.19	18.9 ± 3.4	11.6 ± 1.4	11.55 ± 2.49	23 ± 6.4	13.40 ± 1.98	28.9 ± 4	8 ± 1
Mn	26.4 ± 8.1	57.9 ± 9.5	28 ± 2.5	78.9 ± 6.9	88.9 ± 14.8	22.4 ± 4.1	77 ± 11	28.8 ± 5	26.75 ± 2.96	26.6 ± 2.9	343 ± 20
Cu	4.8 ± 2.1	6.8 ± 1.5	3.95 ± 1.94	2.11 ± 0.99	9.8 ± 1.4	10.9 ± 2.4	2.1 ± 0.8	8.5 ± 1.5	11 ± 3.4	2.16 ± 0.96	1.8 ± 0.6
Fe	516 ± 31	671 ± 20	671 ± 24	620 ± 20.9	1046 ± 33	512 ± 11	848 ± 21	296.6 ± 20	764 ± 21	301 ± 21	145 ± 33

[a] Co was not detected in any of the analyzed samples. [b] Not detectable.

Table 3. Distribution Levels (Mean ± SD) of Heavy Metal Contents[a] in Egyptian Spices and Medicinal Plant Samples (Fruity Group)

metal	caraway, 15	anise, 15	fennel, 15	coriander, 15	dill, 5	black pepper, 5
Pb	6.4 ± 1.1	1.6 ± 0.4	1.8 ± 0.4	4.33 ± 1.25	1.64 ± 0.55	1.1 ± 0.6
Cd	1.36 ± 0.89	2.4 ± 0.6	2.2 ± 1.96	1.69 ± 0.32	1.7 ± 0.7	1.16 ± 0.55
Cr	20.5 ± 11.9	21 ± 1.4	20.5 ± 4.2	33.75 ± 8.68	21.96 ± 4.74	11.19 ± 1.16
Ni	1.3 ± 0.09	*b*	*b*	0.71 ± 0.13	*b*	*b*
Sn	0.09 ± 0.06	*b*	*b*	0.05 ± 0.02	*b*	*b*
Zn	26.16 ± 2.44	62.46 ± 6.2	68.8 ± 4.46	10.69 ± 3.53	63.49 ± 11.96	35 ± 3.2
Mn	23 ± 4.1	36 ± 4.5	31.8 ± 3.68	9.88 ± 2.68	23.3 ± 4.6	118.5 ± 6.35
Cu	8.45 ± 6.1	11.4 ± 1.1	11.1 ± 6.3	2.78 ± 0.78	10.66 ± 1.23	8.4 ± 2.18
Fe	190 ± 8.4	101.6 ± 11.36	114 ± 8	161 ± 23	289 ± 21	26.96 ± 4.14

[a] Co was not detected in any of the analyzed samples. [b] Not detectable.

Table 4. Distribution Levels (Mean ± SD) of Heavy Metal Contents[a] in Egyptian Spices and Medicinal Plant Samples (Flowery Group)

metal	chamomile, 66	karkade, 6	saffron, 5
Pb	6.19 ± 1.88	0.5 ± 0.1	1.1 ± 0.5
Cd	1.3 ± 0.6	1.34 ± 0.3	1.2 ± 0.6
Cr	21.6 ± 8.1	17.38 ± 1.88	17 ± 3.9
Ni	2.78 ± 0.88	1.5 ± 0.9	*b*
Sn	0.08 ± 0.02	0.06 ± 0.02	*b*
Zn	24.7 ± 6.8	9.25 ± 1.33	24.40 ± 2.98
Mn	53 ± 14	289± 30.3	43.6 ± 4.4
Cu	8.88 ± 1.38	*b*	11.3 ± 2.4
Fe	825 ± 14	203 ± 29	247 ± 7

[a] Co was not detected in any of the analyzed samples. [b] Not detectable.

level of Cu (11.3 μg/g) was detected in saffron. Data proved also that karkade samples contained the lowest mean levels of Pb (0.5 μg/g), Ni (1.5 μg/g), Sn (0.06 μg/g), Zn (9.25 μg/g), and Fe (203 μg/g). As can be seen from the presented data, saffron samples contained the lowest mean levels of Cd, Cr, and Mn, which recorded as 1.2, 17, and 43.6 μg/g, respectively. However, the lowest mean level of Cu (8.88 μg/g) was detected in chamomile samples.

