



Temporal changes in soil chemical compositions in acidified mountain forest soils of Czech Republic

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Abstract

The study aimed to investigate the temporal changes of pH, sorption complex, and structure of soil organic matter through the forest soil profile under beech and spruce forests located in Jizera mountains (Czech Republic) and affected by natural and anthropogenic acidification. Soil samples were collected in four different years (2008, 2013, 2015, and 2020) in each horizon: fermented horizons (F), humified horizons (H), organo-mineral horizons (A), and subsurface mineral horizons (B) (cambic or spodic). The cation exchange capacity (CEC), base saturation (BS), exchangeable element contents, pH, and soil organic carbon content (SOC) were determined. The infrared soil spectra were used to calculate indices of potential wettability, aromaticity, and decomposition. Our results showed that most nutrients and aliphatic compounds were retained in the uppermost soil layers. The aromaticity of organic matter increased with depth, while polysaccharides, regarding the decomposition compound, disappeared through the soil horizons. In a long-term observation, SOC content had constantly increased under beech, while spruce remained stable in the organic horizons. Exchangeable element contents increased in each horizon, except for Al and Fe; their content quickly decreased in F horizons and slowly decreased in H horizon under both forest tree species, while the deeper horizons remained constant, but increased in A horizon under spruce. Continuously increasing base cations concentrations in sorption complex of both forest tree species during the study period revealed the effect of forest stand types on acid deposition reduction and mitigation. The temporal changes in CEC, BS, SOC, and soil wettability are more intensive in beech than in spruce forest floor.

Keywords Soil properties · Infrared spectroscopy · Forest ecosystem · Environmental acidification

Introduction

Globally, climate change, soil degradation, and carbon loading in the atmosphere have been concerned and have drawn attention from environmental research and policy agendas (FAO 2017). Forest disturbances including natural and anthropogenic disturbances (e.g., drought, deforestation,

agriculture, and forest fire) are one of the most important in responding to global climate change (Sommerfeld et al. 2018). Acid depositions are also included to affect the forests negatively. In the mid-nineteenth century, industrialization and coal mining were rapidly increased with poor regulation and lacked abatement technology; as a result, the nitrogen and sulfur contained in coal ranging from 1 to 15% had been combusted and emitted (Oulehle et al. 2007; Achilles et al. 2021) and caused a large area of European forests and soils severely disturbed and damaged (Krug and Frink 1983; de Vries et al. 2014). Acid deposition adversely affects the soil chemistry and forest ecosystem by increasing the proton H⁺ concentration, leaching of base cations, and mobilization of Al and other potential toxic elements into the soil (Likens et al. 1996; Johnson et al. 2018; Pavlů et al. 2021). Moreover, acid deposition has been reported as a driver dismissing the diversity in the terrestrial ecosystem (Duprè et al. 2010). Oulehle et al. (2007), who studied the long-term effect of

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tree species on soil acidification in the Ore Mountains, found that depletion of base cations resulted in a low base saturation of 8% under spruce and 6.5% under beech forest. On the other hand, some other studies about the long-term recovery from acidification found that the forest soil has been improved in cation exchange capacity, base cations, and total carbon and declined Al concentration in the organic horizons. However, in some severely affected sites, the soils were slowly recovered and did not suddenly return to a good environmental condition, as previously seen (Hruška et al. 2002; Oulehle et al. 2017; Hédl et al. 2011; Johnson et al. 2018). Therefore, changes in disturbance dynamics and forest management need to be implemented and observed in order to sustain ecosystem diversity and mitigate climate change.

Beech (*Fagus sylvatica* L.) and spruce (*Picea abies* L.) are the most abundant forest tree species in Central Europe and have been studied for their various influences on long-term changes in soil physical, biological, chemical properties after acidification (Berger and Berger 2012; Pavlů et al. 2018; Tejnecký et al. 2013). Generally, forest tree species differ in the soil properties such as pH, exchangeable cations, nutrient cycle, soil fertility, and SOC stabilization due to their quality and quantity of litter, root exudation, nutrients uptake, and litter decomposition, depositional processes (interception of deposition such as throughfall, bulk deposition, and stemflow) canopy uptake and leaching (Bradová et al. 2015; Habumugisha et al. 2018). For instance, forest litter such as leaves, branches, and roots from beech and spruce varies in its composition, quality, and quantity. It provides different nutrients with different availability for plants, and decomposition rate through soil microorganisms (Han et al. 2021). Many researchers pointed out that the spruce litters are decomposed more slowly than litters found in the beech forest because they contain a high amount of recalcitrant substances (i.e., lignin, resin, waxes, and phenolic compounds) (Nacke et al. 2016; Richardson and Friedland 2016). Slower decomposition leads to slow cycling of nutrients but stabilizes SOC in the soil profile (Cremer and Pritzel 2017). The SOC stabilization takes place according to the abiotic and biotic processes (Campbell and Paustian 2015) and goes through these mechanisms: conservation due to recalcitrance, the interchange of elements, and inaccessibility to the decomposer community due to the occlusion in the aggregate soil (Poirier et al. 2018). The content of nutrients such as nitrogen (N), calcium (Ca), potassium (K), and phosphorus (P) in the coniferous forest soils is also likely lower than in deciduous forest soils (Iwashima et al. 2012) due to less decomposition and mineralization. It is also shown by Berger et al. (2006) that beech had higher Ca content than spruce due to the association with the effect of Ca-pump under beech resulting from transpiration of beech and uptake water from deeper soil horizons, in which

the soil solution contains more Ca because of weathering supply of the bedrock. Oulehle and Hruška (2005) studied the effects of specific tree species (beech and spruce) in the long-term acidified forest and found that beech forest had a higher accumulation of Ca and K in the soil than spruce forest, but lower concentration of Al. Spruce is considered a strong acidifier and can deplete the base cations in the topsoil layer, whereas beech performs as the Ca-accumulator and increases Ca in organic horizons (Daněk et al. 2019). The high concentration of Ca in the soil relatively increased soil pH, nutrients, base saturation (BS), and biodiversity (Iwashima et al. 2012).

The study hypothesized that beech and spruce forests are able to recover effectively from anthropogenically enhanced acidification and promote soil organic matter and soil nutrients in the forest ecosystem in the long term. The study mainly focused on: (1) evaluating the temporal changes of sorption complex, exchangeable elements and structure of SOM through the forest soil profile and (2) differentiating the chemical soil structures of SOM between beech and spruce forest affected by acidification. However, the infrared spectroscopy was used to evaluate the chemical soil structures of SOM, which were indicated by some indices.

