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ORIGINAL PAPER

DETERMINATION OF MAGNESIUM, MANGANESE, COPPER AND ZINC IN INFUSIONS OF INFLORESCENCES AND LEAVES OF *SOLIDAGO CANADENSIS**

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Abstract

The *Solidago canadensis* (Canada goldenrod) species is an invasive plant in Europe and Asia. It grows mainly in anthropogenic environments, including those with a high content of heavy metals in soil. Goldenrod is also a honey plant, and its inflorescences and leaves are used in herbal medicine. The aim of the study was to determine the content of metals: Mg, Mn, Cu and Zn, in inflorescences and leaves of *S. canadensis* and in water extracts prepared from them. Material for the research was obtained in Poland. The content of the metals was determined by atomic absorption spectrometry. It was found that leaves of *S. canadensis* contained more Mg and Zn, but less Cu compared with inflorescences. The content of the metals in water extracts was proportional to their content in the raw material, except the content of Mn in leaves. Among the metals studied, the highest extraction of Cu and Mg was noted in infusions of leaves and inflorescences, while Zn achieved the highest extraction in infusions of inflorescences. Mn was extracted to the least extent both in leaves and inflorescence infusions. Infusions of goldenrod leaves provided similar amounts of Mg, Cu and Zn as tea infusions. Infusions of inflorescences provided more Cu and Zn than infusions of chamomile flowers. Considering the use of *S. canadensis* in phytotherapy, water extracts from goldenrod leaves and inflorescences had a negligible share of the analysed metals (on average about 1% or less) in the recommended daily allowances (RDAs).

Keywords: Canada goldenrod, water extracts, magnesium, trace metals, recommended daily allowances.

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INTRODUCTION

Solidago canadensis (L.) – Canada goldenrod, imported to Europe from North America in the 17th century, is one of the most common invasive plant species. Currently, it covers the whole Europe (Weber 2001). It also shows strong invasive potential in Asia (Mei et al. 2006, Weber et al. 2008). The species tolerates a wide range of habitat conditions (Jin et al. 2004, Huang et al. 2007). It enters both habitats with little influence of anthropogenic factors, and those highly transformed by humans (Korniak et al. 2012). The optimal habitat for *Solidago canadensis* is the fertile, moist clay soil (Chen et al. 2013). The plant grows better at higher soil nitrogen levels (Lu et al. 2005). Quite many studies concern the allelopathic effect of *S. canadensis* (Abhilasha et al. 2008, Yuan et al. 2013), as well as the impact of the plant on reducing the biodiversity of inhabited ecosystems (Gioria, Osborne 2014, Fenesi et al. 2015).

Goldenrod is a honey plant (Amtmann 2010, Janiszewska et al. 2012). The species representing the *Solidago* genus also show antibacterial and antimutagenic activity (Deepa, Ravichandiran 2010, Kołodziej et al. 2011). For this reason, they are used as raw material in herbal medicine as the so-called "blood-purifying" agents in gout, rheumatism, arthritis, eczema, and other skin disorders (Kołodziej et al. 2011). Herbal infusions with *Solidago* spp. are especially recommended for the treatment of infections and inflammations, to prevent the formation of renal calculi, and to help remove urinary gravel (Melzig 2004). They also have an antitussive effect (Sutovská et al. 2013). In phytotherapy, *Solidago* flowers and leaves are mainly used (Polish Pharmacopoeia 2008).

Although *Solidago* has been widely studied regarding its use in medicine, there is no literature on the content of metals in infusions of Canada goldenrod. It should be emphasised that *S. canadensis* tolerates a wide range of habitat conditions (Jin et al. 2004, Mei et al. 2006). Because the plant is tolerant to high contents of heavy metals in soil (Antonijevič et al. 2012), it is likely to be intoxicated with these elements (for example: Mn, Cu, Zn).

Hence, we decided to analyse the content of selected metals (Mg, Mn, Cu, Zn) in the leaves and flowers of *S. canadensis* and their infusions. The results of the metal content analysis in *S. canadensis* leaves were compared with commonly used infusions of green teas (Brzezicha-Cirocka et al. 2016) and black teas (Brzezicha-Cirocka et al. 2017) and those regarding the heavy metal content in *S. candensis* inflorescences were confronted with commonly used infusions of chamomile (*Matricaria chamomilla*) flowers (Mirosławski, Paukszto 2018). *Matricaria chamomilla*, like *S. canadensis*, can grow on soils contaminated with heavy metals (Grejtovský et al. 2006, Pieczka et al. 2019).

