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Contents of nutrients and arsenic in litterfall and surface humus in mature nudal beech stands subjected to different emission-immission loads

Katarína Gašová*, Margita Kuklová, Ján Kukla

Department of Soil and Plant Ecology, Institute of Forest Ecology of the Slovak Academy of Sciences, Ľ. Štúra 2, 960 53 Zvolen, Slovak Republic

Abstract

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The aim of the paper was to compare the litterfall and surface humus (Oo) quality in nudal beech stands with different emission-immission load from the Al smelter Žiar nad Hronom. The study was carried out in the Kremnické vrchy Mts (control stand, 18 km from the emission source) and the Štiavnické vrchy Mts (stressed stand, 1.5 km from the emission source), both in Central Slovakia. The contents of arsenic and nutrients (Ca_t, Mg_t, K_t, Na_t), with exception of calcium in the beech litterfall from the stressed stand were markedly higher (by 4.3%, 23.9%, 2.1% and 87.9%, respectively) compared to the samples taken from the control plot. On the contrary, the surface humus samples (with the exception of Na in the necrotic Oon subhorizon) from control plot were richer in nutrients. However, significant differences ($p < 0.01$) between the plots were observed for Ca content in the litterfall as well as in the surface humus. The concentrations of As in Oo samples from the stressed stand mainly increased with the organic horizon depth (mg kg^{-1}): necrotic Oon 1.10 < fermentation Oof 3.1 < humification Ooh 55.6. The results showed, that As amounts detected in subhorizon Ooh and in beech litterfall from the stressed stand were higher than the limit values, thus indicating that the environment of the Žiar territory is exposed to persistent negative impacts of industrial activities.

Keywords

arsenic, litterfall, mature beech forests, nutrients, surface humus

Introduction

Litterfall is a fundamental component in nutrient cycling, and it primarily means the transfer of organic matter and mineral elements from vegetation to the soil surface (Vitousek and Sanford, 1986). It serves as an input-output system for nutrients, and for controlling the rates at which forest litterfall and decays processes regulate energy flow, primary productivity and nutrient cycling in forest ecosystems (Liao et al., 2006). For example, nutrient contents in the litterfall represent annually per hectare approximately 40–55 kg of N, 2–3 kg of P, 7–16 kg of K, 41–73 kg of Ca and 6–7 kg of Mg in young oak stands in the Czech Republic (Novák et al., 2014). Forest stands and other plant vegetation affect physical, chemical and biological soil properties, both directly and indirectly. Differences in the litterfall quality are reflected in the properties of the upper soil layers and the surface humus, which has been proved by studies by several authors (Neirynck et al., 2000; et al., 2002, 2003; Bieńkowski et al., 2006; Wulf and Naaf, 2009; Langenbruch et al., 2012, etc.). Decomposition is very important in the biochemical nutrient cycle, in which the nutrients are mineralized and turn

*Corresponding author:
e-mail: gasova.k@gmail.com

again available to plants (Šimková et al., 2014). The study of Xiaogai et al. (2013) revealed, that the nutrient content in soil positively correlates with the litter substrate quality, showing that higher amounts of soil nutrients are related to a good quality of litterfall, and lower contents to a poor litter quality.

Industrial and agricultural procedures have resulted in an increased mobilisation and deposition of potentially toxic metals, which presents a major threat to the environment and to the human health (Shotyk et al., 1998; Novák et al., 2003). From an ecological point of view, the acid rains considerably affect the soil properties. The declining value of soil reaction leads to toxic elements release (Dubová and Bublinec, 2006). The environment quality in the Žiarska kotlina basin is considerably influenced by industry and agriculture. The long-term production by the Al smelter SNP Ltd caused that the soil in the central part of region was contaminated anthropogenically. Air pollutants from industrial plants also appear to be significant sources of soil pollution and there is also noticeable soil contamination caused by traffic pollutants along the transport corridors (Izakovičová et al., 1998).

