

METAL POLLUTION OF FOREST PHYTOMASS FROM URANIUM INDUSTRY IN CZECH REPUBLIC AND ITS ECOLOGICAL MANAGEMENT PERSPECTIVES

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Abstract

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The paper is focused on the issue of metals migration within the forest environment affected by deep mining of metals and the possibility how to immobilize them using an environment-friendly method. First, the paper presents the information about metal content in the tree leaves in alluvial recipients polluted by metals from uranium deep mining at Dolní Rožínka, the Czech Republic. X-ray fluorescence analysis of dried leaves results showed the increased content of Cu, Fe, Mn, Ni, Rb, Sr, Zn and U; it corresponds to the most seriously polluted areas in the world comparing with the scientific literature. However, statistically, we did not succeed to demonstrate in none of areas of interest the element heterogeneity between the upper, middle and lower streams segments. Element habitat homogeneity can be caused by current stand species composition where *Picea abies* L. dominates and this fact results in the negative impact on the soil pH since it is a primary factor of metals immobilization in the ecosystem and their transformation into toxic variations. Within the area of interest, there is demonstrated positive effect of reconstruction of forest stands, which are close to the dominating deciduous trees, especially *Fagus sylvatica* L. This management change in the selected interested forest stands can result in Ca supply of up to 39 kg.ha⁻¹ from strictly natural sources, which might be a perspective alternative to liming.

Keywords: metals, forest, *Fagus sylvatica*, *Picea abies* L., stream, pH

INTRODUCTION

The research is focused particularly on tree leaves analyses in alluvial environment affected by long-term uranium mining in the Czech Republic and proposing a relevant methodology how to immobilize pollutants in the environment using exclusively management modification and changing forest stand composition or applying afforestation

of new suitable forest stands on non-forest land. Samples for further analyses were taken in the site with current mining activities within the mining area Rožná-Rozchody (deposit Rožná) and the mining area Olší-Drahonín (deposit Olší) where mining was terminated. The research provides the original information about metal content in phyto-mass; any similar research had not been realized so far in

the uranium mining at Dolní Rožínka, the Czech Republic.

The mining on Rožná deposit started in 1957 (Címalá 1997); this deposit has been in continuous operation up to now: it is also the only uranium mine in operation in Central Europe (Government Decree no. 565/2007). The pollution recipient caused by uranium mining in Rožná deposit is found in the Nedvědička stream; in Olší deposit it is found in the Hadůvka stream. Waters of various origin sources, i.e. U, Ra, Cu, Zn, and Ni discharge into the Nedvědička; predominantly, it is treated water from the mine water decontamination plant and leachate from two sludge basins (Váša *et al.* 2015). The Hadůvka stream has been a recipient of treated mine water since 1996; that time, a pumping station and mine water treatment plant were constructed there.

Having finished the mining and flooded the mine in Olší locality, the groundwater flow vitalized, the Hadůvka water level increased and high uranium water concentration up to 2.2 mg.l^{-1} occurred (Hájek and Koscielniak 1997).

The primary issue of mining sites and industrialized regions consists in pollutants deposits (Ianculescu *et al.* 2009), which might be affected by inappropriate forest tree species composition: metals are mobilized and transformed into a toxic shape (Hruška *et al.* 2009). Therefore, in this respect, the utmost importance is put on the forest ability to adjust soil as well as streams pH.

