Recent carbon and nitrogen accumulation and acidification in soils of two Scots pine ecosystems in Southern Germany

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Abstract Changes of the soil chemical status during the recent 22–30 years at two historically degraded forest sites in southern Germany (Pfaffenwinkel, Pustert) stocked with mature Scots pine (*Pinus sylvestris* L.) stands were studied by repeated soil inventories conducted in 1974, 1982–1984, 1994, and 2004 on replicated control plots of fertilization experiments, allowing a statistical analysis. Additionally, the nutritional status of the stands at all plots was monitored from 1964 until 2004 by annual or bi-annual analysis of current-year foliage, and stand growth was assessed by repeated stand inventories carried out in 3- to 9-year intervals. For both sites, a statistically significant systematic decrease of the forest floor C/N ratio between 1974 and 2004 from 35.4 to 29.2 (Pfaffenwinkel) and from 36.5 to 23.0 (Pustert) was observed. The soils at both sites also showed a considerable accumulation of organic carbon (210 and 400 kg C ha^{-1} year^{-1} for Pfaffenwinkel and Pustert, respectively) and nitrogen (13 and 18 kg N ha^{-1} year^{-1}). In addition, the mineral topsoil at both sites has acidified considerably, indicated by significantly decreased pH values (Pustert only; mean decrease 0.1 units per decade), base saturation, and base cation stocks. The trend of N enrichment and base cation loss in the soils is mirrored by the trends of stand nutrition at both sites, which are characterized by improved N nutrition and reduced supply with K, Mg (Pustert only), and Ca. The results confirm findings of other studies indicating a recent N eutrophication and acidification of forest soils in Central Europe and southern Scandinavia. Since soils with historic degradation due to earlier non-sustainable forest utilization are widespread in Central Europe, the results obtained on our study sites probably apply for large forested areas, suggesting a significant potential of Central European forests to sequester atmospheric carbon and nitrogen not only in stand biomass, but also in the soil.

Keywords Carbon sequestration ·
N eutrophication · Pinus sylvestris · Soil acidity ·
Soil monitoring

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Introduction

Since the end of the last glaciation, the soils in humid regions of the Northern hemisphere have experienced a significant loss of "base cations" (i.e., Ca^{2+}, Mg^{2+}, K^+, Na^+) due to leaching with the soil seepage water (Mazzarino et al. 1983; Ulrich 1995) and a significant accumulation of carbon and nitrogen concomitantly with the establishment of a vegetation cover. Particularly at sites with poor bedrock, where mineral weathering could not compensate for base cation loss by leaching, the pH and exchangeable base cation pools in the topsoil gradually have decreased. In European forests, which often have been utilized for centuries for various purposes, such as wood, charcoal, or potash production, or as grazing ground, the natural trend of C and N accumulation in the soil often has been reversed, and natural soil acidification has been accelerated significantly by the export of C, N, and base cations with harvested biomass (cf. Kreutzer 1976; Ulrich 1995; Olsson et al. 1996). At many places, forest soils have been seriously degraded by non-sustainable historic forest utilization (e.g. Kreutzer 1972; Glatzel 1991). Starting with the industrialization, and skyrocketing after World War II (Ulrich 1989; Driscoll et al. 2001), anthropogenic N and S deposition in Central Europe, southern Scandinavia, and the eastern part of the USA counteracted or reversed the historic N depletion of forest ecosystems (von Zezschwitz 1985; Aber et al. 1989; Billet et al. 1990; McNulty et al. 1991; Buberl et al. 1994; Meiwes et al. 2002). Moreover, except for regions in the vicinity of fly ash-emitting power plants which experienced considerable soil alkalinization (Konopatzky and Freyer 1999), atmospheric N and S deposition accelerated the acidification and base cation depletion of forest soils (e.g. Hallbäcken and Tamm 1986; Federer et al. 1989; Hallbäcken 1992; Johnson et al. 1994; Wesselink et al. 1995; Likens et al. 1996; Jönsson et al. 2003). At the same time, excessive nutrient exports from Central European forests by over-exploitation, such as litter-raking and forest pasture were stopped. At present, large forested areas in Europe and in the northeastern USA are considered to experience a combination of N accumulation and soil acidification, i.e. a loss of acid neutralizing capacity (exchangeable "base cations") in the topsoil (Aber et al. 1998; Kreutzer et al. 1998). This development is considered to be of major environmental relevance since it infers in the long run the risk of imbalanced stand nutrition (e.g. Prietzel and Kölling 1998), and a destabilisation of forest stands (Hofmann et al. 1990; Likens et al. 1996). Further consequences are a loss of rare plant species adapted to low N availability (Hofmann et al. 1990; Rodenkirchen 1993), a perturbation of freshwater ecosystems, and a deteriorated groundwater quality (Aber et al. 1998). At the same time, forest growth in large areas of Europe has increased significantly in recent decades (Spiecker 1999), resulting in an increased transfer rate of base cations from the soil into stand biomass. The latter process promotes the rate of temporal soil acidification during the development of forest stands (Hallbäcken and Tamm 1986; Olsson et al. 1996), and results in increased soil acidification when the stand biomass is exported by harvest (Kreutzer 1976; 1979; Staaf and Olsson 1991; Johnson and Todd 1998), unless the exported base cations are replaced by mineral weathering, atmospheric deposition, or fertilization.

The current widespread N enrichment and topsoil acidification in many forests of Central Europe, southern Scandinavia, and North America is unquestioned among scientists, and based on numerous case studies where either (1) soil profiles have been monitored repeatedly (e.g. Hallbäcken and Tamm 1986; Ulrich et al. 1989; Johnson et al. 1994), (2) nutrient input-output balances were calculated (e.g. Markewitz et al. 1998), or (3) comparable sites subject to N and/or S deposition gradients were analyzed (e.g. Hofmann et al. 1990; Spangenberg and Kölling 2002; Prietzel et al. 2004). However, robust proofs of the N enrichment and topsoil acidification trends are scarce due to the rarity of adequate long-term (>20–30 years) forest ecosystem monitoring studies with replicated plots, which are required for a statistical analysis and an exact quantification of observed changes (Hurlbert 1984). This paper presents results of a long-term study, in which the topsoil (forest floor, uppermost 30 cm mineral soil) chemistry, the growth, and the

nutrient status of two mature Scots pine (*Pinus sylvestris* L.) stands in southern Germany (Pfaffenwinkel, Pustert) have been monitored for several decades on replicated control plots of two forest fertilization experiments. The main questions to be addressed in this study were:

- Have the topsoil N and C pools changed significantly within the last 22–30 years?
- Have the topsoils acidified significantly during that period?
- Are the changes in soil chemistry reflected in the long-term development of the nutritional status of the pine stands?

Materials and methods

Study sites

The investigation was conducted in the Scots pine (*Pinus sylvestris* L.) stands Pfaffenwinkel and Pustert in Upper Palatia (Bavaria, Germany). Important properties of the sites and stands are given in Table 1. The stand Pfaffenwinkel is located 5 km SE of the town Waldsassen. It has a cool, subcontinental climate. The geological parent material is intensively weathered tertiary phyllite (saprolith) covered by pleistocenic periglacial solifluidal debris. From this material acidic, nutrient poor Dystric Cambisols with silt loam texture have developed. The soil (Table 2) is intensively weathered and had been heavily degraded by historic litter-raking, forest pasture, and over-exploitation. The forest floor is a mor

with wide C/N and C/P ratios. The soil is acidic down to more than 1 m depth; in the uppermost 70 cm of the mineral soil the base saturation is less than 25%. In 1964, the site was densely stocked with 86-year-old Scots pines of poor quality and growth (site index IV.6 according to Wiedemann 1943). In the period 1985–1987, the atmospheric S deposition at Pfaffenwinkel was 47 kg S ha^{-1} year^{-1}, and the deposition of NO$_3$–N was 8 kg ha^{-1} year^{-1} (Fischer 1989). In 2004, the atmospheric S input was only 15 kg ha^{-1} year^{-1}; the atmospheric N deposition was 24 kg ha^{-1} year^{-1}, consisting of 19 kg ha^{-1} year^{-1} NO$_3$–N and 5 kg ha^{-1} year^{-1} NH$_4$–N. Accepting the same relation between NO$_3$–N and NH$_4$–N in 1985–1987 as in 2004, the total N deposition in 1985–1987 can be calculated as 10 kg N ha^{-1} year^{-1}. The decrease in atmospheric S deposition and the increase in N deposition at Pfaffenwinkel in the last 20 years reflect the general deposition trends in Central Europe.

