ASSESSMENT OF HEAVY METALS CONTENT IN SOME MEDICINAL PLANTS AND SPICES COMMONLY USED IN ROMANIA

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Abstract

The present study was designed to investigate the level of six heavy metals (arsenic, cadmium, copper, iron, mercury and lead) in both wild-growing and packaged samples from twelve commonly used medicinal plants and spices in Romania. The concentrations of all heavy metals, except mercury and lead, were within permissible limits in the analysed plant samples. The wild-growing sample of *Tilia cordata* and some packaged samples of *Origanum vulgare* and *Thymus vulgare* contained higher levels of mercury than 0.1 mg/kg. The concentrations of lead exceeded the limit recommended by the EU (0.3 mg/kg) for some samples of *Achillea millefolium*, *Matricaria chamomilla*, *Mentha piperita*, *Ocimum basilicum*, *Origanum majorana* and *Tilia cordata*. The findings of the study highlight the need for constant monitoring of heavy metals residues in order to ensure the quality and safety of herbal products.

Rezumat

Subiectul studiului prezent l-a constituit evaluarea nivelelor a 6 metale grele (arsenic, cadmiu, cupru, fier, mercur și plumb) în probe colectate din flora spontană și probe ambalate sub formă de ceaiuri medicinale aparținând la 12 specii de plante medicinale și condimente frecvent utilizate în România. Concentrațiile tuturor metalelor grele au fost mai mici față de limitele permise, cu excepția valorilor obținute pentru mercur și plumb. Proba colectată din flora spontană pentru *Tilia cordata* și unele probe ambalate de *Origanum vulgare* și *Thymus vulgare* conțin nivele de mercur mai mari de 0,1 mg/kg. Conținutul în plumb a depășit limita recomandată de UE (0,3 mg/kg) în cazul unor probe de *Achillea millefolium*, *Matricaria chamomilla*, *Mentha piperita*, *Ocimum basilicum*, *Origanum majorana* și *Tilia cordata*. Rezultatele studiului evidențiază nevoia unei monitorizări constante a prezenței și nivelului metalelor grele în vederea asigurării calității și siguranței la administrare a produselor din plante medicinale.

Keywords: heavy metals, medicinal plants, spices, contamination

Introduction

Medicinal plants and plant-derived products play a significant role in the strategies for the assessment of global health and their use is rapidly rising. WHO reported that about 80% of the population in developing countries relies on traditional herbal medicine for their primary healthcare [29]. These findings impose major concerns on the quality and safety of herbal products in order to provide appropriate and effective therapy [12, 15, 16]. Among the main safety risks related to herbal medicines is contamination with heavy metals. The medicinal plants may be contaminated with toxic heavy metals during cultivation, harvesting or processing stages [25]. Also, the environmental pollution that includes industrial waste, traffic emissions and some agricultural activities can affect significantly the heavy metals content in medicinal plants. The uptake and accumulation of heavy

metals in vegetal tissues can directly affect plant growth and can clinically pose major threat for the human health. Excessive levels of these elements can cause central nervous system toxicity (mercury, lead, arsenic), produce kidney and liver damage (mercury, lead, cadmium, copper), or respiratory dysfunction (cadmium), and have adverse effects on skin, bones and teeth (nickel, cadmium, copper, chromium) [2, 11]. Also, large amounts of iron might cause abdominal and skin injuries [2]. Thus, the assessment of metal contamination in herbal raw materials is an important step in the control of plants' quality. Nevertheless, there is limited data in terms of heavy metals contamination of medicinal and culinary plants used in Romanian traditional medicine or diet. Taking all this into account, the present study was designed to investigate the level of six heavy metals (arsenic-As, cadmium-Cd, copper-Cu, iron-Fe, mercury-

Hg and lead-Pb) in some commonly used medicinal plants and spices in Romania.

