Assessment of phytotherapeutic mixtures as a potential route of absorption of some metal compounds

Jolanta Kowol¹, Jerzy Kwapuliński¹, Ewa Nogaj¹, Magdalena Babuśka-Roczniak², Barbara Brodziak-Dopierała¹, Wojciech Roczniak², Anna Szady³, Sylwia Cypcer⁴

¹ School of Pharmacy with Division of Laboratory Medicine, Department of Toxicology, Medical University of Silesia, Sosnowiec, Poland

² Jan Grodek Higher Vocational State School, Medical Institute, Sanok, Poland

³ Central Hospital, Tychy, Poland

⁴ Pharmacy Academic, Sosnowiec, Poland

Kowol J, Kwapuliński J, Nogaj E, Babuśka-Roczniak M, Brodziak-Dopierała B, Roczniak W, Szady A, Cypcer S. Assessment of phytotherapeutic mixtures as a potential route of absorption of some metal compounds. J Pre-Clin Clin Res. 2016; 10(2): 115–121. doi: 10.5604/18982395.1227568

Abstract

Introduction. Nowadays, herbalism seems to arouse much interest. Approximately 80% of the world's population are willingly take plant-derived drugs. It should be remembered that both herbs and their products may have not only beneficial but also harmful components. Moreover, medicinal plants frequently exhibit the capacity to selectively accumulate toxic elements.

Objective. To assess the potential burden of phytotherapy with medicinal plants used in chosen herbal mixtures.

Materials and method. A total of 76 species of medicinal plants from all over Poland and contained in herbal mixtures applied in selected diseases were investigated with respect to the content of heavy metals. Argon plasma atomic emission spectrometry (AES) was applied, using an ICP-ULTIMA spectrometer.

Results. The study plants showed various cumulative properties, depending on plant species and the effect of environmental purity on the metal content in raw materials. Daily doses of metal absorption due to phytotherapy in various diseases were determined. The levels of Ni, Co and Cr accumulation were similar to that of Pb.

Conclusions. Herbal therapies using the medicinal plants studied did not exceed daily norms of the investigated metals. However, as these metals may also occur in other food products, a prolonged use of plant-derived drugs can be an additional burden, especially in the case of renal failure.

Key words

mixture of medicinal plants, heavy metals, daily intake, anaemia, haemorrhagic diathesis, selected diseases, calculus of urinary tract, chronic bronchial asthma

INTRODUCTION

Herbal medicine (phytotherapy) had been widely used until the second half of the 19th century but then decreased due the development of the pharmaceutical industry. When the synthesis of new curative compounds became possible, synthetic medicines almost completely replaced natural drugs [1, 2, 3]. Only in the 20th century it was observed that synthetic drugs could produce some side-effects, such as drug dependence, or cause other diseases when chronically applied [1, 3]. Therefore, there began the re-emergence of natural drugs [1–7].

At present, phytotherapy is experiencing a revival [1, 2, 3, 8, 9, 10]. Estimations show that about 80% of the world population use nature-derived drugs, particularly extracts from plants or their components [2]. Phytotherapy thrives in Europe, as evidenced by the recognition of herbal preparations as medicinal agents by the European Economic Community [2, 8, 9]. The development of phytotherapy is also certified by associations of phytotherapist physicians in Western Europe, new scientific journals and specialized herbal pharmacopoeia, such as the British Herbal Pharmacopeia [2, 8].

Author for correspondence: Wojciech Roczniak, Jan Grodek Higher Vocational State School, Medical Institute, Mickiewicza 21, 38-500 Sanok, Poland E-mail: wojciech_roczniak@interia.pl

Received: 30 June 2015; accepted: 17 November 2016

Herbs and herbal preparations can also contain substances with undesirable effects [11, 12]. Toxic elements in plants capable of selective accumulation can cause damage to the genetic material of the plant. This leads to a decrease in the volume of the active substance, and at the same time, causes a reduction in the therapeutic value. It may also result in the accumulation of elements in the human body which eventually disturb physiological processes [9]. The progression of environmental degradation leads to abnormal quantities of toxic metals in plants [1–7, 11, 13–17]. In natural conditions, the element concentration varies a great deal, depending on plant species, variations, their parts or developmental stage. Trace elements are the fewest in fruits and grains, whereas roots and succulent leaves of vegetables or legumes show the highest level.

Due to herbology development and environmental pollution, the content of elements, especially those which are toxic, should be monitored in medicinal plants [3, 8, 10].

Standards adopted by the Ministry of Health and Social Welfare in 2003 determined the permissible exposure limit for adults of lead in herbal and fruit teas at 1 mg/kg, cadmium 0.1 mg/kg, mercury 0.2,mg/kg and arsenic 0.3mg/kg. The limits for children are even more restrictive: 0.1 mg/kg for lead, 0.01 mg/kg for cadmium, 0.005 mg/kg for mercury and 0.1 mg/kg for arsenic.

Studies on 76 medicinal plant species from all over Poland showed differentiation of cumulative properties of various plant species, and the impact of unpolluted environment on metal content in raw material. The highest lead content $(12\mu g/g)$ was found in Melisa officinalis L., Equisetum arvense L., Marrubim vulgare L. and Taraxacum officinale Web. The selective capability of cadmium accumulation $(3.3\mu g/g)$ was shown in Thymus pulegioides L., Helichrysum arenarium L. and Acorus calamus L. A lower content $(2.5\mu g/g)$ was noted, among others, in Asperula tinctoria L., Althaea officinalis L., Artemisia vulgaris L., and Plantago major. The smallest quantities of cuprum were found in Tussilago farfara L. (80.32 $\mu g/g$), Asperula tinctoria L. (32.00 $\mu g/g$), Chelidonium maius L. and Crataegus oxyacantha L. (15.7 $\mu g/g$).

The levels of Ni, Co and Cr accumulation were comparable to that of Pb. The current study shows that the concentration of heavy metals in the majority of species exceeded the level accepted as 'normal' [4–7, 16–20]. Medicinal plants can be used in various forms: as homogenous herbs aimed to treat specific disorders, herbal mixtures, syrups, teas and herbal and fruit teas or spices.