The stand Pustert is located 50 km north of the city of Regensburg. Compared to Pfaffenwinkel, the climate is warmer (Table 1) and less continental. The geological parent material is pleistocenic solifluidal silt-loam covering intensively weathered cretaceous sediments. The soil is a Stagnic Albeluvisol with a topsoil of silt loam texture and clayey subsoil (Table 2). Also the Pustert site has been heavily degraded by historic litter-raking, forest pasture, and over-exploitation. As with Pfaffenwinkel, the forest floor is a mor with wide C/N and C/P ratios. The soil at Pustert is characterized by impeded drainage in the subsoil, resulting in temporal water logging

Table 1 Short characterization of the studied sites and stands

	Pfaffenwinkel	Pustert
Elevation	528–542 m a.s.l.	477–482 m a.s.l.
Slope aspect	5% NW	3% NW
Mean annual air temperature (growing season: May–September)	5.8°C (12.1°C)	7.2°C (14.7°C)
Mean annual precipitation	615 mm	650 mm
(Growing season: May–September)	325 mm	380 mm
Water regime	Fairly dry	Seasonal change from wet to dry
Soil type	Dystric Cambisol (formerly litter-raked)	Stagnic Albeluvisol (formerly litter-raked)
Trees (N/ha)	380	323
Basal area (m²/ha)	22.6	23.3
Height of dominant trees (m)	24.4 (in 1998)	24.2 (in 1999)

Table 2 Important properties of the soils at the sites Pfaffenwinkel and Pustert in 1999

Horizon	Depth (cm)	Coarse fragments (%)	Texture	C_{org} (g kg^{-1})	Total N (g kg^{-1})	ECEC (mmol$_c$ kg^{-1})	BS (%)	pH (CaCl$_2$)
Pfaffenwinkel								
O	9–0	NA	NA	440.6	14.3	285	35	2.7
AE	0–4	0.5	Silt loam	37.5	1.6	94	11	3.0
Bw	4–17	10	Silt loam	15.8	0.94	55	8	3.7
Bw2	17–37	10	Silt loam	3.3	0.63	29	14	4.0
Bw3	37–60	10	Silt loam	1.4	0.56	27	16	4.0
BC	60–67	30	Silt loam	0.8	0.41	16	23	4.0
2BC	67–73	5	Silt loam	0.9	0.35	17	20	3.9
3BC	73–93	25	Silt loam	0.5	0.18	12	30	4.0
4BC	93–113	70	Silt loam	0.2	0.25	10	32	3.9
Pustert								
O	5–0	NA	NA	380.3	15.5	251	51	3.3
Ah	0–2	10	Silt loam	83.2	5.26	85	30	3.2
AE	2–4	10	Silt loam	33.7	3.11	56	16	3.3
E	4–27	15	Silt loam	8.1	0.48	38	10	3.8
Bt	27–44	20	Loam	2.8	0.27	78	14	3.8
2Btg	44–65	20	Clay	1.9	0.34	119	47	3.9
2BC	65–80	25	Clay	1.6	0.28	112	54	3.9

ECEC effective cation exchange capacity (batch extraction with 0.5 M NH$_4$Cl), *BS* Base saturation, *NA* Not analyzed

during winter and spring. It is less deeply acidified than the soil at Pfaffenwinkel; the base saturation in the subsoil (44–80 cm) is about 50% (Table 2). In 1964, Pustert was densely stocked with 77-year-old Scots pine of poor quality and growth; however, with a site index of III.8 (Wiedemann 1943), the growth of the stand was better than at Pfaffenwinkel. In 2004, the atmospheric S input at Pustert was 10 kg ha^{-1} year^{-1}. The N deposition was 30 kg ha^{-1} year^{-1}, consisting of 21 kg ha^{-1} year^{-1} NO$_3$–N and 9 kg ha^{-1} year^{-1} NH$_4$–N. Thus, the stand at Pustert receives 30% less S, but 30% more N by atmospheric deposition than the stand at Pfaffenwinkel.

In 1964, at both sites fertilization experiments had been established to study several techniques to increase stand productivity at poor sites in Northern Bavaria (Rehfuess and Schmidt 1971; Preuhsler and Rehfuess 1982). Every experiment included three randomly distributed control plots, 30 m × 40 m in size, which were separated from the fertilization variants by buffer areas. As the various fertilization variants, also the control plots have been investigated intensively during the last 40 years. The investigation included (1) a repeated assessment of the chemical status of the topsoil (forest floor + uppermost 30 cm mineral soil) in 10-year intervals, (2) an annual or bian-

nual assessment of the nutritional status of the Scot pines by foliar analysis, and (3) repeated stand inventories between 1964 and 2004.

Soil inventories

On the three control plots of each experimental site, in fall 2004 another soil inventory was carried out, with the methods being exactly the same that had been used in earlier inventories carried out in 1974, 1984, and 1994 at Pustert, and in 1982 and 1994 at Pfaffenwinkel (Prietzel et al. 1997). At each plot, an orthogonal grid net was set up, comprising 20 grid knots. At each knot, a metal frame (size 20 cm × 20 cm) was carefully pushed into the forest floor until the mineral soil was reached. The forest floor within the frame down to the uppermost mineral soil was sampled, taking utmost care to sample the forest floor completely, but to avoid any inclusion of mineral soil. Live roots and ground vegetation biomass were removed from the samples on site. Then the mineral topsoil down to 30 cm depth was sampled with a stainless soil column cylinder auger (inner diameter 5 cm). Because the content of coarse fragments in the topsoil of both sites was small (Pfaffenwinkel: 0–10%, Pustert 10–15%; Table 2), augering was

not confounded. The core was divided into 10 cm sections (0–10 cm, 10–20 cm, and 20–30 cm). For each plot, the soil material sampled at five randomly selected grid knots was pooled by layer, resulting in four pooled samples for each horizon per plot. In total, for each study site 12 samples per horizon were available for analysis. All samples were dried to constant weight at 65°C, and sieved (<2 mm mesh size). Coarse fragments and the fine earth <2 mm were weighed separately, allowing the calculation of bulk densities and element pools. From each sieved sample, a subsample was finely ground for analysis of total C and N. From the sieved samples, the pH value in 0.01 M $CaCl_2$ (soil:solution ratio 1:10 for forest floor, and 1:2.5 for mineral soil samples) was measured with a glass electrode under constant stirring after an equilibration time of 2 h. Exchangeable cations and H^+ were analyzed after 2 h batch extraction with 0.5 M NH_4Cl and subsequent filtration of the extract (0.45 μm cellulose acetate; Schleicher & Schuell Comp.). The concentration of Ca^{2+}, Mg^{2+}, K^+, Na^+, Al^{3+}, Mn^{2+}, and Fe^{2+} in the extracts was analyzed by ICP-OES (Perkin-Elmer Optima 3000). Exchangeable H^+ was calculated from the pH value measured with a glass electrode, taking into consideration H^+ produced by Al^{3+} hydrolysis. From ground samples, the concentration of total C and N was analyzed with a LECO analyzer CHN–2000. Since all samples were free of carbonate, total C is equal to organic C. All samples were analyzed in duplicate with the same methods and instruments as in the inventory 1994. The comparability of data which had been obtained with different analytical methods in earlier phases of the 40-year monitoring period (before 1994) was tested rigorously and assured by Prietzel et al. (1997); additionally, archived soil samples were reanalyzed in this study to confirm the consistency of analytical methods. Since most data were not normally distributed, the non-parametric Kruskal–Wallis H test, followed by a post-hoc Nemenyi test was used for testing the statistical significance of observed differences between soil samples taken in a given horizon at different inventories. All statistical computations were performed using the software package SPSS 12.1. for Windows.

