



Long-term patterns in soil acidification due to pollution in forests of the Eastern Sudetes Mountains

Radim Hédli^{a,*}, Petr Petřík^b, Karel Boublík^c

^aDepartment of Vegetation Ecology, Institute of Botany, Academy of Sciences of the Czech Republic, Lidická 25/27, CZ-602 00 Brno, Czech Republic

^bDepartment of Geobotany, Institute of Botany, Academy of Sciences of the Czech Republic, Zámek 1, CZ-252 43 Pruhonice, Czech Republic

^cFaculty of Environmental Sciences, Czech University of Life Sciences Prague, Kamýcká 129, CZ-165 21 Praha 6 – Suchbátka, Czech Republic

ARTICLE INFO

Article history:

Received 20 December 2010

Received in revised form

1 June 2011

Accepted 4 June 2011

Keywords:

Long-term change

Revisitation study

Soil acidity

Tree composition

Altitudinal gradient

ABSTRACT

Soil acidification was assessed in the Eastern Sudetes Mountains (Czech Republic) between 1941 and 2003, i.e. before and after the period of major industrial pollution (1950s–1990s). The twenty sites included in our study were distributed along a gradient of altitude ranging 1000 m. Values of pH have decreased in 80–90% of the pairs of samples after the six decades, on average by 0.7 for pH-H₂O and 0.6 for pH-KCl. Organic matter increased in the topsoil, probably reflecting a change in decomposition conditions. The most important finding is that the acidification varies along the joint gradient of altitude/tree layer composition, and displays a changing pattern in three soil horizons (A, B and C). Contrary to expectations, most acidified were soils in beech forests at lower elevations.

© 2011 Elsevier Ltd. All rights reserved.

1. Introduction

Soil acidification is a complex of changes in soil properties including pH decrease, release of base cations from the sorption complex (e.g. Porebska et al., 2008) and changes in distribution patterns of aluminium ions (Borůvka et al., 2005). Anthropogenic acidification has deeply affected soils in large areas in Europe and North America (Federer et al., 1989; Likens et al., 1996; Berge et al., 1999). It has had far-reaching consequences not only for the chemistry of soils but also for that of surface waters (Veselý et al., 1998). It has caused changes in biodiversity, which were documented in particular for vegetation (e.g., Falkengren-Grerup, 1989; Baeten et al., 2009). Species sensitive to higher acidity and base cation leaching are especially prone to extinction of local populations (Falkengren-Grerup and Tyler, 1993; Hédli, 2004a).

Acidification largely resulted from the sulphur dioxide and nitrogen oxides emissions from brown coal burning. It peaked between the 19(40)s–50s and the 1990s (Hruška and Cienciala, 2003; Kopáček and Veselý, 2005). The most severely affected area was the “Black Triangle” in the Polish–German–Czech borderland, where the enormously high loads of acidifiers and other pollutants resulted in vast forest die-off (McNeill, 2000). After the decline of heavy industry and the desulphurization of emissions from coal

power plants since the early 1990s, the sulphur loads rapidly decreased. There was an almost 90% reduction in SO₂ emission between 1990 and 2006 resulting in sulphur deposition decrease by 75% (Hruška, 2007; see also www.chmi.cz). Nitrogen deposition, however, remained at the same level further contributing to environmental acidification.

Besides airborne depositions, management practices represent the second main group of factors responsible for acidification in forest ecosystems (Yanai et al., 1999). Tree species composition is of great importance (Van Dobben and de Vries, 2010). In highly acidified areas of Central Europe, the widespread forestry practice of planting Norway spruce has probably largely contributed to soil acidification (Oulehle and Hruška, 2005). The removal of base cations from ecosystems through timber extraction is another important acidifying factor (Dahlgren and Driscoll, 1994; Hruška and Cienciala, 2003). As much as one-third of the long-term base cations loss can be in timber (Hruška et al., 2002). Finally, acidifying compounds of nitrogen are released from decomposing biomass. Acidification thus occurs during ecosystem succession, in other words forest ageing is associated with a decrease in soil pH even under pollution-free conditions.

Using mathematic models, the acidification/recovery process can be effectively reconstructed in the past and forecasted for the future (Krám and Bishop, 2001). However, current anthropogenic acidification has been a decades-long process and despite the fact that modelling and biological proxy-data (e.g. Vrba et al., 2003) can

* Corresponding author.

E-mail address: radim.hedli@ibot.cas.cz (R. Hédli).

provide indirect insights, within-site re-measurements remain the only direct evidence of the long-term changes in soil chemistry (Hruška and Cienciala, 2003). The usage of repeated measurements is therefore twofold: for the calibration of models and as the direct evidence.

With few exceptions (Falkengren-Grerup, 1987; Hédl and Rejšek, 2007), the usual time span of repeated studies is not longer than a decade or two (e.g., Dahlgren and Driscoll, 1994; Jönsson et al., 2003; Oulehle et al., 2006). Only a handful of studies extending the 50 year-period of high pollution impact were published from Sweden (Hallbäcken and Tamm, 1986a,b; Tamm and Hallbäcken, 1986, 1988), Finland (Ahokas, 1997), Belgium (De Schrijver et al., 2006), and the Ukrainian Carpathians (Houška and Schejbalová, 2004; Oulehle et al., 2010); potentially see also a study by Billett et al. (1990). Their results convincingly showed the patterns in long-term acidification. However, they describe areas under relatively low loads of acidifying pollutants, and possess some important constraints (data only from topsoil, a narrow environmental range, or only indirect comparison). Moreover, the focus of most acidification research projects is on small territories with homogeneous conditions, such as small catchments or water bodies (e.g. Veselý et al., 1998; Oulehle et al., 2006). An integral survey from a heavily acidified larger region has been missing so far.

In the present study, we aim to identify patterns in soil acidification after six decades, repeating a soil acidity survey from the early 1940s. We hypothesize that (1) acidification increases with altitude, where in higher elevations higher precipitation and acid-litter producing vegetation have enhanced the acidification process, and that (2) acidification was more intensive in the topsoil than in the subsoil where bedrock material helps to buffer acidification. In addition, we assessed the potential bias produced by the not exactly known position of the original soil pits.

2. Material and methods

2.1. Study area

Our study area includes two orographical subunits of the Czech part of the Eastern Sudetes mountain range, the Hrubý Jeseník Mountains (Altwatergebirge) and the Rychlebské hory Mountains (Reichensteiner Gebirge). It adjoins the area affected by high airborne acidifier deposition in the past decades, however, without a resulting forest die-off in the study area itself (Fig. 1).

The total area is about 1200 km², geographical delimitation is 50°27'–49°53' N and 16°50'–17°26' E. Elevations range from about 350 to 1492 m a.s.l. (Mount Prádel/Altwater). Average annual air temperatures range about 1–8 °C from top to foothills, average annual precipitation varies roughly between 800 and 1200 mm. Geologically the prevailing bedrock is metamorphic siliceous rocks – gneiss, schist, mica-schist, phyllite, amphibolite. Soils are moderately acidic cambisols (=inceptisols), podzols (=spodosols) and their mutual transitions (Appendix 1).