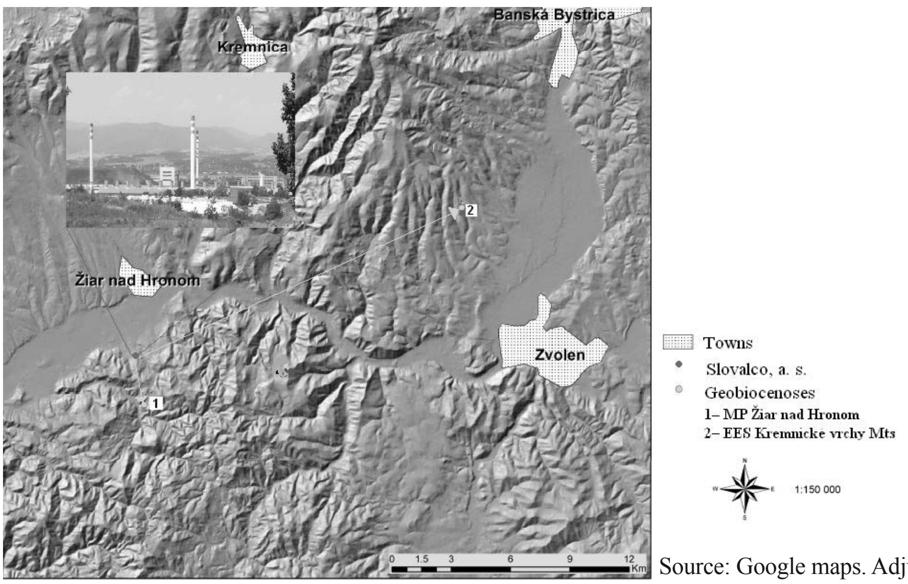
Despite the pollutants-reducing measures implemented, the damage to the surrounding environment seems a long lasting problem (Slovalco, 2014). In general, aluminium smelters emit into the environment many substances: fluorine compounds, aluminium, polycyclic aromatic hydrocarbons (PAH), polychlorinated dibenzo-p-dioxins, dibenzofurans, and traces of heavy metals (Wannaz et al., 2012; Jamnická et al., 2007). Arsenic, one of the trace metals, creates adverse effects on the environment and on human health, due to its toxicity and bioaccumulation. The major cause of human As uptake is through drinking water contaminated from either natural geological sources or from anthropogenic activities like mining, agricultural sources, combustion processes and metal production (Adriano, 2002).

Several studies from the Slovenské stredohorie Mts show that the available forms of soil macronutrients were higher in the control beech stand (Kremnické vrchy Mts) compared with the stressed stand (Žiar nad Hronom), with the differences significant for Ca and Mg. In the plant assimilatory organs, somewhat higher macronutrient contents were found in the stressed stand, with the exception of Ca (Kuklová et al., 2015). Pollutants wash out the base cations (Ca, Mg, K, Na) from the top soil layers and accelerate the primary mineral weathering. The decline in soil organic matter and in available nutrients indicate severe devaluation and extensive soil degradation processes in the Slovak Republic (Kobza and Gašová, 2014). Therefore, the objective of this study was to verify whether different immission loads from the Al smelter affect the (1) macronutrient (Ca_t, Mg_t, K_t, Na_t) and (2) arsenic contents in litterfall and in surface humus (Oo) in mature nudal beech stands.

Materials and methods

Study site

The research was performed on two monitoring plots situated at different distances from the emission source – the Al smelter Žiar nad Hronom. The monitoring plot (MP) Žiar nad Hronom (stressed plot) is located at a distance of 1.5 km from the emission source; the Ecological Experimental Stationary (EES) Kremnické vrchy Mts (control plot), 18 km from the emission source (Fig. 1).



Source: Google maps. Adjusted by Gašová

Fig. 1. Location of the research plots and the emission source.