Materials and Methods

Plant material

A total of forty-two samples belonging to six medicinal plants and six spices commonly used in Romania were analysed. For each medicinal plant species, four samples were analysed as follows: a sample collected from

spontaneous flora of Galați County, Romania, and three samples provided by different indigenous producers as herbal teas (packaged samples). In the case of spices, for each plant species, three samples were analysed, namely: a sample collected from spontaneous flora of Galați County, Romania, and two samples purchased from local suppliers. Table I shows the herb samples investigated in this study and some characteristics of the plant species [26].

Table I

Samples and some characteristics of selected medicinal plants and spices

Plant species	Samples		Common name	Part used	Main beneficial effects
	indigenous producers	spontaneous flora			
Medicinal plants					
<i>Achillea millefolium</i> L.	AM1, AM2, AM3	AM4	yarrow	flowers	- eupeptic - cholagogue - carminative - anti-inflammatory - spasmolytic
<i>Calendula officinalis</i> L.	CO1, CO2, CO3	CO4	marigold	flowers	- cholaretic - anti-inflammatory - spasmolytic - wound healing - antimicrobial
<i>Hypericum perforatum</i> L.	HP1, HP2, HP3	HP4	St. John's wort	aerial parts	- cholaretic - anti-diarrheal - antidepressive - wound healing - antimicrobial
<i>Matricaria chamomilla</i> L.	MC1, MC2, MC3	MC4	chamomile	flowers	- spasmolytic - carminative - anti-inflammatory - sedative - antiulcer
<i>Ocimum basilicum</i> L.	OB1, OB2, OB3	OB4	basil	aerial parts	- eupeptic - spasmolytic - carminative - diuretic
<i>Tilia cordata</i> Mill.	TC1, TC2, TC3	TC4	linden	flowers	- expectorant - spasmolytic - sedative
Spices					
<i>Anethum graveolens</i> L.	AG1, AG2	AG3	dill	leaves	- carminative - eupeptic - diuretic
<i>Mentha piperita</i> L.	MP1, MP2	MP3	peppermint	leaves	- cholaretic - cholagogue - spasmolytic - carminative - anti-diarrheal
<i>Origanum majorana</i> L.	OM1, OM2	OM3	marjoram	aerial parts	- eupeptic - spasmolytic - carminative - sedative - diuretic
<i>Origanum vulgare</i> L.	OV1, OV2	OV3	oregano	aerial parts	- eupeptic - spasmolytic - carminative - expectorant - antimicrobial

Plant species	Samples		Common name	Part used	Main beneficial effects
	indigenous producers	spontaneous flora			
Spices					
<i>Petroselinum crispum</i> (Mill.) Fuss.	PC1, PC2	PC3	parsley	leaves	- carminative - diuretic - antihypertensive - haemostatic
<i>Thymus vulgaris</i> L.	TV1, TV2	TV3	thyme	aerial parts	- eupeptic - spasmolytic - carminative - expectorant - diuretic

Sample processing and heavy metals analysis

The content of heavy metals in selected plants and spices was determined using atomic absorption spectrometry after wet digestion of the samples with 65% HNO₃ and 30% H₂O₂ [30]. The analysis of As, Cd and Pb was done using atomic absorption spectrometry with graphite furnace atomization (SpectrAA240Z spectrophotometer with GTA120 furnace, Varian, The Netherlands) [23, 25]. Flame atomic absorption spectrometry was performed for the analysis of Fe and Cu content (SpectrAA240FS spectrophotometer, Varian, The Netherlands) [23]. The Hg concentrations were determined by cold vapour atomic absorption spectrometry (SpectrAA240FS spectrophotometer, Varian, The Netherlands) [24]. High purity standards for heavy metals (Merck, Germany) were used as reference analytes and the calibration standards were prepared by appropriate dilution of their stock solutions. The concentrations of heavy metals were expressed as mg/kg.

Statistical analysis

Four determinations were performed for each sample and the mean and standard deviation were calculated.