Materials and methods

Site description and soil sampling

The study was carried out in the Jizera Mountains, in the northern part of the Czech Republic along the border with Poland. The region was heavily affected by acid deposition and is one of the most damaged areas in Czech Republic during the 1980–1990 period because of the nearby coal-fired power plant (black triangle – Czech-German-Polish border) (Moldan and Schnoor 1992; Suchara and Sucharová 2002; Sucharova et al. 2011). Mean annual deposition rates in recent years ranged between 2.6 and 8 g NO₃⁻/m² and between 2.6 and 20 g SO₄²⁻/m², while before 1990 the deposition of these acidifiers reached levels higher than 5 g of sulfur and 3 g of nitrogen /m² (CHMI 2023a, b). However, Kopáček and Veselý (2005) reported that S deposition declined linearly from 1990 to 2000, while N deposition declined rapidly during 1989–1994 and then slowly afterward. The composition of the forest tree species in Jizera Mountains in between 2012 and 2020 ranged from 7.7 to 14.9% in beech forest and from 48.8 to 69.1% in spruce forest (Podrázský et al. 2014; Zemědělství 2021). The sampling locality is called Paličnick (50.8683900N, 15.2527000E), where it has been monitored for a long time (Borůvka et al. 2009; Bradová et al. 2015; Tejnecký et al. 2010, 2013); therefore, it was selected for this research as a long-term observation. The elevation of the sampling locality ranges

from 635 to 680 m a.s.l. The average annual precipitation is between 600 and 1200 mm, and the average temperature is roughly between 5 and 9 °C (Remrova and Císlerová 2010; Balcar et al. 2012). However, there was a dry climatic condition in 2015 during the study period observed by Ionita et al. (2017) and shown in Fig. 1. The main vegetation is dominated by the European beech (*Fagus sylvatica* (L.)) within the natural ecosystem of the occurrence of perennial grasses in ground vegetation (*Calamagrostis arundinacea*, *Calamagrostis villosa*), and spruce monoculture forest (Norway spruce: (*Picea abies* (L.) Karst.) along with perennial grasses (*Calamagrostis arundinacea*, *Calamagrostis villosa*). The average height of the forest stands is about 32.4 m in beech forest and 28.6 m in the spruce forest (Bradová et al. 2015). Additionally, the soils were characterized as Aluminic Cambisol under beech forest and Entic and Haplic Podzol under spruce (Tejnecký et al. 2010, 2013, 2014). Soil bedrock was determined as medium-grained porphyritic biotite granite to granodiorite (Cháb et al. 2007). The thickness of the soil layers is very variable under both forest tree species due to slope and the randomly selected soil pits for sampling in each year (supplementary information, table S1).

The samples were taken in different years (2008, $n = 24$; 2013, $n = 24$; 2015, $n = 56$; and 2020, $n = 24$). Three individual soil pits (50×50 cm) were dug at each species sites (spruce and beech) in 2008, 2013, and 2020, but seven soil pits were dug in 2015. The soil samples were taken in different soil horizons: fermented horizons (F), humified horizons (H), organo-mineral horizons (A), and subsurface mineral horizon (B) (cambic horizon under beech, spodic humusos-sequioxidic horizon and cambic horizon enriched with iron oxides under spruce). The soil pits were randomly selected, and the distance of the pits was at least 15 m. The soil samples were air-dried and sieved to 2 mm particles. The soil samples for measurements with infrared spectroscopy were

milled to fine fractions using Fritsch Analysette 3 Spartan Pulvensette miller, Idar-Oberstein, Germany.

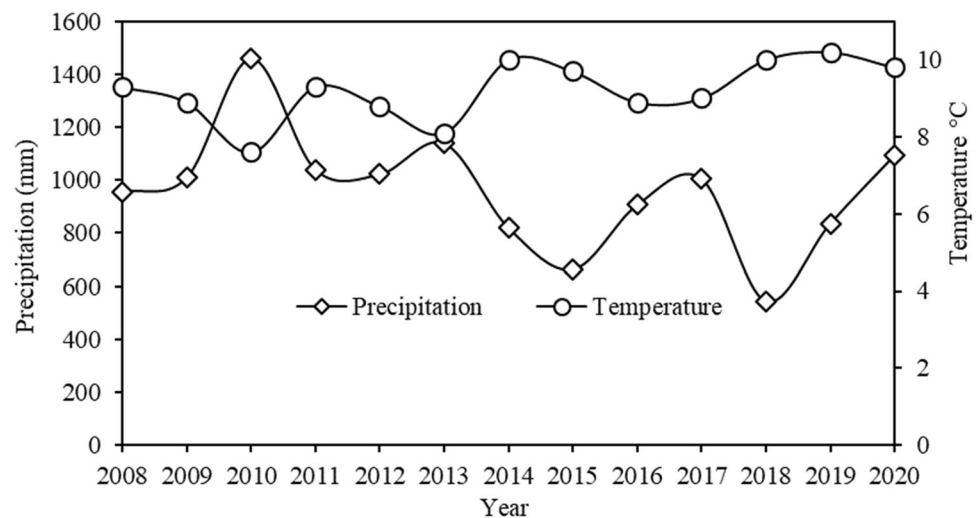
Soil characteristics analysis

The active pH ($\text{pH}_{\text{H}_2\text{O}}$) was determined in a ratio of 1:4 (soil:water, w:v) for organic horizons and 1:2 for mineral horizons. The suspension was shaken for 5 min and then measured using the pH-electrode SenTix 21 (Inolab pH level 21, WTW, Germany). The SOC content was determined by rapid dichromate oxidation ($\text{K}_2\text{Cr}_2\text{O}_7$) techniques following Tyurin's method (Sparks 1996). The total cation exchange capacity (CEC) was determined according to the standard ICP Forest methods (Cools and De Vos 2016). The exchangeable cations were measured in 0.1 M BaCl_2 (2.5:30 w/v ratio) extract of the soil using AAS (SpectrAA Varian 280FS, Australia) and ICP-OES (iCAP 7000, Thermo Scientific, USA). Exchangeable acidity (EA) was calculated from the sum of Al^{3+} , Fe^{3+} , Mn^{2+} , and H^+ , and base cation (BC) was calculated from the sum of Ca^{2+} , Mg^{2+} , Na^+ , and K^+ . The cation exchange capacity was calculated by summing up all cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Al^{3+} , Fe^{3+} , Mn^{2+} , and H^+). The quality control and quality assurance of all laboratory procedures were done according to standard protocol.

Diffuse reflectance infrared Fourier transform spectroscopy

The milled soil samples were measured using an infrared spectrometer (Nicolet iS10) and OMNIC 9.2.41 software (Thermo Fisher Scientific Inc., USA). Due to the darkness of forest soil samples, the spectra were recorded by 120 scans in wavenumbers ranging from 4000 to 400 cm^{-1} at a resolution of 4 cm^{-1} . The measured reflectance was converted to Kubelka–Munk units (KM), and the gold mirror was used as the background of the spectra (Thai et al. 2021).

Fig. 1 The average temperature (°C) and precipitation (mm) under beech and spruce from 2008 to 2020 in the locality—Hejnice meteorological station (CHMI 2023a, b)