From the monitoring data revealed that the heavy metal contents in the samples under investigation were recorded at different levels. If we compare the obtained results with the data of Kim et al. (1994), who investigated the contents of heavy metals in 291 samples of medicinal plants, grown in unpolluted sites, we can note the evident increase of all heavy metals under investigation. They reported that the contents of Cd, Cu, Pb, Zn, Cr, Ni, and As in the plants were 0.39, 6.64, 0.82, 27.78, 1.45, 0.73, and 0.28 μg/g, respectively. They reported also that the average contents of Cd, Cu, Pb, Zn, Cr, Ni, and As in the soil were 0.20, 3.40, 2.24, 11.93, 0.97, 3.99, and 0.59 μg/g, respectively. On the other hand, Gravel et al. (1994)and Janusz et al. (1995) determined the contents of heavy metals in an industrialized region and reported that the plants growing in there had higher contents of heavy metals than plants growing in a less industrialized region.

The current results show that the levels of various metals were higher than those given by Wong et al. (1986, 1993) and Listow and Petrow (1990). The first authors found that few medicinal plant samples contained relatively higher concentrations of the metals Cd and Pb. On the other hand, the results obtained by the second authors showed that 68% of crude drug samples contained <0.5 ppm of Cd and 65% had <0.5 ppm of Pb.

A similar investigation was conducted for plants obtained from Saudi Arabia, Bahrain, and Iraq as Arabian countries. In Saudi Arabia, the levels of Pb and Cd in parsley samples collected from nonpolluted areas were 0.75−2.1 and 30−125 μg/g, respectively (Al-Kathiri and Al-Attar, 1997). These authors reported also that Pb and Cd were detected at levels 8.6−14.6 and 625 μg/g, respectively, in polluted areas. In Bahrain, Al-Saleh and Chudasama (1994) reported that a large portion of the plants examined contained high concentrations of toxic metals and some of them exceeded the limits of toxicity; the reported data indicate a potential health hazard. However, in Iraq, Jawad et al. (1986) observed amounts of 243−850 ppm of Fe in spices and 228−651 ppm of Fe in medicinal plants.

Accumulation of Pb in different parts of plants growing around six Egyptian roads was determined in early studies by Nasralla and Ali (1985). They reported that leafy plants contained higher levels of Pb than fruit samples. They added that the extent of contamination with Pb depended on traffic densities and distance from the road. As definite permissible limits could not be found in the European Pharmacopoeia or the German Pharmacopoeia, the researchers took limit values of the so-called Zentrale Erfassungs und Bewertungsstelle für Umweltchemikalien (ZEBS) as a point of reference. The national ZEBS regulation offers maximum allowable values for herb-like food products, such as grains and vegetables (Table 5). It should be noted that the limits

Table 5. Maximum Allowed Levels of Heavy Metals in Foodstuffs, According to the ZEBS Regulation[a]

		maximum allowed level per type of foodstuff (mg/kg)				
metal	date of ZEBS	grain	rye/ rice	fruit/root vegetables	leafy vegetables	sprout vegetables
lead	1979	0.5		0.5	1.2	1.2
	1986		0.4	0.25	0.8	0.5
cadmium	1979	0.1		0.1	0.1	0.1
	1986		0.1	0.1	0.1	0.1
mercury	1979	0.03				
	1986		0.03	0.05	0.05	0.05

[a] Schilcher et al., 1987.

usually refer to the fresh weight of foodstuffs destined for consumption, whereas medicinal herbs are generally tested in their dried form (Schilcher, 1985). Comparing the obtained results given in this investigation with the ZEBS limits, spices and medicinal herb samples were found to exceed the limits. Failure to meet these limits does not imply, however, that a real health risk is involved. Comparison of test results with the ZEBS values should merely be considered as a tool of quality assurance for the timely detection of potentially undesirable contamination. In the experience of Schilcher and associates (Schilcher et al., 1987; Peters and Schilcher, 1986), herbal drugs occurring wild show more anomalous values than cultivated herbs, in particular with respect to lead levels. The reason is, of course, that drugs grown wild are more difficult to control for all the potential ways of environmental pollution.

