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ORIGINAL PAPER

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Monitoring of metallic micronutrients and heavy metals in herbs, spices and medicinal plants from Austria

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Abstract Medicinal, aromatic and spice plants grown in different regions of Austria were monitored as to their Cd, Cu, Fe, Mn, Pb and Zn contents. Since the plants were grown under common field conditions, the essential elements were within the usual ranges for plant material. The contamination level with the toxic heavy metals, Pb and Cd, can be classified as normally low. Most samples contained less than 0.2 mg kg^{-1} Cd and less than 1.5 mg kg^{-1} Pb on a dry weight basis. Comparison with previous investigations suggests that contaminations with Pb occur rather by chance, whereas enhanced Cd values are restricted to some species having a tendency to accumulate this heavy metal. Some such species are St. John's wort, poppy, yarrow, chamomile and absinth. Careful choice of growing site and appropriate soil management can reduce the Cd uptake of these critical species. These precautions are important when larger amounts of the product are consumed.

Keywords Drug quality · Heavy metal · Cadmium uptake · Elemental composition

Introduction

As in other countries, interest in growing aromatic, medicinal and spice plants has increased in Austria in recent years. These plants are produced on a rather small scale in various regions [1]. The producing regions are concentrated in the northern parts of Austria, mainly northern Lower Austria and Upper Austria. The growers are organised in local associations. The requirements include the production of high quality raw material. Good Agricultural Practice regulations should help in achieving this purpose [2].

The composition of foods and food additives and some herbal products including mineral and micronutrient elements is compiled in comprehensive tables [3]. However, many medicinal and spice plants are not listed in such publications. Furthermore, these plants represent a multitude of different species that are grown in quite different regions, so that not only are big differences in the constitutive essential micronutrients to be expected, but also in the level of contamination with toxic heavy metals. As the consumer desires products free from potentially harmful constituents, especially in products related to health, the monitoring of toxic heavy metals in medicinal plants is part of the quality control in the pharmaceutical industry [4, 5].

Therefore, it is of interest to study these trace metallic elements in the respective plants of known origin. A new set of data is now presented in addition to previous reports from 1989 [6] and 1996 [7] in order to evaluate medium to long-term trends.

Material and methods

Plant material was obtained from local grower associations in Lower and Upper Austria, from "Saatbau Linz", Upper Austria, from the "Landesversuchsanlage für Spezialkulturen in Wies", Styria or directly from growers. The samples were taken from the growing seasons 1999 and 2000. Some plants were also collected in the botanical garden of the Veterinary University of Vienna.

Plant analysis. The air-dried plant material with a remaining moisture of 4–8% was finely powdered using a Cyclotec 1093 sample mill from Tecator (Höganäs, Sweden) and stored in plastic vials until analysis. Small fatty seeds such as poppy, caraway etc. were directly digested without prior grinding. The wet digestion of about 0.3 g plant material with 3 ml concentrated HNO_3 and 0.5 ml H_2O_2 was carried out in PTFE beakers in an Ethos 900 microwave oven, from MLS Mikrowellen-Laborsysteme (Leutkirch im Allgäu, Germany). An adapted heating program ensured the complete mineralisation of the plant material. Bidistilled water was added to the digested material to the 10 ml level and the solution subjected to atomic absorption spectroscopy on a Hitachi Z-8100 system (Hitachi, Tokyo, Japan) with Zeeman background correction. The

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elements Cu, Mn, Fe and Zn were recorded with the flame technique using standard analytic conditions. Cd and Pb were measured with the graphite furnace technique. For Pb, we used the standard addition method. Analytical quality control was carried out with the use of the reference material V-10 hay powder, obtained from the International Atomic Energy Agency, Vienna.

Results and discussion

The variability in the elemental composition of selected medicinal plants is presented in Table 1, and Table 2 shows examples of the composition of further species.

Essential micronutrients

Copper levels ranged from 3 to 34 mg kg^{-1} with most samples between 8 and 16 mg kg^{-1}. Peppermint leaves and poppy seeds turned out to be Cu rich whereas the lowest Cu levels were recorded in some leaves, such as yarrow and thyme.

The greatest variability was observed in Fe content. The highest values were above 1,000 mg kg^{-1}, whereby leaf and herb drugs showed higher Fe contents than products derived from other plant parts. Products low in Fe are yarrow herb and some fruit drugs such as caraway and fennel.