Results and Discussion

The regulatory guidelines for heavy metals content in plants are presented in Table II. The results of the analysis of heavy metals in the selected medicinal plants and spices are summarized in Tables III and IV, respectively.

Arsenic (As). It is a highly toxic non-essential metalloid that significantly affects human health leading to gastrointestinal and hepatic disorders, hypotension, polyneuropathy, loss of sight and carcinogenic effects [19]. Plants are exposed to As mainly from anthropogenic activities such as mining and smelting, the use of As-based pesticides in agriculture and of As-contaminated groundwater for irrigation purposes [7]. In our study, the level of As varied from less than 0.01 to 0.15 mg/kg in medicinal plants samples (Table III) and from less than 0.01 to 0.78 mg/kg in spices, respectively (Table IV). Maximum concentrations of As were found in *Tilia cordata* and *Petroselinum crispum* packaged samples, respectively (TC3; PC1); in all samples of *Hypericum perforatum*, As was undetectable. In general, we noticed that the investigated plant samples contained extremely low concentrations of As. Only some samples of spices, mainly *Petroselinum crispum* (PC1, PC2), *Thymus vulgare* (TV2) and *Origanum vulgare* (OV2, OV3) showed higher values but all were below the acceptable limit (Table II and Table IV). The levels of As identified in this work are much lower than those noticed in other studies. Oprea *et al.* reported for samples of some medicinal plants from two areas belonging to the Baia Mare municipality, Maramureş County (Romania), concentrations of As ranging from 0.11 to 5.33 mg/kg [17]. These large amounts of As are primarily associated with specific metallurgical industry and soil pollution of Baia Mare [17], but contamination with As is important due to the fact that Romania is one of the countries having problems due to the natural arsenic groundwater [28].

Table II

Acceptable limits of the heavy metals in medicinal plants/culinary plants (mg/kg)

Limits	Arsenic (As)	Cadmium (Cd)	Mercury (Hg)	Lead (Pb)
European Pharmacopoeia [6]	-	1	0.1	5
FAO/WHO [30]	1	0.3	-	10
EU [13]	-	0.2	-	0.3

Cadmium (Cd) is a non-essential hazardous heavy metal that causes serious effects on the kidney, liver, vascular and immune system. Prolonged exposure to high-doses of Cd results in the Itai-Itai disease that is characterized by severe renal tubular dysfunction, osteomalacia and osteoporosis [20]. The combustion of fossil fuels, the use of phosphate fertilizers, non-

ferrous smelters, mines and sewage sludge application are the major sources for the Cd contamination of plants [11]. This metal presents high rates of soil-to-plant transfer such that it is a contaminant found in most human foodstuffs [20]. The levels of Cd were within allowable limits (Table II) in all samples of medicinal plants (0.01 - 0.05 mg/kg) (Table III) and

spices (0.02 - 0.12 mg/kg) (Table IV). The highest concentration of Cd was noticed for the sample of *Anethum graveolens* collected from spontaneous flora (0.12 mg/kg) (AG3). All samples of spices harvested from spontaneous flora have higher values of Cd levels compared to their packaged variants of different

origin (0.01 - 0.06 mg/kg vs. 0.05 - 0.12 mg/kg) (Table IV). Cd was undetected in all samples of *Hypericum perforatum*, the sample of wild-growing *Tilia cordata* (TC4) (Table III) and some packaged samples of *Origanum majorana* (OM1) and *Petroselinum crispum* (PC1) (Table IV).