It can be concluded that some types of metal such as Cu, Mn, and Zn are the natural essential components of coenzymes and they are important for growth, photosynthesis, and respiration. Other metals such as Pb and Cd had no biochemical or physiological importance, so they are considered as very toxic pollutants (Sovljanski et al., 1989). The relatively high concentrations of Pb and Cd found in different samples are certainly due to irrigation with contaminated water as well as the addition of some fertilizers and herbicides. In addition, it was reported that the contamination of plants with Pb depends on several factors, for example, traffic densities and distance from the road (Bosque et al., 1990). These authors also suggested that Pb accumulated in plants through both foliage and root systems, but Pb absorption through foliage is more pronounced at locations close to the emission source of Pb vapor and fine particles. On the other hand, heavy metal contents of medicinal plants depend on the plant species and climatic factors (Sovljanski et al., 1990). Yoo-Seung and Song-Kyung-Sik (1991) mentioned that there was no significant correlation between heavy metal contents of soils and cultivated medicinal plants.

Behavior of Heavy Metals in the Medicinal Plants during Processing. With respect to the effect of the plant processing on its metal contents, Table 6 shows that tin was not detected in the two different extracts in all samples. Also, we find that the investigated metals were transferred from the plant tissue into the used water at different ratios, which depend on the metal, the plant, and the applied extraction method. The highest concentrations (expressed as a percent of the initial concentration before processing) of Ni, Pb, Cd, Cr, Fe, Zn, Mn, and Cu transferred from the plant tissues to the water by the first method (in boiling water) were 72% (chamomile), 76% (chamomile), 78% (spearmint), 80% (spearmint), 80% (karkade), 82% (spearmint), 84% (anise), and 87% (caraway), respectively. In the second method (in hot water), the highest concentrations of Ni, Cd, Pb, Cu, Cr, Zn, Mn, and Fe (expressed as a percent of the initial concentration before processing) that were transferred into the water were 46% (spearmint), 48% (anise), 57% (tea), 64% (caraway), 65% (anise), 67% (caraway), 69% (chamomile), and 69% (caraway) in the same order. We can conclude that higher concentrations of heavy metals were transferred into the water by boiling than by immersion in hot water (tea method).

These results are in agreement with those of Ali (1983, 1987), who concluded that the residual concentration in herbal drugs after its extraction with boiling water may be reduced. Passage of metal into the tea was >50% in only 12% of the Pb assays and 8% of the Cd test. Schilcher et al. (1987) reported that washing the plants by water may remove ~15−30% of heavy metal contaminants. Soliman et al. (1997) reported that heavy metal contents in vegetables were affected by washing. The removal percentages were 100% (Cd), 25−100% (Pb), 50−100% (Cr), 76.9−100% (Co), 11.1−36.71% (Ni), 5.2−20.6% (Cu), 12.7−19.5% (Mn), and 1.1−12.5% (Fe). Also, amounts of Pb and Cd could be removed by washing the samples as reported by Igwegbe et al. (1992) and Battaglia et al. (1984).

Conclusion. Heavy metals are present in spices and medicinal plants at different concentrations, which, in some cases, exceeded the permissible levels. This could be attributed to the use of contaminated irrigation water, the addition of some fertilizers and herbicides, and also contamination from traffic. Sewage sludge, industrial activities, fuel, and automobile tires can also be significant metal sources. Heavy metals can accumulate in plants through both foliage and root systems. On the other hand, heat treatments of medicinal plants by both hot and boiling water can extract different concentrations of the plant metal content into the used water. Boiling in water extracted higher metal