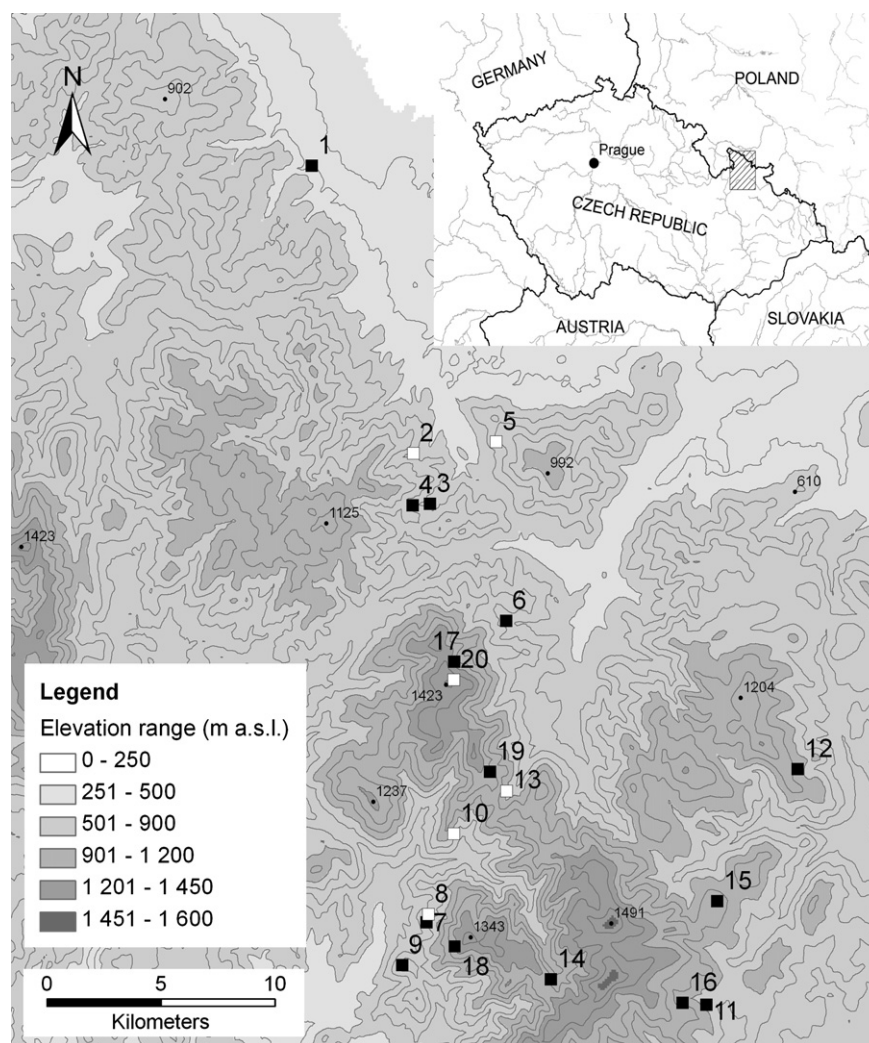


Fig. 1. Study area. Hatched cut-off (right map) delimits the area on the Czech–Polish boundary with resampled soil pits. Numbers in the left map correspond to the serial numbers of study sites. White squares denote sites with assessment of the potential relocation bias (recent pH heterogeneity). Elevation ranges correspond to the vegetation belts used in the Czech Republic (lowland, colline, submontane, montane, supramontane, subalpine).

Forests are the prevailing land cover type. The Jeseníky Protected Landscape Area, representing about 3/4 of the study area, is ca. 80% forested (www.nature.cz). Most forests are commercially managed for timber production. This concerns all sampling sites except for the one at the highest elevation (Fig. 1), which is a part of the nature reserve founded in 1903. Tree composition varied in the past millennia and human impact can be traced back to prehistory (Rybníček and Rybníčková, 2004; Tremblé et al., 2008). Without forestry interventions, the main tree dominants would be European beech (*Fagus sylvatica* L.), Silver fir (*Abies alba* Mill.) and Norway spruce (*Picea abies* (L.) Karsten). Climax spruce forests are usually assumed to be natural in the upper forest belt at elevations from about 1100–1200 to 1350 m a.s.l. (upper tree-line), however, according to palynological evidence spruce forests are relatively recent and anthropogenic in origin potentially enhanced by a colder oscillation in the Little Ice Age (Rybníček and Rybníčková, 2004). Nowadays forests at all altitudes are mainly Norway spruce plantations. However, there is still a relatively high proportion of European beech averaging about 20% and locally reaching twice that percentage.

2.2. Reference dataset

In the 1930s–60s, the German forest ecologist Friedrich K. Hartmann carried out an extensive study devoted to the variability of forest types in the mountainous areas of Central Europe (Hartmann and Jahn, 1967). The geographical range of this study extends from the Rhine to the western edge of the Carpathians. This by now half-forgotten monograph constitutes an outstanding source of high-quality primary data on forest ecosystems in Central Europe from before the major pollution impact. The species composition and structure of vegetation was recorded in several hundred sites. Soils from selected sites were pedologically characterized using classical soil pits. Profiles were described and in most cases samples taken from soil horizons were denoted A, B and C, reflecting the stratification typical of forest soils (Appendix 1).

2.3. Resampling methodology

Each one of Hartmann's sites was described in a standardized way and localized by the number of the forestry compartment in which it lay. Boundaries of forest compartments have not changed since the 1940s, but their numbers have. Old forestry maps deposited in the Opava and Olomouc archives were consulted and Hartmann's original plots localized. For determination of the resampling area, data on elevation, slope exposition and inclination were used, as well as notes on local topography. Tree species composition had to match the original description, i.e. the beech vs. spruce proportion had to be in accordance with the composition 60 years ago (Appendix 1). Sampling areas delimited in this way were sized about one hectare [see Fischer and Stöcklin (1997) for another example of this approach].

Twenty sites fulfilling the criteria were re-visited in autumn 2003. The sites are evenly distributed in the study area. One soil pit was dug at each site reaching the depth indicated by Hartmann (usually 1 m). The stratigraphy of the soil horizons described by Hartmann was carefully confronted with the field situation at present. Fifty-seven new soil samples were taken, each corresponding with the individual horizon type and the depth of sampling described in the 1940s.

2.4. Relocation bias assessment

Due to inexact re-localization of the original soil pits, spatial heterogeneity could have obscured temporal changes in the soil chemistry observed. The bias assessment consists of the comparison of variability of values detected at present and related to the old ones. For approaches to assess the relocation bias, see Fischer and Stöcklin (1997) and Ross et al. (2010). We assessed this bias in six sites, where two transects in horizontal and vertical directions regarding the slope orientation were established, starting from the soil pit. The basic idea was that if the old values were within the range of the values from transects, this would indicate that either acidification did not take place or there really is a bias due to spatial heterogeneity in soil pH higher than the rate of potential soil acidification. As pH varies mostly in upper horizons, soil samples only from A-horizons were taken in increasing distances of 2, 8, 15, 30, 60, ending at 100 m from the soil pit, following the maximum uncertainty of relocation. This resulted in 72 samples (see Appendix 2). It has to be noted that "distance" on transects is a relative variable as it has no relation to the real position of the old soil pit, which remains, after all, unknown within the 1 ha area (see also Section 2.7).

