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NUTRITIONAL VALUE AND PHYTOTHERAPEUTIC RELEVANCE OF SOLIDAGINIS HERBA EXTRACTS OBTAINED BY DIFFERENT TECHNOLOGIES

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Solidago canadensis L., (Asteraceae) has been used in European phytotherapy as a urological and antiphlogistical remedy for centuries. The behaviour of dissolution of mineral elements into different tinctures and aqueous extracts obtained from Solidaginis herba was investigated in connection with their quercetin glycoside and organic acid amount. Commonly applied aqueous and alcoholic extracts were analysed for Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, Na, P, Pb, S, Ti, V and Zn content. The concentrations of the minerals examined were determined by inductively coupled plasma emission spectrometry (ICP-OES). Determination of the flavonoids in Solidaginis herba and extracts was carried out by a spectrophotometric method, as required by the German Pharmacopoea. For the study of the flavonoid composition of crude drug, the HPLC technique was applied.

To determine the relative nutrient contribution of these pharmaceuticals to the diet, data obtained were combined with flavonoid content particulars, then a comparison with U.S Recommended Dietary Allowances (RDA) was made. For evaluation of the phytotherapeutic relevance, K/Na ratio was also calculated.

It has been found that the pharmaceuticals examined are important sources of potassium, chromium, manganese, calcium, magnesium, phosphorus and lower sources of iron and zinc, assuming a daily intake of 1–2 l aqueous extracts as recommended for urological diseases. Flavonoid content of the different Solidaginis herba extracts ranged from 62.4 mg l^{-1} to 305.2 mg l^{-1}.

Keywords: *Solidago canadensis* L., phytopharmaceutical preparations, flavonoids, mineral elements

A number of natural foodstuffs, especially fruits, vegetables and several phyto-medicines used in adjuvant therapy contain substantial quantities of agents that have a considerable role in the prevention of various diseases. Such compounds include vitamins, trace elements and a variety of other agents (flavonoids and other polyphenols, etc.) with antioxidant properties. Enrichment of these components in our daily diet may offer a possibility for increasing health protection (HERTOG et al., 1993).

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Although herbs have already been used as foods or for medicinal purposes for centuries, we may be witness to a renewed interest in pharmaceuticals in recent years. Besides the discovery of new biologically-active molecules by the pharmaceutical industry, public interest turned to the adoption of crude plant-extracts for therapy (HOUGHTON, 1995; CRAIG, 1999).

Medicinal plants and phytopharmaceuticals have been used successfully in the therapy of the urinary tract with parallel administration of synthetic drugs. Regarding the long-term efficacy, tolerability and effectiveness of this medical treatment, the validation of traditional methods and careful quality control is needed (BALES et al., 1999).

Representatives of *Solidago* species have been used in European phytotherapy for centuries as a component of urological and antiphlogistical remedies. *Solidago canadensis* L. (Asteraceae) contains a wide range of active ingredients, such as flavonoids, saponins, phenol-carboxylic acids and several mineral elements, which are responsible for its characteristic anti-inflammatory, spasmolytic and diuretic properties (HÄNSEL et al., 1994; BADER, 1999).

The antioxidant and lipid peroxidation-inhibiting potential of these highly evolved plant components predominantly resides in the radical scavenging capacity and in the ability for the chelation of metals. Probable mechanisms and structure-activity relationships were put forward as an explanation for the role of flavonoids as food protectants (BORS et al., 1990; MOREL et al., 1994).

Since flavonoids are assumed to be responsible for these multivarious effects, it was interesting to study the accurate composition of extracts obtained by different technologies. Nevertheless, the characteristic effect of pharmaceuticals cannot be attributed only to the organic constituents, but also to the complex effect of organic and inorganic compounds.

The aim of our work was to study the dissolution behaviour of some organic ingredients and micronutritients in various commonly applied plant extracts, and to evaluate it regarding their nutritional value and phytotherapeutic relevance. For this purpose, we analysed the metal ion content and the quantity of phenolic constituents dissolved from Solidaginis herba.