Assessment of stand nutrition

From 1964 until 2004, the nutritional status of the stands was assessed annually (Pfaffenwinkel) or bi-annually (Pustert) by foliar analysis. Needles were sampled during the period of winter dormancy from the uppermost crown of 12 dominant trees at each of the three unfertilized plots at both experimental sites. From current-year needles, representative foliage samples were taken and oven-dried at a temperature of 65°C. After the 100-needle pair masses had been determined, the samples were finely ground and their concentration of N, P, K, Mg, and Ca analyzed. Until 1986, the N concentration was analyzed by the Kjeldahl method, later with a Heraeus Macro N analyzer (1987–1990) and with a LECO analyzer CHN–2000 (since 1991). Until 1991, the foliar concentration of P, Ca, Mg, and K was analyzed after combustion at 450°C in a muffle oven and digestion of the residue in HCl, either by colorimetry (P: molybdenum-blue-method), with a flame emission spectrophotometer (Ca, K: Eppendorf ELEX 6361) or by AAS (Mg: Perkin Elmer 380). From 1992 on, the foliage samples were subject to a 6 h pressure-digestion with concentrated HNO_3 at 160–180°C; subsequently the concentration of P, K, Mg, and Ca in the digests was determined by ICP-OES (Perkin Elmer Optima 3000). The comparability of data achieved with different methods during the 40-year investigation period was assured by repeated analyses of reference samples and in case of systematic shifts correction of the data (Prietzel et al. 1997).

Calculation of element sequestration in the pine stands

At both study sites, repeated stand inventories were conducted in 3- to 9-year intervals (Pfaffenwinkel) and in 5-year intervals (Pustert). In these inventories, the height and the diameter at breast height (1.3 m; dbh) of all trees at the studied plots were measured, and the standing volumes of the stands were calculated. Volume increments during the periods between two inventories were calculated from the observed volume changes, including the volume that had been removed from the stand in the course of

thinning operations conducted immediately after each inventory. Volume increment data were converted into annual increment data of merchantable wood (i.e., stem volume over 7 cm diameter at the smaller end). The amount of C, N, P, K, Mg, and Ca tied up per annum in the growing merchantable wood was calculated by converting the annual volume increments of merchantable wood into annual mass increments, accepting a density of 440 kg dry matter per m^{-3} for Scots pine wood (Rademacher et al. 1999), and multiplying the annual mass increment of merchantable wood with the concentration of the respective elements in merchantable wood. The concentration of organic C in wood was accepted to be 500 mg g^{-1} dry matter, the concentrations of N, P, K, Ca, and Mg were accepted to be equal to the values reported by Kreutzer (1976) for a mature Bavarian Scots pine stand (stand age 130 years), as summarized in Jacobsen et al. (2003) (N: 0.85 mg g^{-1}; P: 0.05 mg g^{-1}; K: 0.55 mg g^{-1}; Mg: 0.20 mg g^{-1}; Ca: 0.80 mg g^{-1}). The biomass change during growth of mature Scots pine stands can almost entirely be assigned to the increase in merchantable wood and coarse root biomass, whereas the masses of other tree compartments remain fairly stable (Kreutzer 1976; Rademacher et al. 1999; cited in Jacobsen et al. 2003). Therefore only changes in merchantable wood and coarse root biomass were included in our element sequestration calculation. According to Jacobsen et al. (2003), the mass of coarse roots is about 30% that of the merchantable wood. Thus, the total annual C, N, P, K, Mg, and Ca sequestration of the stands was estimated by multiplying the amount of the respective elements tied up in the annual increment of merchantable wood by the factor 1.3.

Results

### Concentrations and stocks of organic carbon and total nitrogen in the topsoil

Between 1982–1984 and 2004, the concentration of organic carbon in the forest floor of the studied soils has not changed systematically (Fig. 1a, f). The N concentration, however, increased systematically and significantly (Fig. 1b, g), resulting in a significant decrease of the forest floor C/N ratio (Fig. 1c, h). In contrast to the C/N ratio, neither the C/P ratio (Fig. 1d, i), nor the C/Ca ratio has decreased significantly in the forest floors of both sites.

The concentration of organic C in the uppermost 20 cm of the mineral topsoil increased slightly at Pfaffenwinkel (Fig. 2a), and significantly at Pustert (Fig. 2b). The same trends were observed for the stocks of organic carbon in the topsoils (forest floor + uppermost 30 cm mineral soil), which increased slightly at Pfaffenwinkel (Fig. 3a) and significantly at Pustert (Fig. 3c). The increase of the topsoil C stock was more pronounced (+404 kg C ha^{-1} year^{-1}) and statistically significant at Pustert. For Pfaffenwinkel, which has a considerably larger organic matter stock than Pustert particularly in the forest floor, with an annual increment of +210 kg C ha^{-1} year^{-1} the increase rate was only half that of Pustert and due to the larger spatial variation of the original C stock not statistically significant. At both sites, the nitrogen stocks in the forest floor of Pfaffenwinkel and Pustert and of the entire topsoil of Pustert increased significantly (Fig. 3b, d) at an average rate of +13 kg N ha^{-1} year^{-1} for the Pfaffenwinkel and +18 kg N ha^{-1} year^{-1} for the Pustert topsoil. For Pustert also a significant increase of the N stock in the mineral topsoil was observed.