2.5. Soil horizons

Three types of soil horizons were distinguished. They follow the denotation used by Hartmann and correspond to basic concepts used today (ISSS–ISRIC–FAO, 1998). Horizon types were consistent between old and new samplings within a soil pit (see Appendix 1 for description). First, "horizon A" was the topsoil organic-mineral horizon underneath the humus layers. Second, "horizon B" was the subsoil mineral horizon, which includes various horizon types and subtypes. In this study, they were cambic, spodic and occasionally gleyic horizons (site 14). Third, "horizon

C" was the subsoil mineral horizon of weathered bedrock, i.e. usually very skeleton-rich. Occasionally gleyic features were observed in the subsoil (sites 12, 14, 16, 17, 20).

2.6. Laboratory analyses

Soil samples were air-dried and sieved to 2 mm fraction and were subjected to measurements of three soil acidity parameters: i) pH in water suspension (pH-H₂O), ii) pH in KCl solution suspension (pH-KCl), iii) exchangeable acidity. In addition, the analysis of total carbon content changes was performed, as this parameter can be influenced by acidification. We followed the methodology originally used by Hartmann 60 years ago (Hartmann and Jahn, 1967, pp. 275–280). For pH measurements, they used glass electrodes. The original measurements were done after the Second World War from the dry-stored samples (for validity of this approach see Blake et al., 2000). From each sample, 10 g of soil was suspended in 25 ml of distilled water or in 1 N KCl solution. The exchangeable acidity was measured using 100 g of soil and 250 ml 1 N KCl solution, and after one hour of stirring, sedimentation and filtration, the 125 ml of the resulting solution was titrated by Methyl red and 0.1 N NaOH per 100 g of dry mass. The total carbon content was analysed in a dry way, by weighing the dry samples, burning them at 800 °C for half an hour in a furnace, cooling and weighing again. Other soil parameters provided by Hartmann (content of Ca, Mg, K and P) were not re-analysed because of unavailable equipment and unclear analytic procedures. All measurements were carried out in the Laboratory of the Department of Geology and Pedology at the Mendel University of Agriculture and Forestry in Brno.

2.7. Data analysis

Paired analysis was applied to old and new samples regardless horizons. Differences were statistically tested by a two-sample test for dependent observations. Assumptions for normality of distributions were found valid using the Brown–Forsythe test for homogeneity of variances. Old and new values within the four parameters were correlated in order to test for the evenness of change in particular pairs of samples. Moreover, the four parameters were correlated using changes in old–new pairs of values. Regressions of pH-H₂O values against altitude for old and new values were constructed in order to visualize the altitude-dependent acidification patterns then and now. This was done separately for the three types of horizons. All the testing and graph drawings were performed in the statistical program STATISTICA, version 9 (©StatSoft, Inc. 1984–2010).

Next, a GLM analysis was performed to test for complex influences of the main factors and their interaction with time. The following independent (explanatory) variables were used in repeated measurements ANOVA provided in STATISTICA 9: altitude (in metres above sea level) and horizon (categories by types of horizons A, B and C). Tree composition (beech to spruce proportion) was strongly correlated with altitude (Fig. 2); confounding of the two factors was confirmed with regression analysis ($R^2 = 0.75$, $p < 0.001$). We therefore decided to omit tree composition from GLM keeping in mind that its net effect remained undistinguished. The four soil parameters were dependent (explained) variables, each with two repetitions (original and repeated values).

Bias by spatial variability due to the not exactly known position of the original soil pits was assessed visually. We argued that the distance-based method by Fischer and Stöcklin (1997) was not applicable as the actual position of the old pit was unknown within a 1 ha area. We therefore treated each new value as equal, regarding its distance from the soil pit. Distribution of values in all old measurements ($N = 19$), new measurements from soil pits ($N = 19$), and from the transects at the six sites ($N = 68$) were compared using boxplots constructed using STATISTICA 9. To see the individual profile patterns, values of pH-H₂O and pH-KCl from both horizontal and vertical transects were visually related to the old and new values from respective soil pits (see Appendix 2).

3. Results

3.1. Relocation bias assessment

Due to the not exactly known position of the original soil pits, the potential bias by erroneous placement of the new soil pits was assessed using six of the sampling sites. The old values were generally higher than values of both types of new samples. The latter did not differ much from each other (Fig. 3). Looking at profile-specific patterns, no clear pattern could be observed in relation to the distance from the soil pit both for pH-H₂O and pH-KCl (not shown). Except for site 20 all recent values are lower than the old ones, which is congruent with the conclusion on general pH decrease. We may assume that potential error in plot relocation was negligible and did not bias the observed acidification patterns.

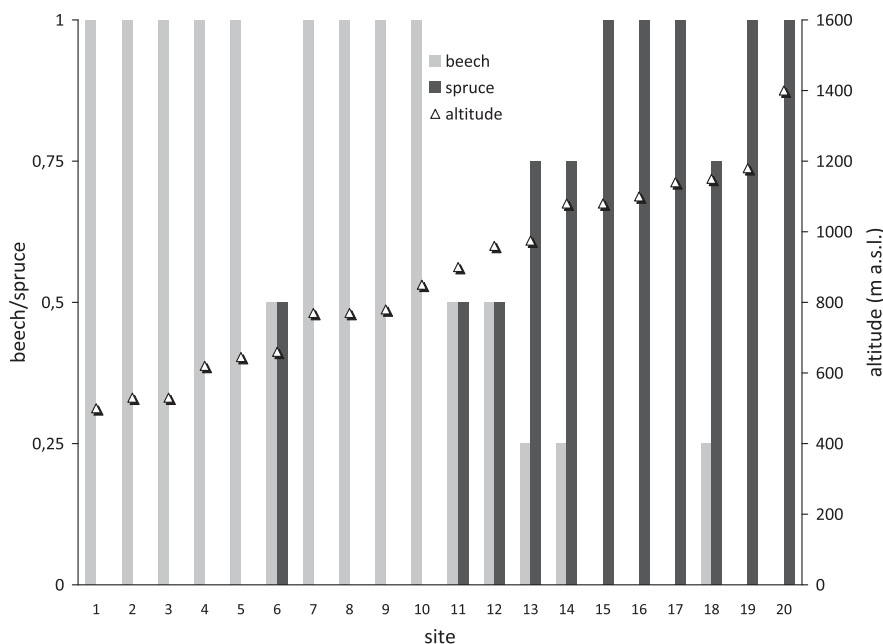


Fig. 2. Twenty sampling sites are distributed along a joint gradient of altitude and dominant tree species. European beech *Fagus sylvatica* (light grey bars) is gradually replaced by Norway spruce *Picea abies* (dark grey bars). Site numbers correspond to those in Appendix 1.