The potential wettability index (PWI), aromaticity index (iAR), and decomposition index (iDEC) were determined using DRIFT spectra. The adsorption bands ranging from 2948 to 2920 cm^{-1} ascribed the asymmetric C–H stretch vibrations, and the adsorption bands ranging from 2864 to 2849 cm^{-1} attributed to the symmetric stretch vibrations. The adsorption band at 1710 cm^{-1} ascribed C=O groups in ketones, carboxylic acids, and amides. The adsorption bands ranging from 1640 to 1600 cm^{-1} referred to carboxylate and aromatic groups (Gerzabek et al. 2006), and OH bending vibrations of water molecules in hydration layers of soil phyllosilicates (Leue et al. 2010). The potential wettability index (PWI) was evaluated according to the adsorption band of the alky C–H groups – A (2948–2920 cm^{-1}), indicating the relative hydrophobicity, and adsorption band of the C=O groups—B (1740–1698 and 1640–1600 cm^{-1}), which is ascribed to hydrophilicity. PWI was calculated as a ratio by summing up the intensity of the C–H and C=O groups ($\text{PWI} = \text{A}/\text{B}$) (Ellerbrock et al. 2005). The higher index value indicated the lower wettability of the soil (Haas et al. 2018). Index of aromaticity (iAR) was calculated from the absorption band of aliphatic bands intensity – AL (3000–2800 cm^{-1}) and aromatic bands intensity – AR (1520 cm^{-1}), ($\text{iAR} = \text{AL}/(\text{AL} + \text{AR})$) (Cunha et al. 2009). The higher mean values of iAR expressed a lower proportion of aromatic compounds in the soil. The index of decomposition (iDEC) was based on the absorption band of carboxylate and aromatic groups – X (1640–1600 cm^{-1}) and polysaccharides—Y (1030 cm^{-1}). The iDEC was calculated by summing up the intensities of the C=O group and polysaccharide intensity and then performed as a ratio ($\text{iDEC} = \text{X}/\text{Y}$) (Artz et al. 2006). The high value of iDEC indicated finer particles and more decomposed organic matter because of the loss of intensity of polysaccharides (Preston et al. 1987; Haberhauser et al. 1998).

Data analysis

The temporal changes of pH, sorption complex, exchangeable elements and structure of SOM during the study period and through the soil horizons were statistically assessed using analysis of variance (ANOVA) at error level of 0.05 (confidence level of 95%). *T* test was used to differentiate the chemical compositions between the forest tree species (beech and spruce). The statistical differences among the soil horizons (F, H, A, B) and between forest types were presented in alphabetical letters using the Tukey test. To run the data correctly, the homogeneity of variances was applied to check its normality. These statistical analyses were evaluated using software IBM SPSS (version 26, New York, USA). Besides that, a correlation matrix and principal component analysis (PCA) were also performed to access co-variations and evaluate the relationships among the

chemical compositions using the R studio program (Jolliffe and Cadima 2016; Girona-García et al. 2018).

Results and discussion

Soil pH

The forest soils were found to be more acidic in the organic horizons than in the subsurface mineral horizons (F, H, A, < B) under both forest tree species. The result showed same pattern as the study by Borůvka et al. (2009). However, more acidic nature in the organic horizons and with time in Fig. 2 resulted from litter decomposition and acid deposition (Hedl et al. 2011; Yang et al. 2015). The SOM decomposition was considered to lower soil pH due to the interaction and exchange with Al (Cremer and Prielzel 2017) and increased amount of H^+ resulted in more acidic soil in the uppermost layer (Zaidey et al. 2010). As a comparison between the forest tree species, the soil pH found no significant difference, but the pH value in spruce soil was found to be lower than under beech forest soil in all years and horizons (Fig. 2) which is in accordance with findings of Daněk et al. (2019).

The cation exchange capacity and exchangeable elements

The cation exchange capacity statistically decreased with depth ($F > H > A > B$) in all years under beech and spruce (Fig. 2). The decrease in CEC revealed that less cations were retained in lower horizons than in the uppermost soil layers and relatively corresponded with high organic material inputs and SOC content during litterfall. Soil organic carbon, generally, plays a vital role in CEC in cation binding and soil sorption complex in the organic and mineral horizons (Ciarkowska and Miechówka 2019; Cremer and Prielzel 2017). Our results (Fig. 5) confirmed that CEC was strongly correlated ($r = 0.83$, $P < 0.001$) with SOC, indicating that CEC increased with increased SOC concentration. Besides that, during the plant uptake, it brings more nutrients to accumulate in the uppermost horizons because of the high proportion of nutrients absorbed in the roots and recycled nutrients through litterfall, stemflow, and throughfall (James et al. 2016; Johnson et al. 2018; Langenbruch et al. 2012; Oulehle et al. 2007).

Regarding the time changes, the CEC increased significantly from 21.6 ± 3.5 to 33.7 ± 3.6 $\text{cmol} + \text{kg}^{-1}$ in F horizon, 13.9 ± 2.8 to 15.2 ± 0.8 $\text{cmol} + \text{kg}^{-1}$ in H horizon, 7.6 ± 2.8 to 11.3 ± 0.8 $\text{cmol} + \text{kg}^{-1}$ in A horizon, and 4.1 ± 1.9 to 7.2 ± 1.1 $\text{cmol} + \text{kg}^{-1}$ in B horizon under beech. Remarkably, the CEC in 2015 was observed to have the lowest values in F and H horizons under beech. It was similar to the CEC in

