

Acidification of forested podzols in North Belgium during the period 1950–2000

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Abstract

Acidification of forest soils in Europe and North America has been an important concern over the last decades. The last area-covering survey of forest soil acidification in Flanders (North Belgium) goes back to 1985 [Ronse A, De Temmerman L, Guns M, De Borger R. Evolution of acidity, organic matter content, and CEC in uncultivated soils of North Belgium during the past 25 years. *Soil Sci*; 146, (1988), 453–460] and highlighted a significant acidification of the upper layer (0.3–0.4 m) of forested podzols during the period 1950–1985. The present study aimed to assess (1) to what extent further acidification of forested podzols occurred during the period 1985–2000 at different depths and (2) whether the average annual acidification rate accelerated or slowed down between 1985 and 2000 compared to the period 1950–1985.

Average soil pH–KCl values of podzols in northern Belgium dropped during the period 1985–2000. This decline extends to a depth of about 50 cm but was most pronounced and significant in the A horizon. In the A₀, A₁ and A₂ horizons, average pH dropped with 0.2, 0.3 and 0.1 units, and in the B_{tr} and C horizons with 0.1 units. No change in average pH value occurred in the B_h horizon. Average annual acidification rate of the A₁ horizon was significantly higher in the period 1985–2000 than in the period 1950–1985.

Changes in pH occurred in the entire soil profile during the period 1950/67–1985 likely because sulphate was the major form of acid deposition before 1985. After 1985, acid sulphur deposition decreased with more than 50% in North Belgium. In contrast, ammonium deposition almost doubled between 1950 and 1980, which may explain why soil acidification between 1985 and 2000 has been restricted to the upper soil horizons.

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1. Introduction

Acidification of forest soils in Europe and North America has been an important concern over the last decades. A large number of recent studies report a significant forest soil acidification over the past 50 years (e.g. Ahokas, 1997; Drohan and Sharpe, 1997; Dai et al., 1998; Hovmand and Hansen, 1999; Van der Salm and De Vries, 2000; Miller et al., 2001; Evers et al., 1974). Podzolic soils, which are naturally acid and depleted in nutrients, are particularly sensitive to acidification because of the low cation exchange capacity and the relatively small amounts of weatherable minerals (Wiklander and Anderson, 1972). Such sandy, podzolic soils are widespread in Flanders, the region of the present study. In addition, acidification of these podzols may be stimulated by the prevailing cover of pine plantations, past forest management practices and intensive cattle breeding in the studied region.

First of all, the prevailing forest types on podzols in the study area are secondary plantations of Scots pine (*Pinus sylvestris* L.) and Corsican pine (*Pinus nigra* ssp. *laricio* Maire) dating from the past two centuries (Afdeling Bos and Groen, 2001). Plantations of coniferous species are known to lead to notable modifications of the soil (Ovington, 1955; Challinor, 1968; Perala and Alban, 1982; Nihlgard, 1972; Binkley and Valentine, 1991; Berger et al., 2003). Compared to broadleaved species, coniferous litter is more resistant to biological degradation (Howard and Howard, 1990; Johansson, 1995) and conifers are found to be more efficient in collecting particles and cloud droplets from the atmosphere, enhancing acid deposition (Brown and Iles, 1991; Draaijers, 1993; De Schrijver et al., 2004).

A second factor that can affect soil acidification under the Flemish pine plantations is the past management, including clear cutting at 30–50 year intervals, total tree use and litter raking (Burny, 1999). All mentioned management practices are known to influence soil acidity especially because of base cation removal (Nilsson et al., 1982; Van Breemen, 1988; Dahlgren and Driscoll, 1994; Prietzel et al., 1997; Adams et al., 2000).

Thirdly, in the middle of the 20th century, intensive livestock husbandry has developed as the main agricultural activity on sandy soils in Flanders, caus-

ing high emissions of ammonia. Average acid deposition in Flanders is among the highest in Europe (UNECE and EC, 2003) and in spite of an important decrease in acid deposition (–12% in 2000 in comparison with 1990), all medium and long-term targets formulated in the Protocol of Göteborg and the European NEC directive are still exceeded (Mensinck et al., 2001).