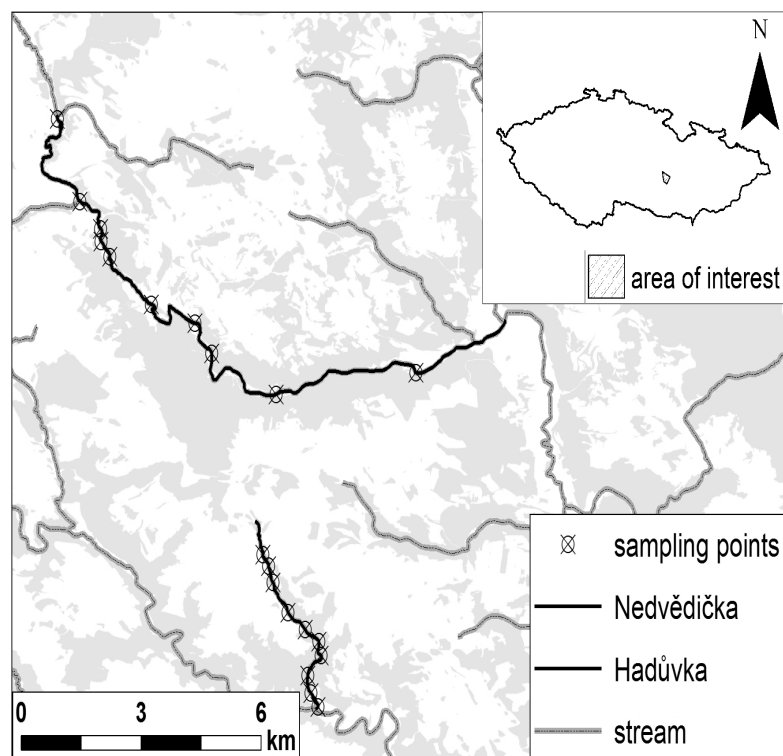
The acidic environment increases solubility and mobility of metals in soil and sediments, their toxicity increases and makes them available for plants: those are, in particular, Mn, Cr, Se, Co, Pb, As, Ni, Zn, U and Cu (Harter 1983; Sandifer, Hopkin 1996; Violante *et al.* 2010). In case of areas of interest, thus there are mobilized particularly Al, Cu, Ni, Zn and U.

Speaking about the prospective closing the Rožná mine, it is advisable to ensure the long-term sustainable ecosystem protection against heavy metals depositions resulting not only from groundwater outflow. This fact can be accomplished applying a relevant woody plants composition. Forest structure with dominating *Picea abies* L. cannot lead to improving the current situation (Souček and Tesař, 2008).

MATERIALS AND METHODS

The research is located in two deposit regions: Rožná-Rozchody (site 1 = Nedvědička stream) and Olší-Drahonín (site 2 = Hadůvka stream).

The streams Nedvědička and Hadůvka are of a stream type: they create the alluvial floodplain in a very limited range. The Nedvědička bedrock consists of amphibolites, gneiss, schists and migmatites. The geological bedrock of the Hadůvka valley consists of amphibolites, migmatites, gneiss (WMS map server, 2016); in the lower parts of the stream there are found syenites (own research). Their flow depends almost entirely on atmospheric



1: The area of interest of Nedvědička (site 1) and Hadůvka (site 2) stream on the forest area background

precipitation (Váša *et al.* 2010). The average annual rainfall reaches 600–650 mm (Tolasz 2007). The average spring temperatures fluctuate within the range of 6–7 °C, summer ones 13–14 °C, autumn 6–7 °C and winter –3 až –2 °C. The average Hadůvka annual flow equals 0.019 m³.s⁻¹ (Šenk 2001), and Nedvědička 0.212 m³.s⁻¹ (Veselý 2006). The forest environment of areas of interest is made up of a commercial forest where coniferous species dominate, in particular, *Picea abies* L. (Úhul 2016). The narrow strip along the Nedvědička and Hadůvka water course is covered with developed fast growing vegetation and ameliorative stands related to higher groundwater subsidies (in particular *Alnus glutinosa* L., *Acer platanoides* L.; personal observation).

Samplings for further analyses were carried out within the time interval October 2014 and 2015. The autumn sampling is represented by wide range of selection and higher concentrations of chemical elements present in samples (Tyler, Olsson 2006). There were sampled leaves from streamside stands, which accumulate high amounts of potential contaminants, metals. Sampled trees in the specific area can be classified as natural stands. The entire deciduous tree species found in the site 1 and site 2 (Fig. 1) streamside stand were sampled (*Aesculus hippocastanum* L., *Alnus glutinosa* L., *Acer platanoides* L., *Acer negundo* L., *Acer pseudoplatanus* L., *Carpinus betulus* L., *Cornus sanguinea* L., *Corylus avellana* L., *Fraxinus excelsior* L., *Prunus avium* L., *Quercus robur* L., *Salix fragilis* L., *Sambucus nigra* L., *Sorbus aucuparia* L.). The aim of research consists in assessing the vegetation impact on the catchment area as a whole and not only its segment represented by a particular species having specific requirements for its environment. The site 1 as well as site 2 is equally represented by 10 sampling points. In terms of site 1, samples were taken from 30 trees within 13,456 m stream segment of interest (443–365 m a.s.l.); considering site 2, samples were taken from 20 trees within 4,015 m stream segment of interest (412–367 m a.s.l.). The higher number of

trees in site 1 is considered as a compensating factor in the large catchment area and longer Nedvědička stream.