OBJECTIVE

The aim of the current study was to assess daily doses of chosen metals absorbed via phytotherapy on the basis of the established content of heavy metals in 20 plant species included in herbal mixtures applied in the course of: anaemia, haemorrhagic diathesis, bronchial asthma, urinary tract calculi and chronic cholelithiasis. A potential burden of the human body was determined in the course of phytotherapy. The scope of the presented information seems to exhaust the issue. The literature was carefully selected, taking into account only unpolluted sites of plant origin, the long duration of earlier research into metal content in the respective morphological parts, and the possibility to compare the results with Polish and foreign centres from the aspect of quality assessment.

MATERIALS AND METHOD

Medicinal plants. Twenty species, 9 pieces of each, were collected simultaneously from sites approximately 20 m distant from a road, a forest clearing and meadows at least 500 m distant from the road. The plants were collected in 2 vegetation periods (2013 and 2014), after the florescence time. The following species were investigated:

- Wild rose Rosa canina
- Bramble Rubus plicatus
- Mullein runner Verbascum thapsiforme
- Black elder Sambucus nigra
- Bilberry Vaccinium myrtillus
- Stinging nettle Urtica dioica
- · Common yarrow Achillea millefolium
- Plantain Plantago major
- Plantain lanceolata Plantago lanceolata
- Dandelion Taraxacum officinale
- Coltsfoot Tussilago farfara
- Cowslip Primula officinalis
- Wormwood absinth- Artemisia absinnthium
- Lawn daisy Bellis perennis

- Field horsetail Equisetum arvense
- St. John's Wort Hypericum perforatum
- Common mantle Alchemilla vulgaris
- Fragrant violet Viola odorata
- Cinquefoil goose Potentilla anserine
- Glechoma hederacea Glechoma hederacea

Each plant was washed and then divided into parts: roots, stems, flower shoots, leaves, flowers, fruit or roots and allspice. The air dried material was minced in a grinder with plastic blades (each part separately). Samples of about 1.0g each were treated with 5 cm² spectrally pure (65%) HNO₃ (V) (MERCK), then left for 24h and twice mineralized with 5 cm² HNO₃ (V) wet in a sand bath. The obtained mineralisate was quantitatively transferred to volumetric flasks of 25 cm³ capacity, and made up to the mark with redistilled water.

The content of metals in the tested samples was determined by Atomic Emission Spectrometry (AES) in argon plasma, using a JCP-ULTIMA spectrometer (Jobin Yvon Horiba) at a 10/15µm gap in a certified laboratory of the 'Laziska' power plant in Silesia, Poland. The range of the determined metals was as follows:

- Cd 0.006-0.5mg/l
- Cr 0.007-1.0mg/l
- Cu 0.009–1.0mg/l
- Mn 0.006-1.0mg/l
- Ni 0.008-1.0mg/l
- Pb 0.02-1.0mg/l
- Zn 0.017-5.0mg/l
- Fe no data
- Na 0.3-5000mg/l
- Ca no data
- Mg no data.

The recovery effectiveness ranged from 94% – Cd, Cr, 96% – Cu, Mn, Ni, Pb, 99% – Zn, Fe, 107% for Na, ca, Mg, depending on the element. The accuracy of determination of the respective elements was 0.01 μ g/g, with the relative error varying between the elements (0.9% for Cd to 3.8% for Cr).

The accuracy of the assays was monitored with the standard addition method by the National Methodology Institute as well as using a certified reference material with a comparable content of the tested metal in relation to the analyzed material for plants -CTA-VTL-2 (ICH-TJ). Differences of 2–4% were noted. In addition, intercalibration (6 reference samples of the tested plant) was conducted in the Department of Environmental Monitoring (in Katowice). Results of the analyses showed 3–5% differences, compared to the intercalibration results. The validation procedure was also applied in cooperation with the Department of Organic Chemistry, Technical University of Gliwice in Silesia.

RESULTS

The possible impact of medicinal plants on selected diseases was considered with regard to some of the most commonly used herbal mixtures. Daily Intake was estimated by application of the amount of raw material, and according the Polish Farmakopea heavy metals contents equals the geometric mean. Complete results of statistical analysis are included in the research documentation.. Jolanta Kowol, Jerzy Kwapuliński, Ewa Nogaj, Magdalena Babuśka-Roczniak, Barbara Brodziak-Dopierała, Wojciech Roczniak. Assessment of phytotherapeutic...

Anaemia. Anemia is a disease in which the haematocrit ratio decreases, and the erythrocyte count and haemoglobin per unit volume of blood are below the norm. There are a few types of anaemia which can be caused by:

- severe blood loss;
- deficiency or disturbed absorption of substances indispensable in the production of erythrocytes (i.e. Fe, vitamin B₁₂ and others);
- accelerated breakdown of red blood cells (haemolytic anaemia);
- failure to restore stem cells in the bone marrow (aplastic anemia).

The treatment should supplement the deficiencies of haematopoietic factors and eliminate the causes of the disease. Phytotherapy can be applied as an adjuvant therapy in the form of mixture I or II.

Mixture I (Tab. 1). Nettle leaf – Folium Urticae 50.0 g Nettle root – Radix Urticae 30.0 g Yarrow – Herba Millefolli 30.0 g Herb Centaurium erythraea – Herba Centaurii 20.0 g Herb wormwood – Herba Absinthii – 10.0 g

Table 1. Exposure during consumption of herbal mixture I in anaemia

Preparation and application: pour a glass of boiling water over one tablespoon of herbs, leave for half-an-hour, filter, and drink twice a day [21]. Table 1 presents maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does not exceed the necessary daily intake. The limit for the concentration of Cd in dry plant materials was not exceeded only in nettle leaf, whereas in the remaining components of the mixture, the Cd content was above the set limit. Pb concentration, however, was not exceeded in any case.