The last area-covering survey of forest soil acidification in Flanders goes back to 1985 (Ronse et al., 1988). This survey highlighted a significant acidification of the upper layer (0.3–0.4 m) of forested podzols during the period 1950–1985. In 1985, most values of the topsoil pH were situated in the Fe/Al buffer range while the deeper horizons were inside the cation exchange buffer range. The first aim of the present study was to assess to what extent further acidification of forested podzols occurred during the period 1985–2000 at different depths. Our second aim was to find out whether the average annual acidification rate accelerated or slowed down between 1985 and 2000 compared to the period 1950–1985, considering the significant emission reductions during the period 1990–2000.

2. Materials and methods

2.1. Data collection

To investigate soil acidification, about 100 unfertilised soils were sampled in 1985 to determine soil pH (Ronse et al., 1988). These values were compared to pH values measured in the period 1950–1967 within the framework of the Belgian soil cartography (Ronse et al., 1988). On both sampling occasions, spatial coordinates, topography, vegetation, and soil profile characteristics (colour, thickness and texture of the horizons) were reported. Soil was sampled from the different horizons in a profile pit.

In 2000, the subset of forested podzols (originally 67 of 100 locations) from Ronse et al. (1988) was reconsidered. Only 33 of the former 67 forested sites on sandy podzols were encountered in an undisturbed state. Thirty-four sites were deforested, built-over or cultivated. Four of the remaining forested sites were cut and replanted between 1985 to 2000. These sites were not considered in the analysis since harvesting

operations lead to more favourable soil moisture and temperature conditions for growth of microorganisms, decomposition of resistant litter can be accelerated, and immobilized cations can be released, influencing soil pH (Hendrickson et al., 1989; Bock and Van Rees, 2002; Pennock and Van Kessel, 1997). Of the 29 remaining plots, 22 were situated under coniferous forest and 7 under deciduous forest.

Based on the available information, the sampling sites were relocated. However, digging the soil profile pits in 1988 disturbed the original sample points. Therefore, we took five soil samples in undisturbed soil with a gouge auger. One sample was taken as close as possible to the original site and four additional samples were taken at a 2 m radius around it, one in each cardinal direction. To assess local variability of soil pH, samples were not pooled but stored and analysed separately.

Each auger sample was divided into soil horizons, and subsamples of each horizon were taken after measuring the thickness of each horizon (Table 1). Within the eluvial- or A horizons, distinction was made between the upper mineral horizon A₁ and the leached mineral horizon A₂. Eluvial or B horizons were those enriched with humus (B_h) or iron (B_{ir}). The parent material was indicated as the C horizon.

2.2. Sample preparation and chemical analyses

Samples were dried for 48 h at 50 °C before sieving (soil samples) or grinding (litter samples). Soil pH–KCl was measured since this potential acidity is less dependent on the soil moisture content and consequently less variable than pH–H₂O (Thomas, 1996). The pH was measured using a glass electrode (Orion, model 920A) after extracting 8 g air-dried soil or litter with 20 ml KCl solution (1 M) during 15 min,

Table 1
Mean thickness (cm) of the soil horizons of podzols (*n*=29) in 1950/67, 1985 and 2000

Level	Horizon	Depth
0	A ₀	–
1	A ₁	0–11
2	A ₂	11–24
3	B _h	24–35
4	B _{ir}	35–44
5	C	44–

similarly to the previous research of Ronse et al. (1988).

2.3. Quality control

The quality of chemical analyses was checked by including repeated measurements of reference samples. Each 15 samples, a certified sample of pH=6 was analysed. The results were rejected if pH value was higher than 6.02. Then, the previous 15 samples were re-analysed after calibration.

2.4. Statistical analysis

For further statistical analyses, the 5 replicate pH values were averaged for each soil horizon of each sampling point since standard deviation was very low (coefficient of variation=4.23%). Before averaging, pH values were converted into H⁺ concentrations. Average soil pH of each soil horizon was obtained by calculating the $-\log$ (average H⁺ concentration).