Acidification status of the topsoil

At Pfaffenwinkel, the pH values were very low already in 1974 (forest floor: 2.7; mineral topsoil: 3.0–3.8), and remained fairly constant in the monitoring period between 1974 and 2004 (Fig. 4a). At Pustert, the pH in 1974 was considerably higher (forest floor: 3.3; mineral soil: 3.7 to 4.1); however, in the mineral soil, it decreased systematically and significantly by 0.3–0.4 units within the following 30 years (Fig. 4b). A systematic and significant decrease was also observed for the base saturation in the entire topsoil of Pfaffenwinkel (Fig. 5a) and in the mineral soil of Pustert (Fig. 5b). The decreases were particularly pronounced in the mineral soil. In Pustert, the base saturation of the mineral topsoil decreased from 11–12% to 5–8%; in
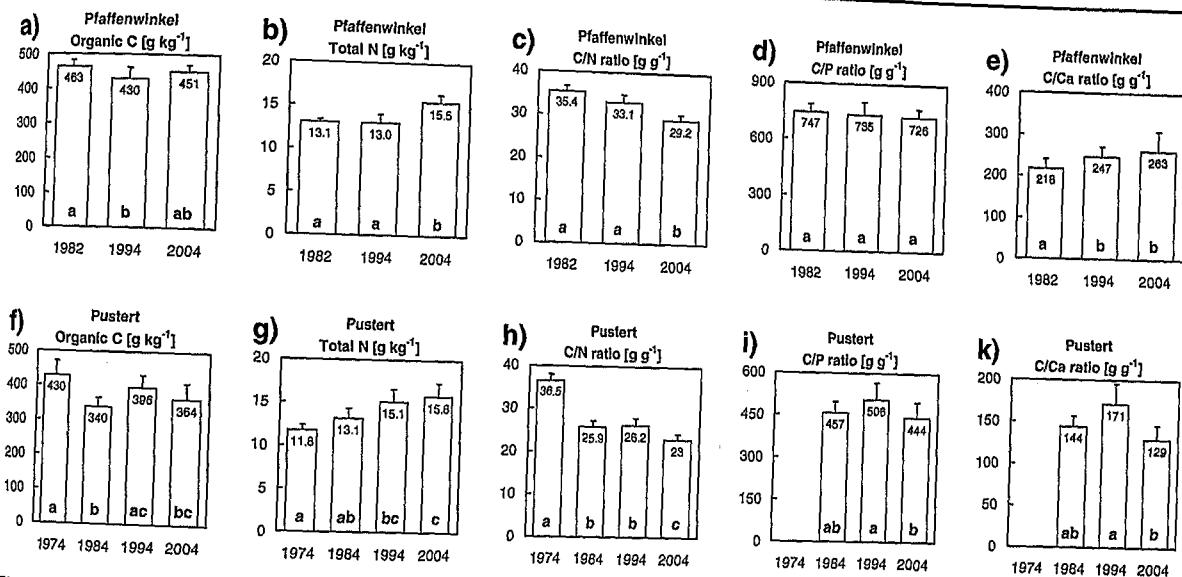
Fig. 1 Concentration of (**a, f**) organic C, and (**b, g**) total N, as well as (**c, h**) the C/N ratio, (**d, i**) the C/P ratio, and (**e, k**) the C/Ca ratio in the forest floor of the soils Pfaffenwinkel and Pustert in soil inventories conducted in 1974, 1982–1984, 1994, and 2004. Significant ($P < 0.05$) differences between different years are indicated by different letters
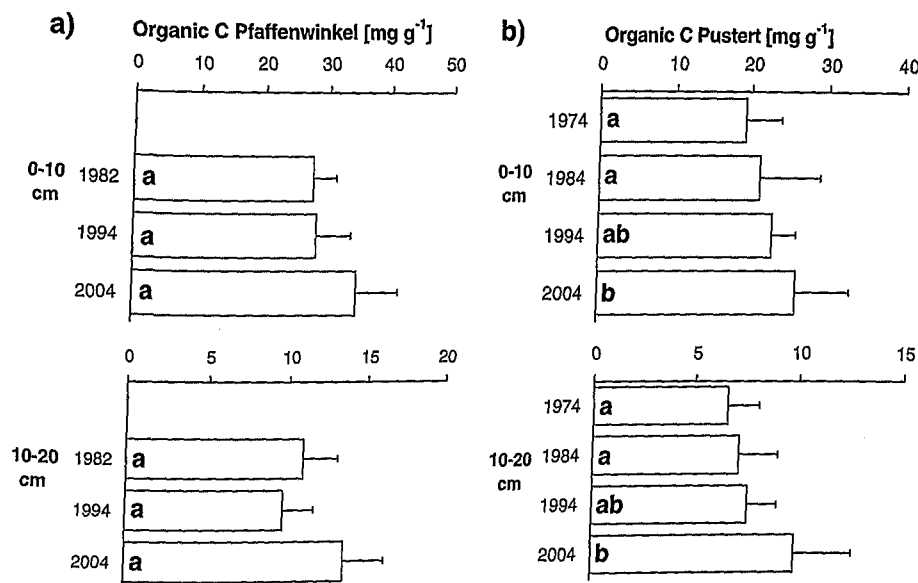

Fig. 2 Concentration of organic C in the uppermost 20 cm of the mineral soil of Pfaffenwinkel (**a**) and Pustert (**b**) in soil inventories conducted in 1974, 1982–1984, 1994, and 2004. Significant ($P < 0.05$) differences between different years are indicated by different letters

Pfaffenwinkel from 6–8% to 2–4%. Consequently, the stocks of exchangeable Mg, and K decreased significantly in the uppermost mineral soil, and also in the topsoil as a whole (Fig. 6). At both sites, also the stocks of exchangeable Ca in the mineral topsoil decreased. However, the decrease was statistically significant only for the Pustert mineral soil due to a large spatial variation of exchangeable Ca stocks in Pfaffenwinkel. Whereas the stock of exchangeable Ca in the forest floor of Pfaffenwinkel did not change systematically during the investigation period, a

Fig. 3 Organic carbon and nitrogen stocks in the forest floor and the mineral topsoil (uppermost 30 cm) of Pfaffenwinkel and Pustert in soil inventories conducted in 1974, 1982–1984, 1994, and 2004. Significant ($P < 0.05$) differences between different years are indicated by different letters
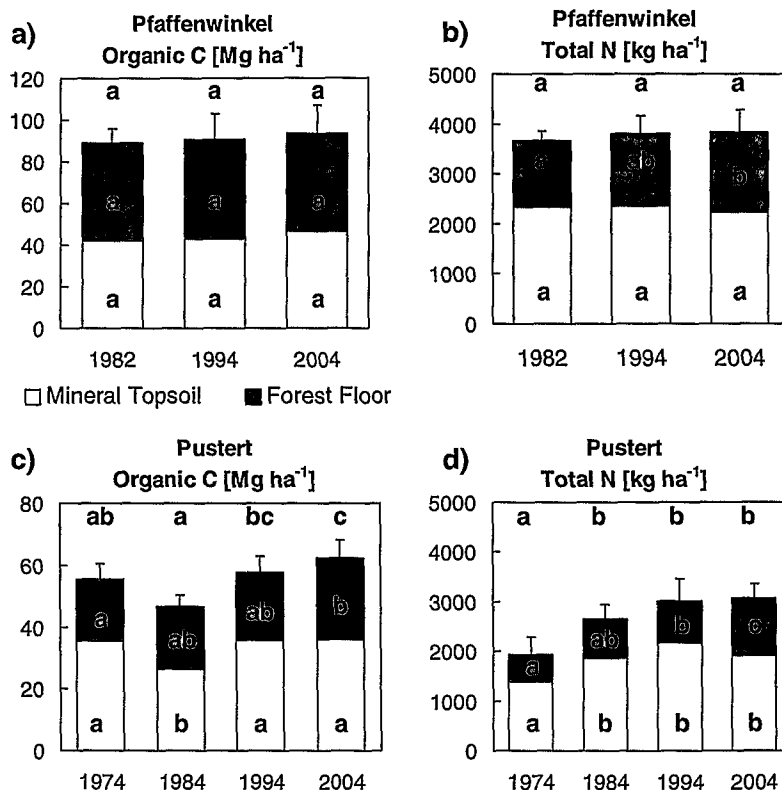
systematic and significant increase of the exchangeable Ca stock in the forest floor of Pustert was observed.

Within 22 years, the topsoil of Pfaffenwinkel (forest floor +uppermost 30 cm mineral soil) lost 38 kg Ca ha^{-1} (16% of the stock in 1982), 14 kg Mg ha^{-1} (40% of the stock in 1982), and 5 kg K ha^{-1} (6% of the stock in 1982). In the Pustert topsoil (forest floor +mineral down to a depth of 20 cm), the stock of exchangeable Mg decreased by 10 kg ha^{-1} (31% of the stock in 1974), the stock of exchangeable K by 30 kg ha^{-1} (28% of the stock in 1974). For the stock of exchangeable Ca at Pustert, the losses of the mineral topsoil were almost completely compensated for by increased Ca stocks in the forest floor, resulting in a small overall loss of 4 kg ha^{-1} (2% of the stock in 1974). In summary, the topsoils of both sites experienced significant losses of exchangeable K and Mg. For Pfaffenwinkel also the stock of exchangeable Ca has probably decreased by 15%, even though the trend was not significant. Furthermore, for both soils a redistribution of exchangeable base cations from

the mineral soil into the forest floor could be noticed.