Mixture II (Tab. 2). Nettle leaf – Folium Urticae 50.0g Rose hip – Fructus Rosae 50.0g Blackcurrent leaf – Folium Ribis nigri 50.0g Parsley leaf – Folium Petroselini 20.0 g Dandelion root – Radix Taraxaci 20.0 g

Preparation and application: pour a glass of boiling water over 2 tablespoons of herbs, simmer covered for 10 min. and filter; drink twice a day before a meal [21].

| Dianat | Amount of raw | | | | | Metal conte | ent [µg/g of | f dry weight] | | | | |
|--|---------------|------|------|--------|--------|-------------|--------------|---------------|--------|---------|----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.23 | 0.26 | 6.93 | 28.7 | 0.58 | 1.4 | 37.07 | 181.05 | 119.58 | 38075.2 | 5022.48 |
| Urticae folium Nettle leaf | 50.0 | 11.5 | 13 | 346.5 | 1435 | 29 | 70 | 1853.5 | 9052.5 | 5979 | 1903760 | 251124 |
| | Daily | 1.24 | 1.4 | 27.28 | 154.41 | 3.12 | 7.53 | 199.44 | 974.05 | 643.34 | 204844.6 | 27020.94 |
| | 1.0 | 2.53 | 0.67 | 8 | 35.85 | 1.79 | 2.5 | 58.51 | 253.46 | 79.67 | 5228.56 | 2041.63 |
| Urticae radix Nettle root | 30.0 | 75.9 | 20.1 | 240 | 1075.5 | 53.7 | 75 | 1755.3 | 7603.8 | 2390.1 | 156856.8 | 61248.9 |
| | Daily | 8.12 | 2.15 | 25.68 | 115.08 | 5.75 | 8.03 | 187.82 | 813.61 | 255.74 | 16783.68 | 6553.63 |
| | 1.0 | 2.57 | 0.61 | 9.08 | 33.88 | 2.1 | 5.3 | 47.04 | 113.58 | 363.84 | 8977.62 | 2168.03 |
| Millefolii herba Yarrow | 30.0 | 77.1 | 18.3 | 272.4 | 1016.4 | 63 | 159 | 1411.2 | 3407.4 | 10915.2 | 269328.6 | 65040.9 |
| lanow | Daily | 8.2 | 2 | 29.1 | 108.8 | 6.7 | 17 | 151 | 364.6 | 1167.9 | 28818.2 | 6959.4 |
| | 1.0 | 1.13 | 0.62 | 11.07 | 31.68 | 1.4 | 1.25 | 26.44 | 130.1 | 128.27 | 8691.01 | 2879.58 |
| Absinthii herba Wormwood | 10.0 | 11.3 | 6.2 | 110.7 | 316.8 | 14 | 12.5 | 264.4 | 1301 | 1282.7 | 86910.1 | 28795.8 |
| womwood | Daily | 1.2 | 0.7 | 11.8 | 33.9 | 1.5 | 1.3 | 28.3 | 139.2 | 137.2 | 9299.4 | 3081.2 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [µg] | | 17.7 | 6.25 | 103.86 | 412.19 | 17.07 | 33.8 | 566.56 | 2291 | 2204 | 259746 | 43615 |

Table 2. Exposure during consumption of herbal mixture II in anaemia

| Plant | Amount of raw | | | | | Metal conte | ent [µg/g of | dry weight] | | | | |
|--|---------------|-------|------|-------|-------|-------------|--------------|-------------|--------|---------|----------|---------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.23 | 0.26 | 6.93 | 28.7 | 0.58 | 1.4 | 37.07 | 181.05 | 119.58 | 38075.2 | 5022.48 |
| <i>Urticae folium</i> Nettle leaf | 50.0 | 11.5 | 13 | 346.5 | 1435 | 29 | 70 | 1853.5 | 9052.5 | 5979 | 1903760 | 251124 |
| | Daily | 1.8 | 2.1 | 55.4 | 229.4 | 4.6 | 11.2 | 296.6 | 1448.4 | 956.6 | 304601.6 | 40179.8 |
| <i>Rosae fructus</i> Dog rose | 1.0 | 0.084 | 0.05 | 3.14 | 72 | 0.94 | 1.29 | 8.14 | 26.2 | 36.89 | 9845.83 | 1628.25 |
| | 50.0 | 4.2 | 2.6 | 157 | 3600 | 47 | 64.5 | 407 | 1310 | 1844.5 | 492291.5 | 81412.5 |
| Dog lose | Daily | 0.7 | 0.4 | 25.1 | 576 | 7.5 | 10.3 | 65.1 | 209.6 | 295.1 | 78766.6 | 13026 |
| | 1.0 | 3.21 | 0.69 | 15.57 | 25.14 | 1.089 | 2.27 | 44.45 | 177.55 | 1997.78 | 5318.59 | 1296.57 |
| <i>Taraxaci radix</i> Dandelion root | 20.0 | 64.2 | 13.8 | 311.4 | 502.8 | 21.78 | 45.4 | 889 | 3551 | 39955.6 | 106371.8 | 25931.4 |
| Danuelloli Tool | Daily | 10.1 | 2.2 | 49.2 | 79.4 | 3.4 | 7.2 | 140.5 | 261.1 | 6313 | 16806.7 | 4097.2 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [μg] | | 12.6 | 4.7 | 129.7 | 885 | 15.5 | 28.7 | 502.2 | 2219 | 7565 | 400175 | 57303 |

Table 2 shows maximal metal quantities consumed by a patient during phytotherapy, assuming that 1 tablespoon of the prepared mixture is 7.5g [22]. The limit of the concentration of Cd in dry plant materials was exceeded only in dandelion root, whereas the limit content of Pb was not exceeded in any case.

118

Haemorrhagic diathesis. This is a state of massive haemorrhage which can be divided into 3 types, depending on causal factors:

- plasma haemorrhagic diathesis (lack of blood plasma agents indispensable for proper blood coagulation);
- thrombocytopenic haemorrhagic diathesis (lack of blood clotting agents of lamellar origin);
- vascular haemorrhagic diathesis (normal coagulation, but fine petechiae are found and bruises occur after applying pressure).

