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Degradation of Forest Soils in the Vicinity of an Industrial Zone

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Abstract

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Forest soils near industrial zones can be endangered by acid deposition and by dust deposition containing potentially toxic elements (PTEs). Soils of the study area are acidified and the surface enrichment with Cd, Cu, Pb, and Zn reflects anthropogenic contamination. Two forms of all PTEs were evaluated: potentially mobilized (2M nitric acid extraction) and mobile (0.01M $CaCl_2$ extraction) – the most toxic form. Negligible amounts of Cu and Pb were found in the mobile form. Pb mobilization is decreased by co-emitted bases and Cu mobilization is mainly controlled by soil reaction. These elements represent just a potential risk for the ecosystem. The mobile forms of Cd, Zn, and Mn account for approximately 30% of potentially mobilized forms in organic horizons. These elements could pose a problem to ecosystem vitality. Cd is toxic at small concentration and its content in mobilized form approaches the critical load. Cd can be considered the most dangerous element in the study area. Zn concentration is not reaching the limit value. Mobilization of Zn and Pb is mainly controlled by Ca and Mg content. The highest concentrations of Mn were found in the mineral horizons. It predicates a geogenic origin. The lowest percentage of the mobile form is in mineral horizon and its mobilization is controlled mainly by pH. Based on these results, a direct damage of forest by PTE contamination in the Silesian Beskids can be excluded. Lower contamination level along with acid condition and P deficiency could act as a permanent stress factor. Stressed forest is more predisposed e.g. to frost or insect damage.

Keywords: acidification; contamination; toxic elements

Heavy industrial activities such as metallurgy are inseparably bound to coal combustion. The combustion of fossil fuels is the main source of acidificants (CO_2, SO_2, NO_X) of the atmosphere. Additionally, the smelting of ores together with coal combustion are sources of metal-rich dust. Subsequently, potentially toxic elements (PTEs) (As, Cd, Cr, Cu, Hg, Mn, Ni, Pb, Zn) associated with these processes come into contact with the soil as deposited dust particles. The deposition of mineral acids with negligible complexing potential (H_2SO_4, HNO_3) leads to the mobilization of potentially toxic elements (Stevens *et al.* 2009). The relationship between their concentrations in soil solution and soil pH was observed and proven by many researchers (McBride *et al.* 1997; Römkens & Salomons 1998; Pierzynski *et al.* 2000). However, the influence of soil pH on their behaviour is strongly modified by the presence of complexing fractions of soil organic matter (Hernandez *et al.* 2003).

Although the emissions from industrial production can potentially be transported over long distances (Steinnes *et al.* 2005; Steinnes & Friedland 2006), the majority is usually deposited in close vicinity to the source (John *et al.* 1976; Fernandez-Turiel *et al.* 2001; Rawlins *et al.* 2006). In the case of forest soils, deposited atmospheric contaminants generally affect the surface horizons, i.e., organic horizons; these surface soil layers are the most dynamic part

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of the forest environment (Fisher & Binkley 2000) and their contamination and acidification leads to damage of the whole forest ecosystems (Koptsik *et al.* 2004; Jamnická *et al.* 2007).

The study area (the Silesian Beskids, Figure 1) is an example of an area with prevailing combined effects of contamination and acidification of forest soils and forests in places showing evidence of recently deteriorating health conditions (Šrámek *et al.* 2008). Kulhavý and Klimo (1998) suppose that the principal direct factors of forest discolouration and defoliation are probably of biotic or meteorological nature, soil condition can act as a long-term predisposing stress factor. These problems exist mainly in spruce forests and similar forest degradation symptoms were reported from the Polish part of the Silesian Beskids (Bytnerowitz *et al.* 1999; Grodzki 2007).

This study focuses on the behaviour and fate of metals (Cd, Cu, Pb, Zn, and Mn), whose direct phytotoxic and long-term harmful character is known (Merian 1991; Adriano 2001; Brady & Weil 2008). The chemical form of the soil elements plays an important role in their potential bioavailability and toxicity (Kabata-Pendias & Pendias 1992; Adriano 2001) and therefore needs to be studied in detail. This paper aims to qualify negative aspects of soil degradation by acidification and contamination in the study area. The contamination by PTEs, whose toxicity is most pronounced in their mobile forms and their mobilization is influenced by soil condition, is focused. The results could be applied in the studies evaluating tree vitality or their damage.