To compare the average soil or litter H⁺ concentrations in 1950/67, 1985 and 2000, Wilcoxon signed rank tests were performed (Neter et al., 1996). We choose this non-parametric test because the differences in H⁺ concentrations between years for each horizon did not meet the conditions of normality. For the set of three pairwise comparisons, a Bonferroni correction was applied if differences in H⁺ concentrations were compared between years. In that case, a confidence coefficient of 0.017 was obtained through division of the family level 0.05-confidence coefficient by three (Neter et al., 1996).

To compare the average annual acidification rate between the periods 1950/1967–1985 and 1985–2000, the data were expressed in terms of increase of H⁺ concentration per gram of soil and per year. Therefore, for each sampling point, the H⁺ concentrations per gram of soil were divided by the exact time span (number of years) between measuring dates. For this dataset, the conditions of normality were met and the average annual acidification rate between both periods was compared using a paired samples *t*-test (Neter et al., 1996). Historical data of litter pH–KCl were only available for the year 1985, so no comparison of the average annual acidification rate between periods was possible for the litter layer.

All statistical analyses were made with SPSS 11.0 for Windows.

3. Results

3.1. Comparison of pH values and H^+ concentrations between sampling years

The average pH-KCl of the humus layer (A_0) decreased from 3.0 to 2.8 (Fig. 1) between 1985 and 2000. The pH of the A_1 horizon declined 0.1 unit between 1950/67 and 1985, and 0.3 units between 1985 and 2000. In contrast to the A_1 horizon, the pH in the A_2 horizon declined more between 1950/67 and 1985 (0.3 units) than between 1985 and 2000 (0.1 unit). Similarly to the A_2 horizon, the pH of the B horizons and the C horizon declined more between 1950/67 and 1985.

The H^+ concentrations in 2000 significantly increased compared to 1950/67 throughout the complete mineral soil profile (Tables 2 and 3). Between 1985 and 2000, H^+ concentrations increased significantly in

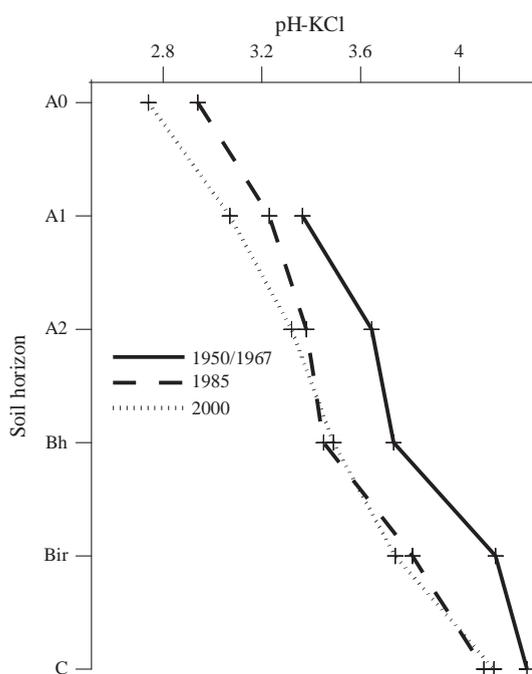


Fig. 1. Evolution in time of pH-KCl in different horizons of 29 podzol soils.

Table 2

Average H^+ concentrations and their standard deviation (10^{-6} mol g^{-1} soil) in different horizons of 29 podzol soils

Level	1950/67	1985	2000
A_0	–	2.56F1.97	4.25F2.07
A_1	1.06F1.12	1.32F0.83	2.27F1.06
A_2	0.48F0.53	0.95F0.86	1.21F1.02
B_h	0.45F0.48	0.88F1.16	0.82F0.77
B_{ir}	0.16F0.06	0.36F0.86	0.48F0.67
C	0.14F0.17	0.21F0.19	0.18F0.14

the three A horizons (Table 3). In the deeper horizons, no significant change in H^+ concentration was detected in this period. Changes in H^+ concentrations did not differ between coniferous and deciduous plots (numbers not shown).