The description of the nudal beech forests and climatic characteristics on the study plots is in Table 1. The plots are situated in the 3rd forest altitudinal zone, the temperate climate region with an average annual temperature of 6–7 °C (EES Kremnické vrchy Mts) and 7–8 °C (MP Žiar nad Hronom) and an average annual precipitation of 700–800 mm on both plots (Miklós et al, 2002). The stand forming species is mono-dominating *Fagus sylvatica* L. In the undergrowth synusia of both phytocoenoses, mesotrophic herb species dominate, which is characteristic for the forest type group *Fagetum pauper* (Zlatník, 1976). The pH_{H2O} value of soil (0–5 cm) on the MP Žiar nad Hronom (4.6–4.7) is approximately by one unit lower in comparison with pH_{H2O} on the EES Kremnické vrchy Mts (5.8–6.3) (Kuklová et al., 2015).

Table 1. Basic information on studied forest ecosystems

Geobiocenoses	G1	G2
Study site	MP Žiar nad Hronom	EES Kremnické vrchy Mts
Geographic coordinates	48°32´01´´, 18°51´53´´	48°38´08´´, 19°04´18´´
Altitude (m)	450	500
Exposition	NNW	WSW
Slope (°)	5–10	15–20
Vegetation altitudinal zone	3rd oak-beech	
Edaphic-hydric order/ suborder	Waterlogged/ alternately waterlogged	Leading/ normal
Edaphic-trofic order/ interorder	A/B hemioligotrophic	B mesotrophic
Group of forest geobiocoen types	*Fagetum pauper superiora*	*Fagetum pauper inferiora*
Stand age (years)	90	110
Stand density	0.7–0.8	0.8
Stand canopy (%)	90–100	90
Parent rock	Rhyolitic tuff, tuffite	Andesite tuffaceous agglomerates
Soil type/subtype	Stagnic Cambisol (WRB)	Andic Cambisol (WRB)

Data collection

On each monitoring plot, there were diagonally installed 5 litterfall traps, each with a capture area of 0.5 m^2 located at a height of about 1.5 m above the ground. The litterfall was sampled depending on the physiological state of the beech stand and on the dynamics dying and falling away. The sampling was dated to: 1 September 2014, 15 September 2014, 24 September 2014, 16 October 2014, 31 October 2014, 12 November 2014, 1 December 2014, 15 December 2014.

The soil units were determined and classified according to the Societas pedologica slovaca (2014) and the IUSS Working Group WRB (2014). The surface humus samples were taken from the subhorizons Oon – necrotic, Oof – fermentation, Ooh – humification, on square miniplots (0.1 m^2), in three random repetitions on a plot. The samples were collected from patches with predominant *F. sylvatica* species. The thickness of Oo varied: plot G1 (MP Žiar nad Hronom) Oon: 2–3 cm, Oof: 1–2 cm, Ooh: 1 cm, plot G2 (EES Kremnické vrchy) Oon: 2 cm, Oof: 1–2 cm.

The dry weight was obtained by drying the samples for 48-hours at a temperature of 80 °C to a constant weight measured with a precision of 0.002 g. The total content of As in samples was determined by an AAS-GTA using a Thermo iCE 3000 Series. Chemical analysis of arsenic was performed at the Central Forestry Laboratory of the National Forest Centre in Zvolen. Concentrations of Ca_t, Mg_t, K_t, and Na_t were obtained by extraction with *aqua regia* and subsequent using atomic absorption spectrometry (GBC SensAA, Braeside, Australia).

Data analysis

The analyses were performed with using a Statistica 9 software (Tulsa, USA), and the variability of the measured characteristics between the monitoring plots was tested with an ANOVA model and the Tukey test.