Trends in foliar nutrient concentrations

The trend of N accumulation in the soils of both sites, but also the stronger trend of N enrichment in the Pustert soil compared to Pfaffenwinkel is mirrored by the long-term changes in foliar N concentrations of the pine trees (Fig. 7a). The pines at Pustert show a significant increase in foliar N levels throughout the entire 40-year period of monitoring. At Pfaffenwinkel, the foliar N concentration showed an increasing trend from 1965 to 1994, followed by a decreasing trend from 1992 to 2004 (Fig. 7a, Table 3). According to Mellert et al. (2004a), the decrease of the foliar N concentration in the pines of Pfaffenwinkel in the period between 1992 and 2004 was probably caused by repeated severe drought events, which were associated with an impeded N mineralization and/or N uptake by the trees. Also the reduction of the exchangeable Ca, Mg, and K stocks in the topsoils of both sites were reflected by decreasing
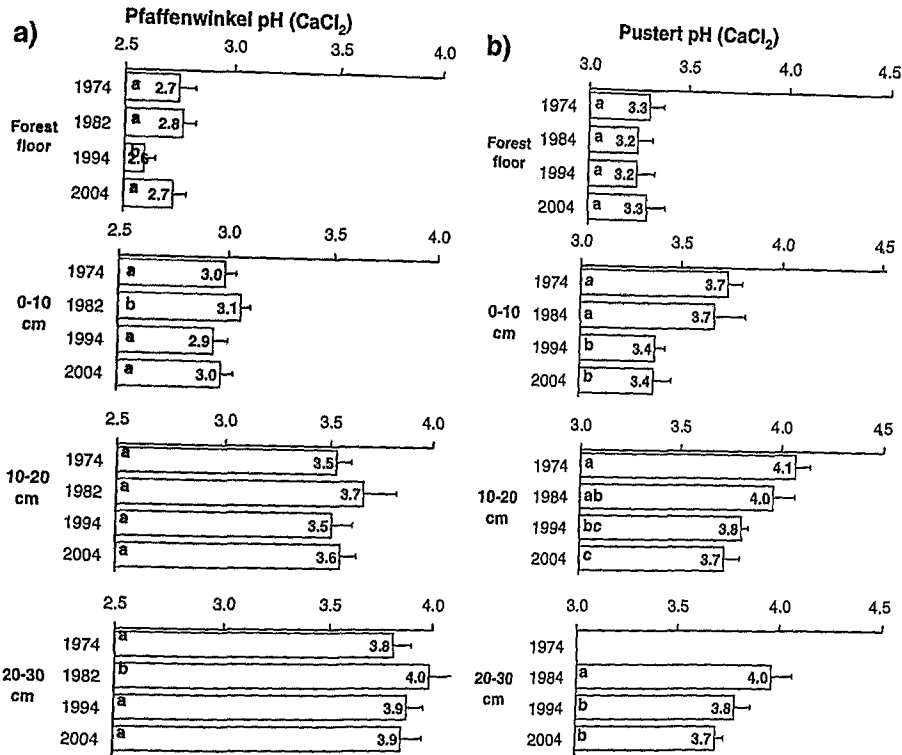
Fig. 4 pH value (0.01 M CaCl₂) in the forest floor and the mineral topsoil of **(a)** Pfaffenwinkel and **(b)** Pustert in soil inventories conducted in 1974, 1982–1984, 1994, and 2004.
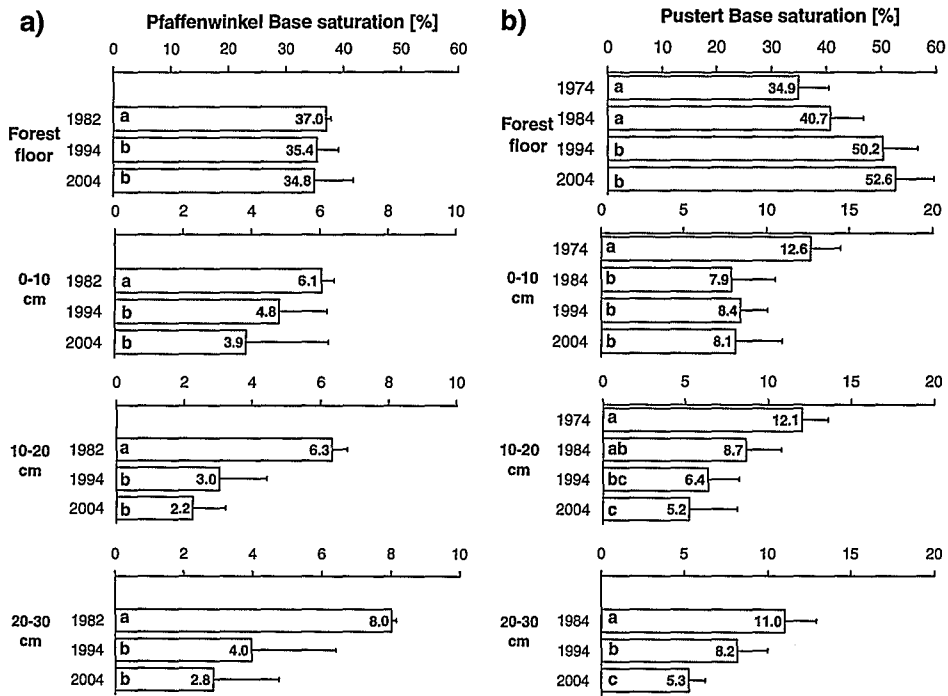
Significant ($P < 0.05$) differences between different years are indicated by different letters

time trends of the foliar concentrations of Mg (Pustert only), K, and most pronouncedly Ca of the pines at both sites (Fig. 7b–d; Table 3).

Discussion

For both soils, a significant N enrichment during the recent 22–30 years, indicated by increased N concentrations and decreased C/N ratios in the forest floor and significantly increased topsoil N stocks, could be demonstrated. The decreased forest floor C/N ratio is probably the effect of an N enrichment of the forest floor organic matter due to elevated atmospheric N deposition (Pfaffenwinkel 2004: 24 kg ha⁻¹ year⁻¹; Pustert 2004: 30 kg ha⁻¹ year⁻¹) rather than the effect of increased humus decomposition. The latter would be associated not only with a decrease of the C/N ratio, but also of the C/P and C/Ca ratios due to preferential C losses by microbial respiration. However, only the C/N ratio, but not the C/P ratio (Fig. 1d, i), nor the C/Ca ratio

has decreased significantly in the forest floors of both sites, reflecting the increased N concentration in the pine foliage as major litter source (Prietzel et al. 1997; Prietzel and Kölling 1998; Table 3). Similar results have been reported for many other forested sites in Europe (e.g. von Zezschwitz 1985; Billet et al. 1990; Buberl et al. 1994). Interestingly, another long-term study conducted in a Norway spruce stand in the Solling Mts. (northern Germany) reports considerable increases in forest floor N stocks, but no systematic decrease in the forest floor C/N ratio between 1968 and 2001 (Meiwes et al. 2002). The N enrichment of the soils at Pfaffenwinkel and Pustert has already been reported for the period between 1974 and 1994 in an earlier study (Prietzel et al. 1997); it continued in the following 10-year period from 1994 to 2004. At our study sites, this enrichment can be attributed to a general recovery of the ecosystems from former intensive N depletion by litter-raking. The recovery is promoted by elevated atmospheric N deposition, which is much higher in Pustert (2004:

**Fig. 5** Base saturation in the forest floor and the mineral topsoil of (a) Pfaffenwinkel and (b) Pustert in soil inventories conducted in 1974, 1982–1984, 1994, and 2004. Significant ($P < 0.05$) differences between different years are indicated by different letters
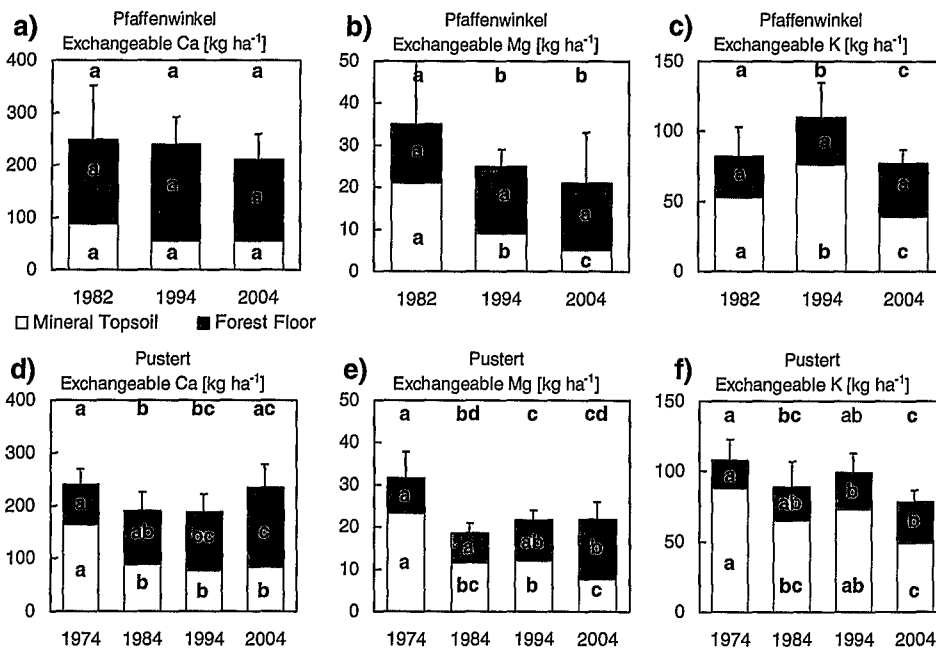
Fig. 6 Stocks of exchangeable Ca, Mg, and K in the forest floor and the mineral topsoil (uppermost 30 cm; Pustert: 20 cm) of (a) Pfaffenwinkel and (b) Pustert in inventories conducted in 1974, 1982–1984, 1994, and 2004. Significant ($P < 0.05$) differences between different years are indicated by different letters

Fig. 7 Foliar concentration of nitrogen, potassium, magnesium, and calcium in current-year needles of the pine stands Pfaffenwinkel and Pustert in the period 1964 through 2004
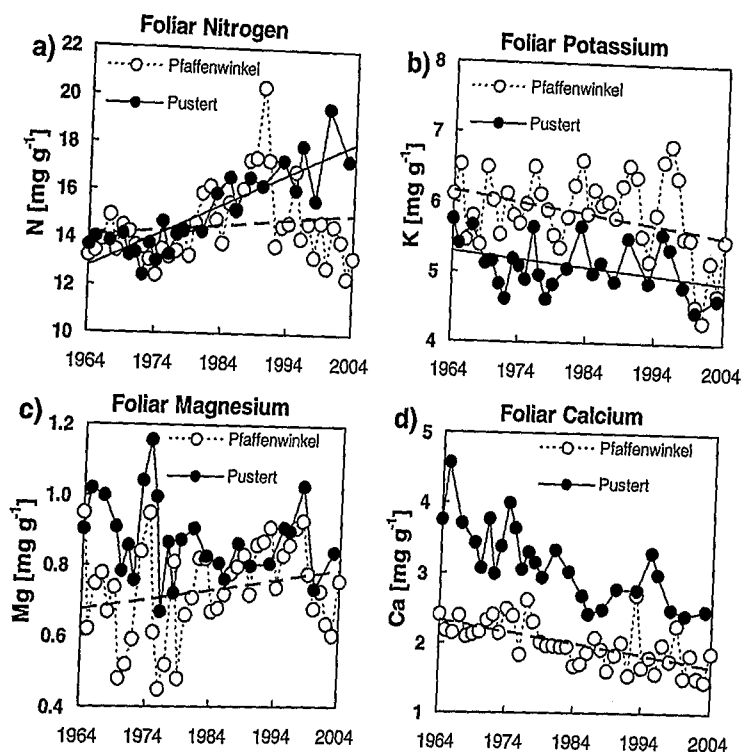
30 kg N ha^{-1} year^{-1}) than in Pfaffenwinkel (2004: 24 kg N ha^{-1} year^{-1}). Consequently, the N accumulation rate in the Pustert topsoil with 18 kg N ha^{-1} year^{-1} is larger than that in the Pfaffenwinkel soil (13 kg N ha^{-1} year^{-1}; Table 4); the same is true for the increase of foliar N levels in the pines at Pustert compared to those at Pfaffenwinkel (Table 3). The overall rate of N accumulation in the ecosystems Pfaffenwinkel and Pustert (topsoil and coarse woody biomass) is 17 kg N ha^{-1} year^{-1} for Pfaffenwinkel and 22 kg N ha^{-1} year^{-1} for Pustert (Table 4). The mean atmospheric N deposition for the period between 1984 and 2004 was calculated/estimated as arithmetic mean value of the deposition measured in 1984 (10 kg N ha^{-1} year^{-1}) and 2004 (24 kg N ha^{-1} year^{-1}). The calculated mean N deposition for the period between 1984 and 2004 is 17 kg N ha^{-1} year^{-1}, which is 70% of the N deposition measured in 2004 (24 kg N ha^{-1} year^{-1}). Accepting a similar ratio for Pustert, where deposition data are available only for 2004, the mean atmospheric N deposition at Pustert for the period between 1984 and 2004 can be estimated as

0.7 × 30 kg N ha^{-1} year^{-1} = 21 kg N ha^{-1} year^{-1}. The fact that the estimated N deposition rates at both sites match well with the calculated rates of N accumulation in the respective ecosystems indicates that at both sites the deposited N is completely retained in the ecosystem and not lost by leaching with the soil seepage water. This is also supported by results of seepage water analyses carried out at Pfaffenwinkel by Fischer (1989) over a 2-year period (1986–1987), in which the NO$_3^-$ concentration in the seepage water was zero. However, it is to be expected that particularly at Pustert continuously high levels of atmospheric N deposition will probably result in future NO$_3^-$ leaching from the topsoil and associated increased topsoil acidification, because the C/N ratio of the forest floor at that site has already decreased to a level, at which increased NO$_3^-$ export is likely to occur (Gundersen et al. 1998; MacDonald et al. 2002).

The soils at both sites also experienced an increase in their organic carbon stocks in the forest floor and in the mineral topsoil. Starting from considerably smaller stocks at the beginning of

Table 3 Linear regression equations y $(mg\ g^{-1}) = a + b \times t$ (t Julian year) describing the temporal trends of the foliar concentration of N, P, K, Mg, and Ca in current-year needles of the Scots pine stands Pfaffenwinkel and Pustert in the periods 1965–2004 and 1995–2004

	Intercept (a)	Slope (b)	Coefficient of determination R^2	Standardized beta	Significance (β)
Pfaffenwinkel					
1965–2004					
N	−30.016	0.022	0.027	0.163	0.315
P	2.555	0.000	0.001	−0.035	0.828
K	36.349	−0.015	0.098	−0.314	0.049
Mg	−6.951	0.004	0.122	0.349	0.027
Ca	33.623	−0.016	0.357	−0.597	0.000
1995–2004					
N	567.909	−0.277	0.434	−0.659	0.035
P	114.331	−0.056	0.766	−0.875	0.001
K	438.756	−0.217	0.563	−0.750	0.012
Mg	55.491	−0.027	0.548	−0.740	0.014
Ca	37.612	−0.018	0.046	−0.215	0.550
Pustert					
1965–2004					
N	−257.694	0.138	0.752	0.867	0.000
P	9.713	−0.004	0.418	−0.647	0.000
K	20.078	−0.008	0.057	−0.239	0.249
Mg	5.658	−0.002	0.053	−0.231	0.267
Ca	75.141	−0.036	0.559	−0.747	0.000
1995–2004					
N	−323.659	0.171	0.125	−0.354	0.559
P	26.485	−0.013	0.360	−0.600	0.285
K	258.616	−0.127	0.740	−0.860	0.061
Mg	30.813	−0.015	0.202	−0.449	0.448
Ca	205.455	−0.101	0.668	−0.818	0.091