3.2. Changes in soil parameters

Soil acidity has decreased in 80–90% of compared pairs of soil samples across horizons both in pH-H₂O and pH-KCl (Fig. 4). The mean decrease was 0.69 for pH-H₂O and 0.56 for pH-KCl (cf. Table 1), with the maximum individual decrease at site 13 (2.78 for pH-H₂O and 2.28 for pH-KCl). Exchangeable acidity did not change significantly in general; only in the A-horizon a weak increase was detected. Organic matter content increased significantly (Table 1). Pairs of old and new pH values were positively correlated (correlation coefficient = 0.53 for pH-H₂O at $p = 0.001$ and 0.39 for pH-KCl at $p = 0.01$) indicating evenness of the acidification process and proving the robustness of resampling methodology at the same time. Moreover, changes in pH seem to be positively correlated with changes in organic matter content in horizon A (Fig. 5),

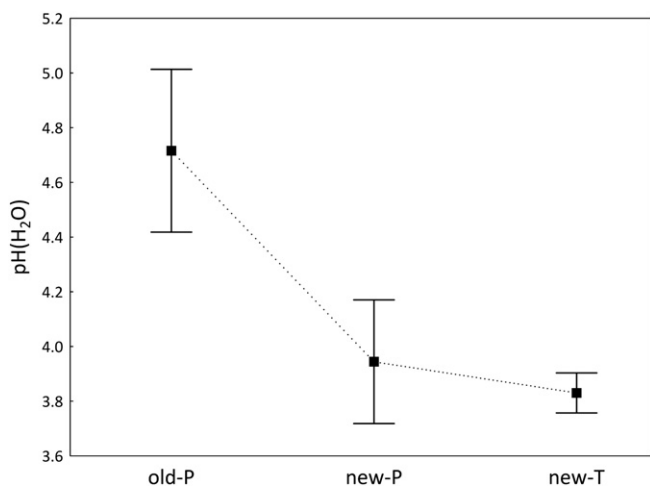


Fig. 3. Estimation of relocation error relating distribution of pH-H₂O values in A-horizon. Distribution of 1941/42 values related to soil pits (old-P), 2002/2003 values from soil pits (new-P), and 2002/2003 values from transects in sampling areas of six sites (new-T). Means and 95% confidence intervals are shown.

however the trend is not significant ($R^2 = 0.24$; $p = 0.07$). A similar pattern could not be observed in horizons B and C.

The results of repeated measurements ANOVA (Table 2) showed that the effects of the three factors (time, altitude and horizon) were largely independent. Soil horizon had a statistically highly significant effect on all four parameters. Altitude had a significant influence on pH-H₂O and organic matter but not on pH-KCl and exchangeable acidity. Time as a factor statistically significantly contributed to pH change but not to the other two parameters. Interaction between time and either altitude or horizon was statistically significant in organic matter, where an increase in the A-horizon could be observed, while in B- and C-horizons the changes were not distinct.

Altitudinal patterns in pH-H₂O and pH-KCl differed concerning the three horizon types (Figs. 6 and 7). In topsoil A-horizons, pH decreased sharply at low altitudes with beech forests (500–850 m, Fig. 2), while it did so less at higher elevations and remained the same in the highest-elevated site with a natural spruce forest (1000–1400 m). In mineral B-horizons this pattern was similar but less pronounced (pH-H₂O, Fig. 6) or dissipated (pH-KCl, Fig. 7). The soil-forming C-horizons have been acidified about evenly at all altitudes (pH-H₂O, Fig. 6) or showed a reversed acidification pattern (pH-KCl, Fig. 7) compared to A-horizons.

4. Discussion

4.1. Patterns in soil acidification

Acidification studies combining long-term comparison (over five decades) before and after pollution impact, landscape scale, and relatively long environmental gradients are rare (cf. Tamm and Hallbäck, 1986, 1988). There are a number of variables that can potentially modify acidification patterns. The most important finding of our study is that the observed strong acidification varies with a combination of the joint gradient of altitude/tree composition and soil horizontal stratification.

Hypothesis 1 on the differing rate of acidification along the altitudinal gradient is partly supported by the results. However,

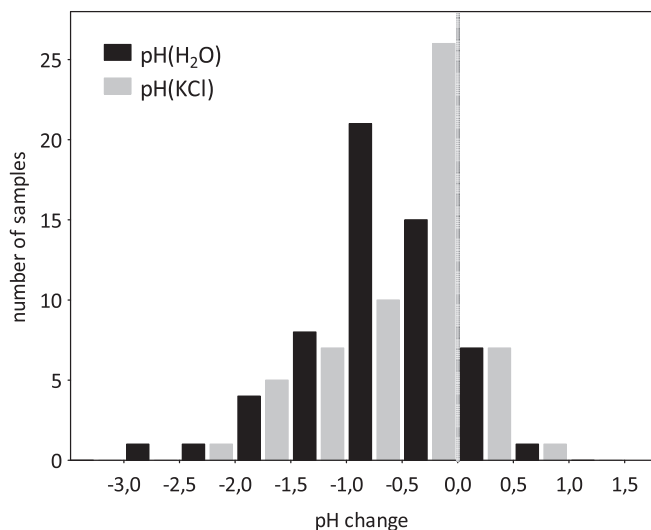


Fig. 4. Changes in pH values for pairs of old (1941/42) and new (2002/03) samples regardless of horizons. In most cases, pH decreased. Statistical values are for pH-H₂O: $N = 58$, average = -0.6909 , SD = 0.6478 , Max = 0.75 , Min = -2.78 ; for pH-KCl: $N = 57$, average = -0.5635 , SD = 0.6417 , Max = 0.88 , Min = -2.28 .

acidification was more pronounced at lower sites with beechwoods, while in coniferous habitats at high elevations it tended to dissipate. This is very much contrary to expectations. We interpret this as the effect of naturally low pH of sites at high elevations already before the pollution impact. Long-term influence of coniferous forests was considered a factor co-forming strongly acidified soils by Earl-Goulet et al. (1998) or Carnelli et al. (2004). Similarly, we assume that the presence of acidic soils in the high-elevated sites in the Eastern Sudetes is connected with the long-lasting natural acidification by spruce dominating the vegetation for at least three to four centuries (Rybníček and Rybníčková, 2004; Tremil et al., 2008).

Hypothesis 2 on the various magnitude of acidification between the soil horizons is supported. Differences in acidification patterns for topsoil were more pronounced than in different layers of subsoil. This demonstrates that considering soil acidification we need to “think vertically” (e.g. Brahy et al., 2000; Hédl and Rejšek, 2007). The least affected soil parameter, exchangeable acidity, increased reflecting acidification in the A-horizon, while remaining stable in both subsoil horizons. It seems that the acidification processes interfered again in the A-horizon, as increased organic matter content may be due to less severe pH decrease or its possible increase. Changes in acidity influence the content of base cations (namely that of Ca) which in turn affects humus accumulation in the topsoil (Likens et al., 1998).

Current acidification trends in Europe demonstrate the amelioration of soil and surface water chemistry due to a decrease

Table 1

Descriptive statistics and results of *t*-tests for dependent samples comparing old and new pairs of samples. Statistically significant changes were found for pH (decrease in both pH-H₂O and pH-KCl) and organic matter content (increase); $p < 0.05$ are in bold.