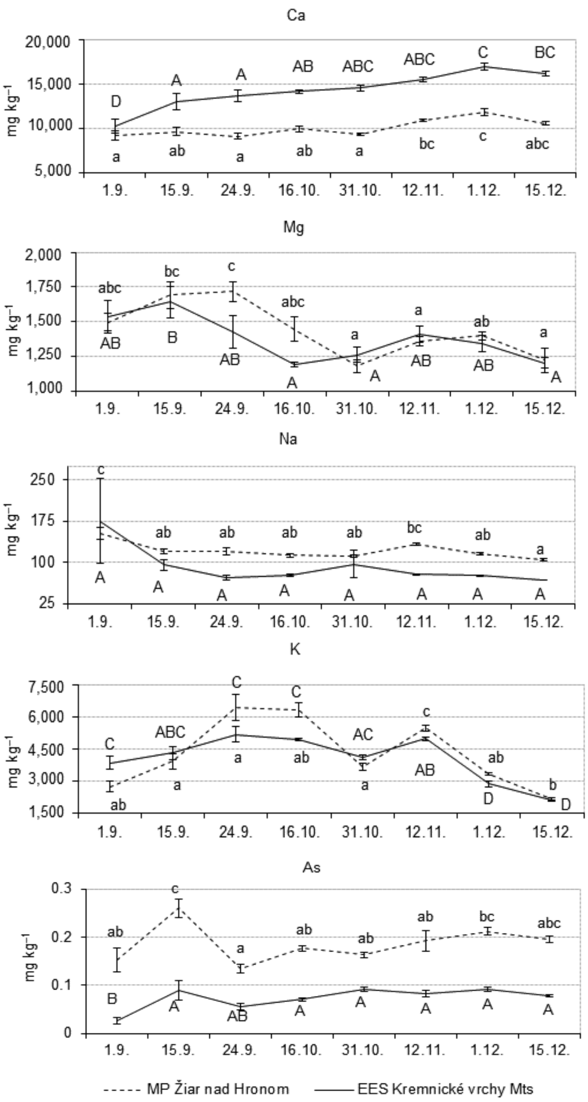


Fig. 2. Dynamics of average contents of nutrients and arsenic in beech litterfall in 2014.

Results and discussion

The results indicate that the higher Ca content was in beech leaves litterfall on the control plot EES Kremnické vrchy Mts (1.4 times more than on the stressed plot by Žiar nad Hronom). In contrast, the mean contents of magnesium, sodium, potassium and arsenic were higher on the MP Žiar nad Hronom (1 time, 1.6 times, 1 time and 5 times, respectively). The calcium in leaves is shown in Fig. 2.

The difference in the average Ca content in leaves between the compared plots was 29.72% and it was statistically significant ($F_{(1,14)}$ = 26.852; p = 0.00014) (Table 2). The values of Ca content in the surface humus show that Ca content decreased with depth on the stressed plot (Fig. 3). The average Ca contents in the subhorizon Oon were 11,720.09 mg kg^{-1} and 19,962.38 mg kg^{-1} on the MP Žiar nad Hronom and the EES Kremnické vrchy Mts, respectively. The difference was big, representing 41.29% and it was statistically significant ($F_{(1,4)}$ = 43.749, p = 0.0027). The average Ca content was higher in the subhorizons Oof than in the subhorizons Oon for both monitoring plots. The difference in Ca content in Oof subhorizons between the compared plots was nearly 45% ($F_{(1,4)}$ = 41.434, p = 0.003). The lowest average Ca content was found out in the humification subhorizon (Ooh) (Table 2). The results show that lower Ca content in beech leaves and in surface humus of the stressed stand was apparently due to lower pH of the top soil. This fact is in agreement with the data reported by Geisler et al. (1998) who state that the soil pH significantly affects the Ca availability to plant species. For instance, Šimková (2014) found an amount of 16,602 mg kg^{-1} of Ca in surface humus of a native beech stand in the Štiavnické vrchy Mts which is by 43.5% more than the Ca content in our stressed stand and by 18.9% less than in our control beech stand. Carnol and Bazgir (2013) found approximately equal amounts of Ca in beech litterfall of the Duke's forest (south-eastern Belgium) compared to the control samples, this is, however by 29% more than displayed samples from our stressed plot. According to Pelíšek (1964), simultaneously with predominance of calcium, soil became enriched with Ca and Mg. Similar finding were attained in our study beech ecosystems, where Ca content exceeded Mg content by approximately 87% and 91% on the MP Žiar nad Hronom and EES Kremnické vrchy Mts, respectively.

