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Temporal changes of soil characteristics on Lítov spoil heap, Czech Republic

Enkhtuya Enkhtaivan[1], *Oldřich Vacek*[2], *Petra Vokurková*[1], *Marko Spasić*[1], *Radim Vašát*[1], *Ondřej Drábek*[1]*

[1]*Department of Soil Science and Soil Protection, Faculty of Agrobiology, Food and Natural Resources, Czech University of Life Sciences Prague, Prague, Czech Republic*

[2]*Department of Landscape Architecture, Faculty of Agrobiology, Food and Natural Resources, Czech University of Life Sciences Prague, Prague, Czech Republic*

**Corresponding author: drabek@af.czu.cz*

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Abstract: This study aimed to examine the changes in selected soil properties at Lítov spoil heap (Sokolov, Czech Republic) and compare the current situation with the situation described twenty years ago. A total of 110 soil samples were taken at Lítov at the same sites as in 1998. The analyses of basic soil characteristics involved: exchangeable soil pH (pH_{KCl}), organic carbon content (C_{ox}), quality of humic substances (A_{400}/A_{600}), exchangeable acidity (E_a), and two types of aluminium contents in the soil. Changes in all soil characteristics between 1998 and 2018 were statistically evaluated, compared, and visualized using Geographical Information Systems (GIS). We have observed an increase of pH_{KCl}, C_{ox} and a slight improvement in humus quality compared to the results from 1998. The temporal changes of soil characteristics were evident in the whole area, and the influence of reclamation methods was also pronounced. Soil development close to the regional common natural conditions was found in the area where agricultural reclamation measures (i.e., covering with topsoil) were carried out. Furthermore, afforestation – mainly by deciduous trees – supported the improvement of soil characteristics favourable for plant growth. High pyrite content and marshland were identified as the main causes that led to vegetation cover mortality.

Keywords: acidification; aluminium; anthropogenic soil; mining; pH; reclamation

Surface lignite mining has a massive negative impact on soil and leads to the destruction of soil properties. It impairs the aesthetic appearance of the landscape and the soil structure, and it removes local native vegetation and results in the disappearance of wildlife (Parrotta & Knowles 2001; MacDonald et al. 2015). As a result of mining, soil tends to have adverse physical properties and nutrient deficits, and favours run-off and erosion, with significant environmental consequences, e.g., transported particles may bear toxic compounds such as potentially toxic elements (Pająk & Krzaklewski 2007; Echevarria & Morel 2015). In addition, surface mining significantly changes humus quality, organic carbon content, and concentrations of potentially toxic elements. Soil organic matter content is generally very low, thus limiting pedologi-

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cal processes of great importance (such as aggregation) and the supply of suitable physical properties and nutrients for plants (Echevarria & Morel 2015). Nowadays, an effort is made to restore the disturbed landscape as much as possible and keep nature healthy, fertile, and stable. In agreement with Czech legislation, that effort is concentrated on landscape reclamation and revitalization (Borůvka & Kozák 2001). Soil quality is among the most important parts of restoring a functional ecosystem after mining (Liu et al. 2017). Reclamation type is the key factor that determines the anthropogenic soil formation rate (Hendrychová 2008). Apart from reclamation, unreclaimed sites left to spontaneous succession also exist.

Significant changes in pH concerning site age have been reported. Some researchers (Frouz et al. 2001; Šourková et al. 2005) have studied spoil heaps in the Sokolov mining basin afforested by alders (*Alnus glutinosa*, *Alnus incana*). They reported that pH had changed from alkaline on young sites to slightly acidic on older sites. pH dropped by 0.8 under deciduous trees but by 2.7 under coniferous monocultures over 28 years of development (Kabrna 2011). Similarly, during succession, pH (H_2O) in the topsoil layer has been shown to decrease from an initial value of 8 to about 6.5 (Frouz & Nováková 2005; Frouz et al. 2008).

We hypothesized that anthropogenic soils have significantly changed characteristics over a twenty-year period (dynamic evolution). Moreover, this development seems to be influenced by soil vegetation cover.

Our study aimed to investigate the changes of selected soil properties on Lítov spoil heap and compare the current situation with the situation twenty years ago. Another goal was to evaluate this spoil heap's temporal development and propose necessary measures leading to overall sustainable vegetation cover.

MATERIAL AND METHODS

Study area and sampling scheme. Lítov, a spoil heap created after extensive lignite mining operations had ended, was selected as a study site for this experiment. The location and sampling sites of the Lítov heap are presented in Figure 1. This site allows studying different soil characteristics and analysing the temporal development of new anthropogenic soils. Lítov was created as an external dump of lignite mines Medard and Libík. The overburden (parent) material of the Sokolov area primarily consists of tertiary (cypress) clays and its pH usually ranges around 8–9 (Jačka et al. 2021).