MATERIAL AND METHODS

Study area. The study area is located in a mountainous region in the eastern part of the Czech Republic (Figure 1) known as the Silesian Beskids. The area is roughly delimited by the Czech-Polish state border in the east, the Olše River in the south-west, and Hluchová and Střelma streams in the north. The altitude ranges between 400 and 990 m in the mountains and the area is characterized by relatively high annual precipitation (up to 1300 mm). Average annual temperature is 6–7°C. Geologically, the area belongs to the External Western Carpathians, especially to the Krosno Nappe Group of the Flysch Belt which is characterized by alternating sandstone and claystone layers (Plašienka 2002). Prevailing soil types are Cambisols and in the highest parts of the mountains Haplic Podzols. Forests in this area are mainly composed of monocultures or mixtures of spruce (*Picea abies* L.) and beech (*Fagus sylvatica* L.) and less commonly of fir (*Abies alba* Mill.).

This region is influenced by nearby industrial zones, firstly by Třinec ironworks located about 10 km from the centre of the study area, and secondly by the Ostrava-Karviná black coal mining and smelting industrial zone (approximately 40 km). In the beginning of the 1980s, Třinec ironworks emitted 27 000 t SO_2 and 12 000 t dust per year. These emissions were decreased by the end of century to 2000–3000 t SO_2 and approximately 1000 t of dust per year. Since then, emission levels stagnate (Třinecké železárny a.s. 2012).

Methods. Soil samples were collected from 25 soil pits spread over the whole study area (Figure 1). Haplic Podzol was identified at three topmost situated soil pits and other soils were classified as Cambisols (IUSS 2006). Soil samples (97) were collected from sufficiently deep horizons (Table 1). In 5 cases, two samples of cambic horizon were collected as there was a slight colour change in the profiles. The average values obtained by measurements of these two samples were then statistically analyzed. The samples were air dried at room temperature and sieved through a 2 mm mesh before analyses.

Basic soil characteristics were determined by the following methods: humus quality was assessed by the ratio of absorbances of pyrophosphate soil extract at the wavelengths of 400 and 600 nm (A_{400}/A_{600}, Pospíšil 1981). Available nutrient contents (P, K, Ca, Mg) were measured in Mehlich III extract (Mehlich 1984). Elements detection was performed by UV

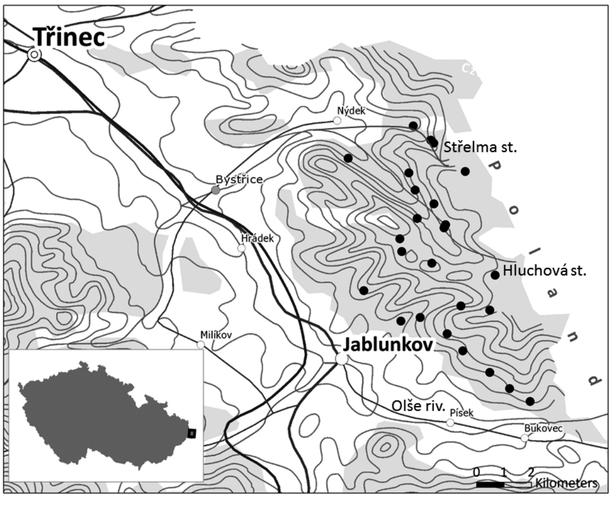

Figure 1. Map of the Czech Republic and a detail of the study area with indicated soil pits

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Table 1. Description of soil samples collection

Soil horizon	Abbreviation	Average thickness (cm)	Collected samples amount
Organic litter horizon	L	2	0
Organic fermented horizon	F	3	25
Organic humified horizon	H	4	25
Organomineral humic horizons A	A	2	3
Eluvial albic horizons	Ep	8	3
Cambic horizon	Bv	20	30
Spodic horizons	Bspod	10	4
Cambic horizon with stagnic colour pattern	Bvm	11	2
Substrate horizons	C	not measured	5

VIS for P (SPECORD 50 PLUS, Analytik Jena AG, Jena, Germany), and by AAS for K, Ca and Mg (GBC 932 AB PLUS, GBC, Braeside, Australia). Cation exchange capacity (CEC) was determined by the Bower method with Na^+ as an index ion (Heese 1998) determined by AAS (Varian SpectrAA-200, VARIAN, Mulgrave, Australia). Hydrolytic acidity (Ha) was determined by the Kappen method (Kappen 1929), whereby sodium acetate reacted with soil to produce acetic acid which was then titrated with sodium hydroxide. Soil pH active in water suspension (pH_{H_2O}) and exchangeable in 1M KCl extract (pH_{KCl}) was determined potentiometrically.