Parameter	Mean		SD		<i>t</i> -Test for dependent samples <i>N</i> pairs	<i>t</i> -Statistics		<i>p</i> -Value
	Old	New	Old	New		<i>t</i> -Statistics	<i>p</i> -Value	
pH (H ₂ O)	5.1	4.4	0.6	0.5	58	8.12	<0.001	
pH (KCl)	4.4	3.8	0.6	0.5	57	6.63	<0.001	
Exchangeable acidity (ml 0.1 N NaOH)	17.8	20.5	20.2	17.1	50	-0.86	0.396	
Organic matter content (%)	8.9	12.6	6.2	10.1	44	-3.21	0.002	

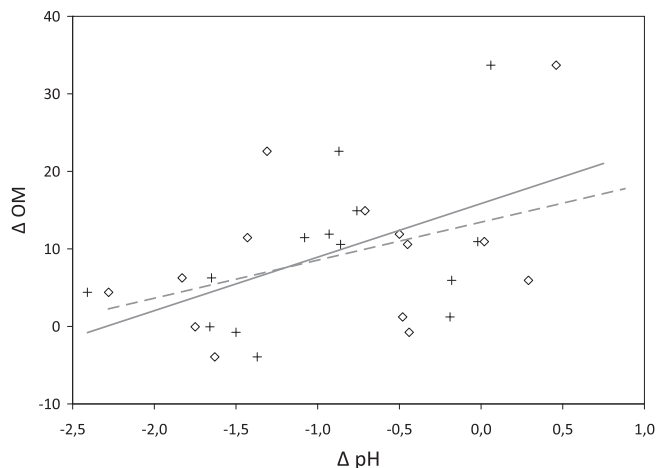


Fig. 5. Correlation between change in pH and change in organic matter content (OM; %) from the pairs of old (1941/42) and new (2002/03) samples of A-horizons. Crosses and solid regression line denote pH-H₂O, diamonds and dashed lines denote pH-KCl. General increase in OM is positively linked with pH change.

in acid pollution, of sulphur in particular (Woodin, 1989; Evans et al., 2001; Navrátil et al., 2007). Ecosystems have been slowly recovering from long-term acidification, which may be indicated by increasing concentration of dissolved organic carbon in stream waters (Evans et al., 2005). However, ongoing anthropogenic acidification can still be observed even in natural forests far from the main sources of pollution (Oulehle et al., 2010). Only a substantial reduction of acidic depositions could lead to a significant recovery of heavily damaged soils and ground waters (Hruška et al., 2002). Monitoring the properties of acidified soils including comparisons of their current state with historical data thus remains an important direction of environmental pollution research.

4.2. Relocation bias assessment

The observed soil changes include both real changes (i.e. temporal variability) and the influence of relocation errors (i.e. spatial variability). These two cannot be separated, only the latter can be minimized by restricting the area within which the old pits were most probably located. Analyzing the data, one can only assess whether the relocation error could have distorted the results or not (e.g., Ross et al., 2010). In this study, we showed that the relocation errors did not distort the observed temporal changes (at least concerning the soil pH in A-horizons). It was not possible to tell which of the pits were actually wrongly relocated because each pit had the same chance to be imprecisely relocated within its own area. It is therefore methodologically correct not to exclude any site from the dataset *a posteriori*, because the observed change could be equally due to the real change and relocation error at each site.

The size of forest units (compartments) into which Hartmann's plots were assigned is relatively small, estimated to about one hectare at most. Possible error is thus within a few dozen metres considering the delimitation by the size of forest compartment, altitude, slope and orientation, as well as information on local topography. Our results however quite clearly indicated that the influence of the relocation error was most probably negligible regarding the temporal changes in soil parameters. We observed rather large current variation in pH (about 1 unit) within an area of about 1 ha, yet it often remains within half a unit of pH. Tamm and Hallbäck (1986, 1988), who used data by Hesselman from the 1920s, were able to re-localize their soil pits with a precision of up to several hectares. We consider our study to be its methodological analogy and the spatial bias comparable.

Table 2

Results of the repeated measurements ANOVA for the four soil parameters. Test statistics are represented with *F* and *p* values, effects with *p* < 0.05 are printed in bold.

Effect	pH (H ₂ O)		pH (KCl)		Exc. acidity		Org. matter	
	<i>F</i>	<i>p</i>	<i>F</i>	<i>p</i>	<i>F</i>	<i>p</i>	<i>F</i>	<i>p</i>
Altitude	12.15	0.001	0.53	0.470	0.00	0.953	7.80	0.008
Horizon	31.34	<0.001	28.56	<0.001	31.69	<0.001	51.97	<0.001
Time	15.31	<0.001	6.16	0.016	0.32	0.575	0.010	0.940
Time × altitude	3.12	0.083	0.55	0.464	0.10	0.754	1.15	0.289
Time × horizon	0.42	0.661	0.69	0.508	1.20	0.312	10.67	<0.001

We would like to comment on two extreme outliers from the observed distributions. First, pH values have increased at site 14 the A-horizon by 0.7 and 0.8 for pH-H₂O and pH-KCl, respectively; and by about 0.1–0.2 in subsoil horizons for pH-H₂O only. The soil pit at site 14 in was both in the 1940s and 2000s in a water-logged slope with a gleyic soil, which could have undergone a different process than the rest of the sites. Second, an exceptionally large pH-H₂O

decrease (more than 2 units) was observed for site 13 (see Appendix 1). The historical soil pit may have been located within a small base-rich patch, while the recent pH values were in pre-vailing base-poor conditions. The recent value from the pit sample did not depart from the transect values.

4.3. Effects of vegetation on acidification and vice versa

We were not able to distinguish the influence of the two dominant tree species due to their strong correlation with altitude.

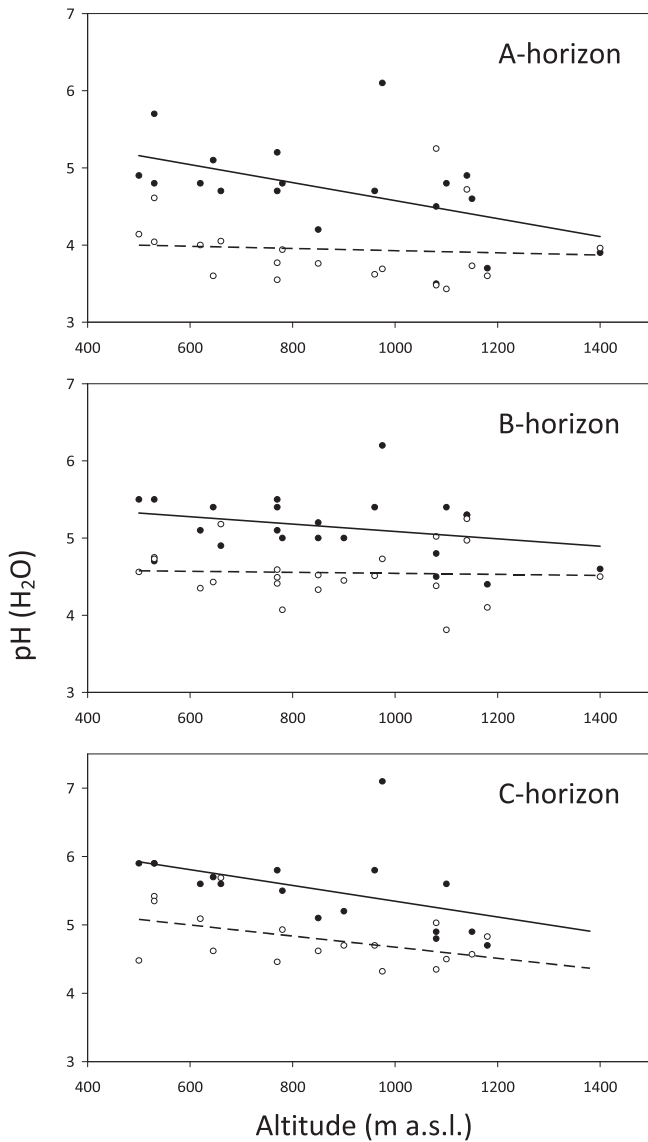


Fig. 6. Acidification patterns follow a combination of altitudinal gradient and horizontal soil stratification, as revealed in regressions of pH (H₂O) values on altitude. Values from 1941/42 are in black and solid regression lines, values from 2002/03 are in open symbols and dashed regression lines.