1. Materials and methods

1.1. Reagents, standards and plant material

Methanolic solutions (200 μg ml^{-1}) of the following pure commercial substances were employed: chlorogenic acid, rutin, hyperoside, isoquercitrin, queritrin and quercetin from Sigma. Methanol and acetonitrile of HPLC grade, and all other chemicals of analytical-reagent grade were from Carlo Erba and REANAL, respectively. HPLC grade water was prepared by double distillation. All solvents were filtered through 0.5 μm (Millipore) membranes and degassed in an ultrasonic bath, while Solidaginis herba extracts were passed through C-18 cartridge before use.

Plant material was collected before full flowering on abandoned farmlands in the vicinity of Budapest (Hungary) and identified as *Solidago canadensis* L. (Asteraceae) in the Department of Pharmacognosy, Semmelweis University, where a herbarium specimen has also been deposited. Aerial parts were used for the extractions as Solidaginis herba according to the HUNGARIAN STANDARD (1986). The air-dried herba was extracted with a Soxhlet apparatus, than concentrated and purified for HPLC analysis and total flavonoid content measurement.

1.2. Preparation of samples

Air-dried herba was extracted with methanol in a Soxhlet apparatus for 6 h, then concentrated and purified for HPLC analysis and measurement of total flavonoid content. Decoction, infusion, maceration and different tinctures were used to obtain aqueous and alcoholic extracts from plant drugs.

For the preparation of extracts, solvents and drug were used in the ratio of 1:40 (2.5 g of drug and 100 ml of solvent).

– Decoctum solidaginis – the drug was boiled in bidistilled water for 5 min. The mixture was filtered while hot.
– Infusum solidaginis – the drug was infused with boiling water, and filtered after cooling.
– Maceratum solidaginis – the drug was macerated in bidistilled water at room temperature for a day, then filtered.
– Tinctura solidaginis – the drug was steeped in diluted (40% v/v, 70% v/v, 96% v/v) ethanol for six days, then filtered.

1.3. Determination of flavonoid content

Total flavonoid content of the dried Solidaginis herba and extracts were determined by spectrophotometry according to the instructions of the German Pharmacopoea (DAB, 1999), after acidic hydrolysis with HCl and complexation with $AlCl_3$. Glycosides and aglycones were jointly determined in aglycon form, and flavonoid content was calculated as hyperoside.

HPLC conditions. HPLC separation was performed with a Hypersil ODS (5 μm) reverse-phase C-18 column (250×4 mm) on Able Jasco HPLC system, consisting of a JASCO PU-980 gradient pump, JASCO PU-980 UV-VIS detector combined with a RHEODYNE 7725 (20 μl) injector and IBM-PC. Separation was achieved at ambient temperature with a flow rate of 1.0 ml min^{-1}. The gradient began with 14 v/v% acetonitrile (eluent A) in acetic acid 2.5 v/v% (eluent B), and was held at this concentration for the first 15 min. This was followed by a linear gradient to 35 v/v% acetonitrile over the next 30 min and then a sharp transition to 100 v/v% acetonitrile over the next 2 min. Data were collected at 360 nm. Peaks were identified by co-chromatography with standard addition and according to UV spectra and relative retention times.

1.4. Determination of element content

The element content of the samples was determined by inductively coupled plasma optical emission spectrometry (ICP-OES).

ICP-OES conditions. Type of instrument: Atom Scan 25 (Thermo Jarrell Ash), a sequential plasma emission spectrometer with a generator (2 kW, 27.12 MHz) exciting argon plasma to 8000–10000 °K. The optical system consists of a Czerny-Turner vacuum monochromator and two photoelectron multipliers. The detection limit was equal to the values given by Thermo Jarrell Ash. Standard solutions (prepared from Merck ICP standards) were in the matrix of the samples.

The samples (0.5 g of drug or 20 ml of evaporated extract) were digested with HNO_3 (10 ml). After digestion the samples were diluted to 25 ml, which were analysed for 23 elements (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, Na, P, Pb, S, Ti, V, Zn) in three repeated measurements. Three times 3 s integration time, blank substraction and background correction were applied.

1.5. Statistical analysis

Mean values and standard deviation (SD) were calculated from the results. One-way analysis of variance (ANOVA) was applied for comparison of the mean values.

2. Results and discussion

Flavonoids are natural compounds present in food items of plant origin, which considerably contribute to the antioxidant defence of the human body. The main polyphenol dietary sources are fruits, beverages (fruit juice, wine, herbal teas) and, to a lesser extent, vegetables. Total intake is approximately 1 g/day. Phenolic acids participate in about one-third and flavonoids in the remaining two-thirds of the total intake. The most abundant flavonoids in the diet are flavonols, anthocyanins and their oxidation products (SCALBERT & WILLIAMSON, 2000).

