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ORIGINAL PAPER

ACCUMULATION OF CHROMIUM, ALUMINUM, BARIUM AND ARSENIC IN SELECTED ELEMENTS OF A FOREST ECOSYSTEM IN THE PRZEDBABIÓGÓRSKIE MOUNTAIN RANGE IN THE WESTERN CARPATHIANS*

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ABSTRACT

Large spore mushrooms and trees are frequently used in bioindication, the reasons being their widespread occurrence and substantial capacity to accumulate trace elements. The aim of the research was to compare the bioaccumulation coefficients of As, Ba, Al, and Cr in *Lactarius salmonicolor* L., and in *Abies alba* M. needles, collected in the Przedbabiogórskie Mountain Range. In 2015, samples of soil, mushrooms and *Abies alba* needles were collected from 17 sites. All the collected samples were dried and homogenized. Laboratory samples of the mushrooms and needles were subjected to dry mineralization in an open system. The soil samples were mineralized in *aqua regia*. Concentration of elements in the solutions was determined by atomic emission spectrometry. Based on the results, bioaccumulation factors of individual elements were calculated. The bioaccumulation factors were calculated by dividing the concentration of a given element in the dry matter of mushrooms and needles used in the research by the content of the same element in the soil. In addition, the correlation coefficient between the content of the above elements in the soil, mushrooms and needles was computed. The content of the elements in forest soils was low, typical for unpolluted areas. Concentrations of the elements in *Lactarius salmonicolor* L. and in *Abies alba* M. needles were in the following decreasing order: As>Cr>Ba>Al. The values of bioaccumulation factors of the elements in needles can be arranged from the lowest as follows: Al>Cr>As>Ba, whereas in the mushrooms the order was Al>Ba>Cr>As. No statistically significant relationship between the organic carbon content in the soil and the level of accumulation of the elements in the fungal fruiting bodies and fir needles was observed. The content of elements in the soil had the largest effect on the amount of these elements accumulated in *Abies alba* needles and in biomass of *Lactarius salmonicolor*.

Keywords: bioaccumulation coefficient, trace elements, needles, mushrooms, fir, forest soils.

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INTRODUCTION

Mushrooms are a very important part of forest ecosystems. In addition to their participation in the decomposition of dead organic matter and in soil-forming processes, mushrooms select trees by eliminating weak specimens. Mycorrhizal mushrooms have an effect on the availability of micro- and macronutrients for trees. They are food for numerous forest animals such as wild boar, deer, roe deer. Owing to their substantial capacity to bioaccumulate elements, they play an important role in the biogeochemical cycle in forest ecosystems. Mushrooms are part of the human diet, and thereby they can affect the amount of trace elements incorporated into the human food chain. Owing to their considerable content of micronutrients, vitamins and carbohydrates, they are regarded as a valuable component of the human diet (KALAC 2009, WANG et al. 2014, REIS et al. 2012). The substantial capacity to accumulate trace elements is the reason why mushrooms are very often found to have an elevated content of heavy metals (COCCHI et al. 2006, DAMODARA et al. 2014). On the other hand, because of their ability to accumulate trace elements, mushrooms are often used as a bioindicator of environmental pollution (MELGAR et al. 2014, LIU et al. 2015). The capacity to accumulate large amounts of trace elements, widespread occurrence, abundance and short life cycles of mushrooms make them good bioindicators. However, the amount of trace elements accumulated in mushrooms depends on numerous factors associated with both the biotope and mushroom organisms. Among the factors connected with the biotope, the most frequently mentioned are the total content of elements in the substrate and physicochemical properties of the substrate, such as pH, organic matter content or level of salinity (YAMAÇ et al. 2007). The amount of accumulated elements is also influenced by the species of mushrooms and their age. A problem associated with the use of mushrooms in biomonitoring is that lifespans of mushroom fruiting bodies are highly varied. Some live for several days, while other can last longer, even for many years. Apart from mushrooms, trees are another useful element in the monitoring of environmental quality. Studies on tree leaves usually concern the level of environmental pollution with heavy metals, but they can also involve processes associated with environmental degradation. Changes in the content of heavy metals in plant leaves might arise from an increase in the content of these elements in the forest biotope or they might be a consequence of changes in soil properties (LINDROOS et al. 2007). Oaks, birches, maples and plane trees are among the deciduous trees most often submitted to research, while pine and spruce species are the most popular coniferous trees (ČEBURNIS, STEINNES 2000, ABOAL et al. 2004, NIEMIEC, ARASIMOWICZ 2010, PARZYCH, JONCZAK 2013). Despite its high capacity to bioaccumulate trace elements, fir is not extensively used as a biomonitoring organism because of its limited coverage (ABOAL et al. 2004, GANDOIS, PROBST 2012). A problem in using leaves of coniferous trees is the variability of the

trace element content depending on the age of needles and trees (ČEBURNIS, STEINNES 2000, UCUN ÖZEL, ÖZEL 2012, GIELEN et al. 2016).