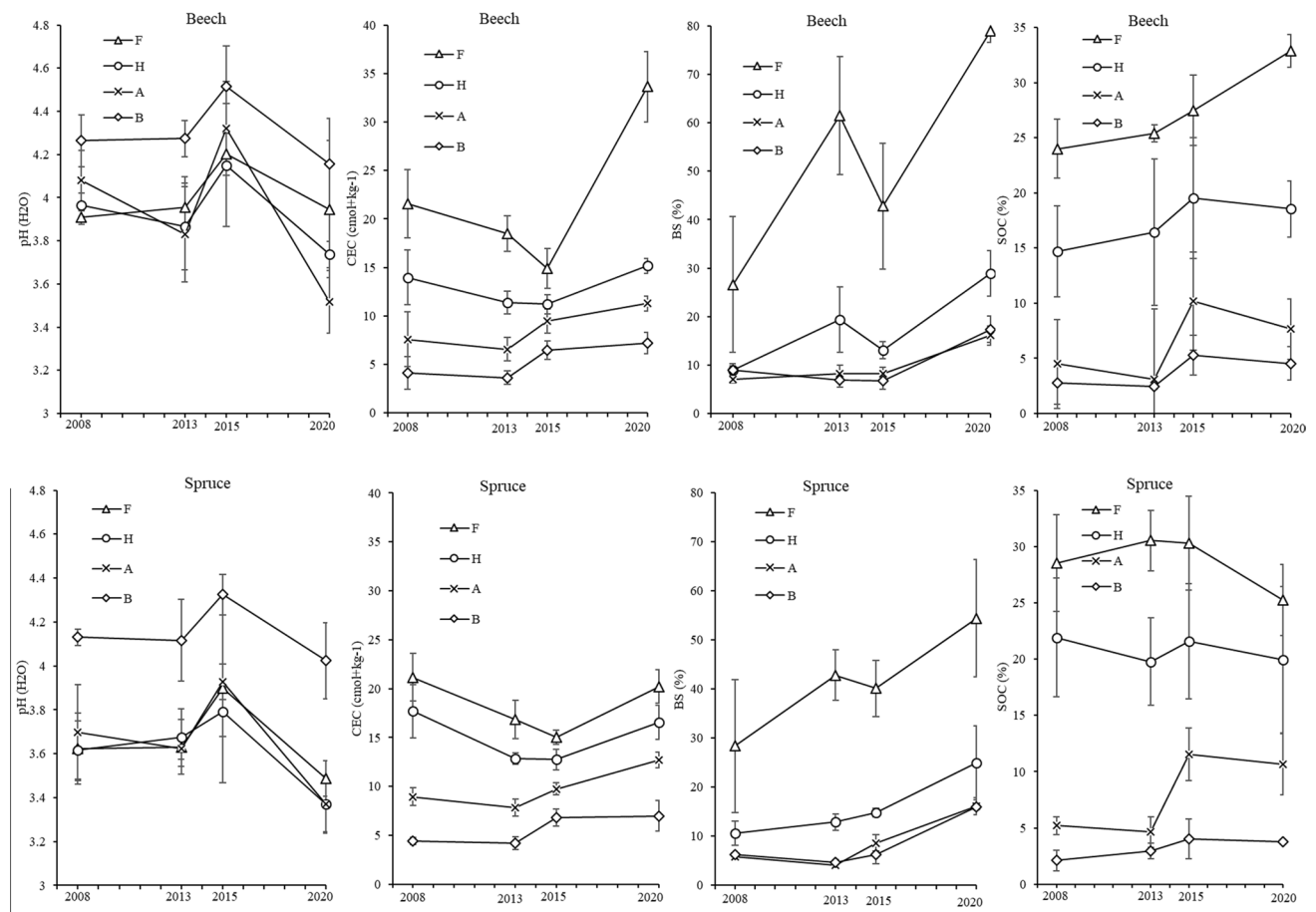


Fig. 2 The distribution and temporal changes of soil chemical characteristics in fermented horizon (F), humified horizon (H), organo-mineral horizon (A), and subsurface mineral horizon (B – spodic or cambic) of the soil profile dug under beech and spruce at Jizera

Mountains (Czech Republic). (Mean and standard deviation; 2008, $n=24$; 2013, $n=24$; 2015, $n=56$; and 2020, $n=24$). Base saturation (BS—%), cation exchange capacity (CEC— $\text{cmol} \cdot \text{kg}^{-1}$), soil organic carbon (SOC—%)

spruce forest, which showed no specific pattern in F and H. The CEC decreased from 2008 to 2015 but recovered slowly in 2020, while A and B showed increasing CEC with time (Fig. 2). The lowest CEC in 2015 in F and H horizons in beech forest was associated also with the decrease of BS. However, the dry climatic conditions (high temperature and low precipitation) in 2015, which is observed in Fig. 1 and by Ionita et al. (2017), decreased nutrients and their recycling due to less adsorption with the organic matter and the decrease decomposition of forest litter (Mondal 2021; Gelybó et al. 2018). For comparison between forest tree species, significant differences in the CEC were observed ($P=0.004$) in F horizon in 2020 (beech > spruce). This result was consistent with the same finding of Mareschal et al. (2010). Tree species compositions affected the soil in various ways (Cremer and Prietzel 2017). Beech forest was considered to have large foliage nutrients and lower foliage lignin content than spruce forest that accelerated litter decomposition and bioturbation and enhance organic layers

with high base cations (Berger and Berger 2012). This corresponds to our results from soil spectra as indices (Fig. 4) and our results in supplementary information (Table S2) that show nutrients under beech such as Ca^{2+} and Mg^{2+} increased significantly from 2008 to 2020 in all horizons, while spruce was relatively constant (no significance) in F and H horizons (Table S2). However, it indicated that spruce used the nutrient stock efficiently, while beech was more likely to hold, cycle, and stock nutrients in the forest floor (Bublinec and Machava 2015; Bagherzadeh et al. 2008). It was in agreement with the research of Matschonat and Falkengren-Grerup (2000), who found Ca^{2+} and Mg^{2+} increased tendency in beech forest after recovering from acid deposition. High acidity and poor decomposition of litters under spruce in the locality contributed to higher Al^{3+} concentration and more mobilization in the forest, leading to depleting the base cations and indicating a slow recovery of nutrients in sorption complex in spruce forest (Oulehle and Hruška 2005; Collignon et al. 2011; Daněk et al. 2019).

Notably, it was observed that the number of exchangeable elements decreased significantly with depths in all years except for Al^{3+} (Table S2). Aluminum content decreased significantly with depths (F, H > A, B) in 2008. This suggested that, in 2008, Al concentration remained more mobilized in the organic horizons due to the atmospheric acid deposition that occurred during 1980–1990. On the contrary, in 2013, 2015, and 2020, Al^{3+} concentration was observed to be accumulated more in H and A horizons under both forest tree species, which resulted from increases in organic acids from the decomposition in the soil and the initial stage of weathering of minerals in mineral phases (Akbar et al. 2010; Iwashima et al. 2012). On the other hand, Dijkstra and Fitzhugh (2003) reported that it may have originated from Al release after SOM decomposition and also from the increased Al dissolution induced by low soil pH. Regarding the long-term study in beech, the Ca^{2+} , Mg^{2+} , K^+ , Na^+ , and Mn^{2+} were statistically changed by their increasing concentrations (2020 > 2015, 2013, and 2008), while Fe^{3+} decreased in F horizon with a range from 1.2 ± 0.3 to $0.25 \pm 0.09 \text{ cmol} + \text{kg}^{-1}$ but increased in H and A horizon ranging from 0.8 ± 0.3 to $1.1 \pm 0.1 \text{ cmol} + \text{kg}^{-1}$ and 0.4 ± 0.1 to $1.0 \pm 0.3 \text{ cmol} + \text{kg}^{-1}$. Despite that, Al^{3+} concentration quickly decreased from 12.4 ± 1.9 to $1.6 \pm 0.5 \text{ cmol} + \text{kg}^{-1}$ in F horizon and slowly decreased from 11.1 ± 2.0 to $7.6 \pm 1.3 \text{ cmol} + \text{kg}^{-1}$ in H horizon, while A and B horizons remained stable (no significance) under beech. There was similar tendency in spruce, the concentration of Al^{3+} significantly decreased from 9.8 ± 2.8 to $3.9 \pm 1.7 \text{ cmol} + \text{kg}^{-1}$ in F horizon and from 11.3 ± 0.9 to $8.3 \pm 1.3 \text{ cmol} + \text{kg}^{-1}$ in H horizons but increased in A horizon ranging from 6.5 ± 0.7 to $7.2 \pm 0.3 \text{ cmol} + \text{kg}^{-1}$. This suggested that a larger part of Al^{3+} was bound in the organic complexes that had been leached from the soil or might be sorbed on soil mineral phases (Borůvka et al. 2009). The higher amount of SOM in the organic horizons (Fig. 2) also indicated that the soil sorption sites were mainly by organic matter that suppressed Al toxicity by decreasing dissolved Al^{3+} activities in surface soils (Berthrong et al. 2009; Borůvka et al. 2009; Dijkstra and Fitzhugh 2003). The mechanism of SOM contributes to the variations of Al^{3+} and Fe^{3+} by exchanging the polyvalent cation-bridging between negative charges of organic matter (Mueller et al. 2012). Likewise, the low concentration of Al^{3+} and Fe^{3+} in F horizon and decreases with time in organic horizons resulted from the direct interaction with atmospheric conditions (temperature and precipitation) (Bradová et al. 2015) and also revealed the effects of forest tree species in long-term soil acidification, less acidic deposition from the atmosphere in the study area, and the podsolization process in spruce forest (Altman et al. 2017; Krug and Frink 1983; James et al. 2016). An increase of Al^{3+} in A horizons with time under spruce resulted from the high acidity in the organic soil solution due to the aluminosilicate

weathering (Oulehle and Hruška 2005). Additionally, continuously increasing nutrients in both forest tree species during the study period revealed the effect of forest tree species on acidity deposition reduction and mitigation.