The analyses were performed manually using ED-XRF (Energy Dispersive X-ray Fluorescence) spectrometer DELTA. The device uses the latest ED-XRF technology, 4 W X-ray tube, and 200 µA current (max). The XRF method is widely used for environmental analyses. It offers accurate and fast analyses of organic material, soils and river sediments (Salminen *et al.* 2005; Zeng *et al.* 2013; Pouzar *et al.* 2006; Crompton 1996; International atomic energy agency 1997; Luke 1968; Yagi *et al.* 2013). Analyses were carried out using dry material. The mixed sample of one leave species was dried at 65 °C until the constant value (Zeng *et al.* 2013); after that it was crushed and homogenized. The measurement itself was conducted in an original disposable cuvette on a static structure.

The concentration differences of selected elements between studied locations (in individual stream segments) were evaluated using the generalized estimating equation (GEE), which is similar to the analogous generalized linear models (GLM) for data taken on the time scale and abnormal distribution (Zuur *et al.* 2009). The individual elements were analyzed using Gamma distribution with inverse link and correlation coefficient "ar1". The correlation coefficient was selected due to the fact the samples had been taken repeatedly along one-dimensional transect (Zuur *et al.* 2009).

RESULTS

The statistically significant difference (GEE) between the upper (sampling sites 1, 2 and 3), medium (sampling sites 4, 5, 6, and 7) and lower stream segment (sampling sites 8, 9 and 10) was not found out. Resulting from anomalous values occurring in spots, the environment of individual investigated localities is characterized as element-similar (Tab. I).

I: Metals concentration in tree leaves; results of XRF analyses, autumn 2014 and 2015 (mg.kg⁻¹ dry weight)

2014						
		% found sampling points	Minimum	Maximum	Average	P-value
Fe	Site 1	100.00	569.00	16,232.00	3,083.27	-
	Site 2	100.00	1,711.00	17,007.00	3,814.40	
	Site 1	100.00	1,225.00	18,759.00	5,259.80	
Mn	Site 2	100.00	1,820.00	33,200.00	10,253.30	***
	Site 1	3.00	166.00	178.00	172.00	
Ni	Site 2	25.00	225.00	302.00	256.60	-
	Site 1	100.00	382.00	1,193.00	736.47	
	Site 2	100.00	542.00	1,500.00	993.70	
Cu	Site 1	100.00	180.00	1,197.00	390.20	-
	Site 2	100.00	182.00	551.00	362.90	
Zn	Site 1	100.00	8.00	107.00	35.70	**
	Site 2	100.00	13.00	57.00	26.00	

2014						
		% found sampling points	Minimum	Maximum	Average	P-value
Sr	Site 1	100.00	53.00	604.00	196.07	**
	Site 2	100.00	84.00	242.00	146.40	
Zr	Site 1	13.00	13.00	21.00	17.00	-
	Site 2	5.00	18.00	18.00	18.00	
Mo	Site 1	43.00	3.00	10.00	4.78	-
	Site 2	50.00	4.00	13.00	6.50	
2015						
Mn	Site 1	57.00	2,400.00	40,800.00	7,864.71	-
	Site 2	50.00	2,800.00	22,300.00	8,720.00	
Fe	Site 1	60.00	1,282.00	6,800.00	2,656.67	-
	Site 2	45.00	1,144.00	26,800.00	4,524.78	
Zn	Site 1	50.00	220.00	3183.00	655.47	**
	Site 2	25.00	182.00	257.00	222.00	
Rb	Site 1	47.00	32.00	330.00	85.93	*
	Site 2	30.00	23.00	62.00	42.67	
Sr	Site 1	100.00	63.00	399.00	190.23	***
	Site 2	100.00	52.00	187.00	102.70	

*** P < 0.001, ** P < 0.01, * P < 0.05, - Wh no statistical significance

DISCUSSION

There is very little information about the metal content in forest species stands leaves. Having used the information available, it can be stated that elements concentration found in samples from the areas of interest, in terms of average parameters, exceed or match the values measured by other authors in localities loaded by increased depositions of elements found (Tab. II). Both in site 1 and site 2 localities there are Fe, Ni, Cu, Mo, Rb, Mn and Sr. Values in Tab. III are exceeded by Zn (site 1, site 2; according to Krpata *et al.* 2009) in maximum parameters (Tab. II). The statistically significant difference was recorded on the locality site 1 in terms of Rb, Sr in 2014 and Zn, Rb and Sr in 2015. There is no comparable available data on Zr content in leaves. We cannot say unambiguously that area with uninterrupted mining is more seriously polluted than area when mining had been finished.