The aim of this research was to compare the bioaccumulation coefficients of As, Ba, Al, and Cr in *Lactarius salmonicolor* L., and in *Abies alba* M. needles, collected in the Przedbabiogórskie Mountain Range.

MATERIAL AND METHODS

The research area consisted of the Jałowiec Mountain Range, which belongs to the Przedbabiogórskie Range, part of the Maków Beskids. The set goal was reached through collecting samples of *Lactarius salmonicolor* from 17 sites (Figure 1). A homogeneous area in terms of habitat conditions

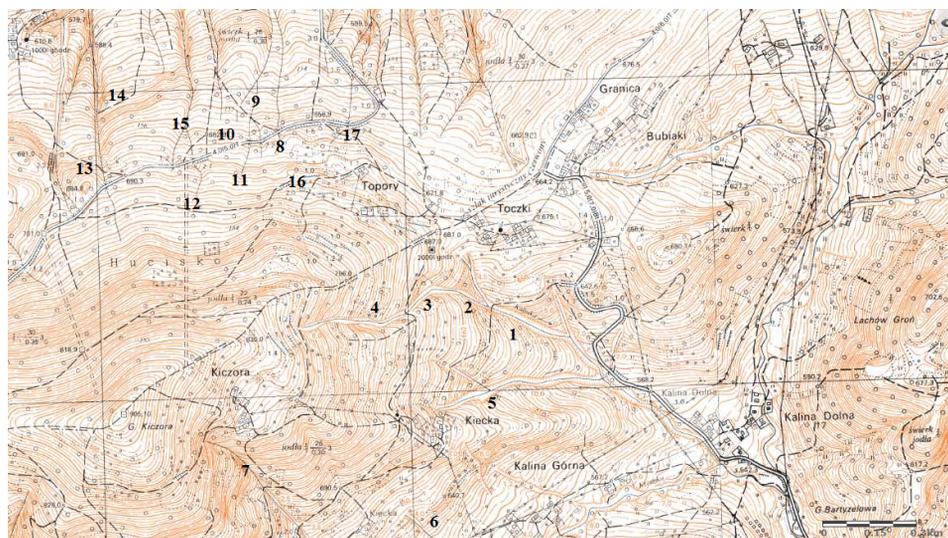


Fig. 1. Research area with established sample collection sites (1,2,3...)

within a 50 m radius was used as a sampling point. A laboratory sample was a bulk sample consisting of approximately 100 subsamples. The mushrooms were collected using a plastic knife. Out of the 17 sampling sites, 16 were located within channels of mountain streams in the Lachówka (from the northern side) and the Skawica river basins (from the southern side). The soil was sampled at the same sites, from the 0-20 cm layer. The soil samples were collected using an Egner's stick. A bulk sample of soil consisted of approximately 20 subsamples. A laboratory sample was prepared by reducing the bulk sample to a mass of approximately 1000 g. In addition, samples of *Abies alba* needles were collected. The needles were collected from plants up to 10 years old, from one-year-old shoots. A laboratory sample, which was simultaneously a bulk sample, had a mass of 500 g. Once the

samples were collected, the mushrooms were washed with distilled water, and then stipes were separated from caps. All the collected samples were dried and homogenized. The ready samples were transported to a laboratory. The laboratory samples of the mushrooms and needles were subjected to dry mineralization in an open system. An analytical sample weighed 3 g. The soil samples were mineralized in *aqua regia* in a quantitative ratio of 1:10 (the soil to reagents). The concentration of elements in solutions was determined by inductively coupled plasma (Perkin Elmer, Optima 7600 DV). Wavelengths that were used to determine the concentration of the elements as well as the detection limit for the methods are provided in Table 1. The quality of the analysis was verified using certified reference material