Magnesium is an essential component of chlorophyll and it affects the assimilation rate. Mg concentrations in beech leaves reached medium variability values on both monitored plots (Table 2). Stronger Mg accumulation was found in the leaves taken from the stressed stand (Fig. 2).

Based on our findings, the Mg content in the surface humus increased with depth (Oon < Oof < Ooh) (Fig. 3, Table 2).

Table 2. Contents (mean ± SE) of nutrients and arsenic in beech litterfall and in subhorizons of soil surface humus (Oon, Oof, Ooh)

		MP Žiar nad Hronom							EES Kremnické vrchy Mts						
		Min	SE	Max	SE	Mean	SE	*cv*	Min	SE	Max	SE	Mean	SE	*cv*
Leaves	Ca	9,070.00	336.09	11,775.00	341.04	10,010.63***	342.43	9.68	10,216.27	785.61	16,945.00	409.22	14,245.16	741.97	14.73
	Mg	1,180.00	46.36	1,715.00	73.56	1,437.50	91.19	13.55	1,190.00	15.00	1,640.00	111.97	1,375.51	56.32	11.59
	Na	104.80	1.68	151.80	11.54	120.68	5.30	12.34	67.20	0.86	174.88	76.83	91.81	12.44	38.33
	K	2,165.00	99.56	6,465.00	624.28	4,276.58	525.23	35.78	2,120.00	28.93	5,159.00	366.26	4,066.38	384.63	26.75
	As	0.13	< 0.01	0.26	0.02	0.19	<0.01	21.28	0.03	< 0.01	0.09	<0.01	0.07	<0.01	30.93
Oon	Ca	11,040.04	210.32	12,224.22	275.68	11,720.09**	352.98	5.22	17,880.90	563.29	22,009.93	225.18	19,962.38	1,191.94	10.34
	Mg	1,313.58	32.64	1,376.90	23.67	1,334.94	20.98	2.72	1,294.13	41.74	1,505.52	24.56	1,384.78	64.56	7.60
	Na	103.44	6.03	133.46	5.16	116.72*	8.84	13.12	81.84	5.22	83.03	4.08	82.59	0.38	0.79
	K	1,492.75	111.46	1,606.96	70.86	1,568.82	38.04	4.20	1,236.83	56.03	1,967.88	135.43	1,694.21	230.16	23.53
	As	0.20	0.01	1.70	0.26	1.10	0.44	71.16	0.10	0.03	0.80	0.17	0.40	0.19	73.59
Oof	Ca	10,650.40	778.21	12,355.89	210.93	11,561.54**	495.78	7.43	18,460.93	1,603.00	23,207.86	2,139.69	21,008.30	1,381.31	11.39
	Mg	1,233.73	33.67	1,459.89	45.90	1,335.28*	66.30	8.60	1,534.16	57.93	1,755.73	206.08	1,634.14	64.87	6.88
	Na	110.21	7.22	146.06	9.30	127.24	10.39	14.14	117.60	4.87	178.27	24.79	152.64	18.13	20.58
	K	945.11	61.39	1,148.50	97.82	1,026.86	62.14	10.34	1,311.50	35.13	1,729.89	105.36	1,490.53	124.44	14.29
	As	1.10	0.18	5.30	0.82	3.10	1.23	68.26	0.40	0.22	4.20	1.74	2.10	1.11	92.76
Ooh	Ca	4,400.40	620.22	5,50	779.41	4846.67		11.86							
	Mg	1,343.77	112.05	1,624.55	210.45	1,410.10		4.42							
	Na	350.52	83.03	397.85	37.84	379.59		6.70							
	K	1,564.61	190.02	1,939.26	214.91	1,690.78		12.73							
	As	34.40	5.86	66.70	7.87	55.60		33.00							

Significant differences in the average element contents in samples (*$\alpha < 0.05$, **$\alpha < 0.01$, ***$\alpha < 0.001$) between compared plots; cv, coefficient of variation (%); SE, standard error (mg kg^{-1}); Min, minimum value (mg kg^{-1}); Max, maximum value (mg kg^{-1}); Mean (mg kg^{-1}).