3.2. Comparison of average annual acidification rate during the periods 1950/67–1985 and 1985–2000

The average annual acidification rate in the A_1 horizon increased from $8F48 \times 10^{-9}$ mol g^{-1} yr^{-1} between 1950/67 and 1985 to $63F56 \times 10^{-9}$ mol g^{-1} yr^{-1} between 1985 and 2000 (Fig. 2). According to a paired t -test, which takes the difference in acidification rate of individual sample points into account, this increase was highly significant. This means that the acidification process in the A_1 horizon significantly accelerated during the period 1985–2000 compared to 1950/67–1985. In the A_2 horizon, the acidification rate remained unchanged between 1950/67 and 2000 ($18F37$ and $17F47 \times 10^{-9}$ mol g^{-1} yr^{-1} for both periods, respectively). In the B_h , B_{ir} and C horizons,

Table 3

Significance level (p) of the difference in H^+ concentrations between the sampling years 1950/67, 1985, and 2000, as given by a Wilcoxon signed rank test

Level	1950/67–1985	1985–2000	1950/67–2000
A_0	–	0.005*	–
A_1	0.000*	0.000*	0.000*
A_2	0.003*	0.008*	0.000*
B_h	0.006*	0.581	0.003*
B_{ir}	0.046	0.109	0.002*
C	0.021	0.023	0.003*

Average soil horizon H^+ concentrations were significantly different at the 95% confidence level (indicated by *) when $p < 0.017$, because of the Bonferroni correction for multiple comparisons.

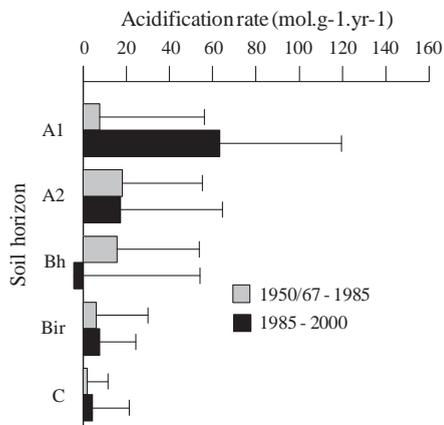


Fig. 2. Average annual acidification rate ($\text{mol g}^{-1} \text{yr}^{-1}$) with standard deviation bars in positive direction during the periods 1950/67–1985 and 1985–2000.

the differences in the acidification rate were also not significant.

4. Discussion

In the present study, soil samples were analysed on pH–KCl giving insight in the potential acidity of the soil. No analyses on actual acidity (pH–H₂O) were performed since the high dependence on soil moisture content can affect interpretation of temporal changes (Thomas, 1996). Based on pH–H₂O values, Ulrich (1983a) distinguished different buffer ranges for interpretation of pH values. Assuming that pH–H₂O and pH–KCl values usually differ about 1 unit in acid soils (Thomas, 1996), it can be concluded that in 2000 the A₀ and A₁ horizons in most surveyed plots were situated in the Al buffer range, which ranges from pH–H₂O 2.8 to 4.2 (Ulrich, 1983a). This means that in 2000 the upper horizons (A₀ and A₁) of the studied podzols have moved into the range where aluminium hydroxides, originating from weathering products of primary silicates (mainly clay minerals), dissolve into the soil solution and leach into the ground water (Ulrich, 1983a). The average pH of the A₂, B and C horizons were still within the cation exchange buffer, which ranges from pH–H₂O 4.2 to 5.0. In 1985, only the average pH of the A₀ horizons was situated in the Al buffer range, while the pH of the entire mineral soil profile was on average situated in the cation exchange buffer range. Evidence for the

shift of podzolic forested soils to the Al buffer range can be found in very high concentrations of aluminium in the soil solution found by Neiryneck et al. (2002) and De Schrijver et al. (2004).