Table III

The content of heavy metals in selected medicinal plants

Plant species/samples		Mean concentration of heavy metals $\pm$ standard deviation (mg $\times$ kg ⁻¹)					
		Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Iron (Fe)	Mercury (Hg)	Lead (Pb)
<i>Achillea millefolium</i>	AM1	0.03 $\pm$ 0.03	0.04 $\pm$ 0.00	11.00 $\pm$ 0.55	180.00 $\pm$ 5.47	0.011 $\pm$ 0.00	0.09 $\pm$ 0.00
	AM2	ND	0.04 $\pm$ 0.00	14.00 $\pm$ 0.67	1490.00 $\pm$ 86.04	0.015 $\pm$ 0.00	0.09 $\pm$ 0.00
	AM3	ND	0.02 $\pm$ 0.00	15.00 $\pm$ 0.86	980.00 $\pm$ 5.47	0.021 $\pm$ 0.00	0.16 $\pm$ 0.01
	AM4	0.05 $\pm$ 0.00	0.02 $\pm$ 0.00	10.00 $\pm$ 0.56	1420.00 $\pm$ 75.66	0.019 $\pm$ 0.00	0.42 $\pm$ 0.01
<i>Calendula officinalis</i>	CO1	0.03 $\pm$ 0.00	0.01 $\pm$ 0.00	4.00 $\pm$ 0.28	13330.00 $\pm$ 111.45	0.027 $\pm$ 0.00	ND
	CO2	0.02 $\pm$ 0.00	0.01 $\pm$ 0.00	13.00 $\pm$ 0.36	14530.00 $\pm$ 103.15	0.008 $\pm$ 0.00	0.25 $\pm$ 0.01
	CO3	0.03 $\pm$ 0.00	0.01 $\pm$ 0.00	16.00 $\pm$ 0.74	21020.00 $\pm$ 114.69	0.013 $\pm$ 0.00	0.14 $\pm$ 0.01
	CO4	0.01 $\pm$ 0.00	0.01 $\pm$ 0.00	24.00 $\pm$ 0.80	11970.00 $\pm$ 100.44	0.015 $\pm$ 0.00	0.24 $\pm$ 0.01
<i>Hypericum perforatum</i>	HP1	ND	ND	11.00 $\pm$ 0.53	730.00 $\pm$ 7.39	ND	ND
	HP2	ND	ND	11.00 $\pm$ 0.59	670.00 $\pm$ 7.35	ND	ND
	HP3	ND	ND	15.00 $\pm$ 1.02	560.00 $\pm$ 5.23	ND	ND
	HP4	ND	ND	17.00 $\pm$ 0.93	5600.00 $\pm$ 64.90	ND	ND
<i>Matricaria chamomilla</i>	MC1	0.06 $\pm$ 0.00	0.01 $\pm$ 0.00	10.00 $\pm$ 0.82	4840.00 $\pm$ 72.85	0.020 $\pm$ 0.00	0.04 $\pm$ 0.00
	MC2	ND	0.02 $\pm$ 0.00	15.00 $\pm$ 1.17	14480.00 $\pm$ 107.26	ND	ND
	MC3	0.01 $\pm$ 0.00	0.01 $\pm$ 0.00	12.00 $\pm$ 1.00	9660.00 $\pm$ 88.10	ND	0.49 $\pm$ 0.02
	MC4	ND	0.02 $\pm$ 0.00	12.00 $\pm$ 0.93	2960.00 $\pm$ 37.86	ND	0.28 $\pm$ 0.01
<i>Ocimum basilicum</i>	OB1	0.04 $\pm$ 0.00	0.01 $\pm$ 0.00	10.00 $\pm$ 0.52	6470.00 $\pm$ 55.48	0.020 $\pm$ 0.00	0.70 $\pm$ 0.02
	OB2	0.01 $\pm$ 0.01	0.01 $\pm$ 0.00	15.00 $\pm$ 1.16	8410.00 $\pm$ 95.44	ND	ND
	OB3	ND	0.01 $\pm$ 0.00	11.00 $\pm$ 0.58	2870.00 $\pm$ 59.25	ND	0.60 $\pm$ 0.02
	OB4	ND	0.01 $\pm$ 0.00	15.00 $\pm$ 1.06	3580.00 $\pm$ 76.01	ND	2.03 $\pm$ 0.08
<i>Tilia cordata</i>	TC1	0.05 $\pm$ 0.00	0.05 $\pm$ 0.00	9.00 $\pm$ 0.56	6390.00 $\pm$ 55.95	0.018 $\pm$ 0.00	0.15 $\pm$ 0.01
	TC2	ND	0.02 $\pm$ 0.00	14.00 $\pm$ 0.90	1010.00 $\pm$ 12.19	0.012 $\pm$ 0.00	ND
	TC3	0.15 $\pm$ 0.01	0.01 $\pm$ 0.00	17.00 $\pm$ 1.02	6820.00 $\pm$ 70.74	0.012 $\pm$ 0.00	0.01 $\pm$ 0.00
	TC4	ND	ND	12.00 $\pm$ 0.75	2230.00 $\pm$ 32.07	0.106 $\pm$ 0.01	0.32 $\pm$ 0.02