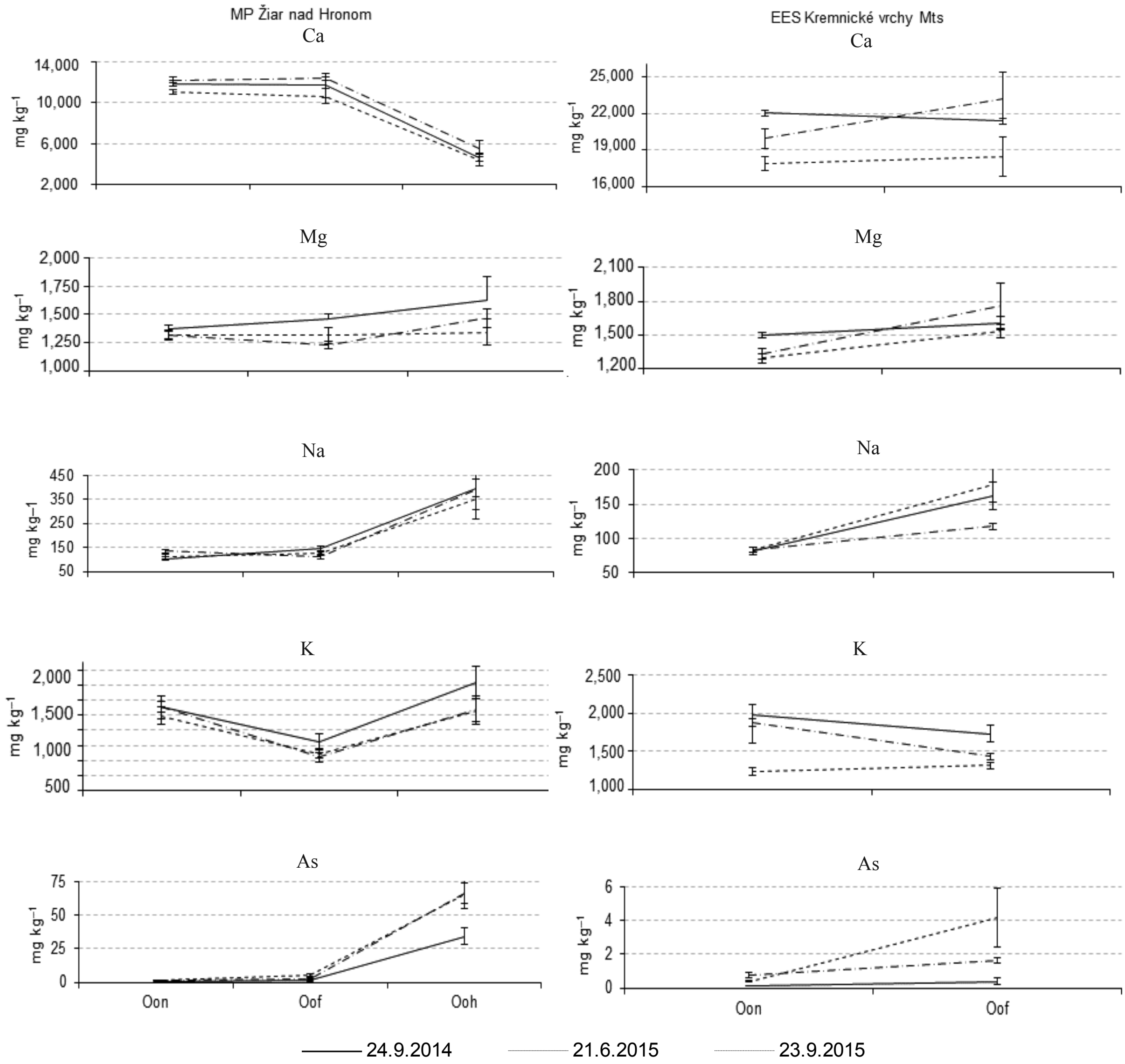


Fig. 3. Dynamics of average content of selected elements in surface humus in 2014–2015.