In 1998, 110 soil samples were collected from the same study area (Figure 1) and analysed (Borůvka

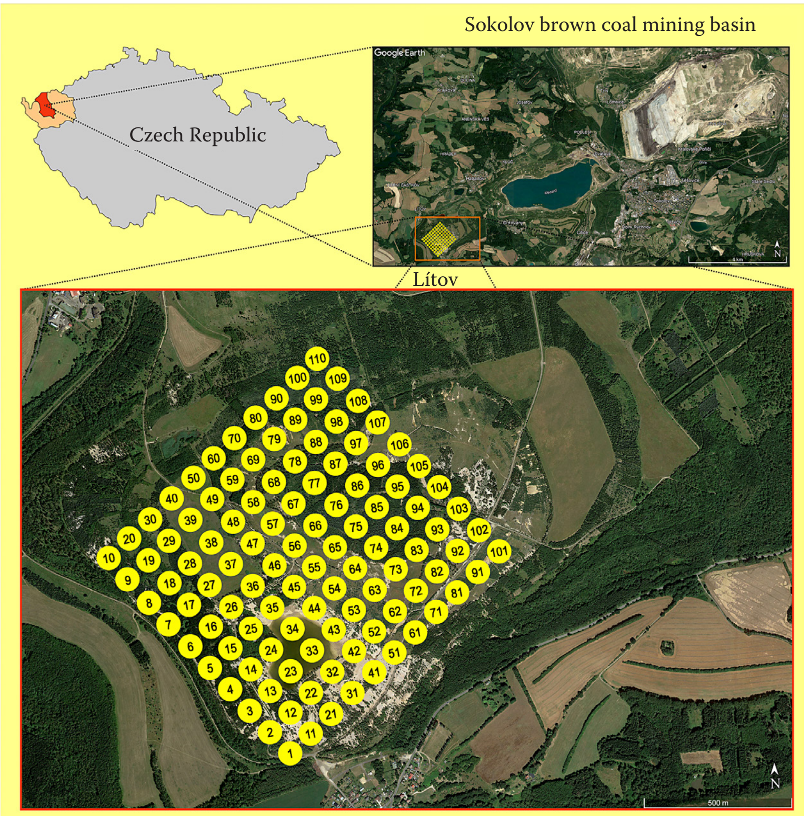

Figure 1. Sampled area with the sampling locations; Lítov spoil heap in the northern Sokolov basin, Czech Republic

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et al. 1999; Borůvka & Kozák 2001). Inspired by their studies, in 2018, a total of 107 composite samples (three of the original sampling sites, No. 23, 33, and 34 on Figure 1, are currently flooded) were collected from a 1.1 km^2 area. Samples were taken by soil auger from the top layer (which encompassed either organomineral (A) horizon developed on top of the mineral parent material (C), or just (C) if no organomineral (A) horizon has formed), 0 to 20 cm. The weight of each sample was approximately 1 kg. As already mentioned by Borůvka and Kozák (2001), the main components of Lítov spoil material were cypress clays, with an admixture of minerals of pyritic nature and brown coal particles.

A map that shows the distribution of different reclamation measures of the study area was done by Borůvka and Kozák (2001) and presented in Figure 2. Agricultural (topsoil covering) and forestry (afforestation with coniferous and deciduous trees) reclamation measures were completed. A certain part of the area was planted with trees, mainly pine (*Pinus* sp.) and alders (*Alnus* sp.), and a small area was turned into a lake whose banks are marshy depending on the season. Part of the area was mixed with natural topsoil, and a small part was covered with topsoil without mixing (Borůvka & Kozák 2001). The elevation of the terrain was initially 450–540 m, and after reclamation, it has increased to 570 m. The rest of the surface was left unchanged to spontaneous succession. The whole study area was characterized by extreme acidity because of the pyrite-containing geological substrate and water in the heap, which make the creation of sulphuric acid possible.

Analytical methods. All laboratory methods measuring selected soil properties, which are the exchangeable soil pH (pH_{KCl}), the quality of humus (A_{400}/A_{600}), the exchangeable acidity (E_a), organic carbon content (C_{ox}), and the content of aluminium in soil (mainly organically bound and so-called labile), were inspired by Borůvka and Kozák (2001) and repeated in our study in the same manner after 20 years. Data for this study were collected using the same methodology to compare with results from the study by Borůvka and Kozák (2001) and to assess the effect of time. All samples were air-dried and homogenized. After that, samples were sieved through a 2 mm sieve. pH_{KCl} and E_a were measured in 1 M KCl extract (1 : 2.5; w : v). The quality of organic matter was described by A_{400}/A_{600} ratio, i.e., the ratio of absorbances of soil in sodium pyrophosphate ($Na_4P_2O_7$) 0.05 M extract (20 mL per

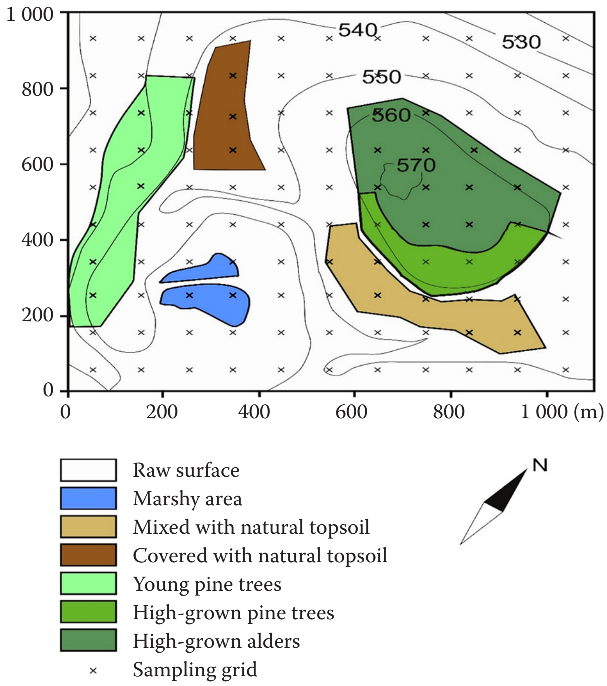

Figure 2. Distribution of vegetation cover and reclamation measures placed on the topographic map of the study area from Borůvka and Kozák (2001)