Mn ranged from 14 mg kg^{-1} to more than 100 mg kg^{-1} in the samples. Higher concentrations were found in leaves and herbs, especially in St. Johńs wort and lovage;

Table 1 Metallic trace elements in selected medicinal and aromatic plants (milligrams/kilogram dry weight)

		Cu	Fe	Mn	Zn	Cd	Pb
Achillea millefolium (herb), *n*=3	Mean	6.8	66.9	66.0	28.5	0.21	1.0
	Median	5.1	67.3	59.1	24.7	0.14	1.0
	SD	3.1	9.1	26.6	10.6	0.16	0.8
	Min.	4.9	57.6	43.5	20.5	0.09	0.2
	Max.	10.3	75.8	95.3	40.5	0.39	1.7
Carum carvi (fruits), *n*=10	Mean	11.2	54.6	29.9	35.5	0.05	0.2
	Median	11.0	54.2	27.7	35.5	0.05	0.2
	SD	2.6	17.8	10.6	4.4	0.02	0.1
	Min.	5.7	33.5	16.4	27.8	0.02	0.1
	Max.	16.3	91.5	54.2	45.2	0.1	0.4
Foeniculum vulgare (fruits), *n*=15	Mean	11.8	60.7	32.6	37.4	0.03	0.4
	Median	12.2	58.0	33.6	33.6	0.03	0.3
	SD	2.5	17.2	6.4	13.0	0.01	0.2
	Min.	5.5	43.8	23.8	19.5	<0.01	0.1
	Max.	15.1	109	45.0	63.5	0.06	1.1
Hypericum perforatum (herb), *n*=6	Mean	7.7	163	76.5	36.5	0.59	0.4
	Median	7.6	165	61.2	38.8	0.64	0.3
	SD	1.3	89.3	51.7	10.9	0.36	0.3
	Min.	6.2	57.1	33.8	20.9	0.15	0.2
	Max.	10.1	303	175.0	47.5	0.98	1.0
Levisticum officinale (leaves), *n*=4	Mean	7.0	248	78.9	24.4	0.6	1.9
	Median	7.5	254	71.7	28.4	0.05	1.5
	SD	1.7	133	36.8	9.9	0.05	1.8
	Min.	4.7	78	48.2	9.9	0.02	0.4
	Max.	8.5	403	123.8	31.0	0.13	4.3
Melissa officinalis (herb), *n*=14	Mean	8.4	544	45.8	32.2	0.02	0.8
	Median	8.9	479	44.1	29.7	0.02	0.8
	SD	2.6	355	17.0	14.3	0.01	0.3
	Min.	5.0	126	21.9	13.5	0.01	0.5
	Max.	13.3	1398	89.1	66.1	0.04	1.6
Mentha piperita (herb), *n*=12	Mean	15.4	305	54.0	28.7	0.05	0.8
	Median	13.9	207	54.1	28.1	0.03	0.5
	SD	8.1	292	13.8	7.5	0.07	0.7
	Min.	6.0	102	32.1	14.4	0.02	0.2
	Max.	34.1	1154	75.1	42.4	0.27	2.4
Papaver somniferum (seeds), *n*=9	Mean	16.9	66.3	57.8	56.9	0.25	0.1
	Median	16.8	60.4	60.8	55.7	0.28	0.1
	SD	2.8	19.9	11.7	7.2	0.14	0.1
	Min.	12.6	48.8	36.8	46.2	0.09	<0.05
	Max.	21.3	104.1	75.3	67.7	0.42	0.4
Salvia officinalis (leaves), *n*=6	Mean	10.0	635	52.8	33.0	0.01	0.8
	Median	9.1	738	51.7	33.5	0.01	0.7
	SD	4.1	298	12.6	9.0	0.01	0.2
	Min.	5.7	152	39.0	21.6	<0.01	0.6
	Max.	17.5	889	67.6	44.9	0.03	1.3

Table 2 Further examples of trace elements in medicinal plants (milligrams/kilogram dry weight). Regions: *1* Waldviertel, *2* Linz, *3* Vienna, *4* Upper Austria, *5* Weinviertel, *6* Styria