The maximum total flavonoid content of 1.5% (15 mg g^{-1}) was measured in *Solidago canadensis* samples collected before full flowering.

For the study of flavonoid composition and dissolution rates of the main compounds, the HPLC technique was applied. Chlorogenic acid, quercetin-3-O-beta-D-rutinoside (rutin), quercetin-3-O-beta-D-galactoside (hyperoside), quercetin-3-O-beta-D-glucoside (isoquercitrin) and quercetin-3-O-beta-D-rhamnoside (quercitrin) were confirmed (Fig. 1, Fig. 2) by retention times and their UV spectra. The main component of the samples was rutin, comprising up to 50% of the flavonoid content, which reflects the similarity of extracts composition.

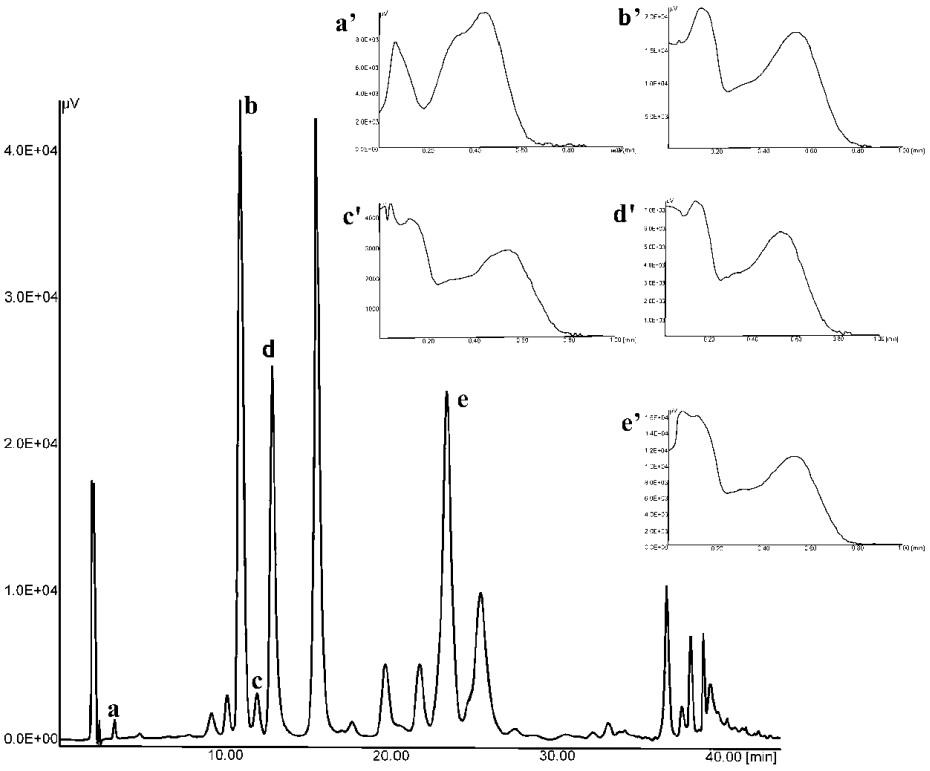

Fig. 1. HPLC analysis of Solidaginis herba. **a-e:** Analysed polyphenolic compounds in Solidaginis herba (a-chlorogenic acid, b-rutin, c-hyperoside, d-isoquercitrin, e-quercitrin); **a'-e':** UV spectra of analysed polyphenolic compounds at 360 nm

In most cases the extraction procedure eliminates the most inactive ingredients such as sugars and lipids. The aim of processing was to obtain an extract rich in active ingredients compared to the genuine plant (MEIER, 1991). Extraction technologies applied in this study yielded the dissolution of flavonoids from 14.9% (Maceratum solidaginis) to 79.2% (Tinctura solidaginis 70 v/v%-ethanol) (Fig. 3). The ingestion of one cup (200 ml) of Infusum solidaginis provides for 5–6% of our daily need of flavonoid. The ingestion of plenty of water is advisable in several cases of urological disorders, which ensures more than half of the recommended flavonoid needs.