Soil organic carbon, indices of soil wettability, aromaticity, and decomposition

In both beech and spruce forests, the content of SOC decreased through soil horizons (F > H > A > B) in each year. Generally, the effect of forest on SOC led to more accumulation in the topsoil layers (F and H) compared to deeper soil profiles. This resulted from the living and dead plant matter accumulating in the uppermost layers (Habumugisha et al. 2018; Langenbruch et al. 2012; Galka et al. 2014).

After a decade of observation, beech and spruce had no significant changes in SOC in the F, H, and B horizons, but A horizons showed a significant increase from 4.5 ± 4.0 to $7.7 \pm 2.8\%$ in beech and from 5.2 ± 0.1 to $10.7 \pm 2.7\%$ in spruce. Remarkably, beech forest showed SOC content increased constantly in average values with time ranging from 24.0 ± 2.7 to $32.0 \pm 1.5\%$ in F horizon, from 14.7 ± 4.1 to $18.0 \pm 2.6\%$ in H horizon, and from 2.8 ± 1.9 to $4.5 \pm 1.5\%$ in B horizon, while spruce remained relatively stable in most horizons (Fig. 2). Besides that, there was a statistical increase of SOC in beech ($P = 0.041$), once all values were combined from soil horizons. It was opposite to spruce, where no significant difference was observed ($P = 0.441$). As a comparison between beech and spruce using a *T* test, there were significant observed differences ($P = 0.032$) in F horizon in 2020, where beech had markedly higher SOC content than spruce (Fig. 2). A lower concentration of SOC under spruce than in beech was not usually found. Numerous studies confirmed that spruce promoted more organic carbon and higher thickness layer of organic horizons in a long-term observation due to the higher recalcitrant substances (e.g., lignin, resin, waxes, and phenolic compounds), which were difficult to be decomposed by microorganisms (Andivia et al. 2016; Grüneberg et al. 2019). It was in contrast to our findings. This resulted from the local environmental condition and different forest stands such as age, density, and canopy forest (Yuan et al. 2013). Bradová et al. (2015), who studied the same locality, reported that the beech forest was older than spruce, by approximately 80 years, and had larger canopy closure (87%) than spruce (40%). The higher density and age of the forest stands in the research area had typically larger basal area of trees, in which it enhanced SOC in the forest soil (Menyailo et al. 2022). It was consistent with the finding of Yuan et al. (2013) who found that SOC was stored higher under the old-growth forest trees. The larger canopy of the beech forest stands, however, reduced the decomposition rate, while an open canopy led to SOC loss due to more favorable soil microclimates, higher exposure to throughfall

and to the warmer condition, which accelerated the decomposition, leaching, and soil respiration by microorganism (Zech et al. 1989; Merabtene et al. 2021; Hanakova-Becvarova et al. 2022; Jandl et al. 2022). It was also documented by Fang et al. (2005) regarding the variation of SOC level with the temperature and by Andivia et al. (2016) about the relationship between SOC and canopy.

Besides that, the constant increases in average values of SOC content in each horizon in beech forest during the study period revealed the larger forest productivities (litterfall) and more SOC inputs that were expected from the compensation from soil acidification by beech forest. The primary sources of organic carbon are not only from the litterfall but also from soil microorganisms, root exudates, and root litters (Andivia et al. 2016; Li et al. 2015; Nickels and Prescott 2021). Roots system is one of the main factors in the distribution of SOC under forest tree species (Tefs and Gleixner 2012). Beech stands, in fact, had a larger and deeper anchored root system, which provided higher belowground biomass and caused SOC to be more distributed into the soil horizons (Laganière et al. 2010; Hansson et al. 2011). Ohno et al. (2014), who observed the molecular composition and biodegradability of soil organic matter, found that deciduous forests had higher root dead mass and root mortality than coniferous forests, and thus enhanced

higher aliphatic content in the forest soil. Together with the root exudate, it improved long-term soil organic carbon storage by adsorption onto the mineral surface under deciduous forests.

On the other hand, the behavior of SOC and other elements is associated with soil wettability, aromaticity, and decomposition, and vice versa (Artz et al. 2006; Cunha et al. 2009; Ellerbrock et al. 2005; Ellerbrock and Gerke 2013). Therefore, our study extended further information about the relationship among the chemical components using DRIFT spectroscopy. The spectra in Fig. 3 clearly showed the differences in the chemical compositions through soil profile under both forest tree species.

The potential wettability index (PWI) declined statistically through the soil profile (F > H > A > B) from each year under both forest tree species. This means that organic horizons had lower wettability (F and H horizons) than the mineral horizons (A and B horizons). A similar result was found by Woche et al. (2017) and Atanassova et al. (2018). The different PWI through the soil profile indicated the importance of SOM inputs in the soil through their vertical transporting processes (root growth and SOM distribution) (Leue et al. 2010). The high PWI in the organic horizon was confirmed by a high SOC content with high hydrophobicity, which lowered soil wettability and reduced the infiltration rate (Goebel

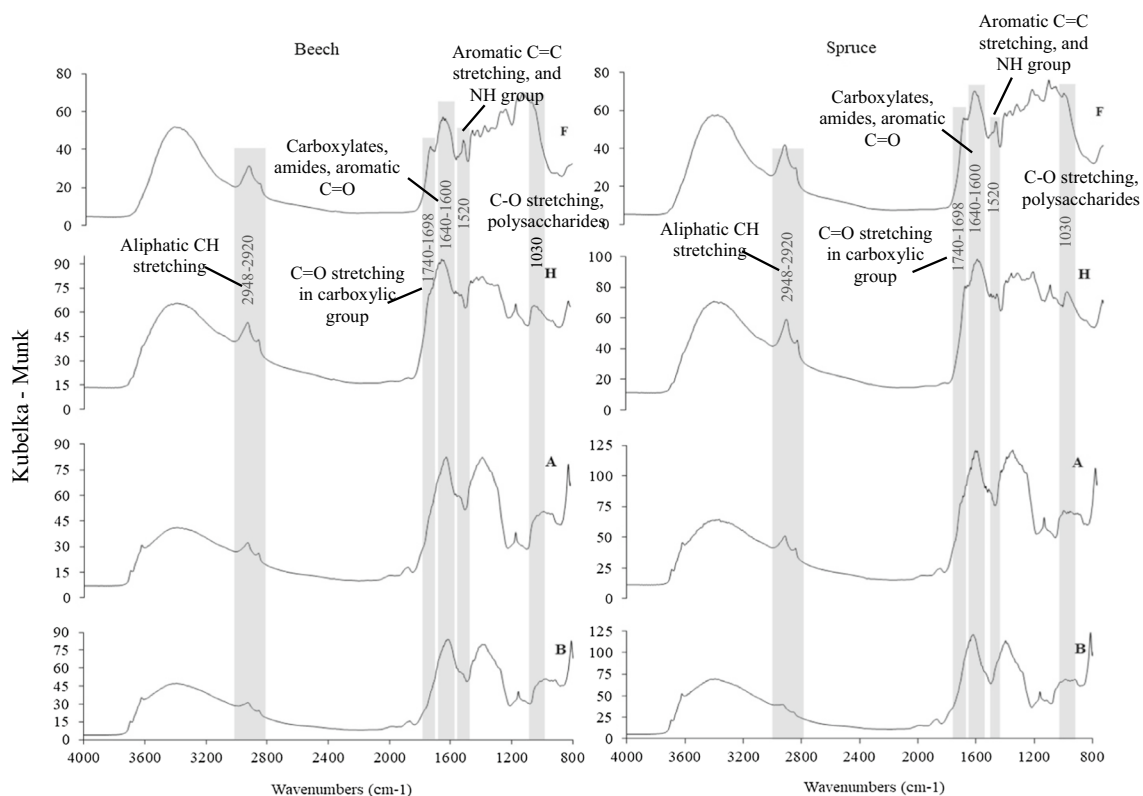


Fig. 3 The representative of average DRIFT spectra of soil in beech and spruce under different soil horizons (Fermented horizons (F), humified horizons (H), organo-mineral horizons (A), subsurface mineral horizons (B—spodic or cambic); $n = 24$) in 2020