1 g soil) at wavelengths 400 and 600 nm, by using Hewlett Packard 8453 spectrophotometer (Pospíšil 1981). Total organic carbon content was determined oxidimetrically by wet combustion using potassium dichromate (Pospíšil 1964). Labile exchangeable Al (Al_{lab}) was determined in 1 M KCl extract by the method proposed by James et al. (1983) when Al^{3+} is bound on eight hydroxyquinolines and determined spectrophotometrically using Hewlett Packard 8453 at a wavelength of 395 nm (Borůvka & Kozák 2001). The determination of mainly organically bound Al (Al_{org}) was done according to the methodology described by Drábek et al. (2003). In the solution, the total content of Al was measured by means of ICP-OES (iCap 7000; Thermo Scientific USA).

Data evaluation. All the statistical analyses were performed using Microsoft Office Excel 2019. Descriptive statistics were calculated for all soil characteristics. Data comparing and changes in soil characteristics between 1998 and 2018 were analysed using a paired-sample *t*-test and correlation matrix. Assumptions for the paired-sample *t*-tests (independence, normality, outliers) were met. The output maps showing the differences of the measured values were processed using GIS (ArcMap Version 10. 5. – ESRI Inc.).

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RESULTS AND DISCUSSION

Compared to the results from 1998 (Borůvka et al. 1999; Borůvka & Kozák 2001), some properties (pH_{KCl}, C_{ox}, E_a, A_{400}/A_{600} and Al_{org}) showed slight improvement (increase in pH_{KCl}, C_{ox}, Al_{org} content and a slight increase in humus quality). The basic statistical analyses of soil properties can be found in Table 1.

We applied a paired *t*-test on basic soil characteristics, and the results are shown in Table 2. In 2018, an increase in values of pH_{KCl}, $C_{ox,}$ and in contents of two different forms of Al (Al_{org} and Al_{lab}) was observed.

With the increase of time after reclamation, salt content, alkalinity, and particle size tend to decrease, while organic matter content tends to increase (Wang et al. 2014). Twenty years of development is a very short period of soil formation. However, the influence of this factor on this area is evident. In general, topsoil covering and afforestation have a predominantly positive effect on changes in reclaimed post-mining sites and gradually improve natural soil conditions. This fact can be found in previous studies, for example, Borůvka et al. (1999), Borůvka and Kozák (2001), Shrestha and Lal (2011), and Spasić et al. (2024), which were dealing with soil development after reclamation measures in lignite spoil heaps.

In 2018, pH values ranged from very strongly acidic (2.3) to neutral (7.0) (Table 1). The most common pH was 5.1. 18% of either grassland or afforested areas indicated neutral soil reaction. However, pH values were mainly very low, 45% of the area was very acidic, and 22% of the whole area showed acidic soil reaction. The lowest pH value was found at the southern border of the area of interest near the water reservoir.

A comparison of pH_{KCl} values frequency is displayed in bar graphs in Figure 3. A paired *t*-test was applied for pH_{KCl} values to analyse a significant difference between 1998 and 2018 in time effects. In 2018, pH_{KCl} values were significantly higher than in 1998. The difference was statistically significant ($P = 0.002$, $^{**}P < 0.01$) between these years.

Soil pH increased by 0.1 to 2 units on 36% of the sampling area (Figure 4). However, almost the same percentage (33%) of samples showed a decrease of 0.1 to 2 units (Figure 4). The average pH increased from 4.1 to 4.6. The increase in pH depended on vegetation, time, and climatic conditions, over the fact published by Borůvka et al. (1999). Asensio et al. (2013) also stated that tree planting (*Pinus pinaster* Aiton) significantly affects the increase in soil pH. These findings do not correspond to the data published by Čížková et al. (2018). These authors did