Contents of potentially mobilized forms of Cd, Cu, Pb, Zn, and Mn were assessed using a 2M nitric acid extraction (1:10, w/v) (Borůvka *et al.* 1996). Contents of mobile forms of these elements (mCd, mCu, mPb, mZn, and mMn) were assessed using 0.01M $CaCl_2$ extraction (1:10, w/v). This solution simulates natural soil solution and element contents approximately correspond to their water soluble and exchangeable contents. Concentrations of elements in both extracts were determined by AAS (Varian SpectrAA-200, VARIAN, Mulgrave, Australia).

Only the largest sample sets (i.e. from F, H and Bv soil horizons) were processed by statistical methods. Contents of P and Mg were often below detection limit (<10 mg/kg) in the Bv horizon. Thus statistical analysis was not performed in these cases. Basic statistical analysis, correlation, regression, factor analysis, and one-way analysis of variance were performed using STATISTICA 9.1 software (StatSoft Inc., USA).

RESULTS AND DISCUSSION

Selected statistical parameters of basic soil characteristics are presented in Table 2. Increasing A_{400}/A_{600} ratio corresponds to decreasing humic:fulvic acids ratio. Quality of organic matter is the lowest in cambic Bv horizon, probably due to higher mobility and easier transfer to mineral soil horizons of fulvic acid (Stevenson 1994). The nutrient pool is sufficient in surface organic horizons with the exception of P, which could be immobilized by Al. Aluminium is released from silicates during acidification process and easy Al-phosphates formation and their difficult dissolution is documented (Sposito 1996). Thus,

Table 2. Selected statistical parameters of basic soil characteristics

	A_{400}/A_{600}	P	K	Mg	Ca	Ha	CEC
		(mg/kg)				(mmol/100g)	
F horizon							
Mean	9.4	39	284	122	1119	98	158
Median	9.1	37	284	123	1110	92	157
Minimum	7.9	18	140	67	547	65	94
Maximum	11.9	64	442	277	1690	200	222
SD	1.0	14	60	38	340	28.4	34.9
H horizon							
Mean	8.0	21	178	58	417	67	87
Median	8.0	19	178	57	388	64	72
Minimum	6.1	11	129	38	165	27	36
Maximum	11.1	58	254	94	877	138	235
SD	1.1	9	32	15	172	23.8	40.3
Bv horizon							
Mean	13.4	N.C.	48	N.C.	117	16	19
Median	12.1	N.C.	44	N.C.	106	16	19
Minimum	7.1	N.C.	20	N.C.	47	5.3	7
Maximum	24.1	N.C.	117	N.C.	245	30	39
SD	4.4	N.C.	21	N.C.	53	6.5	8.0

SD – standard deviation; N.C. – not computed (most values below detection limit; < 10 mg/kg); Ha – hydroloitic acidity; CEC – cation exchange capacity

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P deficiency could negatively affect tree vitality. The pool of all nutrients is far lower in the mineral horizons as was also reported by Novotný *et al.* (2008). Values of CEC decrease with depth. It is obvious the CEC is connected mainly with organic matter in these soils. Values of CEC are thus higher in organic horizons than in mineral horizons. High values of hydrolytic acidity (Ha) are documenting the acid character of these soils. Computed base saturation (BS = (CEC – Ha)/CEC) is approximately 38% in F horizon, 23% in H horizon, and 16% in Bv horizon. These rates are typical for Dystric Cambisols (IUSS 2006).