Carnol and Bazgir (2013) report Mg content by approximately 49% lower in beech leaf litter compared to our results. Ditmarová and Kmeť (2002) studied the health state of beech trees in terms of the latent damage caused by immission load in 1996–2000 on plots in the Slovenské stredohorie Mts. According to these authors, the highest average contents of Mg in assimilatory organs were 2,175.00 mg kg^{-1} (EES Kremnické vrchy Mts) and 2,130.00 mg kg^{-1} (MP Žiar nad Hronom). These Mg values were higher compared to our results. This can be probably attributed to the fact, that the authors analyzed fresh assimilatory organs. Based on the study by Staelens et al. (2011), their oak leaf fall (*Quercus robur* L., *Quercus rubra* L.) was poorer in Mg content, however, their birch litterfall (*Betula pendula* Roth) was richer compared to our beech litterfall.

Higher K accumulation in beech leaves was recorded in the samples from the stressed stand (Fig. 2), with values ranging from 2,165 to 6,465 mg kg^{-1} (Table 2). The K content in our beech litterfall was much higher than those recorded by Kavvadias et al. (2001) in northern Greece, but approximately equal to the K content in fir litterfall reported by the same author. On the other hand, Berger et al. (2009) found K content 1.5 times higher than our results indicate. Potassium is not a structural component of plant litter and it is removed by physical leaching (Swank, 1986).

The average K concentration in the Oon sampled from the Kremnické vrchy Mts was 1.1-times higher compared to the Štiavnické vrchy Mts. However, the difference was statistically insignificant. The average K contents in the subhorizons Oof were 1,026.86 and 1,490.53 mg kg^{-1} on MP Žiar nad Hronom and EES

Kremnické vrchy Mts, respectively. The difference between the compared plots was statistically significant ($F_{(1,4)}$ = 11.141, p = 0.029). The highest K content was detected in Ooh subhorizons (1,690.78 mg kg^{-1}), (Table 2, Fig. 3).

On average, a higher accumulation of K in the surface humus was found out in an altered hornbeam stand in Štiavnické vrchy Mts (2,310 mg kg^{-1}) (Šimková, 2014). Bublinec (1994) reports the value of 2,004 mg kg^{-1} as the average K content in the surface humus in beech ecosystems in the Kremnické vrchy Mts. Compared with our results, the values of K content observed by this author were lower by 6% (control plot) and 16% (stressed plot).

The content of Na in the beech leaf fall from the EES Kremnické vrchy Mts varied from 67.20 to 174.88 mg kg^{-1}. In the leaves from MP Žiar nad Hronom, it ranged from 104.80–151.80 mg kg^{-1} (Table 2, Fig. 2). Higher mean Na content in litterfall is known, for example, from González-Arias et al. (1998) dealing with *Pinus radiata* litterfall from more polluted areas of the Basque Country.

Na content showed an increasing trend with surface humus depth (Fig. 3). The between-plot difference in the average Na content in the subhorizon Oon was statistically significant ($F_{(1,4)}$ = 14.878; p = 0.018). The higher average content was detected in Ooh subhorizon on plot MP Žiar nad Hronom (Table 2).

The results show that higher values of arsenic in beech leaves were found on the MP Žiar nad Hronom compared to the control stand (Table 2, Fig. 2). Porter and Peterson (1975) report some plant species growing on As mine wastes (south-west England, UK) containing average arsenic levels ranging from 350 to 2,040 mg kg^{-1}. Another example is the mean concentrations of As in leaves of plants growing near a copper mine (northern Peru) ranging from 111 to 1,651 mg kg^{-1} (Bech et al., 1997). Temple et al. (1977) found average As contents of 5.80 mg kg^{-1} in grass samples and 7.40 mg kg^{-1} in tree and shrub foliage 700 m away from a secondary lead smelter; with the samples collected at a control site containing < 1 mg kg^{-1}.