Table 1. Basic statistical analyses of soil characteristics

	pH_{KCl}		E_a (mmol/100 g)		C_{ox} (%)		A_{400}/A_{600}		Al_{org} (mg/kg)		Al_{lab} (mg/kg)	
	1998	2018	1998	2018	1998	2018	1998	2018	1998	2018	1998	2018
Mean	4.1	4.6	3.3	1.0	2.4	3.2	6.0	4.0	1 496	3 677	70.2	143.9
Min.	1.6	2.3	0.1	0.1	0.3	0.5	4.7	2.3	282.0	3.9	0.1	0.1
Max.	7.0	7.0	10.7	5.3	3.5	5.9	9.6	12.0	14 476	13 907	295.5	859.9
Median	3.6	4.6	1.6	0.2	2.6	3.1	5.9	3.4	1 036	2 734	44.2	6.1
Mode	2.8	5.1	0.2	0.1	3.1	4.3	–	–	744.0	3.9	–	1.2
SD	1.6	1.4	3.3	1.4	0.9	1.5	0.6	1.7	1 796	2 817	78.0	214.3
CV (%)	39.4	30.1	102.5	135.8	36.9	46.7	10.7	42.9	120.1	76.6	115.4	148.9

pH_{KCl} – exchangeable soil pH; E_a – exchangeable acidity; C_{ox} – organic carbon content; A_{400}/A_{600} – quality of humic substances; Al_{org} – organically bound Al; Al_{lab} – labile exchangeable Al; SD – standard deviation; *CV* – coefficient of variation

Table 2. Paired *t*-test results of basic soil characteristics

	pH_{KCl}	E_a	C_{ox}	A_{400}/A_{600}	Al_{org}	Al_{lab}
P-value	0.002**	< 0.001***	< 0.001***	< 0.001***	< 0.001***	< 0.001***

pH_{KCl} – exchangeable soil pH; E_a – exchangeable acidity; C_{ox} – organic carbon content; A_{400}/A_{600} – quality of humic substances; Al_{org} – organically bound Al; Al_{lab} – labile exchangeable Al; *, **, *** indicate a statistically significant difference at the *P*-value < 0.05, 0.01, 0.001, respectively

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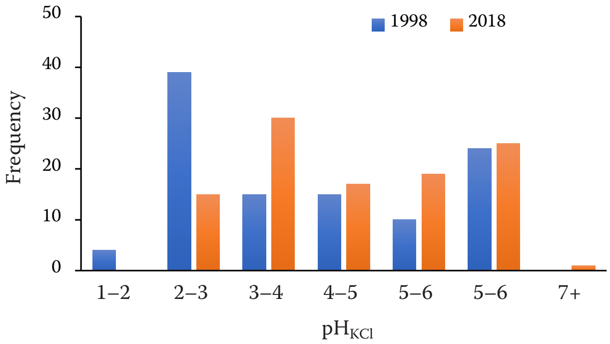

Figure 3. Comparison of exchangeable soil pH (pH_{KCl}) values frequency between 1998 and 2018

not find any changes in pH values depending on time on neither reclaimed nor non-reclaimed spoil heaps (located in Sokolov coal basin, Czech Republic). In contrast, Shrestha and Lal (2011) mentioned the beneficial effect of reclamation measures (replacing overburden, covering with topsoil up to 30 cm) on increasing pH values. They stated that at a depth of 0–15 cm, the pH value increased by four units (from 4.9 to 8.1, i.e., by 31%) on some reclaimed areas compared to the surrounding localities.

Most C_{ox} contents of soil samples were above 2.7%. These samples are thus classified as soils with a very high content of oxidizable carbon. Sample number 79 (Figure 1), where there was no vegetation cover, had the lowest value of C_{ox} (0.5%). The highest value of C_{ox} (5.9%) was determined in sample number 104 (Figure 1), at the site covered with mature coniferous trees. However, a higher value of C_{ox} also can be an effect of brown coal residues. The values of organic carbon content showed a large variation. The organic carbon content could be described mostly as medium to high with an average C_{ox} content of 3.2%. The lowest value of A_{400}/A_{600} was 2.3 in locality number 58 (Figure 1), where humus quality was the finest. Compared to the C_{ox} values from twenty years ago, 46% of the total number of samples showed an increase in the C_{ox} content by more than 1% (Figure 5), and 26% of the measured values increased up to 1% (Figure 5). The average C_{ox} was also increased from 2.7% to 3.2% (very high C_{ox} content).

The reduction of C_{ox} content by more than 1% covered 16% of the whole area (Figure 5). Shrestha and Lal (2011) also found a decrease in C_{ox} content in reclaimed spoil heaps. Borůvka et al. (1999) stated that the lower range of C_{ox} under alder could be explained by the high degree of mineralization due to increased microbial activity. In contrast, Čížková et al. (2018) stated that C_{ox} increases with time and the degree of landfill reclamation. Furthermore, C_{ox} content increases significantly faster on reclaimed post-mining sites than non-reclaimed ones (Čížková et al. 2018). Borůvka et al. (2012) also highlighted the increase in C_{ox} due to topsoil covering. This fact was observed on two spoil heaps from the Czech Republic (Libouš and Pokrok).