The applied herbs are used to seal blood vessels and decrease effusion.

Mixture III (Tab. 3). Rosehip – Fructus Rosae 30.0 g Water pepper smartweed – Herba Polygoni hydropiperis 40.0 g Nettle herb – Herba Urticae 50.0 g Yarrow – Herba Millefolii 50.0 g

Shepherd's purse – Herba Bursae pastoris – 70.0 g Horsetail – Herba Eqiseti 70.0 g Chestnut bark – Cortex Hippocastani 50.0 g

Preparation and application: pour a glass of boiling water over one tablespoon of the mixture, simmer covered for 20 min. and filter; drink three times a day after a meal [21].

Table 3 presents the maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does not exceed the daily human need for these elements. The limit concentration of Cd established for dry plant materials was exceeded in the case of yerba and horsetail herb. Pb content, however, did not exceed the approved limits. Urinary tract calculi. The disease involves the accumulation of deposits of stones, varying in shape, size, chemical composition and hardness, in different sections of the urinary tract. According to their location, the following types of the disorder can be distinguished: nephrolithiasis, urolithiasis and kidney bullous. In the case of urolithiasis, herbs are extremely useful, both in prevention and treatment. Herbal mixtures usually have a diuretic, saluretic, diastolic and sometimes anti-inflammatory and antiseptic effects.

Mixture IV (Tab. 4). Horsetail – Herba Equiseti 40.0 g Thyme – Herba Asperulae 40.0 g Burdock root – Radix Bardanae 40.0 g Elder flower – Flos Sambuci 40.0g Rosehip – Fructus Rosae 40.0 g

Preparation and application: pour a glass of boiling water over one tablespoon of the mixture, cook covered for 3 min. and filter; drink three times a day [21]. Table 4 presents maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does not exceed the daily human need for these elements. The limit of concentration of Cd established for dry plant materials was exceeded in the case of horsetail herb. Pb content, however, did not exceed the approved limits.

Mixture V (Tab. 5). Horsetail herb – Herba Equiseti 25.0 g Herb thyme – Herba Asperulae 40.0 g Burdock root – Radix Bardanae 40.0 g Elder flower – Flos Sambuci 40.0g Fruit of elderberry – Fructus Sambuci 10.1 g Nettle leaf – Folium Urticae 10.0 g

Preparation and application: pour 2 glasses of warm water over 2 tablespoons of the mixture, then cook for 3 min. Leave it for 10 min., filter in to a vaccum flask; rrink half a glass 3 times a day [21]. Table 5 presents the maximal

Table 3. Exposure during consumption of herbal mixture II in haemorrhagic diathesis

| Diana | Amount of raw | | | | | Metal con | tent [µg/g o | f dry weight |] | | | |
|--|----------------------|-------|-------|-------|--------|-----------|--------------|--------------|--------|--------|-----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.084 | 0.052 | 3.14 | 72 | 0.94 | 1.29 | 8.14 | 26.2 | 36.89 | 9845.83 | 1628.25 |
| Rosae fructus Dog rose | 30.0 | 2.52 | 1.56 | 94.2 | 2160 | 28.2 | 38.7 | 244.2 | 786 | 1106.7 | 295374.9 | 48847.5 |
| Dogrose | Dzienna | 0.2 | 0.1 | 5.9 | 135 | 1.8 | 2.4 | 15.3 | 49.1 | 69.2 | 18460.9 | 3053 |
| | 1.0 | 0.17 | 0.18 | 7.28 | 41.07 | 0.6 | 1.19 | 31.57 | 119.11 | 134.16 | 24190.32 | 4302.85 |
| <i>Nettle herba</i> Nettle | 50.0 | 8.5 | 8.8 | 364 | 2053.7 | 30 | 59.3 | 1578.5 | 5955.7 | 6708 | 1209516.2 | 215142.7 |
| Nettie | Dzienna | 0.5 | 0.6 | 22.8 | 128.4 | 1.9 | 3.7 | 98.7 | 372.2 | 419.3 | 75594.8 | 13446.4 |
| | 1.0 | 2.45 | 0.19 | 7.14 | 35.44 | 1.4 | 5.51 | 30.7 | 65.39 | 58.52 | 8537.26 | 1682.51 |
| Millefolii herba Yarrow | 50.0 | 122.5 | 9.5 | 357 | 1772 | 70 | 275.5 | 1535 | 3269.5 | 2926 | 426863 | 84125.5 |
| Turiow | Dzienna | 7.7 | 0.6 | 22.3 | 110.8 | 4.4 | 17.2 | 95.9 | 204.3 | 182.9 | 26678.9 | 5257.8 |
| | 1.0 | 0.88 | 0.45 | 5.89 | 39.06 | 0.73 | 4.99 | 44.34 | 63.41 | 63.18 | 24561.2 | 5537.17 |
| <i>Equiseti herba</i> Equisetum steam | 70.0 | 61.6 | 31.5 | 412.3 | 2734.2 | 51.1 | 349.3 | 3103.8 | 4438.7 | 4422.6 | 1719284 | 387601.9 |
| Equisetum steam | Dzienna | 3.9 | 2 | 25.8 | 170.9 | 3.2 | 21.8 | 194 | 277.4 | 276.4 | 107455.3 | 24225.1 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake fo | or each element [µg] | 12.3 | 3.3 | 76.8 | 545.1 | 11.3 | 48.1 | 403.9 | 903 | 948 | 228190 | 45982 |

Jolanta Kowol, Jerzy Kwapuliński, Ewa Nogaj, Magdalena Babuśka-Roczniak, Barbara Brodziak-Dopierała, Wojciech Roczniak. Assessment of phytotherapeutic...