Current forest at site 2 contains 58.40 % of *Picea abies* L., 24.87 % of *Pinus sylvestris* L. and 2.89 % of *Abies alba* Mill. (Uhál 2016). Deciduous trees occur sporadically (personal observation). However, natural conditions do not support the proper growth of coniferous trees. Souček and Tesař (2008) say that according to the prevailing management set of stands (MSS is characterized by the identical functional focus, natural conditions and stand ratios; relevant details are specified in Decree no. 83/96 Coll.) at the research areas, representation of conifers should not exceed 20 %. Plíva (1987) also said that according to prevailing set of forest types (SFT depends on the ecological relationship and commercial significance of stands), deciduous trees should reach almost 100 %. It follows that the tree

species composition of the forest environment in terms of areas of interest is focused on its commercial use: however, natural ecological forest functions are not considered at all. This situation becomes unsatisfactory in terms of forest impact on soil pH modification (i.e. immobilization metal pollution in ecosystem).

Deciduous trees, which are practically missing in the areas of interest, act on the metals immobilization in the environment in two ways: they modify pH and bind metals of an organic substance from litter. Spur and Barnes (1980) stated that deciduous species are able in one rotation exhaust from the subsoil 3 to 4 times more nutrients (basic cations) than coniferous species. Thus depleted nutrients are transmitted in the form of litter in the upper layers of soil: those are particularly elements such as Ca, Mg, K and partially Na. For some time, they are able to balance (neutralize) the higher sulphur content (from the atmosphere); in case of areas of interest also sulphur deposits from waste water from mining industry. *Fagus sylvatica* L. belongs to woody species characterized by high potential of positive pH soil modification. Kacálek *et al.* (2010) presented pH increase on 12-year beech stands by 0.90 pH units. Hruška and Cienciala (2002) presented the average pH increase for deciduous species comparing to coniferous by 0.70 and at maximum by 1.40 pH units. Further significant factor presented by Hruška and Cienciala (2002) are organic substances in the soil effecting heavy metals immobilization. Heavy metals are bound in organic compounds, which are inaccessible to the plants.

The possible direction for the management of areas contaminated by metals deposition may consist in rearrangement of forest stands and

II: Concentrations of elements in tree dried leaves presented by other authors in contaminated sites and reference localities