Tables 3 and 4 show selected statistical parameters of studied PTEs and their forms together with pH_{H_2O} and/or pH_{KCl}. Generally soil pH could reflect the anthropogenic acidification and low values of pH accompanied by the presence of PTEs could be stress factors for the forest system. For this reason, the actual form pH_{H_2O} is evaluated with mobile PTEs form (Table 3) and potential form pH_{KCl} with potentially mobilized form of PTEs (Table 4). Values of both pH measurements confirm the strong acidic character of the studied soils as was previously mentioned, with slightly increasing values with increasing depth.

Evaluation of PTEs contents is problematic due to the actual absence of official limit values for forest soils and for PTEs mobile form. There exist limits for Cd (0.1 mg/kg), Cu (50 mg/kg), Pb (70 mg/kg), and Zn (100 mg/kg) in 2M HNO_3 extract for agricultural soils only (Act No. 13:1994, available at http://aplikace.mvcr.cz/sbirka-zakonu/ViewFile.aspx?type=c&id=2733). According to the afore-mentioned limits, Pb in surface horizons exceeds the maximum tolerable content and Cd content in F horizon is close to the threshold value. Generally, uncontaminated soils are expected to have naturally present PTEs decreasing in the following order: Cr>Zn>Ni>Cu>Co>Pb>Cd (Kabata-Pendias & Pendias 1992). Any deviation from this order could be explained as human influence e.g. by contamination of soil, as is the case of Pb there. The comparison of PTEs contents in the study area with background values of the uncontaminated site is the next possibility of PTEs loads evaluation. Fiala *et al.* (2008) observed higher amounts of Pb, Zn, and Cd

Table 3. Selected statistical parameters of pH_{H_2O} and mobile forms of studied elements and their differences among soil horizons based on one-way ANOVA results (95% LSD; letters a,b,c in columns represent homogeneous groups)

	pH_{H_2O}	mCd	mCu	mPb	mZn	mMn
		(mg/kg)				
F horizon						
Mean	3.86^b	0.30^a	0.32^a	1.82^b	15.51^a	39.53^a
Median	3.83	0.31	0.29	1.81	15.36	35.37
Minimum	3.55	0.02	0.02	0.73	8.18	9.38
Maximum	4.50	0.46	0.87	3.34	21.08	105.20
SD	0.22	0.11	0.16	0.72	3.61	25.64
H horizon						
Mean	3.81^b	0.21^b	0.41^a	4.02^a	9.82^b	12.34^b
Median	3.87	0.19	0.39	4.03	8.51	10.49
Minimum	3.38	0.12	0.22	2.09	5.11	3.67
Maximum	4.19	0.52	0.64	6.33	20.51	36.50
SD	0.19	0.10	0.11	1.14	3.64	8.46
Bv horizon						
Mean	4.00^a	0.07^c	0.18^b	0.39^c	3.33^c	12.22^b
Median	4.02	0.06	0.18	0.23	2.92	9.15
Minimum	3.60	0.01	0.05	0.04	0.72	0.12
Maximum	4.31	0.23	0.44	1.03	7.22	51.10
SD	0.20	0.05	0.08	0.33	1.31	11.41

SD – standard deviation

Table 4. Selected statistical parameters of pH_{KCl} and potentially mobilized forms of studied elements and their differences among soil horizons based on one-way ANOVA results (95% LSD; letters a,b,c in columns represent homogeneous groups)

	pH_{KCl}	Cd	Cu	Pb	Zn	Mn
		(mg/kg)				
F horizon						
Mean	2.92^b	0.96^a	27.04^a	240^b	50^a	86^b
Median	2.88	0.91	18.96	245	47	76
Minimum	2.61	0.26	9.61	150.9	30.6	20.4
Maximum	3.68	1.87	73.83	323	77	229
SD	0.21	0.34	19.92	51.0	13.4	54
H horizon						
Mean	2.95^b	0.60^b	24.44^a	277^a	28^b	32^c
Median	2.94	0.54	20.47	280	26	31
Minimum	2.64	0.18	11.05	147.4	14.3	7.4
Maximum	3.39	1.05	61.99	424	55	68
SD	0.16	0.21	12.66	74.1	10.1	17
Bv horizon						
Mean	3.68^a	0.38^c	4.13^b	17^c	14^c	184^a
Median	3.66	0.36	3.98	17	13	163
Minimum	3.14	0.08	2.33	9.1	5.5	33.1
Maximum	4.23	0.71	8.17	32	26	522
SD	0.23	0.15	1.27	5.2	5.3	102