The following objectives were adopted in the research:

– determining the content of the following metals: Mg, Mn, Cu, and Zn in inflorescences and leaves of *Solidago canadensis,* as well as in infusions prepared from them;
– determining the correlation between metal content in raw material (inflorescences, leaves) and in infusions;
– estimating the daily dose of metal uptake with infusions from leaves and inflorescences of *S. canadensis* and comparing it with the recommended daily intake of RDAs (daily elemental intake from herbal tea calculated and compared to the recommended daily allowances RDAs).

MATERIAL AND METHODS

Samples

The material of the study was *S. canadensis* inflorescences and leaves collected during the period of intensive flowering of the plant (end of August 2017) in two locations in Poland: in Siedlce (52°13′ - 52°74′ N; 22°13′ - 22°25′ E) and Olkusz (50°15′ - 50°23′ N; 19°26′ - 19°36′ E). Siedlce is in the east of Poland, in an agricultural region characterised by naturally occurring heavy metals (Siebielec 2017). Olkusz is in southern Poland, in an industrial region, including zinc ore metallurgy. The soils in this region are classified as moderately and highly polluted with heavy metals, especially with zinc (Miśkowiec et al. 2015). A total of 50 plant samples were taken for testing (25 in each location).

Preparation of samples and analysis of elements

Twenty ramets were collected for analysis from each site. In the laboratory, the leaves and inflorescence were separated and dried at 60°C to constant weight. The dried samples were homogenised. One-gram samples were pre-mineralised in a muffle furnace at 420°C, followed by wet, multi-stage mineralisation in a high-pressure microwave mineraliser (UniClever TM, BM-1, Plazmatronica, Wrocław, Poland) using 65% HNO_3 and 30% H_2O_2, 3:2, v/v (Ostrowska et al. 1991). After mineralisation, the solution was transferred to 50 ml volumetric flasks.

Infusions were prepared in boiling distilled water (V = 100 ml) from 2 g weighed out amounts of the plant material. Herbs were brewed for 5 minutes. The infusions were filtered through a quality filter paper of medium filtration rate (POCh, Gliwice), and the residue on the filter was washed with hot water. The infusions were then evaporated to reduce their volume to 25 ml. The content of Mg, Mn, Cu, and Zn was determined in the solutions obtained after mineralisation and in infusions by atomic absorption spectrometry (AAS-30, Carl Zeiss Jena, Germany).

Polish Virginia tobacco leaves (INCP - PVTL - 6) prepared and certified at the Institute of Nuclear Chemistry and Technology (Warsaw, Poland) were used as the reference material in the study. The reference material was prepared following the same procedure as the leaves and inflorescences of *S. canadensis*. The results of the reference material analysis are given in Table 1.

Table 1

Comparison of Mg, Mn, Cu, and Zn concentrations in the studied and the reference material (INCP-PVTL-6)

Metal	The reference material (INCP-PVTL-6) (mg kg^{-1})	Found (mg kg^{-1})	Recovery (%)	CV (%)
Mg	2410±90	2290±95	95.02	4.15
Mn	136.0±5.00	139.4±4.88	102.5	3.50
Cu	5.120±0.20	4.970±0.28	97.07	5.60
Zn	43.60±1.40	42.16±2.07	96.70	4.90

Statistical analysis

The data distribution checked with the Shapiro-Wilk test was not normal. Therefore, a nonparametric analysis was used in the study. The Mann-Whitney test was applied to compare the percentage of metal extraction from goldenrod leaves and flowers. The Spearman's correlation coefficients were used as a measure of the strength of relationships between the concentration of metals in dry matter of inflorescences and leaves of *S. canadensis* and in infusions from the two plant parts.

RESULTS AND DISCUSSION

The average dry weight of the aerial part of the plant was 11.06 g (2.73-20.47 g), of which the dry weight of leaves was about 23.6% (14.8-30.8%), and of flowers – 21.6% (14.0-29, 6%) of the weight of the ramets. The dry weight of stems was about 54.8% (40.8-66.9%) of the aerial part of the plant.