		Region	Cu	Mn	Zn	Fe	Cd	Pb
Leaves and herbs	Allium schoenoprasum	3	5.6	24.2	30.2	65.6	0.05	0.16
	Althaea officinalis	3	6.8	38.1	30.3	114.8	0.06	0.77
	Artemisia abrotanum	3	13.1	75.2	49.3	89.4	0.05	1.41
	Artemisia absinthium	4	5.9	47.7	35.2	209.3	0.75	
	Artemisia absinthium	3	13.6	58.7	58.6	144.5	0.06	2.08
	Artemisia dracunculus	3	16.9	64.2	53.5	79.1	0.06	1.25
	Borago officinalis	3	9.3	70.6	39.8	123.2	0.06	0.86
	Fragaria vesca	6	10.2	70.5	40.1	888.0	0.10	0.45
	Malva sylvestris	3	6.7	56.0	65.9	202.1	0.16	0.74
	Ocimum basilicum	3	23.3	65.1	85.2	140.5	0.02	1.34
	Rosmarinum officinalis	3	3.1	38.9	14.2	375.2	0.00	1.45
	Satureja hortensis	3	9.9	36.1	81.3	99.4	0.02	0.6
	Satureja montana	3	12.3	30.7	47.7	73.1	0.01	0.4
	Thymus serpyllum	3	4.1	60.9	32.8	111.5	0.03	0.62
	Thymus vulgaris	3	7.2	84.9	14.4	267.3	0.01	1.12
	Urtica dioica	6	10.5	44.8	30.5	102.0	0.02	0.27
	Mean		9.9	54.2	44.3	192.8	0.1	0.9
	SD		5.2	17.5	20.8	202.7	0.2	0.5
Flowers	Althaea officinalis	3	11.7	21.9	26.3	53.0	0.03	0.26
	Anthemis nobilis	3	12.3	52.5	36.7	159.1	0.06	1.03
	Borago officinalis	3	10.1	54.5	32.5	85.7	0.06	0.77
	Calendula officinalis	3	14.6	21.3	47.6	79.7	0.03	1.1
	Calendula officinalis	6	6.1	49.2	20.8	206.8	0.01	0.29
	Centaurea cyanus	6	10.2	42.0	42.7	181.4	0.09	0.1
	Malva sylvestris	3	6.1	39.1	38.5	159.0	0.06	0.65
	Matricaria recutita	1	6.0	77.7	30.6	91.4	0.23	0.31
	Thymus serpyllum	3	8.5	61.3	58.6	84.4	0.01	0.46
	Mean		9.5	46.6	37.1	122.3	0.1	0.6
	SD		3.1	18.1	11.5	54.4	0.1	0.4
Fruits and seeds	Coriandrum sativum	1	14.9	33.1	55.8	38.5	0.03	0.02
	Pimpinella anisum	5	9.7	29.2	37.1	193.9	0.10	0.21
	Sylibum marianum	1	18.8	23.1	44.4	157.1	0.05	0.12
	Sylibum marianum	1	20.0	31.7	59.6	587.8	0.05	0.37
	Amaranthus hypochondr.	6	7.4	19.1	39.9	142.3	0.05	0.24
	Cannabis sativa	6	17.8	105.1	76.0	158.3	0.04	0.28
	Chenopodium quinoa	6	5.3	19.6	32.4	72.1	0.05	0.3
	Cucurbita pepo	5	12.8	39.4	77.3	74.2	0.03	0.08
	Cucurbita pepo	5	11.9	34.7	94.1	79.7	0.04	0.07
	Linum usitatiss.	5	12.7	25.5	48.4	65.6	0.15	0.07
	Nigella sativa	4	11.1	30.8	68.9	265.7	0.04	0.22
	Nigella sativa	4	10.6	14.2	62.3	90.0	0.06	0.85
	Mean		12.8	33.8	58.0	160.4	0.1	0.2
	SD		4.4	18.2	18.2	157.1	0.1	0.5

lower concentrations, in contrast, in fruits. The Zn levels recorded ranged from 20 to 95 mg kg^{-1}. General differences in Zn accumulation among leafy drugs, flower drugs or fruits and seeds could not be detected. Products higher in Zn were seeds from poppy, pumpkin and Nigella. Furthermore, there was a slight tendency toward higher Zn values in samples from Vienna in comparison to other regions.

Toxic heavy metals

The Cd contents showed some interesting tendencies: more than two-thirds of the samples had less than 0.1 mg kg^{-1} Cd. Few samples had more than 0.2 mg kg^{-1} Cd. The highest Cd concentrations were close to 1 mg kg^{-1}. Samples with higher Cd contents were restricted to specific plant species, mainly yarrow herb, St. John's wort and some samples of poppy seeds. Absinth herb and chamomile flowers also had higher Cd contents (Table 2). A specific region where plants had enhanced Cd concentrations could, however, not be found; the Cd level was more dependent on the plant species than on the origin. On the other hand, within the same region, as for instance in the Waldviertel, plants from a Cd accumulating species both high and low in Cd were observed.

The Pb levels were higher in leaf and herb samples than in fruit and seed drugs (Table 2, Table 3). The latter products showed, in general, Pb contents below 0.5 mg kg^{-1}, and leaf drugs between 0.4 and 1.2 mg kg^{-1}. Flowers had an intermediate position. The highest Pb values were around 2 mg kg^{-1} and were found in *Artemisia* species. Again, a correlation between Pb level and origin was not observable.