SD – standard deviation

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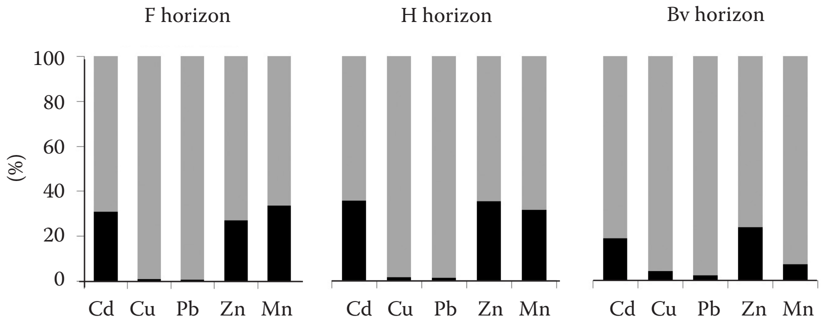

Figure 2. Fraction of mobile forms of elements from the potentially mobilized form in F, H, and Bv horizon

in soils of Jablunkov Pass (southern part of the area studied in this paper) compared to surrounding parts.

The contents of PTEs except Mn are highest in the surface organic horizons. It confirms prevailing anthropogenic origin of the elements that are bound preferably in the surface organic matter (Watmough *et al.* 2004). In the case of Mn potentially mobilized forms are mostly of geogenic origin. The differing affinity for organic matter of the studied elements is apparent from Tables 2 and 3. Mobile forms of Pb and Cu attain the highest concentrations in H horizon. This fact reflects the accumulation of organic matter with bonded elements in this horizon as documented e.g. by Yelpatyevsky *et al.* (1995) or Niemtur *et al.* (2002). On the other hand, concentrations of both Cd and Zn forms, and mobile Mn form are highest in F horizon. This could be explained by their higher bioavailability, since they are somehow incorporated in litter, and by their higher mobility, since they are easily leached during decomposition process and thus they are not accumulated in the in H horizon.

The fraction of mobile form as a proportion of the potentially mobilized form is an important indicator of potential endangerment toward soil organisms and forest ecosystems (Figure 2).

It is clear that negligible amounts of Cu and Pb are in mobile forms. These elements could occur in insoluble minerals of deposited dust. Also, they are known for their affinity to high molecular organic matter and they are strongly bonded together. Thus

Table 5. Correlation analyses among relative amount of potentially toxic elements (PTEs) mobile form and basic soil characteristics (correlation coefficients)

PTEs	pH_{H_2O}	pH_{KCl}	A_{400}/A_{600}	Ha	CEC	P	K	Mg	Ca
F horizon									
$mCd_{\%}$	–0.066	0.126	–0.054	0.168	0.012	–0.066	–0.139	–0.011	0.099
$mCu_{\%}$	0.243	0.110	–0.105	0.005	–0.018	0.131	–0.194	–0.017	0.123
$mPb_{\%}$	–0.280	–0.394	0.204	0.214	0.261	–0.455*	–0.362	–0.600**	–0.538**
$mZn_{\%}$	–0.242	–0.354	0.429*	0.215	0.016	–0.299	–0.208	–0.582**	–0.513*
$mMn_{\%}$	–0.346	–0.230	0.397	0.086	–0.148	0.026	0.075	–0.263	–0.289
H horizon									
$mCd_{\%}$	0.271	0.310	–0.317	–0.178	–0.277	–0.163	–0.200	0.075	0.122
$mCu_{\%}$	–0.519**	–0.588**	0.484*	0.383	0.287	0.464*	0.227	–0.119	–0.325
$mPb_{\%}$	–0.213	–0.295	–0.317	–0.125	–0.444*	0.043	–0.572**	–0.806***	–0.753***
$mZn_{\%}$	–0.205	–0.222	–0.108	0.120	–0.194	0.281	–0.159	–0.319	–0.517*
$mMn_{\%}$	–0.416*	–0.451*	0.316	0.284	0.122	0.317	0.071	–0.126	–0.104
Bv horizon									
$mCd_{\%}$	0.080	–0.186	–0.192	0.169	0.219		0.077		0.443*
$mCu_{\%}$	–0.040	0.273	0.168	–0.098	–0.154		–0.098		–0.354
$mPb_{\%}$	0.267	0.004	–0.262	–0.212	–0.301		–0.254		0.353
$mZn_{\%}$	–0.334	–0.146	–0.084	–0.067	–0.146		–0.004		–0.125
$mMn_{\%}$	–0.546**	–0.740***	–0.368	0.473	0.479*		0.560**		0.481*