The studied parts of *S. canadensis* differed in their content of the analysed metals (Table 2). The leaves of *S. canadensis* contained about twice as much magnesium and fourfold more zinc than inflorescences. However, goldenrod inflorescences contained about twice as much Cu as leaves. The content of Mn in goldenrod leaves and inflorescences was similar (Table 2). The content of Mg and other metals in goldenrod leaves was within the range of values measured in the leaves of green teas (Brzezicha-Cirocka et al. 2016) and black teas (Brzezicha-Cirocka et al. 2017). Compared with the levels in camomile flowers collected in Poland, the average Mn content in goldenrod inflorescences was lower (96.5-112 mg kg^{-1}), zinc content was comparable (42.6-45.76 mg kg^{-1}), while copper content was higher (8.5-8.61 mg kg^{-1}), e.g. Sembratowicz, Rusinek-Prystupa 2014, Pieczka et al. (2019).

The results of metal content measurements in the studied plant parts, presented in Table 2, are within a wide range. This stems from the fact that the test samples were taken from various sites, e.g. located on abandoned farmland, near communication routes, and in the case of Olkusz also in the vicinity of the Bolesław Mine and Smelter. These sites differed in soil pH, organic carbon content, and heavy metal content in the soil. Compared with Siedlce, the soil samples from Olkusz had a higher content of organic carbon, higher soil pH, and higher content of Mn, Cu, Zn (Bielecka, Królak 2019*a*,*b*). The accumulation of metals in plant tissues depends on the type of metal and soil properties, such as soil pH and organic matter content (Fijałkowski et al. 2012). For example, Mn at low soil pH (Siedlce) tends to translocate to aerial parts of the plant, while the high organic carbon content in the soil (Olkusz) limits the translocation of metals (Cu, Zn) from the roots of the plant upwards. A detailed comparison of the metal content in goldenrod leaves and flowers in both locations is presented in two other papers by Bielecka, Królak (2019*a*,*b*).

Table 2

The content of bioelements in dry parts of *Solidago canadensis* in mg kg^{-1} (mean ± SD, median, range) and the percentage of extraction

Metal	Part of plant	Mean±SD	Median	Range	Extraction (%)	The value of the Mann-Whitney test
Mg	I	2360.8±239.3	2321.7	1816.2-2833.5	39.22±7.131	Z=2.23; p<0.026
	L	4591.3 ± 635.7	4449.4	3488.4-6071.7	35.53±9.050	
Mn	I	62.99 ± 33.94	56.53	10.37-157.6	7.922 ± 4.714	Z=6.02; p<0.001
	L	98.66±101.8	44.53	13.62-425.8	19.03 ± 10.42	
Cu	I	12.03±9.391	10.97	7.183-75.66	48.10±13.74	Z=2.10; p=0.036
	L	6.377±2.099	5.706	3.541-11.66	55.05±14.41	
Zn	I	43.35±51.52	30.79	12.36-332.0	43.80±11.12	Z=7.19 p<0.001
	L	177.6±178.9	117.1	26.46-969.8	19.35±12.13	

I – inflorescences, L – leaves, n=100

The percentage of the extraction of metals from inflorescences was in the order Mn < Mg < Zn < Cu and in the range of 7.92 to 48.10%. The percentage of extracted metals from leaves varied from 19.03 to 55.05% in the following order: Mn < Zn < Mg < Cu (Table 2). The percentages of extraction from goldenrod leaves and green and black tea leaves were comparable for Mg. They were lower for Mn and Zn and higher for Cu

(Brzezicha-Cirocka et al. 2016, 2017). The percentage of extraction from goldenrod inflorescences for Zn and Cu was higher than from chamomile flowers, in which Sembratowicz and Rusinek-Prystupa (2014) noted, respectively, 19.95% and 15.28%. In contrast, the percentage of extraction from chamomile flowers for Mn (16.5%) was higher than from goldenrod inflorescences (Diaconu et al. 2012).

The tested metals were extracted into infusions in amounts proportional to the content of the same elements in the raw material, as shown by statistically significant values of the Spearman's correlation coefficients. Absence of a significant relationship was found only between the Mg content in leaves and infusions (Table 3).