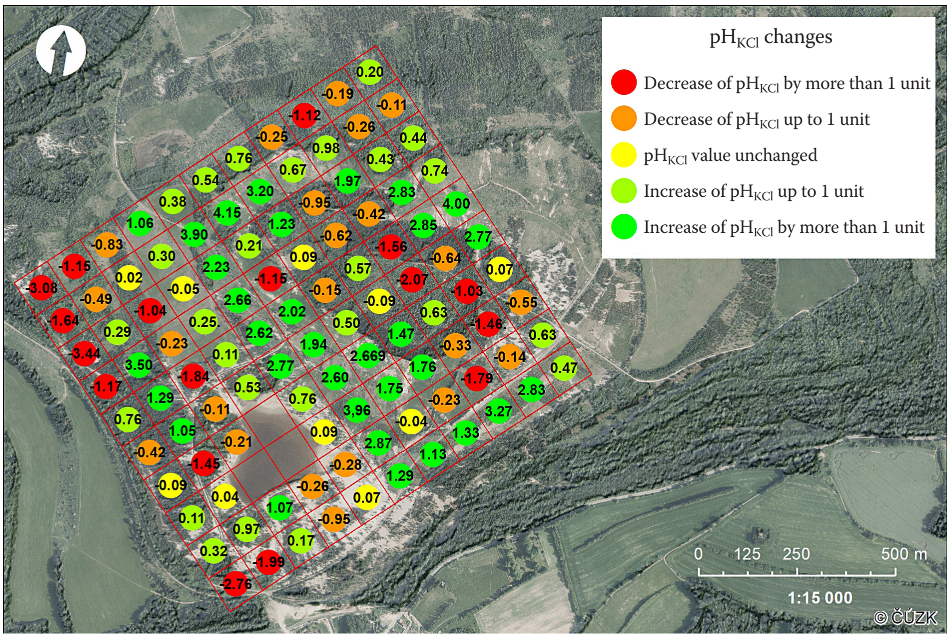

Figure 4. The changes of exchangeable soil pH (pH_{KCl}) (difference between 1998 and 2018) on Lítov spoil heap

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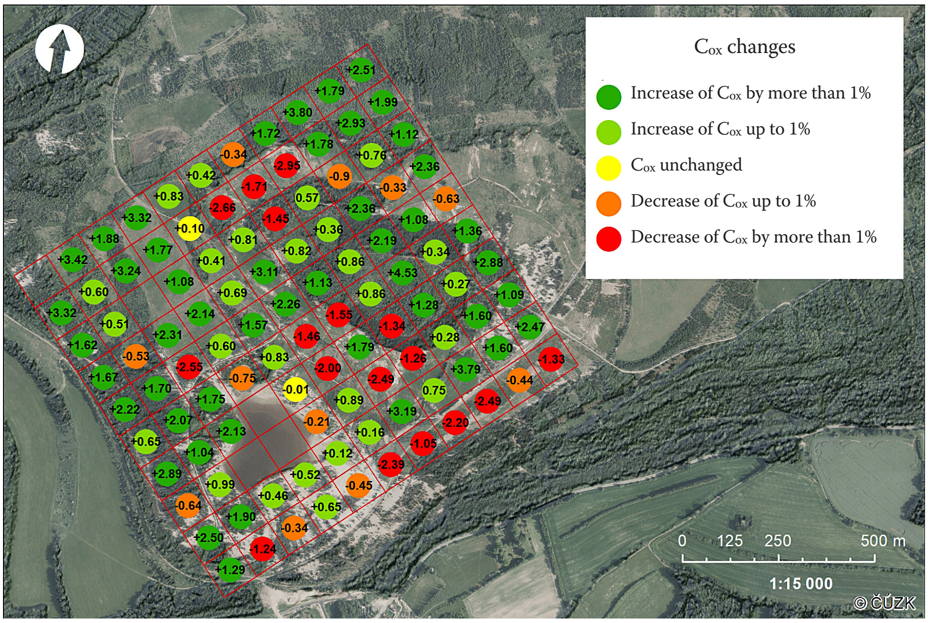

Figure 5. The organic carbon content (C_{ox}) changes (difference between 1998 and 2018) on Lítov spoil heap

According to the A_{400}/A_{600} coefficient, the humus quality was improved. 55% of the whole area showed an improvement in humus quality by up to 3 A_{400}/A_{600} units (Figure 6), and 32% of the samples showed an increase by more than 3 A_{400}/A_{600} units. The influence of vegetation on the C_{ox} content and humus quality was evident. Swab et al. (2017) found that the vegetation mixture, which includes native plants, positively affected the stabilization of humus and potentially improved soil conditions. Borůvka and Kozák (2001) also found that the humus quality was mainly influenced by afforestation (in the given study – afforestation of alders). According to Asensio et al. (2014), tree vegetation, especially eucalyptuses and pines, could significantly increase the mine soils' organic carbon content. The gradual formation