*, **, *** represent significant correlation at α = 0.05, 0.01, and 0.001, respectively

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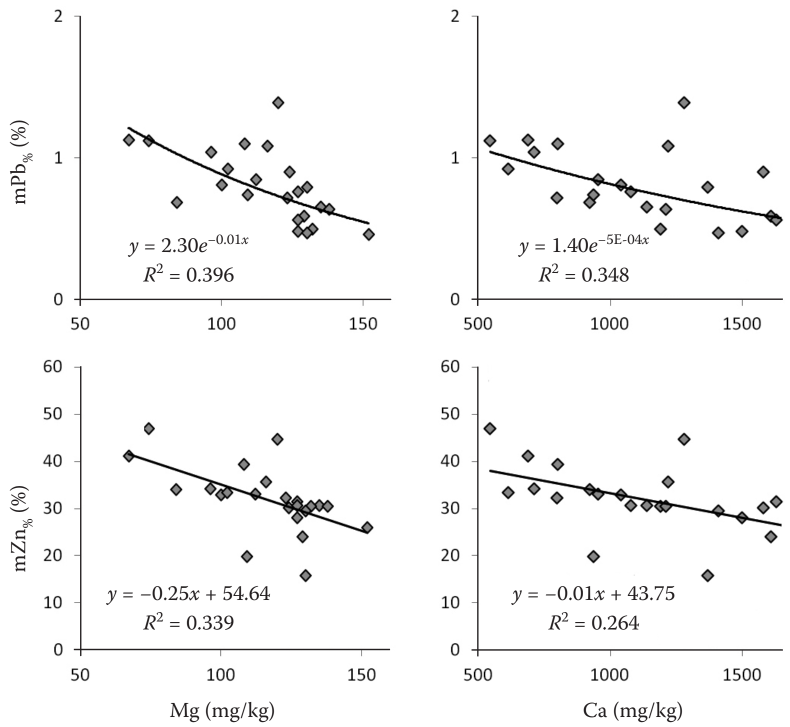

Figure 3. Selected relationships in F horizon with linear or exponential fit described by regression equation and regression coefficient

only small amount can be mobilized (Hernandez *et al.* 2003). Therefore, these elements do not represent high-risk towards the studied ecosystem. Distribution of Cd, Zn, and Mn differs slightly between organic and mineral horizons. Their mobile forms account for approximately 30% of potentially mobilized forms in organic horizons F and H. These elements thus could influence ecosystem vitality. In the mineral Bv horizon, mCd and mZn represent only about 20% and mMn less than 10%.

Correlation analysis (Table 5) documents some weak and several significant relationships among basic soil characteristics and mobile form PTEs percentage of potentially mobilized form (relative