Table 3

Spearman's correlation coefficients between the element content in the herbal raw material (inflorescences and leaves) and in infusions

Metal	Part of plant	Raw material	
		inflorescences	leaves
Mg	inflorescences - infusion	0.40**	
	leaves - infusion		ns
Mn	inflorescences - infusion	0.41**	
	leaves - infusion		0.88***
Cu	inflorescences - infusion	0.47***	
	leaves - infusion		0.46***
Zn	inflorescences -infusion	0.90***	
	leaves - infusion		0.60***

*** $p<0.001$, ** $p<0.01$, *ns* – not significant, $n=100$

The metals studied in this paper are essential for plant growth. These elements participate in metabolic processes, including reduction and oxidation in cells, photosynthesis, and activation of enzymatic reactions. In addition, Zn is necessary for membrane integrity and structural integrity of ribosomes (Kabata-Pendias, Pendias 1999). As in plants, these metals in the human body are involved in the activation of enzymes or are part of them and regulate metabolic processes. Zinc is also responsible for maintaining the stability of cell membranes and the body's immune defence (Jarosz 2012).

The recommended daily intake of these metals by adults, developed for the Polish population (aged over 31 years, given in mg/day/person) Jarosz (2012), is: for Mg – 320 (female) and 420 (male), for Zn – 8 (female) and 11 (male), and for Cu – 0.9 (irrespective of sex) – Jarosz (2012). In the case of manganese, according to *Regulation (EC) No. 1924/2006 on nutrition and health claims made on foods* (2006), the RDA is 2 mg.

The estimation of the percentage share of infusions from inflorescences

Table 4

Estimation of the percentage share of infusions from flowers and leaves of Canada goldenrod in RDA

Metal	RDA mg/day/person		Part of plants	Intake with infusions from 1 cup	%RDA	
	female	male			female	male
				(mg 2 g^{-1})		
Mg[a]	320	420	I	1.840 (1.050-2.760)	0.600 (0.342-0.909)	0.440 (0.110-0.704)
			L	3.221 (1.100-4.904)	1.000 (0.341-1.503)	0.760 (0.261-1.160)
				(μg 2 g^{-1})		
Mn[b]	2	2	I	9.143 (1.905-45.40)	0.460 (0.102-0.270)	0.460 (0.102-2.270)
			L	24.12 (7.604-84.21)	1.20 (0.381-4.200)	1.20 (0.381-4.200)
Cu[a]	0.9	0.9	I	10.46 (4.103-17.92)	1.160 (0.451-1.990)	1.160 (0.415-1.990)
			L	6.722 (4.501-12.30)	0.750 (0.505-1.372)	0.750 (0.505-1.372)
Zn[a]	8	11	I	34.22 (10.82-167.4)	0.420 (0.132-2.100)	0.310 (0.100-1.522)
			L	43.81 (20.62-166.2)	0.551 (0.263-2.113)	0.400 (0.190-1.522)

[a] – Jarosz (2012), [b] – *EC Regulation* (2006), I – inflorescences, L – leaves

and leaves of Canada goldenrod in the RDA was made assuming the consumption of one cup of infusion (200 ml) per day prepared from 2 g of flower or leaf (Table 4).

The research shows that the share of selected bioelements in infusions from inflorescences and leaves of *S. canadensis* compared with the recommended daily requirement of an adult is small (Table 4). A cup of infusion prepared from 2 g of inflorescences and leaves of the plant covers about 1% or less of the daily demand for bioelements. With the exception of manganese, these amounts are comparable to infusions from green teas (Brzezicha-Cirocka et al. 2016) and black teas (Brzezicha-Cirocka et al. 2017). Green teas cover the RDA for Mn in approximately 22% (male) and 28% (female) – Brzezicha-Cirocka et al. (2016), and black teas – in 11.7% (male) and 15% (female) – Brzezicha-Cirocka et al. (2017). For infusions prepared from leaves and inflorescences of *S. canadensis*, the max. RDA value for Mn is 4.2%.

It should be noted that even though the Zn content in *S. canadensis* leaves collected in the vicinity of Olkusz can be high (max. content 969.8 mg kg^{-1}), infusion prepared from goldenrod leaves covers about 2% RDA at the most.

CONCLUSIONS

1. The leaves and inflorescence of *Solidago canadensis* differ in their metal content; the content of metals in water extracts is proportional to their content in the herbal raw material.

2. Goldenrod leaf infusions provide similar amounts of Mg, Cu and Zn as green and black tea infusions.

3. Infusions of goldenrod inflorescences seem to be a better source of Cu than infusions from the leaves of this plant.

4. The research showed that infusions from leaves and inflorescences of *Solidago canadensis* cover the daily demand for selected bioelements to a small extent, even if the herbal raw material is obtained from sites with high metal content, e.g. zinc, in soil.

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