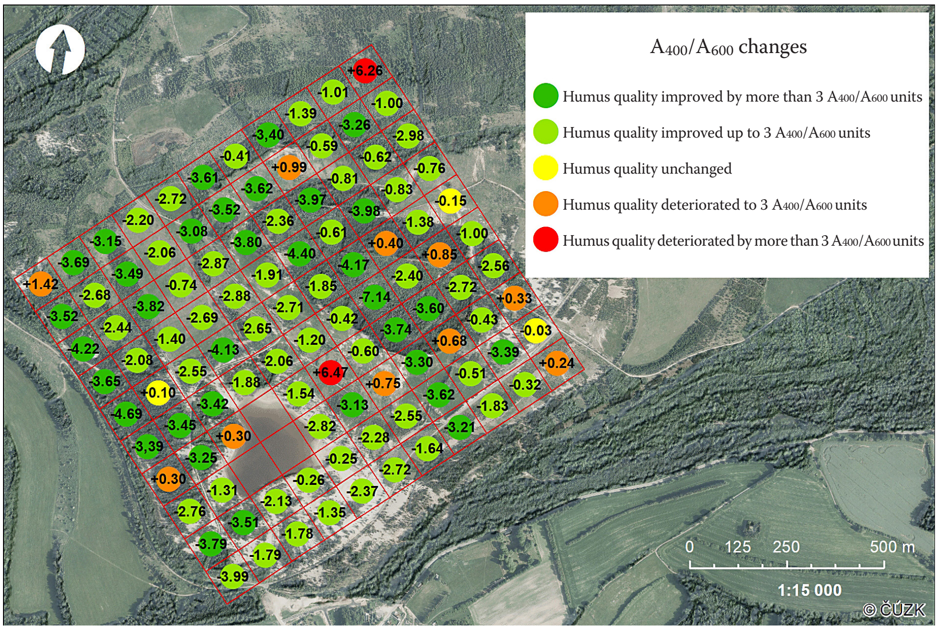

Figure 6. Humus quality coefficient (A_{400}/A_{600}) changes (difference between 1998 and 2018) on Lítov spoil heap

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of more complex soil organic matter led to a decrease of A_{400}/A_{600} values. Borůvka et al. (2012) observed that the natural soil cover also affected an improvement in C_{ox} and A_{400}/A_{600}. Spasić et al. (2023) investigated the influence of various tree species on initial soil-forming processes on post-mining sites in Sokolov region, and Vachova et al. (2022) investigated plant species diversity under different vegetation in this region. Both studies pointed out the positive effects of alders and long-life deciduous trees compared to most conifers tested. In various studies of soil fauna under different vegetation on lignite post-mining sites, it was found that deciduous trees create better conditions for edaphon than conifers (Dunger et al. 2001; Frouz et al. 2001, 2008, 2013; Wanner & Dunger 2001). Dunger et al. (2001) stated that soil biological activity under deciduous reclamations can reach the level of natural woodland soil in only 10 to 20 years, whereas in conifers, biological activity was limited due to the lower decomposing activity.

Most of the area (68%) had exchangeable acidity in a range from 0 to 1 mmol/100 g; meanwhile, the rest of the area (32%) represents very strong E_a (more than 1 mmol/100 g). The locality with the highest E_a value (sample No. 25) was located next to the water reservoir (Figure 1), probably affected by naturally created sulfuric acid. The average E_a value decreased from 3.3 mmol/100 g to 1 mmol/100 g since 1998. 29% of the whole area showed a high exchangeable acidity.

Most of the measured E_a values (46%) showed a decrease by more than 1 mmol/100 g (Figure 7), and 18% indicated a reduction of E_a by less than 1 mmol/100 g (Figure 7). One of the most serious problems on spoil heaps is water, which accelerates rock weathering, and as a result, the rocks disintegrate and release pyrite. This process was thoroughly described by Rimstidt and Vaughan (2003). Jelenová et al. (2018) also found high pyrite content in heaps near Kaňk (Czech Republic). Bussinow et al. (2008, 2012) have investigated wind-blown, pyrite derived acidification after polymetalliferous mining in the forests near Zlaté Hory, Czech Republic, and concluded that although some contamination by potentially toxic elements was present, severe acidification (that led to severe nutrient deficiencies) seemed to be the major process of soil degradation. Pyritic minerals tend to oxidize and form sulfuric acid (Sheoran et al. 2010). The formation of sulfuric acid in the soil environment causes an increase in exchangeable acidity, a decrease in pH value, and soil fertility. At the edge of dumpsites, water outflow, which probably contains a higher amount of sulfuric acid, was observed. Observed mortality of vegetation, including mature pines, was caused by this outflow.

Different forms of Al have different characteristics of mobility, bioavailability, and toxicity in soil. Al is phytotoxic to most plants at soil pH below 5.5 (Schmitt et al. 2016), and a stronger relationship with soil pH was found in the organic horizon (Pavlů et al.