The higher As content was detected in the stressed stand in both subhorizons – Oon and Oof (Fig. 3). The differences between the compared monitoring plots were 60.04% and 33.97%, respectively, however, without statistical significance. The average As content in the subhorizon Ooh represented 55.60 mg kg^{-1}. This was by 98% and 94% more than the As content in the Oon and Oof subhorizons (Table 2). Tang et. al (2015) studied a total As concentration in a timberline area in the east part of the Tibet Plateau. These authors obtained the total mean As concentrations in leaves, litter horizon and soil mineral horizons (A, C) fluctuating as follows (mg kg^{-1}): 0.12 < 16.51 < 26.72 mg kg^{-1}. According to the results presented in our study, the litter horizon tended to accumulate also high concentrations of As in the beech leaves. The results indicate that the As amounts detected in the surface humus (Ooh) on the plot MP Žiar nad Hronom were beyond the limit value for agriculture soils (30 mg kg^{-1}) stated by the Act No. 220/2004. Markert (1996) sets a background value for As in plants as 0.1 mg kg^{-1}. The average As content in beech litterfall on the MP Žiar nad Hronom was by 47.4% higher than the declared value.

For comparison, we present here Zn and Hg concentrations in the surface humus samples taken from the two plots in 2007. Their values indicate that mercury (MP ZH: 0.18–0.65 mg kg^{-1}, EES KV: 0.09–0.12 mg kg^{-1}) and zinc (MP ZH: 41.15–174.77 mg kg^{-1}, EES KV: 71.42–114.00 mg kg^{-1}) in samples from the MP Žiar nad Hronom and the EES Kremnické vrchy Mts were lower than the limit values (Fig. 4).

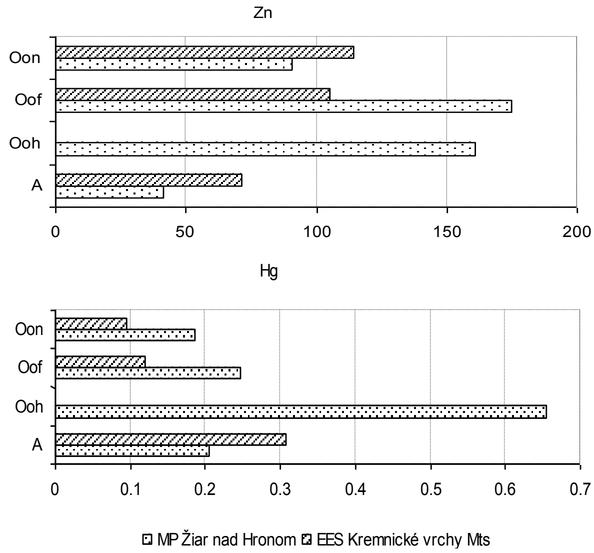


Fig. 4. Content of risk elements in surface humus in 2007 (mg kg^{-1}).

Conclusion

Based on the results, significant differences between plots were observed for Ca content in the litterfall of beech leaves as well as of surface humus. A result of Ca content in surface humus proved that Ca content decreased with depth in the stressed plot. In control stand, there was found more or less increase of Ca content with depth. Mg_t, K_t and Na_t were markedly higher in litterfall of stress stand compared to the samples from control plot. On the contrary, surface humus samples from control plot were richer in nutrients. The concentration of arsenic in samples of surface humus was higher in stressed stand and considerably increased with depth of organic horizon. The results showed, that As amounts detected in surface humus (Ooh) and in beech litterfall of stress stand were higher than limit values, thus pointing at the persistent negative impact of industrial activity on the environment of Žiar territory.

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