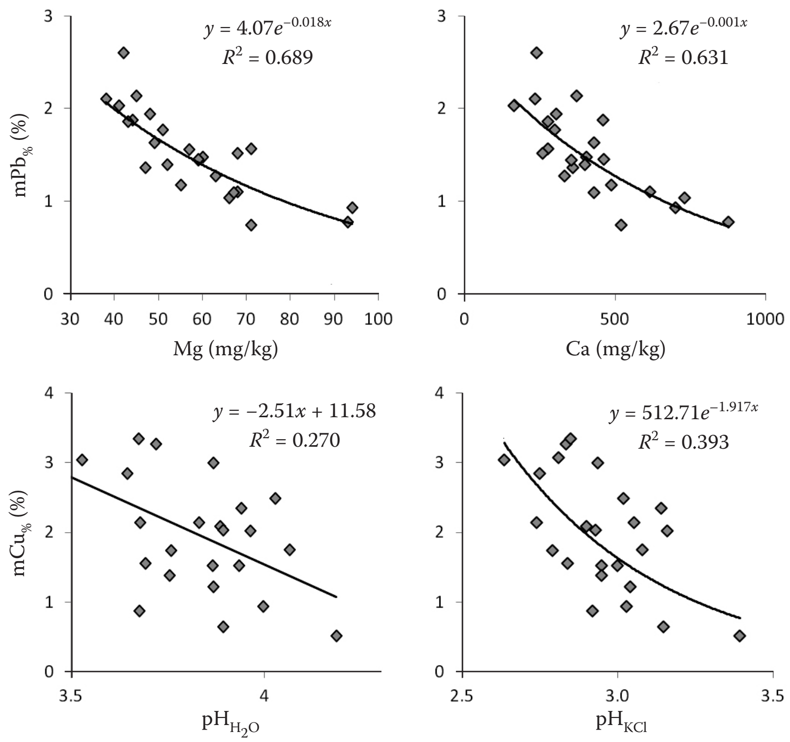

Figure 4. Selected relationships in H horizon with linear or exponential fit described by regression equation and regression coefficient

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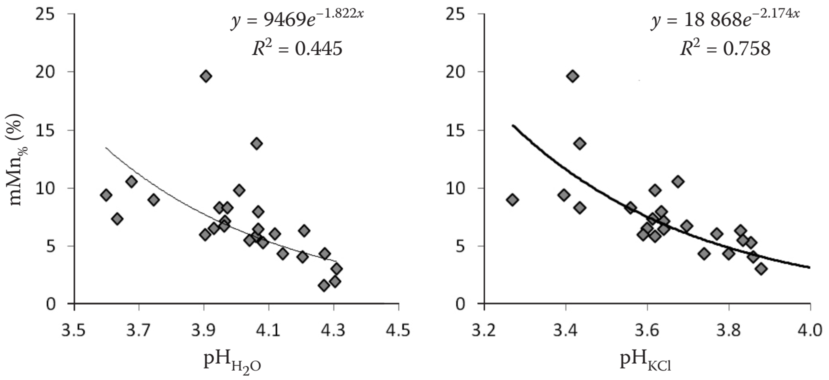

Figure 5. Selected relationships in Bv horizon with exponential fit described by regression equation and regression coefficient

amounts of mobile PTEs; $mCd_{\%}$, $mCu_{\%}$, $mPb_{\%}$, $mZn_{\%}$, and $mMn_{\%}$). These relationships indicate basic soil characteristics ability to control mobilization of PTEs.

The most important relationships are also presented in Figures 3–5. The exponential fit used in several graphs explains the relationships better than do the linear correlations. Relationships between $mPb_{\%}$, $mZn_{\%}$ and Mg, Ca contents are significant in F horizon. A positive effect of increasing Mg and Ca content on Pb and Zn immobilization in soil is apparent. The joint deposition of PTEs with base substances in dusty particles thus particularly prevented stronger damage of ecosystem.

Relationships of $mPb_{\%}$ with Mg and Ca content are stronger in H horizon than in F horizon and stronger are also indirect relationships of relative amount of mobile form of PTEs with soil reaction. There are also significant correlations of $mCu_{\%}$ and $mMn_{\%}$ with both soil reaction types. These results confirm the above-mentioned statement, that decreasing pH increases mobilization of PTEs (McBride *et al.* 1997; Römkens & Salomons 1998; Pierzynski *et al.* 2000). However, soil reaction is not only one control mechanism and does not explain distribution of all studied PTEs in the study area. Increasing humus quality leads to Zn and Cu immobilization in organic horizons, but these relationships appertain to weaker one.

Soil reaction relationship with $mMn_{\%}$ is the most apparent one in Bv horizon. Soil pH increasing leads to Mn immobilization. This relation is also visible in organic horizons, but it is weaker there.