et al. 2005). The process of SOC in soil wettability had few mechanisms. The hydrophobicity decreased wettability by reducing the surface free energy of the soil, restricting the soil from wetting (Turski et al. 2022) and decreasing the rehydration rate (Eynard et al. 2004). However, the SOC improved the soil porosity by holding soil particles together against external disturbances (Eynard et al. 2004). The variation of PWI through the soil profile was also associated with the abundant root system, and high Ca content in the organic horizons that enhanced soil rhizosphere and indicates strong incorporation between Ca and OM, which possibly stabilizes aliphatic contents that further increased PWI (Leue et al. 2010; Ohno et al. 2014; Turski et al. 2022). A strong correlation was highlighted between Ca and PWI ($r=0.58$, $P<0.001$) in Fig. 5.

Regarding a long-term observation, PWI increased significantly from 0.17 ± 0.02 to 0.24 ± 0.01 in F horizon and 0.13 ± 0.02 to 0.20 ± 0.02 in H horizons under beech, while spruce relatively remained stable (no significance) in all horizons. The increases of PWI with time revealed a lower soil wettability and enhanced SOC and aggregate stability (Woche et al. 2017). It can be said that SOM improved PWI (lowering wettability) and high PWI prevented SOC from microbial decomposition (Atanassova et al. 2018). In a comparison between forest tree species, there were significant differences observed in F and H horizons in 2015, where spruce had a higher index, but in 2020, beech had a higher PWI in F horizon. This was consistent with the increase in SOC content in 2020 (Fig. 2). Beech produced more residues and had deeper root system compared to spruce; therefore, it enhanced organic carbon better and sequestered faster into the soil profile. With this, it significantly affected the PWI (Eynard et al. 2004; Ilek et al. 2015). On the other hand, it was observed that there was a strong correlation between PWI and SOC ($r=0.89$, $P<0.001$) (Fig. 5). Hence, the variation of PWI was explained according to the tendency of SOC. This was also supported by Ellerbrock and Gerke (2013) who explained that the SOM compositions regulated the soil wettability and sorption and transfer properties of the flow pathway.

The aromaticity index decreased dramatically with increasing soil profile depths ($F>H>A>B$), which means the aromatic content increased with depths. A similar result was found by Zech et al. (1989) and Silva et al. (2022), showing that the content of aromaticity increased from L to B horizons. It was contrast to the finding of Ohno et al. (2014) where the abundance of condensed aromaticity of SOM was much lower in subsoil under coniferous and deciduous forests due to the adsorption of aromatic compounds to the minerals present in the soil profiles. The decrease or increase of the iAR through the soil profile, generally, depends on the association of plant residue compositions, climate conditions, and microorganism present in the soil

(Koutika et al. 2020; Thai et al. 2022). The lower aromaticity in the uppermost soil layer than in the deeper soil profile under both forest tree species, however, was indicated by the less humified and decomposed organic matter due to the chemical and physical interaction mechanisms (Chen et al. 2013; Corvasce et al. 2006). Ussiri and Johnson (2003) found that the decomposition of forest residues relatively affected the aromaticity index changes through the soil horizons. Nonetheless, the aromaticity in the soil profile had been shown to increase with degrees of decomposition and maturity of OM (Chefetz et al. 1998; Margenot et al. 2015; Veum et al. 2014). Aromatic compounds were less susceptible to become mobile; therefore, they are likely retained in the mineral soil horizon (Bi et al. 2013). Zech et al. (1989) reported that the aromatic compounds in the spodic soil profile were increased due to the leaching of phenolic substances during the podsolization.

After a decade of observation, the iAR changed significantly by increasing from 0.29 ± 0.04 to 0.36 ± 0.01 in F horizon under beech, while spruce had no statistical changes in all horizons. To compare between the two forest tree species, there were significant differences observed in F, H, and A horizons in 2015, where spruce had a higher index than beech, but in the F horizon in 2020, beech had a higher iAR. The changes of iAR by increasing with time in F horizon under beech indicated the degree of SOM decomposition and more aliphatic compounds that dominated in this soil horizon (Pizzeghello et al. 2017). It is also shown in Fig. 5 that iAR was correlated with SOC ($r=0.89$, $P<0.001$) regarding iAR increase with increasing SOC content. The remaining constant iAR under spruce and in subsoil was due to the preferential retention of condensed and substituted aromatic molecules, which are recalcitrant to mineralization processes (Corvasce et al. 2006; Gangloff et al. 2014). Furthermore, aromatic compounds were more likely adsorbed onto the soil particles than mobile in the soil solution through the soil profile (Thai et al. 2022; Ohno et al. 2017). Besides that, the high inputs of root lignin and cool climate in the study sites were considered as a cause of stability in the aromaticity content in the soil under spruce and in the subsoil horizons (Zech et al. 1989; Silva et al. 2022).

The index of decomposition displayed no significant difference through the soil horizons in 2008 and 2013 in beech and spruce in 2008 ($P>0.05$). Contrary to 2015 and 2020, the iDEC significantly changed by increasing with depth under both forest tree species. Similar results were shown by Prasad et al. (2000) that the iDEC increased with depth. The increases of the iDEC through the soil profile indicated that the intensities peak of polysaccharides (1030 cm^{-1}), which indicated one of the primary energy sources contained in the plant residues, were dramatically declined from F to B horizons by various moieties of forest litter substances (Haberhauer et al. 2000). This result was agreed with Nuzzo

et al. (2020) who found that the intensity of polysaccharides spectral band decreased with depth. On the other hand, the carboxylic and aromatic compounds, which were relatively linked with the lignin structures, were less vulnerable and resistant to microbial attack, while polysaccharides were decomposed easily by microorganisms in the organic horizons (F and H horizons) (Tseng et al. 1996; Artz et al. 2006). Additionally, Fig. 5 shows that there was a negative correlation ($r = -0.63$, $P < 0.001$) between iDEC and iAR. In long-term observation during the study period, the iDEC significantly increased from 1.3 ± 0.3 to 1.9 ± 0.2 in B horizon under beech and from 1.4 ± 0.1 to 1.7 ± 0.2 in A horizon under spruce. This resulted from a high amount of inorganic substances in A and B horizons that caused the changes in the intensity peaks of organic matter, which affected the decomposition index (Prasad et al. 2000; Haberhauer et al. 1998). In comparison between the forest stands, there were no significant differences between forest tree species in most of the soil horizons in each year except for F and H horizon

in 2008 and B horizon in 2015, which showed a significant difference ($P = 0.05$, 0.014 , and 0.018 , respectively) between forest tree species (beech > spruce). The higher iDEC found in beech than spruce revealed that beech had more decomposable materials than spruce and provided more variable nutrients in the forest soil.