Acidification of forest soils is a result of natural soil forming processes, forest growth and anthropogenic activities as acid deposition. The average internal proton production for six podzolic forest soils in the Netherlands, mainly as a consequence of organic acid deprotonation, was calculated to range from 0.1 to 0.9 $\text{kmol ha}^{-1} \text{yr}^{-1}$ (De Vries and Breeuwisma, 1985). Proton production as a consequence of forest growth depends on the type of harvesting and was reported to vary from 0.5 to 2.0 $\text{kmol ha}^{-1} \text{yr}^{-1}$ (Ulrich and Matzner, 1983; Van Breemen et al., 1984) in case of total tree use and 0.5 to 0.6 $\text{kmol ha}^{-1} \text{yr}^{-1}$ (Ulrich, 1983c) in case of conventional harvesting, in which only stems are removed. In North Belgium, acidifying deposition in 2000 was found to be on average 4.5 kmol ha^{-1} (Mensinck et al., 2001). Summarizing it can be concluded that soil acidification resulting from atmospheric deposition clearly exceeds internally generated protons on this type of soils. The importance of acid deposition for the acidification of forest soils was also confirmed by Bredemeier (1989), who calculated the internal generation of acidity to account for less than 30% of total proton load.

Changes in pH occurred in the entire soil profile during the period 1950/67–1985, while the main changes between 1985 and 2000 were restricted to the A horizons. We hypothesize that this observation can be related to the different composition of the acidifying atmospheric deposition during the last 50 years. In the eighties, sulphur (S) emissions were generally considered the most important cause of acid deposition. However, since then S emissions and thereby S deposition have constantly decreased over large parts of Europe, whereas NH₃ emissions mostly stayed constant or even increased (de Vries et al., 1991). This general European pattern also holds for the region of the present study. Emissions of SO₂ in Belgium in 1970 were twice as high as in 1950 and in 1985 (Mylona, 1996). Furthermore, S emission and deposition decreased with about 40% in North Belgium between 1990 and 2000 (Mensinck et al., 2001). Deposition of ammonium in Belgium almost doubled between 1950 and 1980 (Asman et al., 1988) and

slightly increased (+5.4%) between 1990 and 2000 (Mensinck et al., 2001). In 2000, the average acid deposition in Flanders amounted to $4496 \text{ mol ha}^{-1} \text{ yr}^{-1}$, of which $1200 \text{ mol ha}^{-1} \text{ yr}^{-1}$ came from SO_x , $1113 \text{ mol ha}^{-1} \text{ yr}^{-1}$ from NO_y , and $2183 \text{ mol ha}^{-1} \text{ yr}^{-1}$ from NH_x (NH_4^+ deposition was converted to deposition of H^+ assuming complete nitrification in the soil) (Mensinck et al., 2001).

The process of sulphate neutralization is not bound to any depth range in the soil (Ulrich, 1983b). The fact that S deposition was the dominant acidifying pollutant can explain why the average pH dropped in the complete soil profile during the period 1950/67–1985. During the period 1985–2000, soil acidification mainly occurred in the upper soil horizons, which may be due to the fact that the acidifying process of ammonium nitrification is aerobic and hence mainly restricted to the upper soil horizons.

Mineral weathering can help offset soil acidification — either directly in the upper soil horizons or indirectly due to weathering in lower soil horizons and redistribution due to plant uptake and litterfall. Our results can be related to the study of Van Ranst et al. (2002), who thoroughly studied the current acid neutralizing capacity (ANC) represented by the base cations and aluminium of the litter layer and the upper 10 cm of the mineral soil of two podzols under coniferous forest in North Belgium. The soil, the vegetation and the deposition load of both plots are comparable to the conditions of most of the plots in our survey. When the total ANC values of the forest floor (67 and 96 kmol ha^{-1} for both plots, respectively) and the upper 10 cm of the mineral soil (1147 and 871 kmol ha^{-1}) (Van Ranst et al., 2002) are compared with the average acid loadings in both plots (between 4.4 and $4.7 \text{ kmol ha}^{-1} \text{ yr}^{-1}$), and assuming that all acid input is buffered in the mentioned upper soil horizons, only 205 to 276 years of buffering capacity of the upper 10 cm is still left. In reality, this period will be longer than calculated since part of the acid will also be buffered in deeper horizons, for which no ANC-calculations were performed.

The critical load for protecting the acid buffering potential of the soil was exceeded in the majority of North Belgian forest ecosystems in 2000 (Mensinck et al., 2001). This clearly indicates the dramatic condi-

tion of North Belgian forest ecosystems and points out the urgent need for further drastic reduction emissions in the near future.

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