Factor analysis was applied to perform multivariate study of the relationships detected by correlation analysis (Figure 6). This analysis allows a more complex view to cross relationships in the soil horizons. Two factors were selected in each soil horizon. These factors accounted for 50.5, 60.0, and 52.8% of the variability in the original data for F, H, and Bv horizons, respectively. The varimax rotation was used. Positive effect of nutrients (bases) content on Pb and Zn immobilization in F horizons is clearly apparent. Situation is different in H horizon. Pronounced Pb immobilization by Ca and Mg is still apparent, but Zn behaviour is already controlled by different mechanisms. Soil reaction influenced especially Cu mobilization and slightly Mn mobilization. Cu and Mn mobility decreases with pH increase or with hydrolytic acidity decrease. These relationships are also connected with CEC and humus quality. The main part of CEC is formed by acid ions (Ha) and lower

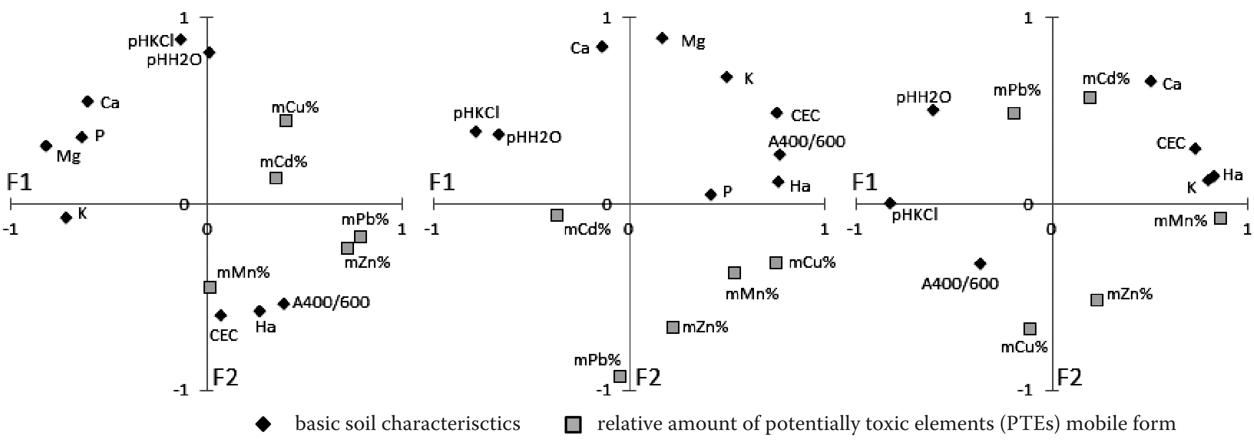

Figure 6. Plots of factor loadings after varimax rotation for F, H, and Bv horizons

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humus quality is typical of acid soils. The relationships of $mMn_{\%}$ and soil acidity indicators are dominant in Bv horizon. No basic soil characteristics control Cd mobilization in the studied soil horizons.

CONCLUSION

The soils of the Silesian Beskids are contaminated by potentially toxic elements, especially by Pb originating from a nearby industrial zone. Pb exceeds the maximum tolerable content in surface horizons. However, high contents of potentially mobilized elements do not represent a direct risk for the whole ecosystem. Only a negligible part of Pb is in mobile form. Pb mobilization is further decreased by co-emitted bases. Toxic effect of Pb is thus probably irrelevant. Also very small contents of Cu occur in mobile (toxic) forms. Mobilization of Cu is mainly controlled by soil reaction. A direct toxic effect is not probable.

A different situation exists for the contents of Cd, Zn, and Mn. These elements are mobilized more easily and their concentrations in mobile forms are higher. Cadmium is toxic even in small concentrations and its content in mobilized form approaches the critical load. No basic soil characteristics significantly control Cd mobilization. The toxic effect of Cd could be higher than of other studied elements and Cd could be comprehended as the most dangerous element in the study area. Zn concentration is not reaching the limit value. Distribution of Zn is controlled by a similar mechanism as Pb. The highest concentrations of Mn were found in mineral horizons. It predicates a geogenic origin. The lowest percentage of the mobile form is in the mineral horizon.

Based on these results, a direct damage of trees by PTE contamination in the Silesian Beskids can be excluded. Lower contamination level together with acid condition and P deficiency could act as a permanent stress factor. Stressed forest is predisposed e.g. to frost or insect damage.

With regard to decreasing emissions, it is important to focus on the stabilization of PTEs deposited to soil in the past. It is necessary to sustain the following conditions: higher pH, higher Ca and Mg content, and relatively high quality and content of organic matter (connected to high CEC). This state could be temporarily achieved by chemical amelioration mainly in the parts of area with visible forest damage.

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