Correlation matrix and principal components analysis (PCA) among the soil properties

Figures 4 and 5 show the correlation matrix and PCA results among the soil properties. The length of the vectors indicated the strength of their contribution to each PC. Vectors pointing to similar directions were positively correlated; vector directions contrary to each other were negatively correlated; directions of vectors at 90° angle were not correlated. The PCA was grouped by year with four different colors for each year. Markers on the same side described a high contribution on the variable dataset with 95% confidence ellipse

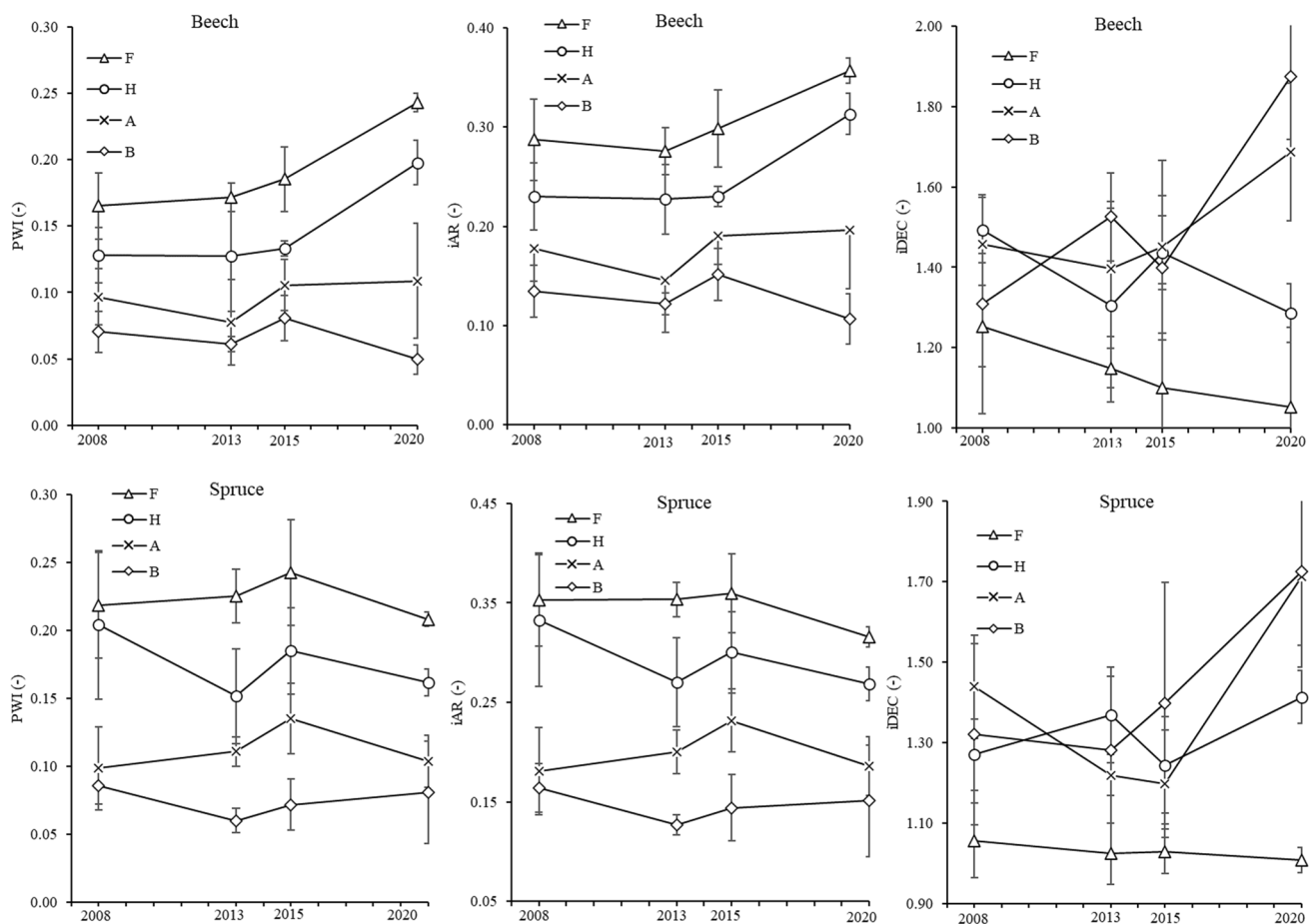


Fig. 4 The distribution and temporal change of the potential wettability index (PWI), index of aromaticity (iAR), and index of decomposition (iDEC) in fermented horizons (F), humified horizons (H), organo-mineral horizons (A), subsurface mineral horizons (B—spo-

dic or cambic) of the soil profile dug under beech and spruce at Jizera Mountains (Czech Republic). (Mean and standard deviation; 2008, $n = 24$; 2013, $n = 24$; 2015, $n = 56$; and 2020, $n = 24$)