Table 4. Exposure during consumption of herbal mixture IV in urinary tract calculi

| | Amount of raw | | | | | Metal cont | ent [µg/g of | dry weight] | | | | |
|--|---------------|-------|-------|-------|--------|------------|--------------|-------------|--------|--------|----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.88 | 0.450 | 5.89 | 39.06 | 0.73 | 4.99 | 44.34 | 63.41 | 63.18 | 24561.2 | 5537.17 |
| Equiseti herba Equisetum steam | 40.0 | 35.2 | 18 | 235.6 | 1562.4 | 29.2 | 199.6 | 1773.6 | 2536.4 | 2527.2 | 982446.4 | 221486.8 |
| Equisetani steani | Daily | 3.3 | 1.7 | 22.1 | 146.5 | 2.7 | 18.7 | 166.3 | 237.8 | 236.9 | 92104.4 | 20764.4 |
| | 1.0 | 0.15 | 0.26 | 10.75 | 19.64 | 0.46 | 2.45 | 43.58 | 102.5 | 66.81 | 5148.71 | 2897.65 |
| Sambuci flos Elder flower | 40.0 | 6 | 10.4 | 430 | 785.6 | 18.4 | 98 | 1743.2 | 4100 | 2672.4 | 205948.4 | 115906 |
| | Daily | 0.6 | 1 | 40.3 | 73.7 | 1.7 | 9.2 | 163.4 | 384.4 | 250.5 | 19307.7 | 10866.2 |
| | 1.0 | 0.084 | 0.052 | 3.14 | 72 | 0.94 | 1.29 | 8.14 | 26.2 | 36.89 | 9845.83 | 1628.25 |
| Rosae fructus Dog rose | 40.0 | 3.4 | 2.1 | 125.6 | 2880 | 37.6 | 51.6 | 325.6 | 1048 | 1475.6 | 393833.2 | 65130 |
| bogrose | Daily | 0.3 | 0.2 | 11.8 | 270 | 3.5 | 4.8 | 30.5 | 98.3 | 138.3 | 36921.9 | 6105.9 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [µg] | | 20.1 | 2.9 | 74.2 | 490.2 | 7.9 | 72.7 | 360.2 | 721 | 626 | 148334 | 37736 |

Table 5. Exposure during consumption of herbal mixture V in urinary tract calculi

| | Amount of raw | | | | | Metal cont | ent [µg/g of | dry weight] | | | | |
|--|---------------|------|------|-------|-------|------------|--------------|-------------|--------|--------|----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.88 | 0.45 | 5.89 | 39.06 | 0.73 | 4.99 | 44.34 | 63.41 | 63.18 | 24561.2 | 5537.17 |
| <i>Equiseti herba</i> Equisetum steam | 25.0 | 22 | 11.3 | 147.3 | 976.5 | 18.3 | 124.8 | 1108.5 | 1585.3 | 1579.5 | 614029 | 138429.3 |
| Equisetani steani | Daily | 2.4 | 1.2 | 15.8 | 104.7 | 2 | 13.4 | 118.8 | 169.9 | 159.3 | 65923.9 | 14839.6 |
| | 1.0 | 0.23 | 0.26 | 6.93 | 28.7 | 0.58 | 1.4 | 37.07 | 181.05 | 119.58 | 38075.19 | 5022.48 |
| <i>Urticae folium</i> Nettle leaf | 10.0 | 2.3 | 2.6 | 69.3 | 287 | 5.8 | 14 | 370.7 | 1810.5 | 1195.8 | 380751.9 | 50224.8 |
| Nettie leaf | Daily | 0.2 | 0.3 | 7.4 | 30.7 | 0.6 | 1.5 | 39.7 | 193.7 | 128 | 40740.5 | 5374.1 |
| | 1.0 | 0.15 | 0.26 | 10.75 | 19.64 | 0.46 | 2.45 | 43.58 | 102.5 | 66.81 | 5148.71 | 2897.65 |
| Sambuci flos Elder flower | 10.0 | 1.5 | 2.6 | 107.5 | 196.4 | 4.6 | 24.5 | 435.8 | 1025 | 66.81 | 51487.1 | 28976.5 |
| Elder nower | Daily | 0.2 | 0.3 | 11.5 | 21 | 0.5 | 2.6 | 46.6 | 109.7 | 71.5 | 5509.1 | 3100.5 |
| | 1.0 | 0.13 | 0.1 | 5.9 | 27.16 | 0.47 | 1.14 | 24.09 | 55.98 | 35.6 | 5291.01 | 1582.58 |
| Sambuci fructus Elder fruit | 10.0 | 0.1 | 0.1 | 6.3 | 29.1 | 0.5 | 1.2 | 25.8 | 59.9 | 34.9 | 5661.4 | 1693.4 |
| Elder fruit | Daily | 0.1 | 0.1 | 6.3 | 29.1 | 0.5 | 1.2 | 25.8 | 59.9 | 34.9 | 5661.4 | 1693.4 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [µg] | | 2.9 | 1.9 | 41 | 185.5 | 3.6 | 18.7 | 230.9 | 533 | 405 | 11773 | 25008 |

metal quantities consumed by a patient during phytotherapy, assuming that 1 tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does nott exceed the daily human need for these elements. The limit for the concentration of Cd established for dry plant materials was exceeded in the case of horsetail herb. Pb content, however, did not exceed the approved limits.

Bronchial asthma. The disease presents with bronchial muscle contraction which makes breathing difficult and leads to expiratory dyspnea, being the main symptom of the disease. Different factors may contribute to the disease. The attack may be caused by an allergen, effort, stress, physical factor or cold air. The basic treatment involves pharmacotherapy, although herbal mixtures can play a supplementary role.

Mixture VI (Tab. 6). Anise root – Radix Pimpinellae 50.0 g Primrose flower – 50.0g Yarrow flower – Flos Millefoli 50.0g Coltsfoot leaf – Folium Farfarae 50.0g Elder flower – Flos Sambuci 50.0g White dead nettle flower Flos Lami albi – 50g Motherwort flower – Flos Leonuri 50.0g Dye less herb – herba Vincae minor 50.0g

Preparation and application: pour a glass of water over one tablespoon of herbs. Cook for 1 minute, leave the mixture covered for 10 min., then filter; drink half a glass 3 times a day [21]. Table 6 presents the maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does not exceed the daily human need for these elements. The limit concentration of Cd established for dry plant material was exceeded in the case of primrose flower, yarrow flower and coltsfoot leaf. Pb content, however, did not exceed the approved limits.