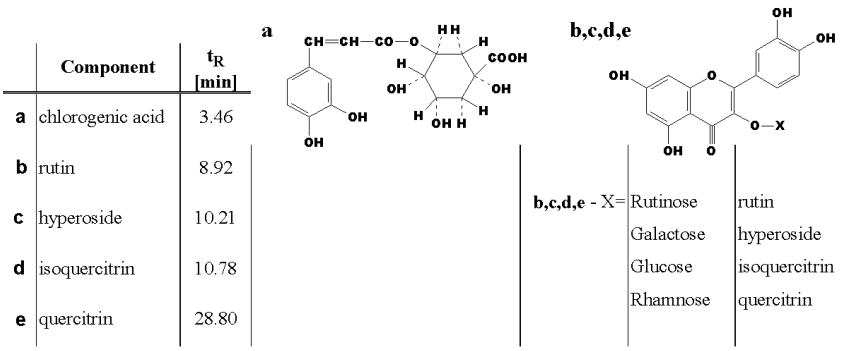

Fig. 2. Identified polyphenol components in Solidaginis herba

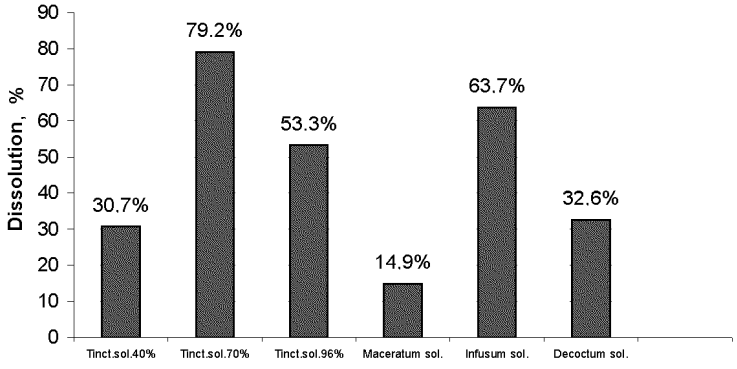

Fig. 3. Dissolution of flavonoids to different extracts prepared from Solidaginis herba

Nowadays, rational phytotherapy requires phytopharmaceuticals of high quality. To satisfy these requirements, the analysis of herbal medicines for trace elements is also very important.

Solidaginis herba investigated in this study – as other medicinal plants used in phytotherapy – is rich in mineral elements. Element composition of the various Solidaginis herba extracts is shown in Table 1.

Table 1. Parameters of the mineral element measurement, and element content of different extracts of Solidaginis herba (mg l^{-1})

Parameters of the element measurements				
Element	Wave-length	Plasma power (W)	Detection limit (mg l^{-1})	Upper conc. applied (mg l^{-1})
Al	396.152	1350	0.020	10
B	249.773	1350	0.002	10
Ba	455.403	1150	0.001	10
Ca	393.366	950	0.0005	10
Co	228.616	1550	0.003	2
Cr	267.716	1750	0.004	2
Cu	324.754	1150	0.002	2
Fe	259.940	1750	0.002	20
K	766.490	1150	0.100	100
Mg	279.553	1150	0.0005	50
Mn	257.610	1150	0.001	5
Na	589.592	1150	0.010	5
P	185.943	1350	0.060	35
S	180.731	1750	0.030	33.33
Ti	334.941	1350	0.002	2
Zn	206.200	1350	0.002	2