Table IV

The content of heavy metals in selected spices

Spices/Samples		Mean concentration of heavy metals $\pm$ standard deviation (mg/kg)					
		Arsenic (As)	Cadmium (Cd)	Cooper (Cu)	Iron (Fe)	Mercury (Hg)	Lead (Pb)
<i>Anethum graveolens</i>	AG1	0.04 $\pm$ 0.00	0.02 $\pm$ 0.00	127.00 $\pm$ 5.68	320.00 $\pm$ 7.19	ND	0.09 $\pm$ 0.00
	AG2	ND	0.04 $\pm$ 0.00	142.00 $\pm$ 6.39	348.00 $\pm$ 9.11	ND	0.08 $\pm$ 0.00
	AG3	0.08 $\pm$ 0.00	0.12 $\pm$ 0.01	133.00 $\pm$ 5.83	320.00 $\pm$ 6.78	ND	ND
<i>Origanum majorana</i>	OM1	ND	ND	163.00 $\pm$ 6.86	657.00 $\pm$ 5.45	ND	0.14 $\pm$ 0.00
	OM2	0.01 $\pm$ 0.00	0.05 $\pm$ 0.00	131.00 $\pm$ 4.70	719.00 $\pm$ 6.61	ND	0.13 $\pm$ 0.00
	OM3	0.07 $\pm$ 0.00	0.05 $\pm$ 0.00	348.00 $\pm$ 7.48	473.00 $\pm$ 3.74	ND	0.95 $\pm$ 0.01
<i>Origanum vulgare</i>	OV1	0.03 $\pm$ 0.00	0.06 $\pm$ 0.00	201.00 $\pm$ 5.78	1375.00 $\pm$ 32.64	0.374 $\pm$ 0.01	ND
	OV2	0.21 $\pm$ 0.00	0.06 $\pm$ 0.00	198.00 $\pm$ 5.77	1460.00 $\pm$ 59.40	0.019 $\pm$ 0.00	0.04 $\pm$ 0.00
	OV3	0.33 $\pm$ 0.00	0.07 $\pm$ 0.00	222.00 $\pm$ 7.71	2471.00 $\pm$ 77.56	0.075 $\pm$ 0.00	ND
<i>Mentha piperita</i>	MP1	0.18 $\pm$ 0.01	0.01 $\pm$ 0.00	309.00 $\pm$ 7.88	6683.00 $\pm$ 40.39	ND	0.58 $\pm$ 0.06
	MP2	0.09 $\pm$ 0.00	0.05 $\pm$ 0.00	237.00 $\pm$ 4.86	2378.00 $\pm$ 12.41	ND	0.18 $\pm$ 0.01
	MP3	ND	0.10 $\pm$ 0.01	157.00 $\pm$ 3.34	673.00 $\pm$ 6.35	0.010 $\pm$ 0.00	1.28 $\pm$ 0.08
<i>Petroselinum crispum</i>	PC1	0.78 $\pm$ 0.01	ND	133.00 $\pm$ 5.61	1360.00 $\pm$ 11.20	0.083 $\pm$ 0.00	0.06 $\pm$ 0.00
	PC2	0.19 $\pm$ 0.00	0.05 $\pm$ 0.00	122.00 $\pm$ 3.44	336.00 $\pm$ 2.38	0.052 $\pm$ 0.00	ND
	PC3	ND	0.07 $\pm$ 0.00	123.00 $\pm$ 4.03	748.00 $\pm$ 5.93	0.026 $\pm$ 0.00	0.11 $\pm$ 0.00
<i>Thymus vulgaris</i>	TV1	0.03 $\pm$ 0.00	0.03 $\pm$ 0.00	191.00 $\pm$ 6.35	1314.00 $\pm$ 12.39	0.131 $\pm$ 0.01	0.07 $\pm$ 0.00
	TV2	0.42 $\pm$ 0.01	0.07 $\pm$ 0.00	131.00 $\pm$ 4.73	1449.00 $\pm$ 11.32	0.036 $\pm$ 0.00	0.09 $\pm$ 0.00
	TV3	ND	0.09 $\pm$ 0.00	169.00 $\pm$ 5.32	2578.00 $\pm$ 13.62	0.024 $\pm$ 0.00	0.11 $\pm$ 0.00