Chronic cholelithiasis. Deposits of gallstones, recurrent inflammation, formation of new stones and bile duct stenosis cause numerous disorders: bile flow into the duodenum is difficult, bilirubin is present in the blood, digestion and lipid absorption deteriorate. An extended period of cholestasis can lead to liver cirrhosis. The regular use of herbs can, after some time, restore the correct composition of bile or hinder the process of stone formation. Jolanta Kowol, Jerzy Kwapuliński, Ewa Nogaj, Magdalena Babuśka-Roczniak, Barbara Brodziak-Dopierała, Wojciech Roczniak. Assessment of phytotherapeutic...

| Diant | Amount of raw | | | | | Metal cont | ent [µg/g of | dry weight] | | | | |
|--|---------------|------|------|-------|--------|------------|--------------|-------------|--------|--------|----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.59 | 0.14 | 6.84 | 25.72 | 0.92 | 0.64 | 47.44 | 89.2 | 132.96 | 5011.02 | 3140.24 |
| <i>Primulae flos</i> Primula flower | 50.0 | 29.5 | 7 | 343 | 1286 | 46 | 32 | 2372 | 4460 | 6648 | 250551 | 157012 |
| i illindid flower | Dzienna | 0.8 | 0.2 | 9.6 | 36.3 | 1.3 | 0.9 | 66.9 | 125.8 | 187.5 | 7065.5 | 4427.7 |
| | 1.0 | 1.71 | 0.15 | 9.16 | 38.86 | 1.65 | 7.32 | 43.66 | 64.35 | 56.78 | 6889.42 | 1816.99 |
| <i>Millefolii flos</i> Yarrow flower | 50.0 | 85.5 | 7.5 | 458 | 1943 | 82.5 | 366 | 2183 | 3217.5 | 2839 | 344471 | 90849.5 |
| | Dzienna | 2.4 | 0.2 | 12.9 | 54.8 | 2.3 | 10.3 | 61.6 | 90.7 | 80.1 | 9714.1 | 2562 |
| | 1.0 | 1.98 | 0.17 | 14.57 | 37.51 | 1.24 | 0.57 | 77.41 | 85.06 | 178.96 | 17465.5 | 3727.24 |
| <i>Farfarae folium</i> Tussilago | 50.0 | 99 | 8.5 | 728.5 | 1875.5 | 62 | 28.5 | 3870.5 | 4253 | 8948 | 873274 | 186362 |
| russilago | Dzienna | 2.8 | 0.2 | 20.5 | 52.9 | 1.7 | 0.8 | 109.1 | 119.9 | 252.3 | 24626.3 | 5255.4 |
| | 1.0 | 0.15 | 0.26 | 10.75 | 19.64 | 0.46 | 2.45 | 43.58 | 102.5 | 66.81 | 5148.71 | 2897.5 |
| Sambuci flos Elder flower | 50.0 | 7.5 | 13 | 537.5 | 982 | 23 | 122.5 | 2179 | 5125 | 3340.5 | 257435.5 | 144882.5 |
| | Dzienna | 0.2 | 0.4 | 15.2 | 27.7 | 0.6 | 3.5 | 61.4 | 144.5 | 94.2 | 7259.7 | 4085.7 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [µg] | | 6.2 | 1 | 58.2 | 171.7 | 5.9 | 16.5 | 299 | 481 | 614 | 48666 | 16331 |

Table 6. Exposure during consumption of herbal mixture V in bronchial asthma

Mixture VII (Tab. 7).

Dandelion root – Radix Taraxaci 50.0g Marigold flower – Flos Calenduale 50.0g Sweet flag rhizome – Rhizoma Calami 30.0 g Daisy flower – Flos bellidis 50.0g Yarrow flower – Flos Millefolii 50.0g Herb celandine – Herba Chelidonii 50.0g St John's Wort – Herba Hyperici 50.0g

Preparation and application: pour half a glass of water over 2 tablespoons, cook for 2 minutes; drink half a glass 3 times a day [21]. Table 7 presents the maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5g [22]. The total daily intake of the respective elements does not exceed the daily human need for these elements. The limit concentration of Cd established for dry plant material was exceeded in all plants included in the above mixture. Pb content, however, did not exceed the accepted limits.

Diabetes. The disease involves disturbed insulin secretion, leading to the destruction of pancreatic cells secreting insulin (insulin-dependent diabetes), or to the reduction in insulin level or in target cell sensitivity to insulin (insulin-independent diabetes). In the case of insulin-dependent diabetes that mainly appears in young people, the primary treatment is insulin administration. However, in elderly patients with insulin-independent diabetes, especially in its initial phase, dietary treatment and herbal therapy can be effective.

Mixture VIII (Tab. 8). Herb goat's rue – Herba Galagae 50.0 g Beans – Pericarpium Phaseoli 50.0 g Bilberry leaf – Folium Myrtylli 50.0 g Dandelion root – Radix Taraxaci 50.0 g Dandelion flower – Flos Taraxaci

Preparation and application: pour a glass of water over 2 tablespoons of the mixture, cook covered for 1 minute,