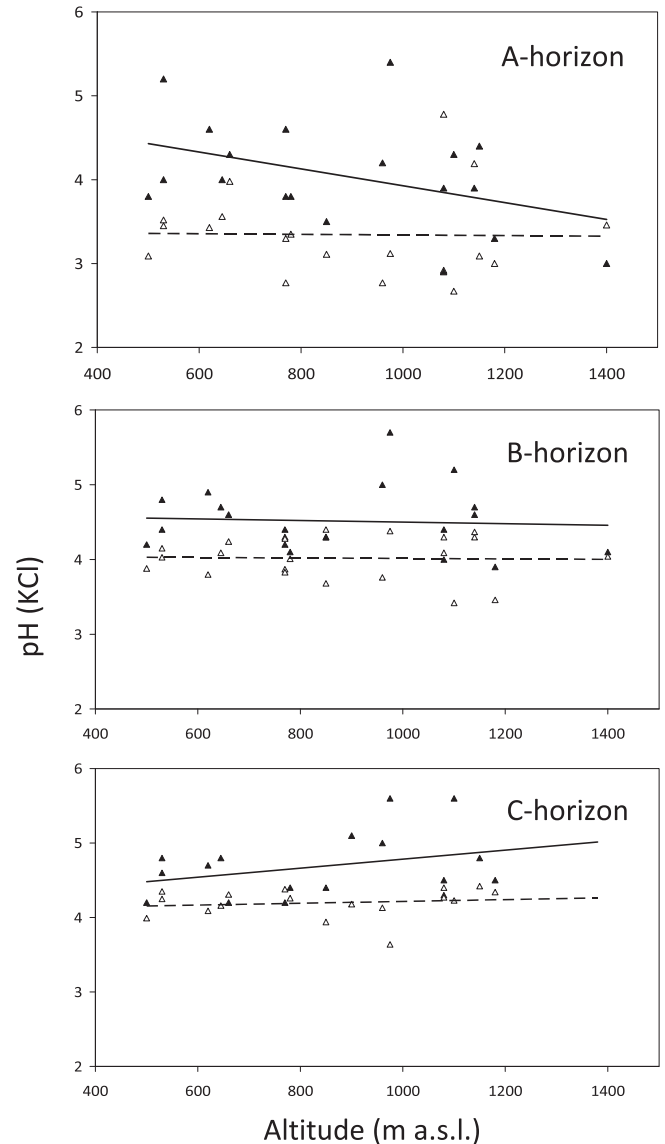


Fig. 7. Patterns in pH (KCl) values on altitude. For explanation see Fig. 6.

However, considering altitude and tree composition as a joint factor, our results contradict the conclusions on the higher acidification effect of Norway spruce in comparison with European beech (Oulehle and Hruška, 2005; Berger et al., 2006). In congruence with the long-term acidification in naturally spruce-dominated forests discussed above, our opinion is that over the historically relatively short period of industrial pollution, the acidification effect by spruce did not much differ from the undoubted acidification by beech (e.g., Kuehne et al., 2008). Focussing on forest understory, Van Dobben and de Vries (2010) reported the high importance of tree composition and the relatively low effect of acidification on understorey species composition in monitoring plots across Europe including the Czech Republic. We argue that the results of this study may report of only short-term differences by tree species, but they are not applicable to long-term processes including overall acidification of the forest cover, regardless of tree species. Airborne pollution then further increases acidification at the scale of about a century.

Forestry management applied between the two surveys could be the second main factor linked with acidification. The observed acidification may be at least partly due to forest ageing (e.g., Hédl, 2004b; Durak, 2010). Relating the forest age, composition and structure between the surveys, we concluded that no larger management actions took place at any of the sites. We further assumed that the effect of small-scale disturbances which may have occurred at some of the sites was negligible. Our conclusion, however, supports the hypothesis on concurrent acidification by forest ageing. This may explain the rather severe decrease of soil pH.

Soil acidity (effectively the soil calcium content) is one of the best predictors for patterns in vegetation (e.g., Falkengren-Grerup et al., 1995; Ewald, 2003). In recent years, one of the most important topics has been the effects of nitrogen deposition on plant diversity (Gilliam, 2006; Bobbink et al., 2010). As shown by studies examining the effects of atmospheric depositions on vegetation, acidification and eutrophication are among the most important factors responsible for diversity losses in the long term (Thimonier et al., 1994; Dupré et al., 2010; Maskell et al., 2010). However, nutrient-rich habitats may have experienced eutrophication but not acidification by nitrogen inputs (Maskell et al., 2010). In forests, acidification is often a combined result of airborne pollution and forest management actions and generally causes decline in species richness (e.g., Baeten et al., 2009; Durak, 2010).

Concerning our study area with the substrates constituted of nutrient poor siliceous bedrocks, long-term exposition to high sulphur and nitrogen deposition loads has resulted in a compositional shift in beechwood vegetation (Hédl, 2004b). Biodiversity decrease in the Eastern Sudetes Mountains is likely to have been caused by base cations depletion, which particularly affected the distribution of sensitive forest plant species (Fabiszewski and Břej, 2000; Hédl, 2004a). Eutrophication by nitrogen may also have occurred, but this was not indicated by the indirect evidence of vegetation changes. Plant species may not reflect environmental changes in straightforward ways.

5. Conclusions and outlook

Strong acidification was the major soil chemistry change in the study area since the 1940s. It was larger at lower (500–1000 m) than at higher (1000–1400 m) altitudes. This contradicts expectations based on other studies from Central Europe, where acidification is enhanced at higher elevations due to higher precipitation and more intense soil cation leaching than at lower sites. Effects of industrial pollution and forest ageing could have acted jointly while the two factors could not have been separated in this study.

Especially sensitive to acidification is topsoil, which has a strong message for forestry management. Foresters should avoid actions leading to the further deterioration of soil conditions (i.e. large clear-cuts, use of harvesters, excessive plantations of coniferous species) in order to mitigate adverse environmental changes in the future. From a methodological point of view, we support the utilization of historical soil data representing valuable information on soil parameters before the major pollution impact.

Acknowledgements

This study was funded by a one-year research grant by the Ministry of Education, Youth and Sport of the Czech Republic, Nr. 1023/3FR 1307, and supported by grants 206/08/0389 from the Grant Agency of the Czech Republic and AVOZ60050516 from the Academy of Sciences of the Czech Republic. P.P. was further supported by grant LC 06073 (Ministry of Education, Youth and Sport) and K.B. by grant 42900/1312/3114 (IGA: FES, CULS). We would like to thank M. Helan for his help with field sampling, V. Koblížková for the laboratory analyses, Z. Münzbergová and M. Hejda for advice with statistical analyses, J. Hruška for consultation on laboratory analytic methods, and M. Adámek for the preparation of Fig. 1. P. Szabó kindly corrected our English and gave valuable comments on the meaning of some of the sentences we have produced. Thanks go also to the handling editor and two anonymous reviewers, whose comments have substantially contributed to reconsidering our ideas.