Table 1

Parameters of the analytical method

Specification	As	Ba	Al	Cr
Wavelengths (nm)	228.8	327.3	220.3	206.2
Detection limit (mg dm ⁻³)	0.0027	0.0097	0.042	0.0059
Content in certified material (mg kg ⁻¹)	0.013	5.641	0.470	12.51
Measured (mg kg ⁻¹)	0.011	5.831	0.534	13.83
Recovery (%)	84.60	103.4	113.6	111.1

(NIST-1515). Table 1 shows results of analyses of the reference material and an estimated value of recovery based on analyses conducted in 4 replications. Bioaccumulation factors of individual elements were calculated based on the above results. They were computed by dividing the concentration of the elements in dry matter of mushrooms and needles used in the research by the content of these elements in the soil. The statistical significance of the correlation coefficient between soil reaction, the content of organic matter and the analysed elements in the soil, as well as the content of these elements in mushrooms and plants were derived. The correlation coefficient was calculated using the t-Student test ($p = 0.01$).

RESULTS AND DISCUSSION

Soils taken from individual sampling points had acid pH, which ranged from 4.22 to 5.68. Soil pH values determined in the authors' own research are characteristic for forest soils. The organic carbon content varied in a wide range between 19.9 and 78.8 g kg⁻¹. Forest soils generally have a low pH and a high content of organic carbon. Processes associated with the accumulation of organic matter in forest soils arise from the accumulation of these elements in forest litter and from their content in the bedrock (JONCZAK et al. 2016). The mean arsenic content in the forest soils studied was

2.179 mg kg⁻¹ and ranged between 1.263 and 3.95 mg kg⁻¹ (Table 2). No considerable differences in the content of this element in individual samples were observed. The relative standard deviation was 33.11%. In the research, the arsenic content determined in the soils was not high and was characteristic for unpolluted soils (DOUSOVA et al. 2016). Much greater diversity was observed in the case of arsenic concentrations in fir needles, where they ranged from 0.1 to 0.225 mg kg⁻¹ at the relative standard deviation of 72.67% (Table 2). The mean content of this element was 0.144 mg As kg⁻¹. Arsenic content in pine needles collected from areas with a high human impact index amounted to 0.77 mg As kg⁻¹ (RUCANDIO et al. 2011). No differences in the content of this element in the stipes and caps of the studied mushrooms were observed. Arsenic content in the studied samples of mushrooms was characteristic for regions unpolluted with this element. Arsenic content in edible mushrooms is a very important parameter of their consumption value. Many authors draw attention to a high capacity of mushrooms to accumulate this element, which is conditioned by the symbiosis of mushrooms and microorganisms (which transform compounds of this element into more assimilable forms (SOEROES et al. 2005, MELGAR et al. 2014). Arsenic content in the studied mushrooms ranged between 0.037 and 0.312 mg As kg⁻¹ (Table 2). No differences in the content of this element in the stipes and caps of *Lactarius salmonicolor* were observed. A similar content of arsenic in mushrooms to the one obtained in this research was found by FANG et al. (2014). LLORENTE-MIRANDES et al. (2014) report an arsenic content in shiitake mushrooms at a level of 0.39 mg As kg⁻¹, with the As content in the substrate at a level of

Table 2

Statistic parameters of the content of elements of the soil, needles and mushrooms

Specification	Unit	Soil	Needles	Stipes	Caps	Soil	Needles	Stipes	Caps
		As				Ba			
Minimum	(mg kg ⁻¹)	1.263	0.225	0.050	0.100	42.13	27.89	1.950	1.688
Maximum		3.950	2.388	0.312	0.225	121.4	83.33	4.210	3.425
Average		2.179	0.724	0.146	0.156	73.01	55.92	3.083	2.691
Standard deviation		0.721	0.526	0.066	0.055	22.97	16.46	0.675	0.527
Coefficient of variation	%	33.11	72.67	45.19	35.68	31.46	29.43	21.91	19.60
		Al				Cr			
Minimum	(mg kg ⁻¹)	4119	117.5	23.39	19.34	6.200	0.738	0.276	0.588
Maximum		11438	547.9	76.29	107.5	23.01	2.538	1.313	1.913
Average		8516	315.5	40.68	45.97	15.954	1.349	0.554	1.022
Standard deviation		1847	121.43	16.63	26.49	4.948	0.491	0.257	0.366
Coefficient of variation	(%)	21.69	38.49	40.89	57.62	31.02	36.40	46.40	35.84