Table 7. Exposure during consumption of herbal mixture VII in chronic cholelithiasis

| Plant | Amount of raw | | | | | Metal conte | ent [µg/g of | dry weight] | | | | |
|--|---------------|-------|------|-------|-------|-------------|--------------|-------------|--------|---------|----------|---------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 3.21 | 0.69 | 15.57 | 25.14 | 1.089 | 2.27 | 44.45 | 177.55 | 1997.78 | 5318.59 | 1296.57 |
| <i>Taraxaci radix</i> Dandelion Root | 50.0 | 160.5 | 34.5 | 778.5 | 1257 | 54.5 | 113.5 | 2222.5 | 8877.5 | 99889 | 265929.5 | 64828.5 |
| Danachon noot | Daily | 21.9 | 4.7 | 106.2 | 171.5 | 7.4 | 15.5 | 303.1 | 1210.9 | 13624.9 | 36272.8 | 8842.6 |
| | 1.0 | 1.04 | 0.26 | 8.02 | 23.82 | 1.34 | 2.47 | 66.98 | 178.62 | 994.34 | 11387.3 | 2381.66 |
| Flos Bellis perennis Daisy flower | 50.0 | 52 | 13 | 401 | 1191 | 67 | 123.5 | 3349 | 8931 | 49717 | 569366 | 119083 |
| | Daily | 7.1 | 1.8 | 54.7 | 162.5 | 9.1 | 16.8 | 346.8 | 1218.2 | 6781.4 | 77661.5 | 16242.9 |
| | 1.0 | 1.71 | 0.15 | 9.16 | 38.86 | 1.65 | 7.32 | 43.66 | 64.35 | 56.78 | 6889.42 | 1816.99 |
| Millefolii flos Yarrow flower | 50.0 | 85.5 | 7.5 | 458 | 1943 | 82.5 | 366 | 2183 | 3217.5 | 2839 | 344471 | 90849.5 |
| Tarlow nower | Daily | 11.7 | 1 | 62.5 | 265 | 11.3 | 49.9 | 297.08 | 438.9 | 387.2 | 46985.8 | 12391.9 |
| | 1.0 | 8.01 | 0.15 | 6.1 | 10.01 | 1.64 | 4.08 | 65.49 | 60.31 | 45.94 | 7726.74 | 1955.24 |
| <i>Hyperici herba</i> St.John's wort | 50.0 | 400.5 | 7.5 | 305 | 500.5 | 82 | 204 | 3274.5 | 3015.5 | 2297 | 386337 | 97762 |
| SLJOIN S WORL | Daily | 54.6 | 1 | 41.6 | 68.3 | 11.2 | 27.8 | 446.6 | 411.3 | 313.3 | 52696.4 | 13334.7 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake for each element [µg] | | 96.3 | 8.5 | 265 | 667.3 | 39 | 82.7 | 1504.3 | 3279 | 21107 | 213616 | 50812 |

Jolanta Kowol, Jerzy Kwapuliński, Ewa Nogaj, Magdalena Babuśka-Roczniak, Barbara Brodziak-Dopierała, Wojciech Roczniak, Assessment of phytotherapeutic.

Table 8. Exposure during consumption of herbal mixture VIII in diabetes

| Diant | Amount of raw | | | | | Metal conte | ent [µg/g of | dry weight] | | | | |
|---|--|-------|------|-------|--------|-------------|--------------|-------------|--------|---------|----------|----------|
| Plant | material [g] | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| | 1.0 | 0.11 | 0.10 | 12.44 | 786.08 | 0.930 | 0.62 | 29.48 | 34.99 | 69.98 | 3235.39 | 1198.29 |
| <i>Myrtylli fructus</i> Bilberry fruit | 50.0 | 5.5 | 5 | 622 | 39304 | 46.5 | 31 | 14.74 | 1749.5 | 3499 | 161769.5 | 59914.5 |
| bildeny nate | Daily | 1 | 0.9 | 112 | 7074.7 | 8.4 | 5.6 | 265.3 | 314.9 | 629.8 | 29118.5 | 10784.6 |
| <i>Taraxaci radix</i> Dandelion root | 1.0 | 3.21 | 0.69 | 15.57 | 25.14 | 1.089 | 2.27 | 44.45 | 177.55 | 1997.78 | 5318.59 | 1296.5 |
| | 50.0 | 160.5 | 34.5 | 778.5 | 1257 | 54.5 | 113.5 | 2222.5 | 8877.5 | 99889 | 265929.5 | 64828.5 |
| Dundenon root | Daily | 28.9 | 6.2 | 140.1 | 226.3 | 9.8 | 20.4 | 400.1 | 1598 | 17980 | 47867.3 | 11669 |
| | 1.0 | 11.54 | 0.1 | 12.31 | 23.33 | 3.79 | 0.45 | 30.35 | 69.74 | 407.66 | 4992.22 | 12458.48 |
| <i>Taraxaci flos</i> Dandelion flower | 50.0 | 577 | 5 | 615.5 | 1166.5 | 189.5 | 22.5 | 1517.5 | 3487 | 20383 | 249611 | 122924 |
| Dandenon nower | Daily | 103.9 | 0.9 | 110.8 | 210 | 34.1 | 4.1 | 273.2 | 627.7 | 3668.9 | 44930 | 12126.3 |
| | | Cd | Cr | Cu | Mn | Ni | Pb | Zn | Fe | Na | Ca | Mg |
| Total daily intake fo | Total daily intake for each element [µg] | | 8.0 | 362.9 | 7511 | 52.3 | 30.1 | 938.6 | 2541 | 22279 | 121916 | 44580 |

then leave it for a few minutes and filter; drink half a glass 3 times a day [21]. Table 8 shows the maximal metal quantities consumed by a patient during phytotherapy, assuming that one tablespoon of the prepared mixture is 7.5 g [22]. The total daily intake of the respective elements does not exceed the daily human need for these elements [23, 24, 25]. The limit of concentration of Cd established for dry plant material was exceeded in the case of dandelion root and dandelion flowers. Pb content, however, did not exceed the accepted limits.

The presented study does not include plant species, which is a poorly accumulated research element. The amount of their occurrence was below the detectability of the method used.

In summary, it can be concluded that phytotherapy in the long term is underrated, yet it is a significant source of additional quantities of selected metals, together with their simultaneous presence in different parts of the environment (soil, food of plant and animal, particulate matter). The average contents of selected metals in the plants investigated can be used as reference values in prospective studies, and test plant species can be used to bioindicative lead, cadmium, nickel and chromium.