Element	Tree leaves	mg.kg ⁻¹	Conc.	Author
Cu	<i>Platanus hispanica</i> Münchh.	38.96	-	Rodríguez-Germade <i>et al.</i> (2014)
	<i>Juglans regia</i> L.	0.34–13.80	UA	Dogan <i>et al.</i> (2014)
	<i>Quercus pubescens</i> Willd.	5.15–6.41	-	Bargagli <i>et al.</i> (2002)
		5.74	+	
Fe	<i>Fagus sylvatica</i> L.	11.00–13.20	-	Maňková <i>et al.</i> (2004)
	<i>Fagus sylvatica</i> L.	163.00–1046.00	-	Maňková <i>et al.</i> (2004)
	<i>Malus domestica</i> Borkh.	123.00	+	
	<i>Prunus persica</i> L.	66.00–124.00	+	Barker a Pilbeam (2007)
		44.00–58.00	-	
	<i>Juglans regia</i> L.	12.72–698.20	UA	Dogan <i>et al.</i> (2014)
	<i>Quercus pubescens</i> L.	112.50–1143.00	-	Bargagli <i>et al.</i> (2002)
		127.00	+	
Mo		0.06	-	
	<i>Citrus</i> L.	0.06–0.09	-	Zekri a Obreza (2015)
		0.10–1.00	+	
		2.00–50.00	-	
	<i>Carya illinoensis</i> Wangenh.	>2.00	+	Heerema (2013)
Mn	<i>Fagus sylvatica</i> L.	836.00–1774.00	-	Maňková <i>et al.</i> (2004)
		458.00	+	
Ni	<i>Tilia tomentosa</i> Moench	1.74–7.27	+	Stanković <i>et al.</i> (2011)
	<i>Phyllanthus</i> L.	1,090.00–60,170.00	Hpa.	Seregin a Kozhevnikova (2005)
	<i>Juglans regia</i> L.	0.13–2.744	UA	Dogan <i>et al.</i> (2014)
	<i>Quercus pubescens</i> L.	3.09–8.21	-	Bargagli <i>et al.</i> (2002)
		4.81	+	
Rb	<i>Fagus japonica</i> Maxim.	10.00	+	Yamagata <i>et al.</i> (1959)
	<i>Quercus crispula</i> Blume	2.00–5.00	+	
	<i>Sorbus commixta</i> Hedl.	4.00	+	
	<i>Acer Tschonoskii</i> Maxim.	6.00	+	
	<i>Acer Mono</i> Maxim.	4.00	+	
	<i>Aesculus turbinata</i> Blume	3.00	+	
	<i>Tilia japonica</i> Miq.	5.00	+	
Sr	<i>Betula</i> L.	15.10–53.90	+	Rodushkin <i>et al.</i> (2015)
		106.00	-	Simon <i>et al.</i> (2011)
	<i>Acer pseudoplatanus</i> L.	54.00	-/+	
		68.00	+	
	<i>Citrus sinensis</i> L.	>7,000.00	-	Wutscher (1999)
Zn	<i>Populus tremula</i> L.	486.00–974.00	-	Krpata <i>et al.</i> (2009)
		340.00	+	
		53.50	-	Maňková <i>et al.</i> (2004)
	<i>Fagus sylvatica</i> L.	30.30–40.50	+	
	<i>Citrus</i> L.	2.00–50.00	-	Zekri a Obreza (2015)
	<i>Carya illinoensis</i> Wangenh	>2.00	+	Heerema (2013)
	<i>Prunus persica</i> L.	17.90	-	Tezotto <i>et al.</i> (2013)
	<i>Malus domestica</i> Borkh.	12.50	-	
	<i>Juglans regia</i> L.	7.362–56.03	UA	Dogan <i>et al.</i> (2014)
		15.00–22.70	-	Bargagli <i>et al.</i> (2002)
	<i>Quercus pubescens</i> Willd.	20.40	+	

“UA” Urban Area; “Hpa” hyper accumulators; “+” reference sites, localities with no load or optimal concentrations; “-” localities affected by pollution, high values

the appropriate desirable management. Having applied site 1 and site 2 areas of interest, we can introduce the basic framework and the benefits of an innovative approach to similarly loaded sites. The basic management stimulus should become a pH soil modification.

The MSS areas were identified in the site 2 locality area suitable for further rearrangement of the entire acreage of 610.69 ha. The active area of the total equals 481.76 ha; it is marked with contour lines from the east and west, with a road network from the north and with the stream Loučka from the south. The active area is understood as the area where the tree species composition is able to effect positively pollutants immobilization in the ecosystem, i.e., without terrain barrier, which can restrict circulation of nutrients in the ecosystem. By Kacálek *et al.* (2010), 12-year beech stand is able at the litter amount (Tab. III) as stated by Dietrich (1963) and Peřina (1960), on the area of 481.76 ha introduce in the environment 8,749.00–18,846.41 kg.ha⁻¹ Ca and 1,218.71–2,625.44 kg.ha⁻¹ Mg. The ration between a spruce monoculture and beech forest is in favour of beech forest in Ca production of 2.13–4.58 : 1 and in Mg production of 2.07–4.46 : 1.

On the site 1 locality it is useful to use the land, which serves currently as production meadows, and convert them to forest land. The total acreage of selected area is 157.89 ha. These newly selected areas in the site 1 locality are situated between the rail and road causeway (they make a hydro-geological barrier), i.e. in the area with no affect of current forest canopy vegetation. 12-year beech stand (by Ca and Mg content as stated by Kacálek *et al.* (2010) and at litter amount as stated by Dietrich (1963) and Peřina (1960) is able to subsidize the ecosystem of an area of 157.89 ha with 2,867.38–6,177.15 kg Ca and 399.42–860.46 kg Mg annually. The Ca (mg.kg⁻¹) litter beech/spruce ratio is 2.13–4.58 : 1 for Mg, then 2.07–4.46 : 1 (calculated by Kacálek *et al.* 2010).