Table 4 Mean annual changes of organic C, total N, Ca, Mg, and K stocks (soil: exchangeable cation pools) in the Scots pine ecosystems Pfaffenwinkel and Pustert during recent decades

	Topsoil[a]	Stand (kg ha^{-1} a^{-1})			Balance
		Shoot	Root	Total	
Pfaffenwinkel (Period 1982–2004)					
C	+210	+2,118	+635	+2,753	+2,963
N	+12.5	+3.6	+1.1	+4.7	+17.2
Ca	−1.72	+3.39	+1.02	+2.69	+4.41
Mg	−0.63	+0.85	+0.25	+1.10	+0.47
K	−0.23	+2.33	+0.70	+3.03	+2.80
Pustert (Period 1974–2004)					
C	+404	+1,666	+500	+2,166	+2,570
N	+18.3	+2.8	+0.9	+3.7	+22.0
Ca	−0.13	+2.6	+0.8	+3.4	+3.27
Mg	−0.33	+0.67	+0.2	+0.54	+0.87
K	−1.00	+1.83	+0.55	+2.38	+1.38

[a] Pfaffenwinkel: forest floor + mineral soil 0–30 cm depth; Pustert: forest floor + mineral soil 0–20 cm depth

the monitoring period, the annual accumulation rate was twice as large in Pustert (404 kg C ha^{-1} year^{-1}; equal to an annual increment of 0.6% of the soil humus pool in 1974 or 20% of the amount of C sequestered per annum in the pine stand) than at Pfaffenwinkel (210 kg C ha^{-1} year^{-1}; equal to an annual increment of 0.3% of the soil humus pool in 1982 or 10% of the C sequestered annually in the pine stand). According to Böttcher and Springob (2001), the carbon pool of soils under pine forests, which in most cases have been regenerated by clearcutting and replanting, generally increases during stand development following an s-shaped course and remains constant or even decreases again for mature pine stands older than 70 to 80 years. For the studied stands, which were 87 (Pustert) and 104 years old (Pfaffenwinkel) at the time of the first soil C inventory, a C accumulation in the soil due to stand aggradation thus is unlikely. The observed C accumulation is rather caused by several other factors: first, at both sites, the soil organic matter pools, particularly those of the forest floors, had been depleted considerably by long-term historic litter-raking, which lasted until World War II. Thus at both sites the forest floor organic matter stocks at the time of the first soil inventory (1974, 1982) were much smaller than the C stocks reflecting the "natural" steady state equilibrium of annual litter input to the soils and humus mineralization (Böttcher and Springob 2001). It can be assumed that after cessation of litter-raking this steady state equilibrium is being approached again, resulting in slowly increasing forest floor organic matter pools until that equilibrium is reached. Second, the increase in soil humus stocks is further promoted by the fact that the growth and thus probably also the net primary production of the stands at both sites has increased considerably in the recent four decades (Rehfuess and Schmidt 1971; Mellert et al. 2004a), shifting the equilibrium towards larger soil C stocks. According to Mellert et al. (2004a), the increased stand growth can mainly be attributed to an increased supply of N derived from atmospheric N deposition (cf. Townsend et al. 1996). This is similar to the effects observed in N fertilization experiments (Mäkipää 1995; Eriksson et al. 1996), where an increased growth and an increased annual litter

input to the soil had been observed. Finally, the N enrichment in the forest floor organic matter is supposed to be associated with a stabilization of the forest floor humus against mineralization (Berg and Matzner 1997; Berg 2000; Hagedorn et al. 2003), also shifting the equilibrium towards larger soil C stocks. A strong increase of forest floor organic matter stocks between 1968 and 2001 was also observed by Meiwes et al. (2002) for a 118-year-old Norway spruce and a 153-year-old European beech stand in the Solling Mts. subject to high atmospheric N and S deposition. Since the amount of litter input at the sites has not changed in the monitoring period, Meiwes et al. (2002) attributed the C accumulation to a reduced microbial humus mineralization rate, which could be caused by increased soil acidity, an increased availability of toxic heavy metals, an increased N content of the organic matter (cf. Berg and Matzner 1997; Berg 2000; Hagedorn et al. 2003), or a combination of these factors. For our sites, an increased acidity of the forest floor can be ruled out as major factor for the accumulation of humus in the forest floor. The pH in the forest floor of both sites was very low already in the first soil inventory (Pfaffenwinkel: 2.7; Pustert 3.3) and has not decreased during the 30 years of monitoring (Fig. 4); at Pustert, the base saturation of the forest floor has even increased significantly (Fig. 5b).

All potential driving factors for a humus accumulation (intensity of former C depletion by litter-raking, atmospheric N deposition) are more pronounced at Pustert than at Pfaffenwinkel, explaining the higher rate of C accumulation in the Pustert soil. The factors mentioned for our study sites (former litter raking, increased N deposition) apply for large forest regions in Central Europe. Therefore, many forest soils here—in contrast to recent results reported from Great Britain (Bellamy et al. 2005)—are likely to accumulate organic carbon rather than losing it and thus to be a relevant sink of atmospheric CO_2, except soils where recent lime application may have resulted in considerable organic matter losses (Ulrich and Keuffel 1970; Marschner and Wilczynski 1991; Kreutzer 1995). However, due to the scarcity of appropriate long-term monitoring plots at the moment only few studies with

robust results about recent changes of C stocks in European soils are available; available data (Meiwes et al. 2002) are confined to the forest floor.

For both soils a significant acidification and base cation depletion could be proven to have occurred during the recent decades: between 1982 and 2004, the topsoil (forest floor + - uppermost 30 cm mineral soil) of Pfaffenwinkel lost 1.7 kg (0.09 $kmol_c$) Ca ha^{-1} $year^{-1}$, 0.6 kg (0.05 $kmol_c$) Mg ha^{-1} $year^{-1}$, and 0.2 kg (0.01 $kmol_c$) K ha^{-1} $year^{-1}$ (Table 4). The losses are probably mainly caused by two processes: First, Pfaffenwinkel has been subject to a considerable historic input of atmospheric S, mainly originating from the highly industrialized Ohre/ Eger valley (Czech Republic). In 1986, the atmospheric S deposition at Pfaffenwinkel was 47 kg ha^{-1} $year^{-1}$ (Fischer 1989), and in 2004 still 15 kg ha^{-1} $year^{-1}$. This is the highest atmospheric S input measured in Bavarian forests in the year 2004 (cf. BayStMLF 2005). Since the S retention potential of the highly-weathered soil at Pfaffenwinkel is weak (Prietzel and Kölling 1999), and a significant portion of the deposited SO_4^{2-} is not accompanied by "base cations" but protons, the elevated S deposition has resulted in significant seepage water losses of SO_4^{2-} (1985–1987: 29 kg (1.82 $kmol_c$) SO_4–S ha^{-1} $year^{-1}$; Fischer 1989), probably accompanied by Al^{3+}, H^+, and base cations. According to our long-term N budget, the annual amount of N retained in the soil (12.5 kg N ha^{-1} $year^{-1}$) and in the pine stand (4.7 kg N ha^{-1} $year^{-1}$) matches fairly well with the mean estimated atmospheric N deposition into the ecosystem during the monitoring period (17 kg N ha^{-1} $year^{-1}$). Moreover, the study of Fischer (1989) showed that the concentration of NO_3^- in the soil seepage water of Pfaffenwinkel was zero. Obviously, at Pfaffenwinkel atmospheric N has been completely retained in the ecosystem, and no N losses by leaching with the seepage water have contributed to the observed topsoil acidification.