Cooper (Cu) is an essential trace element in humans and plants. It is a cofactor of many redox enzymes and plays a significant role in iron metabolism and many physiological processes including antioxidant defence, neuropeptide synthesis and immune function [3]. Also, Cu is essentially involved in normal growth and development of plants [31]. Elevated levels of Cu can cause gastrointestinal symptoms, hepatotoxicity, skin irritation and sensitization, and dysfunction of upper respiratory tract [4]. Sources of Cu contamination in plants could be linked to industrial and mining activities.

The Cu concentrations ranged from 4 to 24 mg/kg in samples of medicinal plants (Table III) and from 122 to 348 mg/kg in spices samples (Table IV). The highest levels were recorded in wild-growing samples of *Calendula officinalis* (CO4) (Table III) and *Origanum majorana* (OM 3) (Table IV). For similar species, the results obtained in this study are higher than other samples from Romania [8] or other regions [9, 10]. Also, all investigated samples of spices have a high Cu content (over 120 mg/kg).

Iron (Fe) is a biologically essential trace element for all living organisms. It is involved in a broad variety of cellular processes, namely electron transport and oxygen supply, cellular respiration, synthesis of DNA and proteins, cell proliferation and differentiation, regulation of gene expression and immunity [4]. Overdoses (above 20 mg/kg) of iron are associated with gastrointestinal disorders, metabolic acidosis, cardiomyopathy, and renal and hepatic failure [4]. The results from this current study revealed a large variation of Fe content in selected plant samples. It was found in a range between 180 and 21020 mg/kg in samples of medicinal plants (Table III) and between 320 and 6683 mg/kg in spices samples (Table IV). The highest Fe level was obtained for a packaged sample of *Calendula officinalis* (CO3). In fact, all samples of *Calendula officinalis* contained large amounts of Fe (over 11000 mg/kg) (Table III). Among investigated species, *Anethum graveolens*, *Origanum majorana* and *Petroselinum crispum* are characterized by a low accumulation of Fe. The *Mentha piperita* and *Hypericum perforatum* presented large variations between samples (even 10 times) (Tables III and IV). In the case of all samples, our results are higher than the reported Fe values for similar species from Mediterranean area [10], United Arab Emirates [5], or even Romania [8].