	Alcoholic extracts			Aqueous extracts		
Element	Tinctura solidaginis (40%-ethanol)	Tinctura solidaginis (70%-ethanol)	Tinctura solidaginis (96%-ethanol)	Maceratum solidaginis	Infusum solidaginis	Decoctum solidaginis
Al	0.521±0.008	0.240±0.003	0.065±0.003	0.883±0.003	0.888±0.011	0.553±0.016
B	0.320±0.003	0.398±0.023	0.11±0.0019	0.341±0.007	0.493±0.026	0.312±0.006
Ba	0.0279±0.0003	0.0162±0.0003	0.0064±0.0001	0.0206±0.0004	0.0246±0.0001	0.0485±0.0003
Ca	25.01±0.72	9.17±0.05	2.36±0.01	37.86±0.46	45.48±1.26	42.64±0.29
Co	0.0013±0.0012	<d.l.	<d.l.	0.0068±0.00121	0.0058±0.0009	0.0077±0.0002
Cr	0.0053±0.0012	0.0049±0.0006	0.0022±0.0003	0.0169±0.0172	0.0143±0.0002	0.0281±0.0014
Cu	0.173±0.014	0.166±0.011	0.029±0.001	0.117±0.004	0.153±0.002	0.154±0.001
Fe	0.208±0.006	0.093±0.001	0.112±0.002	0.414±0.014	0.193±0.007	0.171±0.002
K	228.1±1.5	280.5±3.0	76.95±0.81	335.8±3.6	284.3±2.8	277.2±0.6
Mg	13.01±0.48	23.89±0.19	2.34±0.04	24.31±0.44	25.34±0.22	24.64±0.07
Mn	0.179±0.001	0.0485±0.0012	0.0049±0.0002	0.332±0.005	0.348±0.004	0.431±0.010
Na	1.968±0.071	1.043±0.110	0.994±0.002	0.840±0.008	0.958±0.002	1.841±0.110
P	23.49±2.69	26.59±0.50	4.237±0.057	59.85±1.75	45.41±1.08	43.71±2.60
S	11.29±0.81	13.93±0.08	2.646±0.021	23.82±1.02	20.64±1.00	21.83±0.96
Ti	0.0017±0.0004	0.0015±0.0004	0.0005±0.0002	0.0098±0.0000	0.0033±0.0003	0.0003±0.0001
Zn	0.258±0.071	0.353±0.002	0.055±0.00151	0.1555±0.0045	0.1710±0.0032	0.1732±0.0261

<d.l.: below detection limit

One-way analysis of variance (ANOVA) was used for statistical comparison of results. Element concentrations showed significant difference in various extracts of Solidaginis herba. According to the ANOVA test – except for cobalt ($P = 0.2148$) and chromium ($P = 0.0032$) – the difference between the samples was highly significant ($P<0.001$).

Owing to the contamination of the environment by several intoxicants, toxic elements are often accumulated in plants, therefore, investigation of the heavy metal content in commonly available drugs is essential. In our experiment no toxic elements (As, Cd, Hg, Pb) were measured in higher concentration than the detection limit. The concentration of lithium, molybdenum and vanadium were also below the detection limit, therefore, these elements are not indicated in Table 1.

According to expectations, element dissolution in aqueous extracts exceeded that in tinctures except for copper (12.8–72.9%), sodium (32.0–65.2%) and zinc (9.2–57.2%), while the concentrations of boron (12.4–43.0%) and potassium (17.9–65.9%) approached the values measured in aqueous extracts.

Tinctura solidaginis (96% v/v-ethanol) was the worst source of mineral elements, none of them were measured in prominent concentration. The highest amount of copper (0.166 mg l^{-1}) and zinc (0.353 mg l^{-1}) was found in Tinctura solidaginis (70% v/v-ethanol), while Tinctura solidaginis (40% v/v-ethanol) contained more sodium (1.968 mg l^{-1}) than other extracts.

The highest quantity of aluminium (0.888 mg l^{-1}), boron (0.493 mg l^{-1}), calcium (45.48 mg l^{-1}), magnesium (25.34 mg l^{-1}) was found in Infusum solidaginis, while Decoctum solidaginis contained more barium (0.0485 mg l^{-1}), cobalt (0.0077 mg l^{-1}), chromium (0.0281 mg l^{-1}) and manganese (0.431 mg l^{-1}) than other extracts. Maceratum solidaginis was the best source for the remaining minerals.

Aqueous and alcoholic extracts are frequently used pharmaceuticals in medical practice, even for the prevention of diseases. Due to the high alcohol content of tinctures, different application rules are accepted in therapy. Regarding this, tinctures may be used in smaller doses mainly for prevention and for detoxication of the human body, while aqueous extracts are applied in higher doses as phytotherapeutics. Accurate selection of the pharmaceutical technology required for curing various health problems might result in high amounts of mineral element intake.

Nutritive value of the samples studied was characterised by comparing nutrient concentrations of different extracts and U.S. Recommended Dietary Allowances (RDA, 1989). The differences in the mode of application (tinctures: 15 ml/day, teas: 1–2 l/day divided in 3–4 parts) must also be taken into consideration. Those sources were deemed most satisfactory, which contained at least 10% of the RDA.