0.11 mg As kg⁻¹. The bioaccumulation factor value given by these authors was over 3. The mean bioaccumulation factor of arsenic in the biomass of *Abies alba* needles was 0.332, varying from 0.121 to 0.871 at individual locations. The high value of the factor at those locations resulted from the increased content of this element in the studied organisms rather than from the lower accumulation of arsenic in the soil. The bioaccumulation factor of arsenic in biomass of the mushrooms ranged between 0.027 and 0.137. The mean arsenic bioaccumulation factor in the mushrooms was 0.07. NIKKARINEN and MERTANEN (2004) reported that the value of the bioaccumulation factor for arsenic in *Lactarius trivialis* in unpolluted regions of Finland was 0.053. In most cases, values of the arsenic bioaccumulation factor in various species of mushrooms collected in polluted regions of Yunan province were approximately 0.3, and the content of this element in the substrate was at a level of approximately 5 mg As kg⁻¹ (LIU et al. 2015). These authors determined a several-fold higher arsenic content in mushrooms than detected in our study. The mean barium content in the studied soils was 73.01 mg Ba kg⁻¹, whereas in needles it equalled 55.92 mg Ba kg⁻¹. Amounts of barium determined in fir needles were several times higher than the content of this element determined by GANDOIS and PROBST (2012) in pine needles collected in Madrid. The barium content in *Lactarius salmonicolor* varied from 1.688 to 4.210 mg kg⁻¹. NIKKARINEN and MERTANEN (2004) determined a similar content of this element in unpolluted areas of Finland as determined in this research in *Lactarius trivialis*. No difference in the content of this element in the stipes and caps of the samples studied was observed. The relative standard deviation for barium concentrations in the samples studied was approximately 20%. Mean values of the barium bioaccumulation factor in the biomass of fir needles, stipes and caps of *Lactarius salmonicolor* reached, respectively, 0.766, 0.042 and 0.037 (Table 3). The highest values of this factor were observed at the same research sites where the highest values of arsenic bioaccumulation

Table 3

Bioaccumulation factor for metals in needles, stipes and caps

Specification	Unit	Needles	Stipes	Caps	Needles	Stipes	Caps
		As			Ba		
Minimum		0.037	0.122	0.028	0.017	0.450	0.026
Maximum		0.137	0.871	0.116	0.070	1.395	0.072
Average		0.072	0.334	0.069	0.040	0.803	0.045
Coefficient of variation	(%)	0.029	0.198	0.027	0.014	0.265	0.013
		Al			Cr		
Minimum		0.0022	0.015	0.0027	0.033	0.046	0.016
Maximum		0.0136	0.058	0.010	0.145	0.178	0.088
Average		0.0056	0.038	0.005	0.068	0.091	0.038
Coefficient of variation	(%)	0.0033	0.014	0.002	0.026	0.038	0.019

were determined. In this research, a high bioaccumulation factor value was determined. The value of this factor in *Lactarius trivialis* in Finland was 0.01 (NIKKARINEN, MERTANEN 2004).

The chromium content in the forest soils varied from 6.2 to 23.01 mg kg⁻¹ (Table 2). The mean content of this element determined in fir needles was 1.349, which was double the the content of this element in pine needles from polluted regions of Spain reported by GANDOIS and PROBST (2012). On the other hand, ARASIMOWICZ et al. (2010) give the mean chromium content in pine needles from anthropogenically transformed areas of Krakow at a level of 2.5 mg kg⁻¹. The chromium content in the mushrooms was low (on average, 0.544 and 1.022 mg kg⁻¹, respectively). MELGAR et al. (2014) obtained a similar chromium content in northern Spain, whereas GARCIA et al. (2013) give a concentration of this element in various species of wild growing mushrooms from the same region in northern Spain at a level of 0.67 to 16 mg kg⁻¹, depending on the degree of environmental pollution. These authors found differences in the content of this element depending on the part of mushrooms. Our results show that caps contained twice as much chromium as stipes. ALOUPI et al. (2012) report the chromium content in the milk-cap at a level of 0.1 mg kg⁻¹, whereas SARIKURKCU et al. (2011) determined the chromium content in various wild species of mushrooms from Soguksu National Park to be within the range from trace amounts to over 20 mg kg⁻¹. The latter authors concluded that mushrooms in their research were good bioindicators. Similar conclusions were drawn based on the results of this research. The mean bioaccumulation factor of chromium in the mushrooms amounted to 0.035 for stipes and 0.06 for caps. GARCIA et al. (2013) give much higher values of the bioaccumulation factors for various mushrooms species in northern Spain. NIKKARINEN and MERTANEN (2004) report the chromium content in *Lactarius trivialis* from unpolluted areas to be at a level of 0.12, with the value of the bioaccumulation factor being 0.003 mg kg⁻¹ DM. The content of this element in edible boletus from the same regions was several-fold lower.