Appendix. Supplementary data

Supplementary data associated with this article can be found, in the online, version at doi:10.1016/j.envpol.2011.06.014.

References

- Ahokas, H., 1997. Acidification of forest top soils in 60 years to the southwest of Helsinki. *Forest Ecology and Management* 94, 187–193.
- Baeten, L., Bauwens, B., De Schrijver, A., De Keersmaecker, L., Van Calster, H., Vanderkerkhove, K., Roelandt, B., Beeckman, H., Verheyen, K., 2009. Herb layer changes (1954–2000) related to the conversion of coppice-with-standards forest and soil acidification. *Applied Vegetation Science* 12, 187–197.
- Berge, E., Bartnicki, K., Olendrzynski, K., Tsyro, S.G., 1999. Long-term trends in emissions and transboundary transport of acidifying air pollution in Europe. *Journal of Environmental Management* 57, 31–50.
- Berger, T.W., Swoboda, S., Prohaska, T., Glatzel, G., 2006. The role of calcium uptake from deep soils for spruce (*Picea abies*) and beech (*Fagus sylvatica*). *Forest Ecology and Management* 229, 234–246.
- Billett, M.F., Parker-Jervis, F., Fitzpatrick, E.A., Cresser, M.S., 1990. Forest soil chemical changes between 1949/50 and 1987. *European Journal of Soil Science* 41, 133–145.
- Blake, L., Goulding, K.W.T., Mott, C.J.B., Poulton, P.R., 2000. Temporal changes in chemical properties of air-dried stored soils and their interpretation for long-term experiments. *European Journal of Soil Science* 51, 345–353.
- Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinnerby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.-W., Fenn, M., Gilliam, F., Nordin, A., Padro, L., De Vries, W., 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. *Ecological Applications* 20, 30–59.
- Borůvka, L., Mládková, L., Drábek, O., 2005. Factors controlling spatial distribution of soil acidification and Al forms in forest soils. *Journal of Inorganic Biochemistry* 99, 1796–1806.
- Brahy, V., Deckers, J., Delvaux, B., 2000. Estimation of soil weathering stage and acid neutralizing capacity in a toposequence Luvisol–Cambisol on loess under deciduous forest in Belgium. *European Journal of Soil Science* 51, 1–13.
- Carnelli, A.L., Theurillat, J.-P., Thion, M., Gaëlle, V., Talon, B., 2004. Past uppermost tree limit in the Central European Alps (Switzerland) based on soil and soil charcoal. *The Holocene* 14, 393–405.
- Dahlgren, R.A., Driscoll, C.T., 1994. The effects of whole-tree clear-cutting on soil processes at the Hubbard Brook Experimental Forest, New Hampshire, USA. *Plant and Soil* 158, 239–262.
- De Schrijver, A., Mertens, J., Geudens, G., Staelens, J., Campforts, E., Luyssaert, S., De Temmerman, L., De Keersmaecker, L., De Neve, S., Verheyen, K., 2006. Acidification of forested podzols in North Belgium during the period 1950–2000. *Science of the Total Environment* 361, 189–195.