Logging can withdraw high amount of basic cations from the area (Hruška 2009); this fact contributes to acidification of soil, water in watercourses and its sediments (Hruška *et al.* 2006). However, as stated by Hruška and Cienciala (2002), the loss of Ca and Mg can be reduced by 40 % if branches are kept in the logging site. Klimo *et al.* (2001) stated that while keeping branches and bark in the logging site, Ca loss can be reduced by 23 % and Mg loss by 19 %. The most appropriate method for newly established stand might consist in

applying the selective logging, or these stands could belong to areas with limited or fully prohibited commercial logging.

The change of tree species composition can be an appropriate alternative to liming forests. Liming and further support of microbial activity may result in N and K deficiency in the soil (Ouimet and Moore 2015). Liming is able to increase soil pH. It is a comparatively short effect, however, as stated by Deromea and Saarsalmi (1999), 5 years after application of various fertilizers with various dolomite content, at the dosage of 2,000 kg.ha⁻¹ (Ca 420, 98 and 315 kg.ha⁻¹ and 100.61 and 50 kg.ha⁻¹ Mg), the pH value was almost at the same level as before liming. DEROMEA and SAARSALMI (1999) reported a reduction of Zn, Ni and Cu; regardless the fact, the graphs presented in the results show that locality, which is the most seriously exposed to the pollution from the metallurgical industry, offers the trend to the increased concentration of Zn, Ni and Cu in the soil.

The dose of Ca 420.98 kg.ha⁻¹ (Deromea and Saarsalmi 1999) applied through liming can be reached by beech litter subsidy in 12-year beech canopy stand in 11–23 years; the dose of 315 kg.ha⁻¹ in 8–17 years, the dose of 98 kg.ha⁻¹ in 3–5 years. The Mg subsidy from the litter is lengthier than in case of Ca. The dose of 61 kg.ha⁻¹ Mg as stated by (Deromea and Saarsalmi 1999), can be reached in 11–24 years and the dose of 50 kg.ha⁻¹ in 9–19 years (calculated by Kacálek *et al.* 2010; Dietrich 1963; Peřina 1960). The change of tree species composition comparing to liming, has the advantage of long-term effect: in addition, liming has to be repeated regularly so that the effect of desired metals immobilization and pH increase could be maintained. In terms of long-term effect, this fact may become a financial burden for the forest land owners.

III: Litter amount and its element composition (Ca, Mg) on areas with *Picea abies* L. and *Fagus silvatica* L. stands

Tree species	Litter (kg.ha ⁻¹) ^a	Elements content in litter (mg.kg ⁻¹) ^b		Elements from litter to soil (kg.ha ⁻¹) [*]	
		Ca	Mg	Ca	Mg
Fagus silvatica	3,662.00–7,889.00	4,959.20	690.80	18.16–39.12	2.53–5.45
Picea abies	3,214.00	2,656.60	380.40	8.54	1.22

^aDietrich (1963) and Peřina (1960), ^bKacálek *et al.* (2010), ^{*} calculated by Dietrich (1963) and Peřina (1960) and Kacálek *et al.* (2010)

CONCLUSION

The areas of interest show a significant redistribution of metals in woody species leaves. The analyses showed increased concentrations of Cu, Fe, Mn, Mo, Ni, Rb, Sr, Zn and U comparing to other authors who had been conducting research on areas exposed to metal pollution. The statistically significant difference between the metal content between the upper, middle and lower stream segments was not demonstrated on either of sites in question. Alluvial forest ecosystems, being recipients of mining waste from Dolní Rožínka and Olší, are heavily affected; unfortunately, there is no trend to improve the situation in terms of the distance from the source of pollution. Both site 1 and 2 are seriously metal polluted; therefore management of active mining areas is as important as monitoring after finished mining activities. The measures proposed to model examples are based on the principle of converting the stand composition from the dominated *Picea abies* L. to deciduous forests with prevailing *Fagus sylvatica* L. The proposed forest stand management for the localities affected by deep metal mining also pursue the elimination of basic cations loss due to logging. These procedures, although it is a long-term process, are a convenient alternative to forest liming.

Acknowledgement

This research was supported by the Ministry of Education, Youth and Sports of the Czech Republic under the project CEITEC 2020 (LQ1601).

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