Mercury (Hg) is a highly toxic non-essential metal that is considered to be a dangerous xenobiotic in living cells. The consequences of Hg toxicity mainly as organic mercury compounds include central nervous system damage, alteration of motor function, neuro-behavioural, neurodevelopmental, immunological effects and cardiovascular and kidney injuries. Exposure of plants to Hg derives from natural sources (volcanic eruptions, emissions from the ocean) and anthropogenic activities (burning of fossil fuels, production of

cement, mining, smelting, production of ferrous and non-ferrous metals) [21]. Hg levels were within allowable limits in our samples (Tables II, III and IV) except for a wild-growing sample of *Tilia cordata* (0.106 mg/kg) (TC4) and some packaged samples of *Origanum vulgare* (OV1) and *Thymus vulgare* (TV1) spices (0.374 and 0.131 mg/kg, respectively). Hg was not detected in any of the samples of *Hypericum perforatum*, *Anethum graveolens* and *Origanum majorana*. Also, some packaged and wild-growing samples of *Matricaria chamomilla* (MC4), *Ocimum basilicum* (OB4) (Table III) and packaged samples of *Mentha piperita* (MP1, MP2) do not contained Hg (Table IV). It seems that the accumulation of Hg in plants depends primarily on the plants species, the degree of soil and atmosphere contamination, the season for harvesting [14]. Şenilă *et al.* reported high concentrations of Hg in perennial plants growing in the urban area of Baia Mare, a historical mining and metal processing centre from North-western Romania. The average content exceeded the permissible limit of Hg in plants (0.22 mg/kg compared to 0.1 mg/kg) [27]. Also, Ordak *et al.* showed a high accumulation of Hg in the leaves of the more commonly used perennial plants collected from the vicinity of busy streets in Poland [14].

Lead (Pb) is a highly noxious metal for humans and the environment. It can affect any organ but causes toxic effects mainly on the nervous central system, the cardiovascular, haematological and renal systems. Plants' exposure to Pb results from: mining, smelting, steel industry, electroplating, inorganic fertilizers and pesticides use, fuel combustion, exhaust of automobiles and sewage sludge [22]. The content of Pb ranged from 0.04 to 2.03 mg/kg in samples of medicinal plants used as herbal teas (Table III) and from 0.04 to 1.28 mg/kg in samples of spices (Table IV). The highest levels were found in wild-growing samples of *Ocimum basilicum* (2.03 mg/kg) (OB4) and *Mentha piperita* (1.28 mg/kg) (MP3). The concentrations of Pb were lower compared to the permissible limits defined by the European Pharmacopoeia and WHO/FAO (Table II), but they exceeded the maximum value required by the EU (0.3 mg/kg) for 75% of *Ocimum basilicum* samples, 66.66% of *Mentha piperita* samples and 33% of *Origanum majorana*, *Matricaria chamomilla*, *Achillea millefolium* and *Tilia cordata* samples, respectively (Tables III and IV). In general, the samples of plants collected from spontaneous flora are highly contaminated with lead. However, all values were inline or lower compared to those reported in other studies for similar plants from different regions of the globe [1, 5, 25], or even Romania [18].

Conclusions

In our study, forty-two samples of medicinal plants and spices commonly used in Romania were screened for six heavy metals contents (arsenic, cadmium, cooper,

iron, mercury and lead). The concentrations of arsenic, cadmium were below the permissible limits.

Undetectable levels of arsenic, cadmium, mercury and lead were noticed for *Hypericum perforatum*. The content of mercury crossed the limits for some samples of *Origanum vulgare*, *Tilia cordata* and *Thymus vulgare*. Also, the concentrations of lead exceeded the limit recommended by EU for some samples of *Achillea millefolium*, *Matricaria chamomilla*, *Mentha piperita*, *Ocimum basilicum*, *Origanum majorana* and *Tilia cordata*.

The high load of iron and copper can be associated with the collection site, such as the metallurgical type industrialized area of Galați city, Romania, in the case of wild-growing plants, and additionally, the processing methods for packaged samples. The findings of the study highlight the need of constant monitoring of heavy metals residues or other pollutants in order to ensure the quality and safety of herbal products. Therefore, the present study provides baseline data useful for a comprehensive assessment of heavy metals in medicinal plants from different parts of our country.

Conflict of interest

The authors declare no conflict of interest.

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