In addition to elevated atmospheric S deposition, also the nutrient cation uptake of the growing stand has resulted in a depletion of the soil base cation stocks at Pfaffenwinkel. Interestingly, the overall Ca, Mg, and K

balances of the studied ecosystem compartments (stand + topsoil) were positive (Table 4), indicating an accumulation of base cations. This is probably due to the fact that a considerable part of the cations sequestered in the stand is taken up from deeper soil horizons, where the base saturation is higher (Table 2) and the ion competition by Al^{3+} is lower. Unfortunately, a detailed assessment of the relative contribution of S deposition and stand uptake to the observed topsoil acidification, as e.g. carried out by Markewitz et al. (1998) cannot be made on the basis of the available data, because the respective amounts of base cations taken up by the stand from the investigated topsoil horizons and the non-investigated subsoil are unknown, and also because the atmospheric deposition of Ca and Mg has not been measured.

At Pustert, in the period 1974–2004 the topsoil (forest floor + uppermost 20 cm mineral soil) lost 0.1 kg (0.007 $kmol_c$) Ca ha^{-1} $year^{-1}$, 0.3 kg (0.03 $kmol_c$) Mg ha^{-1} $year^{-1}$, and 1.0 kg (0.03 $kmol_c$) K ha^{-1} $year^{-1}$. Thus, with exception of K, the base cation losses were smaller for the topsoil of Pustert than for the topsoil of Pfaffenwinkel. This may be partly due to the fact that atmospheric S deposition is 50% lower (2004: 10 kg S ha^{-1} $year^{-1}$), and also the annual wood production of the forest stand at Pustert and thus its net base cation uptake is 20% smaller than that of the stand at Pfaffenwinkel (Table 4). Moreover, the soil at Pustert is less deeply acidified, and its base saturation increases to 50% already 50 cm below the surface (ECEC: 119 $mmol_c$ kg^{-1}; thereof 56 $mmol_c$ kg^{-1} base cations; Table 2), whereas at Pfaffenwinkel even at a depth of 100 cm the base saturation is only 32% (ECEC: 10 $mmol_c$ kg^{-1}; thereof 3 $mmol_c$ kg^{-1} base cations). Therefore, a considerable portion of the base cation demand of the stand at Pustert is assumed to be taken up in deeper soil horizons, and a significant part of these base cations is not tied up in bolewood or in coarse roots, but in foliage which is deposited on the soil surface by litterfall ("base pump" function of the stand). The significant relevance of this mechanism at Pustert is indicated by the larger foliar Ca concentrations of the pine foliage (Fig. 7), as well as by the considerably

higher pH value (Fig. 4) and base saturation of the forest floor (Fig. 5) compared to Pfaffen-winkel. The question whether the high atmospheric N deposition at Pustert at the moment results in significant NO_3^- leaching with the soil seepage water, cannot be answered thoroughly. Our N balance (Table 4) suggests that at least most, if not all deposited N (on average 22 kg N ha^{-1} $year^{-1}$ for the period between 1984 and 2004) is still retained in the soil or in the growing stand. Moreover any NO_3^- leached from the topsoil during the winter and spring period of water-logging would probably be transferred into volatile N compounds by denitrification. However, the strong decrease of the forest floor C/N ratio during recent decades to a value of 23.0 in 2004 indicates an increased probability of NO_3^- leaching with the topsoil seepage water (Gundersen et al. 1998; MacDonald et al. 2002), particularly during seasons without water-logged conditions.

The considerable, statistically significant top-soil acidification and base cation depletion documented in this study for Pfaffenwinkel and Pustert is in line with similar results from other studies conducted in Europe and America (e.g. Hallbäcken and Tamm 1986; Federer et al. 1989; Hallbäcken 1992; Johnson et al. 1994; Wesselink et al. 1995; Likens et al. 1996; Jönsson et al. 2003; Bailey et al. 2005). The same is also true for the increased N nutritional status and the decreased supply of base cations in the investigated pine stands (Mellert et al. 2004b). In the long term, the observed trends of decreasing pH, base saturation, and stocks of Ca, Mg, and K and of increasing stocks of nitrogen in the upper horizons of forest soils may result in increased concentrations of Al^{3+} and NO_3^- in the soil seepage water, constituting an environmental risk for stand stability as well as for the quality of forest-derived drinking water and the integrity of freshwater ecosystems. Soil acidification can be mitigated by application of lime or wood ash (Lundström et al. 2003). In large forested areas in Central Europe, dolomitic limestone is applied on the soil surface in regular intervals (6–15 years) to reduce soil acidity and to re-increase base cation stocks in the rooted topsoil. However, particularly at sites with sandy or skeletal soils, where a great

portion of the total soil organic matter stock is located in the forest floor, the application of lime or wood ash is associated with the risk of considerable humus losses caused by accelerated microbial activity (Ulrich and Keuffel 1970; Marschner and Wilczynski 1991; Kreutzer 1995; Persson et al. 1995); yet this may not be the case when small doses of lime or wood ash are applied on N-limited soils (Lundström et al. 2003; Jacobson et al. 2004). This humus loss results in an undesired reduction of the water storage capacity of the soil and an undesired release of CO_2 into the atmosphere. Moreover, for N-saturated acidic soils, the percentage of which is increasing in Central Europe, forest liming often is associated with an increased NO_3^- export into the groundwater (Marschner and Wilczynski 1991; Kreutzer 1995). Another, slow but gentle way to restore anthropogenically acidified soils under coniferous forests is the introduction of broadleaf trees (e.g. beech, oak, maple, lime, or hornbeam) which effectively take up base cation nutrients from the subsoil, and depose a large part of these cations on the soil surface via litterfall ("base pump" effect), resulting in an increased pH and base saturation of the topsoil (Heitz 1998; Augusto et al. 2002; Prietzel 2004). The introduction of broadleaved trees into coniferous forests is particularly effective on sites with significant available base cation pools in the subsoil. It is associated with only modest or no humus losses and with an ecologically desired transfer of soil organic matter from the forest floor into the mineral soil (Heitz 1998; Prietzel 2004). Additionally, the NO_3^- concentration in the soil seepage water is decreased (Rothe et al. 2002; Prietzel 2005) due to reduced interception of atmospheric N by the stand and/or increased sequestration of N in woody biomass.

Conclusions

During the recent 22–30 years, the topsoil of two intensively studied, historically degraded forest sites in southern Germany with mature Scots pine stands on poor soils showed a statistically significant decrease of the forest floor C/N ratio, and a considerable, significant accumulation of organic

carbon (210 and 400 kg ha^{-1} year^{-1}) and nitrogen (13 and 18 kg ha^{-1} year^{-1}). In addition, the mineral topsoil at both sites has acidified significantly, indicated by decreased pH values, base saturation, and base cation stocks. The trend of N enrichment and base cation losses in the soils is mirrored by the temporal trend of stand nutrition at the sites, which is characterized by an improved N nutrition and a reduced supply with K, and Ca (Mg). The results confirm results of other studies, indicating a recent N eutrophication and acidification of European forest soils. Since soils with historic degradation by non-sustainable forest utilization practices subject to elevated N deposition are widespread in Central Europe, the results obtained from the study sites apply for large forested areas, suggesting a significant potential of the Central European forests to sequester atmospheric carbon and nitrogen not only in stand biomass but also in the soil.

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