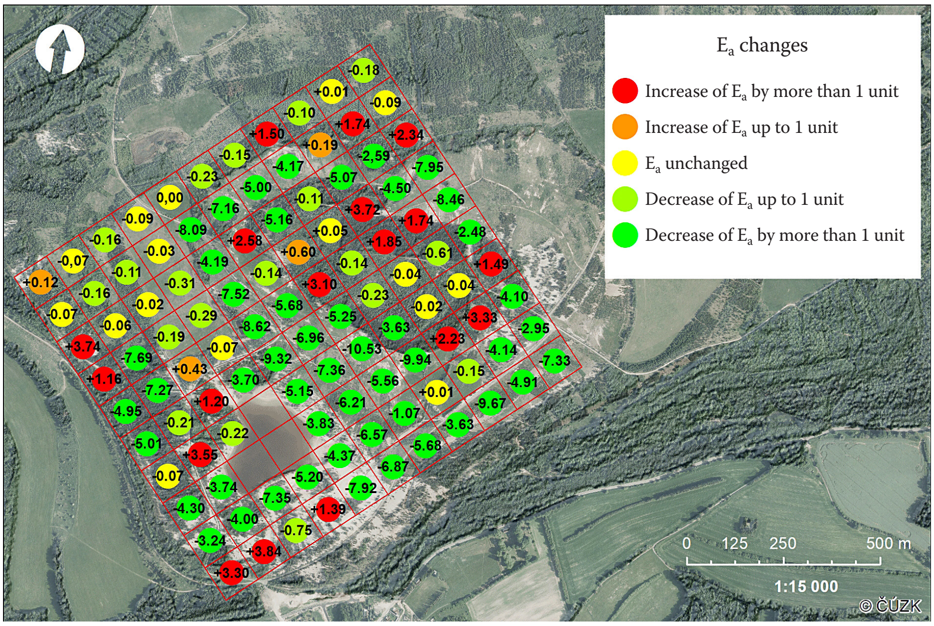

Figure 7. Exchangeable acidity value (E_a) changes (difference between 1998 and 2018) on Lítov spoil heap

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Table 3. Correlation matrix of soil characteristics in 1998 and 2018

	pH_{KCl}	E_a	C_{ox}	A_{400}/A_{600}	Al_{org}
1998					
E_a	–0.87				
C_{ox}	–0.40	0.54			
A_{400}/A_{600}	–0.01	–0.07	–0.39		
Al_{org}	–0.01	–0.11	–0.37	0.13	
Al_{lab}	–0.65	0.70	0.41	–0.10	–0.12
2018					
E_a	–0.43				
C_{ox}	0.06	0.09			
A_{400}/A_{600}	–0.06	–0.07	–0.04		
Al_{org}	–0.41	0.22	0.00	0.03	
Al_{lab}	–0.76	0.56	0.11	0.03	0.40

0.0 to ± 0.2 little correlation; ± 0.2 to ± 0.4 weak correlation; ± 0.4 to ± 0.7 correlated; ± 0.7 to ± 0.9 strong correlation; ± 0.9 to ± 1.0 very strong correlation

pH_{KCl} – exchangeable soil pH; E_a – exchangeable acidity; C_{ox} – organic carbon content; A_{400}/A_{600} – quality of humic substances; Al_{org} – organically bound Al; Al_{lab} – labile exchangeable Al

2019). In contrast, Al_{org} is reported as non-toxic and positively affects the monitored ecosystem (Drábek et al. 2003). Exchangeable and organically bound Al concentrations were higher in the anthropogenically acidified area (Pavlů et al. 2019). Also, Hagvall et al. (2015) and Dang et al. (2016) stated that the content of free Al in soil decreases in the presence of organic materials with a suitable soil reaction (neutral pH).

Both determined forms of Al showed a wide range, and their average values were doubled (Al_{org} 3 677 mg per kg, Al_{lab} 143.9 mg/kg) since 1998 (Table 1). The correlation analyses of determining soil characteristics are shown in Table 3. According to the data from 2018, a closer inverse dependence was presented between the Al_{lab} content and pH_{KCl} (r = –0.76) (Table 3) compared to data from 1998 (Table 3). In contrast, in 2018, correlation analysis of Al_{lab} showed weaker direct dependence with exchangeable acidity (r = 0.56) than in 1998 (Table 3). The weak relationships of Al_{org} to other soil characteristics (Al_{org} with pH_{KCl} r = –0.41, with E_a r = 0.22, and with C_{ox} r = 0.00) were indicated (Table 3) in 2018.

Al_{org} from 2 104 to 4 204 mg/kg covered 36% and 3.9 mg/kg to 2 104 mg/kg in another 34% of the whole area. Sampling points located in partially forested areas (samples No. 65, 84) had minimum values of Al_{org} (Figure 1). These localities also indicated strongly to very strongly acidic soil reactions. 49% of measured Al_{org} values showed an increase up to 3 000 mg/kg (Figure 8), and 32% of all samples showed an increase of more than 3 000 mg/kg (Figure 8). In general, the average value of Al_{org} was doubled after 20 years of soil development (Table 1).