The mean aluminum content in all the studied samples forest soils was 8516 mg kg⁻¹ and ranged between 4119 and 11438 mg kg⁻¹ (Table 2), whereas the concentration of this element in the needles of *Abies alba* varied from 117.5 to 547.9 mg kg⁻¹. The aluminum content determined in fir needles was high, characteristic for environments with an elevated level of human impact. The content of this element in *Picea abies* needles of the Gulf of Bothnia ranged from 55 to 76 mg kg⁻¹ (LINDROOS et al. 2007). The aluminum content in needles of pine from the Madrid agglomeration reached 400 mg kg⁻¹ (RUCANDIO et al. 2011). On the other hand, GANDOIS and PROBST (2012) give the aluminum content in fir needles from unpolluted areas of France at a level of 382 mg kg⁻¹. The mean value of the aluminum bioaccumulation factor for fir needles determined in our research was 0.005.

Based on the results, correlation coefficients between the content of the elements studied in the soil and in selected parts of the biocenosis were

computed. Organisms which are good bioaccumulators, regardless of the type of an environment in which they live, ought to accumulate elements in their tissues in quantities proportional to their concentration in the biotope (GANDOIS, PROBST 2012, NIEMIEC et al. 2015, NIEMIEC 2016). Based on the current research, statistically significant correlations between the arsenic content in the soil and in fir needles as well as in stipes of the studied mushrooms were observed (Table 4). In the case of barium, a significant correlation

Table 4

The correlation coefficient of the content of elements in different parts of the environment and between the properties of the soil

Specification	Needles	Stipes	Caps	Needles	Stipes	Caps
	As			Ba		
Content in soil	0.619*	0.551*	0.422	0.534*	0.281	0.056
pH	-0.138	0.17	0.453	-0.109	0.230	-0.197
Organic mater kontent in soil	-0.326	-0.361	0.125	-0.110	0.077	-0.259
Content in needles	-	0.624*	0.647*	-	0.125	-0.043
	Al			Cr		
Content in soil	0.451	0.300	0.202	0.384	0.301	0.525*
pH	-0.173	-0.509*	0.140	-0.406	-0.274	-0.424
Organic mater kontent in soil	0.411	-0.027	-0.139	0.411	-0.027	-0.139
Content in needles	-	0.081	0.176	-	0.314	0.108

* the significance of the correlation coefficient $p = 0,05$

was observed only between the content of this element in the soil and needles, whereas the amount of chromium in mushroom caps was significantly positively correlated with the content of this element in the soil. No statistically significant relationship was observed between the soil reaction and the content of the elements (except aluminum) in stipes of *Lactarius salmonicolor* (Table 4). No statistically significant relationship was noted between the organic carbon content in the soil and the level of accumulation of all the elements studied in needle and mushroom biomass.

CONCLUSIONS

1. Concentrations of the analysed elements in *Lactarius salmonicolor* L. and in *Abies alba* M. needles can be arranged in the increasing order: As>Cr>Ba>Al.

2. The chromium content in mushroom caps was approximately twice as high as in stipes. In the case of other elements, no differences in their content between the parts of mushrooms were detected.

3. Values of the bioaccumulation factor for the analysed elements in needles can be arranged from the lowest as follows: Al>Cr>As>Ba, whereas the ones determined in the mushrooms were Al>Ba>Cr>As.

4. Bioaccumulation factors for arsenic and chromium in the samples of mushrooms and fir needles were close to the ones determined by other authors in areas with a low level of human impact.

5. The content of elements in the soil had a significant effect on the amount of these elements accumulated in *Abies alba* needles and in biomass of *Lactarius salmonicolor*. No statistically significant relationship between the organic carbon content in the soil and the level of accumulation of the analysed elements in the selected organisms was observed.

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