Table 6. Behavior of Heavy Metals[a] in Medicinal Plants during Processing

	spearmint			caraway			anise			chamomile			karkade			tea		
metal	1[b]	2[c]	3[d]	1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
lead	11.5	8.7	6.1	12.8	9.3	6.2	3.2	2.4	1.5	12.2	9.3	5.9	1.0	0.74	0.43	6.8	5.1	3.9
cadmium	3.6	2.8	1.7	2.6	1.9	1.2	4.8	3.6	2.3	2.6	1.9	1.1	2.6	1.9	1.1	*e*	*e*	*e*
chromium	38.3	30.7	20.2	41.8	33.5	27.8	46.0	32.4	29.1	43.9	35.3	25.9	36.2	25.6	16.9	19.4	14.3	11.1
nickel	2.8	2.0	1.3	2.6	1.8	1.1	*e*	*e*	*e*	5.4	3.9	2.1	3.0	2.1	1.3	3.8	2.4	1.6
tin	0.11	*e*	*e*	0.22	*e*	*e*	*e*	*e*	*e*	0.14	*e*	*e*	0.10	*e*	*e*	0.19	*e*	*e*
zinc	35.3	29.8	20.8	57.1	44.3	35.6	127.8	96.5	83.4	51.2	36.3	32.2	21.6	17.4	13.2	16.0	11.0	8.9
manganese	179.4	145.8	105.9	47.3	38.5	29.8	71.9	60.5	40.4	106.7	87.2	73.1	578.3	470.5	381.1	686.0	504.1	360.3
copper	19.9	15.3	11.7	19.2	16.7	12.2	23.5	18.3	14.8	17.6	11.2	8.9	4.1	3.1	2.3	3.6	2.11	1.7
iron	2073.3	1503.4	1027.7	414.3	326.1	286.3	224.0	167.9	136.5	1720.0	1228.0	1052.0	411.1	329	352.0	290.0	204.	156.6

[a] Cobalt was not detected in all the analyzed samples. [b] Initial concentration (μg/2 g of dried plant sample). [c] Concentration in boiling water (μg/100 mL). [d] Concentration in hot water (μg/100 mL). [e] Not detectable.

concentrations from the plant to the used water than heating in hot water.

LITERATURE CITED

Ali, S. L. Bestimmung der pestiziden Rückstände und anderer bedenklicher veruntreinigungen-wie toxische Metallspuren—in Arzneipflanzen. 2-Mitteilung. Bestimmung von toxischen Metallspuren in Arzneidrogen. *Pharm. Ind.* **1983**, *45*, 1294−1295.

Ali, S. L. Assay of pesticide residues and toxic heavy metal traces in plant drugs and their tea infusions. *Pharm. Ztg.* **1987**, *132* (March 12), 633−638.

Al-Kathiri, M. N.; Al-Attar, A. F. Determination of lead and cadmium in parsley tissues grown in the Riyadh area. *Bull. Environ. Contam. Toxicol.* **1997**, *58* (5), 726−732.

Al-Saleh, F. S.; Chudasama, S. Trace metal concentrations in the medicinal plants of the flora of Bahrain. *Toxicol. Environ. Chem.* **1994**, *46* (1−2), 1−9.

AOAC (Association of Official Analytical Chemists). *Official Methods of Analysis*, 16th ed.; Washington, DC, 1995.

Bataglia, R.; Benggert, H.; Romann, E. Lead levels on vegetables grown in the Town of Zürich. *Mitt. Geb. Lebensmittelunters. Hyg.* **1984**, *75*, 127−137.

Bosque, M. A.; Schuhmacher, M.; Domingo, J. L.; Lobet, J. M. L. Concentrations of lead and cadmium in edible vegetables from Tarragona province, Spain. *Sci. Total Environ.* **1990**, *95*, 61−67.

De Smet, P. A. G. M. *Adverse Effects of Herbal Drugs*; Springer-Verlag: Berlin, 1992; ISBN 3-540-53100-9.

Giron, H. C. *At. Absorpt. Newsl.* **1973**, *12*, 28 [cited from Wong et al. (1993)].

Gorsuch, T. T. *The Destruction of Organic Matter*; Pergamon: Oxford, U.K., 1970 [cited from Wong et al. (1993)].

Gosselin, R. E.; Smith, R. P.; Hodge, H. C.; Braddock, J. E. *Clinical Toxicology of Commerical Products*, 5th ed.; Williams and Wilkins: Baltimore, MD, 1984.

Gravel, I. V.; Yakovlev, G. P.; Petrov, N. N.; Stulovskü, S. S.; Listov, S. A. Content of heavy metals in some species of medicinal plants in the Altaiskü Krai. *Rastitel'nye-Resursy* **1994**, *30* (1−2), 101−108.