Some metallic elements, the micronutrients Cu, Mn, Zn and Fe, are constitutive elements with specific functions in the plant metabolism. They are also essential elements in the human or animal organism.

Table 3 Trace elements in medicinal plants from various Austrian regions (milligrams/kilogram dry weight)

		n	Cu Mean	M SD	Zn Mean	Fe SD	Cd Mean	Pb						
								SD	Mean	SD	Mean	SD	Mean	SD
Herbs and leaves	Vienna	20	9.4	5.3	58.1	24.7	43.3	21.6	142.1	78.9	0.05	0.04	1.1	0.6
	Weinviertel	4	8.2	1.4	57.6	11.1	26.1	13.7	296.7	168.4	0.10	0.09	0.7	0.4
	Waldviertel	10	12.9	8.3	71.8	40.2	37.7	12.8	409.6	360.1	0.28	0.41	1.0	1.3
	Linz	8	11.2	2.0	55.6	17.3	27.0	5.7	287.9	191.8	0.08	0.08	0.5	0.3
	Upper Austria	16	8.6	5.9	43.5	13.6	28.8	5.6	460.5	365.4	0.13	0.24	0.7	0.3
	Styria	6	9.0	2.4	48.4	13.7	36.8	5.3	572.7	367.2	0.04	0.04	0.7	0.3
Fruits and seeds	Weinviertel	15	13.1	2.7	34.0	10.4	50.9	18.3	63.9	37.5	0.06	0.07	0.2	0.2
	Waldviertel	9	15.4	3.4	42.3	17.1	50.6	18.6	135.3	173.2	0.10	0.13	0.1	0.1
	Linz	7	13.2	3.6	39.1	20.5	42.7	14.7	66.3	19.5	0.16	0.17	0.2	0.1
	Upper Austria	8	11.3	3.0	34.0	15.7	43.9	16.0	82.9	76.1	0.07	0.09	0.3	0.2
	Styria	7	11.2	4.1	37.3	30.8	42.7	17.3	89.8	42.1	0.05	0.03	0.2	0.1

Table 4 Cadmium content of selected medicinal plants showing accumulation of this heavy metal. Percentile values (90%) from Kabelitz [5]

Plant part	Cd ($mg.kg^{-1}$)
Birch leaves	0.7
St. John's wort herb	1.3
Mallow leaves	1.2
Willow bark	1.8
Yarrow herb	0.49
Chamomile flowers	0.42
Golden rod herb	0.44
Lin seeds	0.54
Dandelion herb	0.69
Absinthe herb	0.42
Watercress (Nasturtium off.) herb	0.46
Buckwheat herb	0.86

The plants were grown on ordinary agricultural soils and, in comparison to other crops or wild plants, their micronutrient contents were within the usual range [3, 8]. Leaves are generally higher in micronutrients than fruits and seeds. None of the species investigated showed a tendency to accumulate one of the micronutrients. As in most regions, the plants originated from several growing sites and due to the great heterogeneity in the species studied, it was not possible to point out general differences in trace elements based on the provenance of the drugs.

The presence of the toxic heavy metals, Cd and Pb, in plant samples is due to the circumstance that these elements are ubiquitous in trace amounts in the environment and might be taken up by or contaminate plants. To evaluate the contamination with these metals, reference values have been established for various foods in order to point out the usual unavoidable levels of these elements in the product. This way, the occurrence of elevated values may indicate unnatural contamination.

To set adequate reference values, a great number of samples of the considered product must be analysed and the values fixed so that 90–98% of the samples remain below them. In Germany and similarly in Austria, the reference values for leafy vegetables are fixed at 0.8 mg kg^{-1} Pb and 0.2 mg kg^{-1} Cd on a fresh weight basis [9]. For medicinal plants, the proposals for the reference values are 0.2 mg kg^{-1} for Cd and 5 mg kg^{-1} for Pb [4]. Contaminations with Hg, which, however, were not considered in this study, are not a problem [5].