As follows from the above, aqueous extracts are very good sources of chromium (RDA 50–200 μg/day/adult), potassium (RDA 2000–3500 mg/day/adult) and manganese (RDA 2–5 mg/day/adult). In the case of calcium (RDA 800 mg/day/adult), copper (RDA 1.5–3 mg/day/adult), magnesium (RDA 350 mg/day/adult) and phosphorus (RDA 800 mg/day/adult) recommended consumption of herbal teas can ensure the daily needs. Although the concentration of the other elements measured in

aqueous extracts were overmatched, even such small amounts of iron (RDA 10 mg/day/adult) and zinc (RDA 15 mg/day/adult) may ensure the supply of the recommended doses (1–2%).

The diuretic effect of potassium has been known for a long time. The correlation between the diuretic effect and potassium content of pharmaceutical preparations has already been investigated (KANIAS et al., 1979; ABED & BENMERABET, 1981).

Due to the traditional application of *Solidago* species in diuretic preparations, it is worth to investigate potassium and sodium content of different Solidaginis herba extracts (CHODERA et al., 1991).

Our earlier study on official vegetable drugs with proved diuretic action verified that the potassium-sodium ratios in decoctions were above 150. The same relative number for traditionally used diuretic drugs ranged only between 100 and 150 (SZENTMIHÁLYI et al., 1998).

The potassium-sodium ratio was calculated according to the measured element concentrations depicted in Table 1. The highest ratio was observed in Maceratum solidaginis (360) and Infusum solidaginis (299), while a high value of Tinctura solidaginis (70%-ethanol) (267) combined with highest flavonoid content could be observed (Fig. 4).

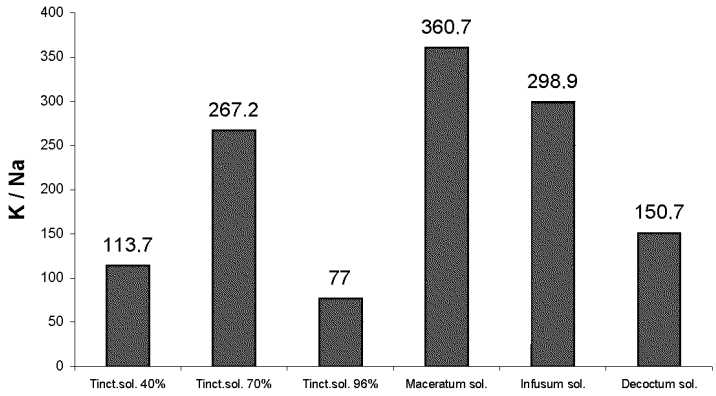

Fig. 4. K/Na ratio in different Solidaginis herba extracts

3. Conclusions

Plants used in traditional medicine have an important role in the preservation of health and in the introduction of new treatments. The pharmacological effect and therapeutical relevance of herbal drugs and extracts are presumably related to the complex effect of organic and inorganic compounds. The application of pharmaceuticals prepared from medicinal plants is usually safe and free from adverse effects, however, careful investigation and quality control is essential.

It may be concluded that high amount of polyphenol constituents, the presence of various flavonoid glycosides and favourable distribution of mineral elements in various extracts may greatly contribute to the successful application of phytopharmaceutics. Flavonoids occurring in glycosidic form in Solidaginis herba extracts have a favourable effect on the hydrophilous character of molecules.

The highest amount of active ingredients measured in alcoholic extracts (Tinctura solidaginis (40% v/v-, 70% v/v-, 96% v/v-ethanol): 12.5 mg 100 ml^{-1}, 30.52 mg 100 ml^{-1}, 21.19 mg 100 ml^{-1}, respectively), support the application of these tinctures in the form of pharmaceuticals. Owing to the hydrophilous character of flavonoid glycosides, the richness of aqueous extracts' in mineral elements and advantageous dissolution rate of potassium and sodium, aqueous extraction can be recommended as the most beneficial method for direct use in phytotherapy. Conditions for the preparation of infusion simultaneously resulted in equally high rate dissolution of flavonoids (26.76 mg 100 ml^{-1}), mineral elements (30–70%) and a beneficial ratio of potassium-sodium in the extract.

Based on the above results, Infusum solidaginis was formed to be the most beneficial phytopharmaceutical preparation studied in our experiments.

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