- Dupré, C., Stevens, C.J., Ranke, T., Bleeker, A., Pepller-Lisbach, C., Gowing, D.J.G., Dise, N.B., Dorland, E., Bobbink, R., Diekmann, M., 2010. Changes in species richness and composition in European acidic grasslands over the past 70 years: the contribution of cumulative atmospheric nitrogen deposition. *Global Change Biology* 16, 344–357.
- Durak, T., 2010. Long-term trends in vegetation changes of managed versus unmanaged Eastern Carpathian beech forests. *Forest Ecology and Management* 260, 1333–1344.
- Earl-Goulet, J.R., Mahaney, W.C., Sanmugadas, K., Kalm, V., Hancock, R.G.V., 1998. Middle-Holocene timberline fluctuation: influence on the genesis of Podzols (Spodosols), Norra Storfjället Massif, northern Sweden. *The Holocene* 8, 705–718.
- Evans, C., Cullen, J., Alewell, C., Kopacek, J., Marchetto, A., Moldan, F., Prechtel, A., Rogora, M., Vesely, J., Wright, R., 2001. Recovery from acidification in European surface waters. *Hydrology and Earth System Sciences* 5, 283–297.
- Evans, C.D., Monteith, D.T., Cooper, D.M., 2005. Long-term increases in surface water dissolved organic carbon: observations, possible causes and environmental impacts. *Environmental Pollution* 137, 55–71.
- Ewald, J., 2003. The calcareous riddle: why are there so many calciphilous species in the central European flora? *Folia Geobotanica* 38, 357–366.
- Fabiszewski, J., Brej, T., 2000. Contemporary habitat and floristic changes in the Sudeten Mts. *Acta Societatis Botanicorum Poloniae* 69, 215–222.
- Falkengren-Grerup, U., 1987. Long-term changes in pH of forest soils in southern Sweden. *Environmental Pollution* 43, 79–90.
- Falkengren-Grerup, U., 1989. Soil acidification and its impact on ground vegetation. *Ambio* 18, 179–183.
- Falkengren-Grerup, U., Tyler, G., 1993. Experimental evidence for the relative sensitivity of deciduous forest plants to high soil acidity. *Forest Ecology and Management* 60, 311–326.
- Falkengren-Grerup, U., Quist, M.E., Tyler, G., 1995. Relative importance of exchangeable and soil solution cation concentrations to the distribution of vascular plants. *Environmental and Experimental Botany* 35, 9–15.
- Federer, C.A., Hornbeck, J.W., Tritton, L.M., Martin, T.W., Pierce, R.S., Smith, C.T., 1989. Long-term depletion of calcium and other nutrients in Eastern US forests. *Environmental Management* 13, 593–601.
- Fischer, M., Stöcklin, J., 1997. Local extinctions of plants in remnants of extensively used calcareous grasslands 1950–1985. *Conservation Biology* 11, 727–737.
- Gilliam, F.S., 2006. Response of the herbaceous layer of forest ecosystems to excess nitrogen deposition. *Journal of Ecology* 94, 1176–1191.
- Hallbäck, L., Tamm, C.O., 1986a. Changes in soil acidity from 1927 to 1982–84 in a forest area of south-west Sweden. *Scandinavian Journal of Forest Research* 1, 219–232.
- Hallbäck, L., Tamm, C.O., 1986b. Resampling of soil profiles in south-west Sweden for pH measurements. *Geologiska Foreningens i Stockholm (Journal of the Geological Society of Sweden)* 107, 291–295.
- Hartmann, F.K., Jahn, G., 1967. *Waldgesellschaften des mitteleuropäischen Gebirgsraumes nördlich der Alpen*. G. Fischer, Stuttgart.
- Hédl, R., 2004a. Retreat of *Dentaria enneaphyllos* L. in Rychlebské hory Mountains, Czech Republic, in perspective of habitat preferences. *Biologia (Bratislava)* 59, 417–423.
- Hédl, R., 2004b. Vegetation of beech forests in the Rychlebské Mountains, Czech Republic, re-inspected after 60 years with assessment of environmental changes. *Plant Ecology* 170, 243–265.
- Hédl, R., Rejšek, K., 2007. Soil changes after forty years of succession in an abandoned coppice in the Czech Republic. *Acta Agronomica Hungarica* 55, 453–474.
- Houška, J., Schejbalová, H., 2004. The dynamics of selected soil properties over 60 years in natural forest ecosystems: general tendencies and internal dynamics. *Ekológia (Bratislava)* 23 (Suppl. 2), 66–79.
- Hruška, J., 2007. Risks to soil biodiversity in EU substantially reduced by 2013. In: Petřík, P., Čámská, K., Dotlačil, L., Hruška, J., Fanta, J., Peterová, P., Poštulka, Z., Roudná, M., Vokasová, L. (Eds.), *Review of Knowledge Base and Biodiversity Research Results from the Czech Republic That Directly Contribute to the Sustainable Use of Biodiversity in Europe*. Institute of Botany, Academy of Sciences of the Czech Republic. www.ibot.cas.cz/biop.
- Hruška, J., Cienciala, E. (Eds.), 2003. *Long-Term Acidification and Nutrient Degradation of Forest Soils – Limiting Factors of Forestry Today*. Czech Ministry of Environment, Prague.
- Hruška, J., Moldan, F., Krám, P., 2002. Recovery from acidification in central Europe – observed and predicted changes of soil and streamwater chemistry in the Lysina catchment, Czech Republic. *Environmental Pollution* 120, 261–274.
- ISSS–ISRIC–FAO, 1998. World reference base for soil resources. *World Soil Resources Reports* 84, 1–92.
- Jönsson, U., Rosengren, U., Thelin, G., Nihlgaard, B., 2003. Acidification-induced chemical changes in coniferous forest soils in southern Sweden 1988–1999. *Environmental Pollution* 123, 75–83.
- Kopáček, J., Veselý, J., 2005. Sulfur and nitrogen emissions in the Czech Republic and Slovakia from 1850 till 2000. *Atmospheric Environment* 39, 2179–2188.
- Krám, P., Bishop, K.H., 2001. Overview of the MAGIC model applications in 1985–2000. In: *Detecting Environmental Change: Science and Society Conference Proceedings*. University College, London, pp. 20–21.
- Kuehne, C., Donath, C., Müller-Using, S.I., Bartsch, N., 2008. Nutrient fluxes via leaching from coarse woody debris in a *Fagus sylvatica* forest in the Solling Mountains, Germany. *Canadian Journal of Forest Research* 38, 2405–2413.
- Likens, G.E., Driscoll, C.T., Buso, D.C., 1996. Long-term effects of acid rain: response and recovery of a forest ecosystem. *Science* 272, 244–246.
- Likens, G.E., Driscoll, C.T., Buso, D.C., Siccama, T.G., Johnson, C.E., Lovett, G.M., Fahey, T.J., Reiners, W.A., Ryan, D.F., Martin, C.W., Bailey, S.W., 1998. The biogeochemistry of calcium at Hubbard Brook. *Biogeochemistry* 41, 89–173.
- Maskell, L.C., Smart, S.M., Bullock, J.M., Thompson, K., Stevens, C.J., 2010. Nitrogen deposition causes widespread loss of species richness in British habitats. *Global Change Biology* 16, 671–679.
- McNeill, J.R., 2000. *Something New Under the Sun: an Environmental History of the Twentieth-Century World*. W.W. Norton & Company, New York, NY.
- Navrátil, T., Kurz, D., Krám, P., Hofmeister, J., Hruška, J., 2007. Acidification and recovery of soil at a heavily impacted forest catchment (Lysina, Czech Republic) – SAFE modeling and field results. *Ecological Modelling* 205, 464–474.
- Oulehle, F., Hleb, R., Houška, J., Šamonil, P., Hofmeister, J., Hruška, J., 2010. Anthropogenic acidification effects in primeval forests in the Transcarpathian Mts., western Ukraine. *Science of the Total Environment* 408, 856–864.
- Oulehle, F., Hofmeister, J., Cudlín, P., Hruška, J., 2006. The effect of reduced atmospheric deposition on soil and soil solution chemistry at a site subjected to long-term acidification, Načetín, Czech Republic. *Science of the Total Environment* 370, 532–544.
- Oulehle, F., Hruška, J., 2005. Tree species (*Picea abies* and *Fagus sylvatica*) effects on soil water acidification and aluminium chemistry at sites subjected to long-term acidification in the Ore Mts., Czech Republic. *Journal of Inorganic Biochemistry* 99, 1822–1829.
- Porebska, G., Ostrowska, A., Borzyszkowski, J., 2008. Changes in the soil sorption complex of forest soils in Poland over the past 27 years. *Science of the Total Environment* 399, 105–112.
- Ross, L.C., Woodin, S.J., Hester, A., Thompson, D.B.A., Birks, H.J.B., 2010. How important is plot relocation accuracy when interpreting re-visitation studies of vegetation change? *Plant Ecology and Diversity* 1, 1–8.
- Rybníček, K., Rybníčková, E., 2004. Pollen analyses of sediments from the summit of the Praděd range in the Hrubý Jeseník Mts (Eastern Sudetes). *Preslia* 76, 331–347.
- Tamm, C.O., Hallbäck, L., 1986. Changes in soil pH over a 50-year period under different forest canopies in SW Sweden. *Water, Air, and Soil Pollution* 31, 337–341.
- Tamm, C.O., Hallbäck, L., 1988. Changes in soil acidity in two forest areas with different acid deposition: 1920s to 1980s. *Ambio* 17, 56–61.
- Thimonier, A., Dupuey, J.L., Bost, F., Becker, M., 1994. Simultaneous eutrophication and acidification of a forest ecosystem in northeast France. *New Phytologist* 126, 533–539.
- Treml, V., Jankovská, V., Petr, L., 2008. Holocene dynamics of the alpine timberline in the High Sudetes. *Biologia (Bratislava)* 63, 73–80.
- Van Dobben, H., de Vries, W., 2010. Relation between forest vegetation, atmospheric deposition and site conditions at regional and European scales. *Environmental Pollution* 158, 92–133.
- Veselý, J., Hruška, J., Norton, S.A., Johnson, C.E., 1998. Trends in the chemistry of acidified Bohemian lakes from 1984 to 1995: I. Major solutes. *Water, Air, and Soil Pollution* 108, 107–127.
- Vrba, J., Kopáček, J., Fott, J., Kohout, L., Nedbalová, L., Pražáková, M., Soldán, T., Schaumburg, J., 2003. Long-term studies (1871–2000) on acidification and recovery of lakes in the Bohemian Forest (central Europe). *Science of the Total Environment* 310, 73–85.
- Woodin, S.J., 1989. Environmental effect of air pollution in Britain. *Journal of Applied Ecology* 26, 749–761.
- Yanai, R.D., Siccama, T.G., Arthur, M.A., Federer, C.A., Friedland, A.J., 1999. Accumulation and depletion of base cations in forest floors in the Northeastern United States. *Ecology* 80, 2774–2787.