CONCLUSIONS

- 1. For the different mixtures estimated values of daily intake and total daily intake for given diseases can be useful as preferences for future comparison, and comparing the degree of exposure during given phytotherapy.
- 2. Long-term phytotherapy may be the source of additional exposure of people to the toxic metals Pb, Cd, Ni and the important physiological elements Cr, Fe, Zn.

REFERENCES

- 1. Senderski EM. Zioła. Praktyczny przewodnik o ziołach i ziołolecznictwie. Warszawa K.E. Liber 2009: 9–17 (in Polish).
- 2. Zarawska-Lamer E, Gierczyk-Kowal B, Niedworok J. Fitoterapia i leki roślinne. Warszawa PZWL 2007: 9-17 (in Polish).
- 3. Wesołowski M, Radecki I. Rośliny lecznicze. Skład pierwiastkowy, źródła składników mineralnych dla roślin, wskaźniki skażenia środowiska metalami ciężkimi. Śarma Pol. 2003; 59: 911–919 (in Polish). 4. Iwanek K, Kowol J, Kwapuliński J. Rośliny lecznicze jako źródło metali
- ciężkich. Problemy Ekologii 1997; 7: 211–214 (in Polish). 5. Ražic S, Onjia A, Dogo S, Slavkovič L, Popvič A. Determination of metal
- content in some herbal drugs. Empirical and chemometric approach. Talanta, 2005; 67: 233–239.

- 6. Kwapuliński J, Kowol J, Ciba J. Ocena potencjalnej intoksykacji organizmu rtęcią podczas terapii ziołoleczniczej mniszkiem lekarskim. Bromat Chem Toksykol. 2000; 33: 277–282 (in Polish).
- 7. Kwapuliński J, Michalewska A, Rochel R, Kowol J. Intoksykacja surowców roślin leczniczych metalami ciężkimi w świetle obowiązujących uregulowań ustawodawczych oraz zaleceń WHO. Problemy Ekologii 2005; 9: 202–204 (in Polish).
- 8. Lutomski J. Ziołolecznictwo. Tradycja i przyszłość. Gdańsk Tower Press 2003: 5-8
- 9. Samochowiec L. Lek roślinny w świetle nowoczesnej farmakoterapii. Post Fitot. 2001; 2: 2-6 (in Polish).
- 10. Srogi K. Zawartość wybranych pierwiastków w surowcach leczniczych dziko rosnących. Bromat Chem Toksykol. 2004; 2: 2-6 (in Polish).
- 11. Błoniarz J, Źaręba S, Ranhama M. Zawartość kadmu i ołowiu w ziołach, preparatach ziołowych oraz naparach wykonanych z tych ziół stosowanych u dzieci i dorosłych. Przegl Lek. 2001; 58: 39–43 (in Polish).
- 12. Michalewska A, Kwapuliński J, Rochel R, Kowol J. Rola transporterów metali ciężkich w intoksykacji roślinnych surowców leczniczych. Herba Pol. 2004; 50: 111-119 (in Polish).
- 13. Kowol J, Kwapuliński J, Fischer A, Królak E, Librowska J. Ekologiczne i fitochemiczne uwarunkowania wpływające na kumulację metali w roślinie. Ekologia i Technika 2010; 18: 94–102 (in Polish).
- 14. Kwapuliński J, Rochel R, Michalewska A. Dystrybucja biopierwiastków w roślinie. Problemy Ekologii 2010; 1: 17-21 (in Polish).
- 15. Kwapuliński J, Rochel R, Michalewska A. Mechanizmy pobierania metali ciężkich przez rośliny z gleby. Problemy Ekologii 2010; 2: 66-71(in Polish)
- 16. Kowol J, Kwapulinski J, Brodziak B, Paukszto A, Bogunia B, Rochel R, Ahnert B. Influence of a transboundary emission on bioavailability of metals of stinging nettle from soil in the area Vistula (Poland). Pol J Environ Stud. 2011; 20: 101–110.
- 17. Kowol J, Paukszto A, Kwapuliński J, Ahnert B, Rochel R, Surma M. Medicinal plants and their sites by metal contamination in the area of the "umbrella effect" as exemplified by Łagisza Power Station (Poland). Fresenius Environ Bull 2009; 18: 1094–1099
- 18. Rochel R, Kwapuliński J, Kowol J, Bogunia M. Migration of bioavailable chemical forms of chromium from soil to medicinal plants. Fresenius Environ Bull. 2009; 18: 1963-1966.
- 19. Smrkolj P, Stibilj V. Determination of selenium in vegetables by hydride generation atomic fluorescence spectrometry. Anal Chim Acta 2004; 512: 11–17.
- 20. Mirosławski J, Wiechuła D, Kwapuliński J, Rochel R, Loska K, Ciba J. Występowanie Pb, Cd, Mn, Ni, Co, Cr w wybranych gatunkach roślin leczniczych na terenie Polski Brom Chem Toksykol. 1995; 28: 363–368 (in Polish)
- 21. Łaszewska A, Kowol J, Wiechuła D, Kwapuliński J. Kumulacja metali w wybranych gatunkach roślin leczniczych z terenu Beskidu Śląskiego W. Schulz Zywier Bruch robit Ecol. 2007; 11: 285–291 (in Polish).
 Janicki K, Rewerski W. Medycyna naturalna. Warszawa PZWL 2001;
- 322-323, 329-330, 395, 399 (in Polish).
- 23. Książkiewicz T. Ziołolecznictwo Ojców Bonifratów dla osób starszych. Warszawa Oficyna Wydawnicza Rytm 2000: 39, 104–105, 107–108, 161-162, 192-193
- 24. Musielińska R, Kowol J, Kwapuliński J, Rochel R. Antagonism between lead and zinc ions in plants. Arch Environ Protect 2016; 42(2): 78–81. 25. Peralta-videa JR, Lopez ML, Narayan M, Saupe G. The biochemistry of
- environmental heavy metal uptake by plants: Implications for the food chain Intern. J Biochem Cell Biol. 2009; 91: 1665-1677.