Igwegbe, A. O.; Belhaj, H. M.; Hassan, T. M.; Gibali, A. S. Effect of a highway's traffic on the level of lead and chromium in fruits and vegetables grown along the roadsides. *J. Food Saf.* **1992**, *13* (1), 7−18.

Janusz, M.; Danutra, W.; Jerzy, K.; Robart, R.; Krysztof, L.; Jerzy, C. The occurrence of Pb, Cd, Cu, Mn, Ni, Co, and Cr in selected species of medicinal plants in Poland. *Bromatol. Chem. Toksykol.* **1995**, *28* (4), 363−368.

Jawad, I. M.; Al-Khafji, S. H.; Ali, A. A. The level of trace metals and other contaminants in some spices and medicinal plants used in Iraq. *J. Biol. Sci. Res.* **1986**, *17* (3), 139−146.

Kim, B. Y.; Kim, K. S.; Lee, J. S.; Yoo, S. H. Survey on the natural content of heavy metal in medicinal herbs and their cultivated soils in Korea. *RDA-J. Agric. Sci., Soil Fert.* **1994**, *36* (2), 310−320.

Krug, H. Gemüseproduktion. *Paul Parey* **1986** (cited from Chizzola, R. Metallic trace elements in herbs and spices grown in Austria. *Acta Hortic.* **1989**, 249).

Listow, S. A.; Petrow, N. W. Heavy metal contents in plants from the USSR. *Dtsch. Apoth.-Ztg.* **1990**, *130* (Nov 29), 2629−2630.

Nasralla, M. M.; Ali, E. A. Lead accumulation in edible portions of crops grown near Egyptian traffic roads. *Agric., Ecosyst. Environ.* **1985**, *13* (1), 73−82.

Peters, H.; Schilcher, H. Schwermetallbelastung von Arzneidrogen und deren Zuberei-tungen. *Planta Med.* **1986**, 521−522.

Schilcher, H. Rückstände und Veruntreinigungen bei Drogen und Drogenzubereitungen. 19. Mitteilung: Zur Wertbestimmung und Qualitätsprüfung von Drogen. *Planta Med.* **1982**, *44*, 65−77.

Schilcher, H. Contamination of natural products with pesticides and heavy metals. In *Topics in Pharmaceutical Sciences 1983*; Breimer, D. D., Speiser, P., Eds.; Elsevier Science Publishers: Amsterdam, The Netherlands, 1983; pp 417−423.

Schilcher, H. Rüchstandsanalytik bei Drogen und Drogenzubereitungen. *Fresenius' Z. Aanl. Chem.* **1985**, *321*, 342−351.

Schilcher, H.; Peters, H.; Wank, H. Pestizide und Schwermetalle in Arzneipflanzen und Arzneipflanzen-zubereitungen. *Pharm. Ind.* **1987**, *49*, 203−211.

Soliman, K. M.; Abou-Arab, A. A. K.; Badawy, A.; Khayria Naguib. Heavy metal contamination levels in Egyptian vegetables and fruits and removal by washing procedure. *Bull. Nutr. Inst. Cairo, Egypt* **1997**, *17* (2), 120−146.

Sovljanski, R.; Obradovie, S.; Kisgeci, J.; Lazie, S.; Macko, V. The heavy metals contens and quality of hop cones treated by pesticide during the vegetation. *Acta Hortic.* **1989**, *249*, 81−88.

Sovljanski, R.; Lazic, S.; MaCko, V.; Obradovic, S. Heavy metals contens in medicincal and spice plants cultivated in yugoslavia. *Herba-Hung.* **1990**, *29* (3), 59−63.

Wong, M. K.; Koh, L. L. *Trace Elem. Res.* **1986**, *10*, 91 [cited from Wong et al. (1993)].

Wong, M. K.; Tan, P.; Wee, Y. C. Heavy metals in some chinese herbal plants. *Biol. Trace Elem. Res.* **1993**, *36*, 135−142.

Yoo-Seung, J. O.; Song-Kyung-Sik. Studies on the heavy metal content in cultivated medicinal plants and their correlations with soils. *Saengyak Hakhoechi* **1991**, *22* (1), 33−35.

Received for review May 14, 1999. Revised manuscript received February 16, 2000. Accepted February 24, 2000.

JF990508P