Nevertheless, some plant species need higher reference values for Cd as they preferentially take up this heavy metal. Therefore, the enhanced reference values for Cd are 0.8 mg kg^{-1} for poppy seeds, 0.6 mg kg^{-1} for sunflower seeds and 0.3 mg kg^{-1} for linseed [9]; proposals are 0.3 mg kg^{-1} for yarrow herb and 0.5 mg kg^{-1} for St. John's wort [4]. Kabelitz [5] gathered Cd and Pb data from more than 12,000 samples originating from quality control analyses in various companies dealing with herbs and medicinal plants and indicated, aside from the minimum and maximum values, the 90th percentile as the critical heavy metal content of a product which should not be exceeded. A selection of plant products higher in Cd is presented in Table 4, showing that the proposed reference values often cannot be met. He stated that a former proposal for medicinal plants used in infusions with limits of 10 mg kg^{-1} Pb and 0.5 mg kg^{-1} Cd on dry weight proposed by Schilcher and Peters in 1990 [10] might also be practicable. The WHO prescribes limits for various medicinal plants of not more than 10 and 0.3 mg kg^{-1} Pb and Cd respectively in the final dosage form of the plant material [11].

When compared with previous surveys of trace elements in medicinal plants from Austria [6, 7], the micronutrients recorded in this study were in the same range. St. John's wort, chamomile and poppy seeds had already been pointed out as Cd rich. In this context it is interesting that St. John's wort showed in the present study higher Fe and Cd levels than in both previous studies. Pb levels are generally higher in leaves than in other aboveground plant parts, which suggests that Pb contaminations occur mainly through the atmosphere as the Pb uptake capacity of plants is low. Some leafy drugs had higher and others had lower Pb values in the present study, but there was no obvious rule, indicating that contaminations with Pb happen rather by chance. A trend in the evolution of Pb and Cd contamination could not be seen. On the one hand, the ending of the use of leaded fuels a decade ago alleviated the contamination situation, but Pb is still in the environment. As the Pb levels are already low, a further decrease cannot be seen or expected. On the other hand, estimations of an input of 12.3 g Pb and 1.6 g Cd per hectare and year through fertilization and deposition from the atmosphere in agrarian ecosystems suggest a slow increase in the contamination [12].

In Austria, soils in the Northern Limestone Alps, in Salzburg and the Tyrol, showed the highest natural Cd contents and high levels of Pb were recorded in soils from Salzburg and the Tyrol [13]. As the plant material investigated did not originate from these regions a low Cd supply from the soil can be assumed. However, the Cd availability to plants is also governed by parameters such as soil pH and the content on organic substances. In this context, some soils in Lower and Upper Austria, mainly on the granite and gneiss tablelands, may also present good Cd availability. The 90th percentile data reported by Kabelitz [5] are in all cases higher than the mean values reported in this study.

The issue of how to deal with Cd-enriching species encompasses different aspects: the Cd uptake capacity may vary greatly with the plant variety considered. This is the case in poppy [14], sunflower [15], linseed [16] and St. John's wort [16, 17, 18]. In the case of species which are widely used, efforts to select and breed lines low in Cd have just begun to be undertaken especially for sunflower [15] and recently for St. John's wort [18].

Another aspect which has to be considered is the plant part or product derived from the plant. The final product used must be free from or low in toxic heavy metals. For instance, during extraction, about 28–32% of Cd and up to 32% of Pb from the medicinal plant could be transferred to the extract [10]. In eastern Europe, some regions are affected by heavy industrial pollution where the soils are contaminated with hazardous elements [19, 20, 21]. Although the heavy metal contents in the plants grown there were high, the distilled essential oils were not contaminated. The conclusion was that cultivation on such soils is suitable for essential oil production.

A further aspect in the toxicological consideration is that teas, tea products and fresh herbs represent less than 1% of the total food intake [5] and that medicinal plants are used occasionally in the case of an illness.

Various precautions may be suitable for minimisation of the uptake of heavy metals by plants. Cd transfer from soil to plant may also be influenced by cutting time. In various herb species, the Cd content of later cuttings was higher than that of earlier ones [16, 22]. In the case of chamomile, sowing in spring yielded higher Cd contents in the flower heads than when the sowing was done in autumn [22]. In trials with various wheat cultivars, Wenzel et al. [23] observed that Cd availability for the plants was governed by Cd in the soil and was modified by sorption to organic matter and the concentration of Ca and Cl in the soil solution. To avoid Cd accumulation in wheat, they proposed a careful choice of the variety and adjusting the soil chemical conditions.

In conclusion, the heavy metal contamination level of medicinal plants in Austria is not critical. However, some products from species with a tendency toward Cd accumulation should be monitored in future. Particularly when they are consumed in higher amounts, they may considerably affect the Cd load of the consumer. This may be the case with poppy seeds in some traditional dishes or the dietetic use of linseeds over a longer period. The careful choice of the growing site and the management of soil conditions in order to avoid enhanced Cd input into the food chain is recommended when growing these two plant species.

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