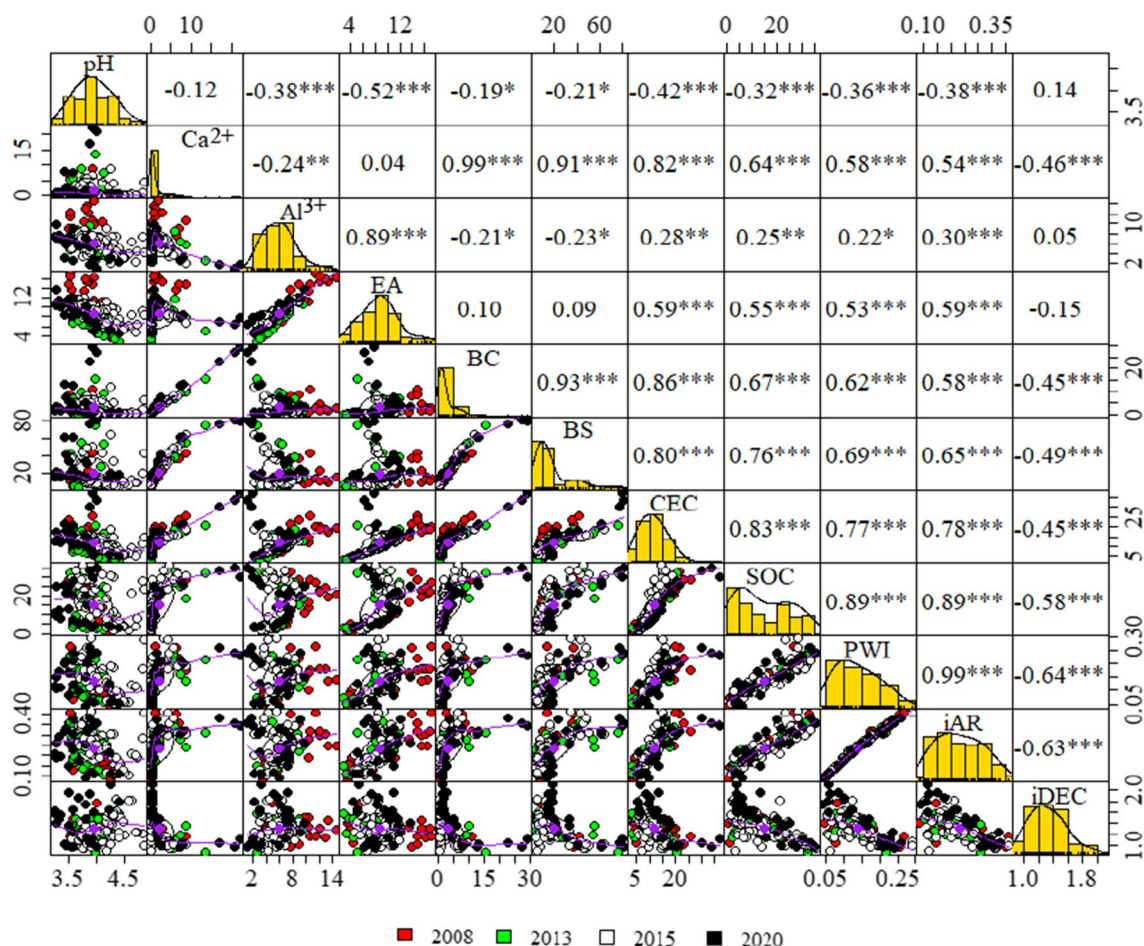


Fig. 5 Correlation matrix plot of all data among the soil properties under forest tree species. (Note: *, **, *** correlation significant at 0.05, 0.01, and 0.001, respectively). (2008, $n=24$; 2013, $n=24$; 2015, $n=56$; and 2020, $n=24$). Ions concentration in sorption complex—(Ca^{2+} , Al^{3+} , exchangeable acidity—EA, and base cation—BC

($\text{cmol} + \text{kg}^{-1}$), base saturation (BS (%)), cation exchange capacity (CEC ($\text{cmol} + \text{kg}^{-1}$)), soil organic carbon content (SOC (%)), potential wettability index (PWI (-)), index of aromaticity (iAR (-)), and index of decomposition (iDEC (-))

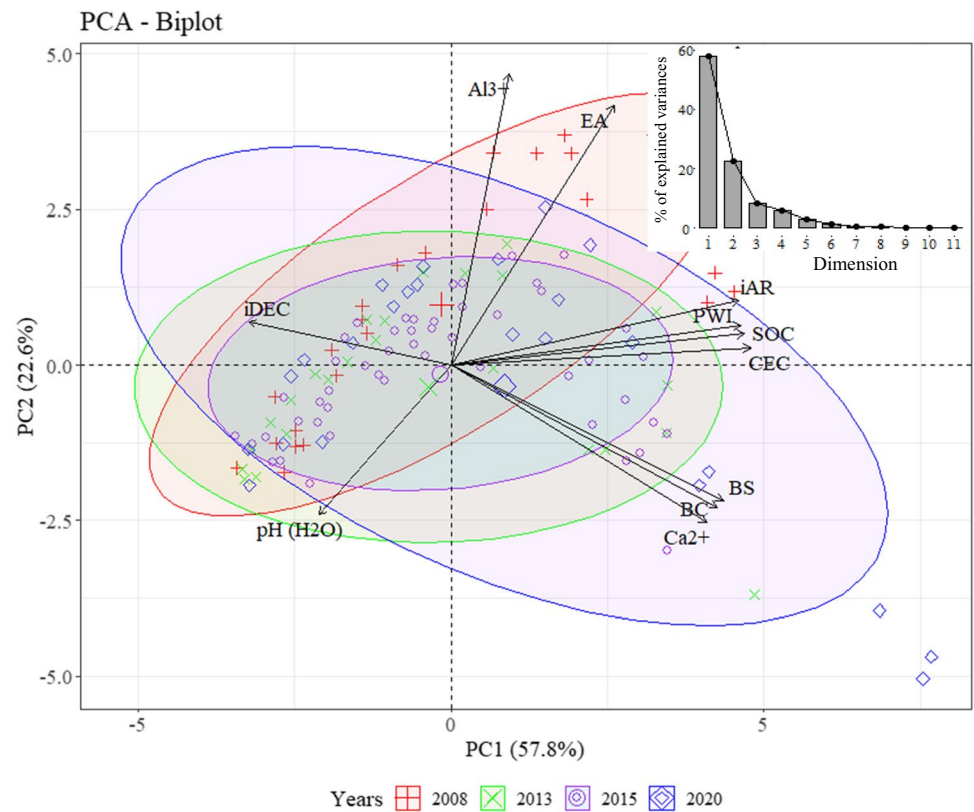
(Escobar-Flores et al. 2019). There were eleven principal components with eigenvalue > 1 . The principal component analysis, which selected the main soil properties, showed the first two PCs as 79.8%. The first axis of PC1 was 57.8%, and the second axis of PC2 was 22.0% of the total variance (Fig. 6). Most of the soil properties such as SOC, PWI, iAR, CEC, BS, BC, and Ca^{2+} positively contributed to PC1 and PC2, whereas the $\text{pH}_{(\text{H}_2\text{O})}$ and iDEC were negatively associated. The correlation matrix, however, showed that PWI, SOC, BS, CEC, iDEC, and iAR were closely correlated with each other. A similar correlation among these properties was described by Ellerbrock and Gerke (2013) and Solly et al. (2020) and further discussion was in our results mentioned above. Surprisingly, the ellipse of the year 2008 (Fig. 6) showed the same direction of EA and Al^{3+} and then gradually moved their directions toward Ca^{2+} , BC, BS, CEC, SOC, PWI, and iAR in 2013, 2015, and 2020. This indicated that soil in 2008 maintained high concentration

and mobilization of Al^{3+} and Fe^{3+} and was also proved the influences of the forest tree species on the recovery and mitigation of the soil properties from acidification in a long-term observation.

Conclusion

The study agreed with the hypothesis that beech and spruce forests are able to recover effectively from anthropogenically enhanced acidification and promote soil organic matter and soil nutrients in the forest ecosystem in the long term after massive decline of atmospheric acids deposit during the 1990s. The cation exchange capacity and soil organic carbon content under both forest tree species were higher in the organic horizons than the mineral horizons. After a decade of observation, it was proven that some soil horizons, especially in F and H horizons, were

Fig. 6 Principal component analysis (PCA) biplot combining the main observed variables that contributed to the distribution and temporal changes of soil properties in all horizons of the forest soil. (2008, $n=24$; 2013, $n=24$; 2015, $n=56$; and 2020, $n=24$)



more pronounced in changing the soil chemical properties under both forest stand types. The exchangeable base cations (Ca^{2+} , Mg^{2+} , Na^+ , and K^+) which were depleted from acidification have been significantly increased with time under both forest stands. Moreover, it was observed that the nutrients were more variable under beech forest in all horizons than in spruce stands due to more decomposable materials, larger canopy and productivity of litter, and less acidity in the soil. However, the drought in 2015 was considered to lower the exchangeable elements in the organic horizons (F and H horizons). Additionally, Al and Fe content quickly decreased in F horizons and slowly decreased in H horizon under both forest tree species. This indicated that Al and Fe were bound in the organic complexes that had been leached from the soil or might be sorbed on soil in mineral horizons. It was also observed that SOC content increased constantly in values under beech, while spruce remained stable. Nonetheless, the soil wettability decreased in F and H and aromaticity index increased in F horizon with time under beech, which signified that SOC improved and aliphatic group proportion increased in organic horizons. As a result, we assumed that beech forest was better in reducing and mitigating of the soil acidification than spruce one. Moreover, beech stand improved soil to more favorable conditions for future plant growth.

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Declarations

Competing interests The authors declare no competing interests.

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