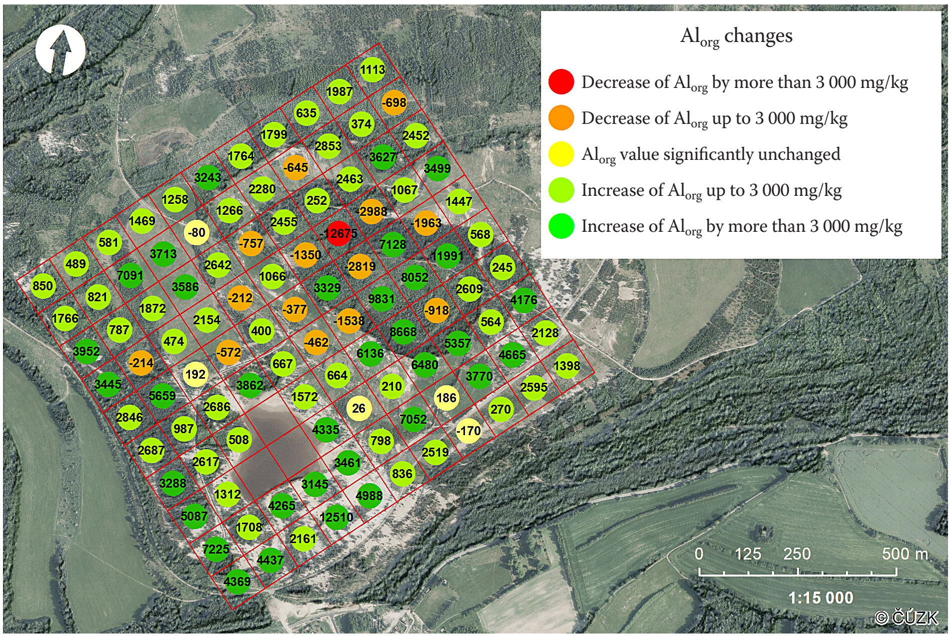

Figure 8. Organically bound Al (Al_{org}) value changes (difference between 1998 and 2018) on Lítov spoil heap

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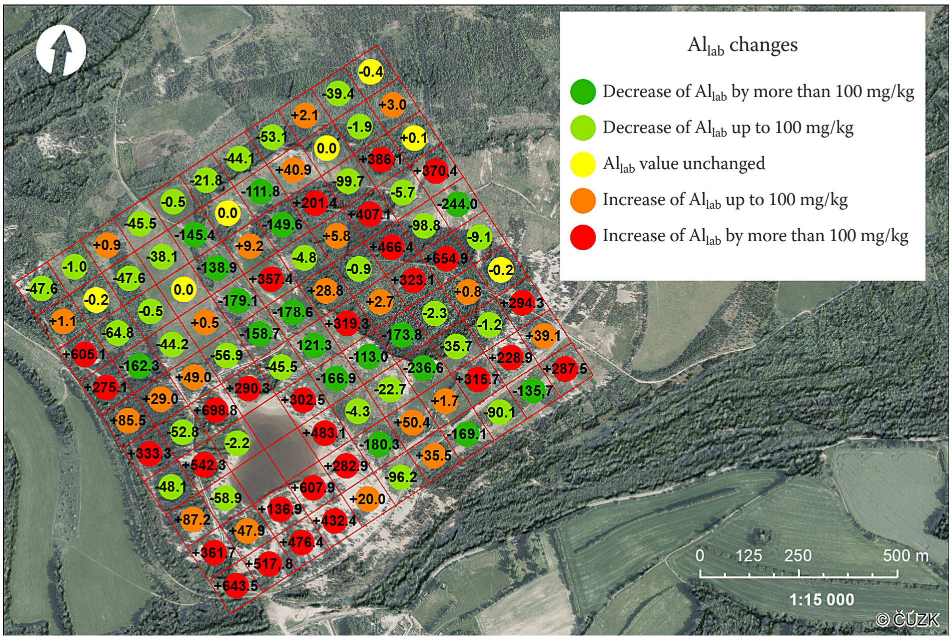

Figure 9. Labile exchangeable Al (Al_{lab}) values changes (difference between 1998 and 2018) on Lítov spoil heap

68% of the territory showed Al_{lab} values ranging from 0.1 to 155.3 mg/kg. The lowest contents of Al_{lab} were determined in areas with a slightly acidic to neutral soil reaction. Bowman et al. (2008) stated that soil acidification accelerates the mobilization of potentially toxic elements (e.g., Al or Mn).

Al_{lab} values' increase was observed in 2018. The results indicated significant differences at levels P = 0.0005 (***P <0.001) between these years.

31% of Al_{lab} measured values were reduced by 100 mg/kg (Figure 9), and 27 % of the samples showed an increase greater than 100 mg/kg (Figure 9). The amount of Al_{lab} decreased with increasing pH values. The average content of Al_{lab} was also doubled (143.9 mg/kg) since 1998. This disturbing increase in Al_{lab} may be due to a strongly acidic soil reaction of the parent material, low C_{ox} content, and coniferous vegetation in certain parts of the study area. Álvarez et al. (2005) stated that aluminium toxicity varies considerably among different species and found the most significant risk of Al toxicity in soils under pine and the lowest under oak. Coniferous stands lead to soil acidification and thus to an increase in Al_{lab} content (Álvarez et al. 2005). Drábek et al. (2003) also found the highest amount of Al_{lab} in soils with the lowest pH_{KCl} and organic matter under forest cover.

According to Kotowski et al. (1994) and Merino et al. (1998), aluminium mobilization depends on acid concentrations and salts and differs between soil horizons. Merino et al. (1998) also found the highest mobilization of Al in the leachates from upper horizons.

CONCLUSION

Humus of greater quality and higher content of organically bound (non-toxic) aluminium were found in areas that were covered by topsoil and afforested (mainly with deciduous trees). On the contrary, high labile (toxic) aluminium content was found under coniferous trees. We observed that in 2018, 45% of the whole area showed a very acidic soil reaction. On these acidic parts of soils, it is possible to carry out liming or afforestation with deciduous trees and promote vegetation cover diversity. The organic carbon content also increased over time. Reclamation measures (e.g., topsoil covering) had affected this rate of increase. The most problematic part of the spoil heap is the marshland, which is located around the water reservoir. This part contains high amounts of pyrite, which produces sulfuric acid. Anthropogenically affected soils are affected by vegetation cover, and soil properties undergo dynamic evolution.

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