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Changes of soil chemistry, stand nutrition, and stand growth at two Scots pine (*Pinus sylvestris* L.) sites in Central Europe during 40 years after fertilization, liming, and lupine introduction

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Abstract Long-term (40 years) effects of two soil amelioration techniques [NPKMgCa fertilization + liming; combination of PKMgCa fertilization, liming, tillage, and introduction of lupine (Lupinus polyphyllus L.)] on chemical topsoil properties, stand nutrition, and stand growth at two sites in Germany (Pfaffenwinkel, Pustert) with mature Scots pine (Pinus sylvestris L.) forest were investigated. Both sites are characterized by base-poor parent material, historic N and P depletion by intense litter-raking, and recent high atmospheric N input. Such sites contribute significantly to the forested area in Central Europe. Amelioration resulted in a long-term increase of pH, base saturation, and exchangeable Ca and Mg stocks in the topsoil. Moreover, significant losses of the forest floor in organic carbon (OC) and nitrogen stocks, and a decrease of the C/N ratio in the topsoil were noticed. The concentrations and stocks of OC and N in the mineral topsoil increased; however, the increases compensated only the N, but not the OC losses of the forest floor. During the recent 40 years, the N nutrition of the stands at the control plots improved considerably, whereas the foliar P, K, and Ca concentrations decreased.

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H. Pretzsch Lehrstuhl für Waldwachstumskunde, Technische Universität München, 85350 Freising, Germany The 100-fascicle weights and foliar concentrations of N, P, Mg, and Ca were increased after both amelioration procedures throughout the entire 40-year period of investigation. For both stands, considerable growth acceleration during the recent 40 years was noticed on the control plots; the amelioration resulted in an additional significant long-term growth enhancement, with the NPKMgCa fertilization liming + being more effective than the combination of PKMgCa fertilization, liming, tillage, and introduction of lupine. The comprehensive evaluation of soil, foliage, and growth data revealed a key relevance of the N and P nutrition of the stands for their growth, and a change from initial N limitation to a limitation of other growth factors (P, Mg, Ca, and water).

Keywords Amelioration · Liming · Base cations · Carbon budget · Long-term study · Nutrition trend · Pinus sylvestris · Soil organic matter · Carbon budget

Introduction

In large areas of the northern hemisphere, forest growth was hitherto often limited by the supply of the trees with nitrogen (e.g., Aber et al. 1989; Tamm 1991), and stand growth thus can be increased significantly by N fertilization (e.g., Kenk and Fischer 1988; Binkley and Högberg 1997). Consequently, managed forests in these regions, particularly in Northern Scandinavia, Canada, and the Pacific Northwest of the USA are routinely fertilized with N, often combined with the application of other nutrients of short supply, primarily P, K, S, Mg, or B (e.g., Persson et al. 1995a; Brockley 2000; Nohrstedt et al. 2000). Numerous fertilization experiments have been carried out in these countries to find out optimal fertilization regimes, and the issue remains a hot topic until today (e.g., Tamm et al. 1995;

Nohrstedt et al. 2000; Nilsen and Abrahamsen 2003; McNulty et al. 2005; Blevins et al. 2006), including new aspects such as the influence of forest fertilization on the large scale N balance (Akselsson et al. 2007), losses of greenhouse gases (e.g., Maljanen et al. 2006), and the sequestration of atmospheric carbon in tree biomass (Iivonen et al. 2006) and in forest soils (Akselsson et al. 2007; Jandl et al. 2002, 2007).

In Central Europe and southern Scandinavia, in contrast, many forests since several decades have received elevated atmospheric N deposition, sometimes exceeding the N demand of the stands. This has resulted in accelerated N accumulation in many Central European and southern Scandinavian forests as indicated by decreased forest floor C/N ratios and/or increased forest floor N contents (Billett et al. 1990; Hildebrand 1994; Prietzel et al. 1997, 2006; Meiwes et al. 2002). Moreover, changes in the composition of the ground vegetation have been observed in many Central European forests, with the abundance of N-demanding species increasing at the expense of species adapted to low N availability (e.g., Hofmann et al. 1990). Furthermore, the growth of Scots pine (Pinus sylvestris L.) stands in Central Europe and southern Scandinavia has increased considerably (Pretzsch 1985; Elfving and Tegnhammar 1996; Spiecker 1999), mostly due to an improved N supply of the stands (Mellert et al. 2004a; Prietzel et al. 2006). Therefore, N fertilization is no longer carried out in managed forests of Central Europe; instead they are routinely treated with granulated dolomitic lime to counteract soil acidification caused by high atmospheric N and S deposition (Hildebrand 1994; Kreutzer 1995; Lundström et al. 2003; Huber et al. 2006).

However, the situation in Central Europe was entirely different 50 years ago. In the 1950s and 1960s, many forests on sites which had been severely degraded by excessive wood harvest, forest pasture, charcoal and potash production, and litter raking, consisted of Scots pine stands with poor growth. Accepting that the poor stand growth had been caused by the historic human depletion of the sites particularly of N, but also of P, K, Mg, and Ca (Ebermayer 1876; Kreutzer 1972; Glatzel 1991), numerous fertilization experiments were established in the 1960s to find out efficient techniques for improving the growth and nutritional status of the degraded stands. Since the most critical nutrient at that time generally was N, in most experiments different approaches of N amendment were tested, again often combined with the application of other nutrients of short supply, including fertilization with gaseous ammonia, urea, ammonium sulfate, ammonium nitrate, or nitrochalk (e.g., Zöttl and Kennel 1962; Heinsdorf 1966; Tamm et al. 1974; Preuhsler and Rehfuess 1982). Another method tested was the introduction of N-fixing plants, mainly lupine (Lupinus polyphyllus L.; Nemeč 1942; Rehfuess et al. 1991). The evaluation of these experiments showed that short-term stand growth of Scots pine could be enhanced mainly by N fertilization, but also by lupine introduction (e.g., Zöttl and Kennel 1962; Rehfuess and Schmidt 1971; Preuhsler and Rehfuess 1982). Assessments of the foliar nutritional status of the stands showed an improved nutrition of pine particularly with the formerly limiting N. The chemical status of the topsoil also was improved significantly, as indicated by a decrease of the forest floor C/N ratio and an accumulation of N, P, and exchangeable K, Mg, and Ca. However, for most experiments only short-term reactions have been reported, covering at best the first 10 or 20 years after the amelioration treatments. Reports describing longer-term reactions (30 or 40 years after amelioration) are rare. In this paper, the longterm effects of two amelioration procedures, (1) superficial application of NPKMgCa fertilizer + lime, and (2) combination of PKMgCa application, liming, tillage, and lupine sowing, on topsoil chemistry, stand nutrition, and stand growth are compared for two mature Scots pine stands in southern Germany. The sites had been historically degraded and seriously depleted in N, P, and base cations. In recent decades they have received considerable N and S input by atmospheric deposition (Prietzel et al. 1997, 2006). Earlier papers (Rehfuess and Schmidt 1971; Preuhsler and Rehfuess 1982; Makeschin et al. 1985; Rehfuess et al. 1991) have reported short-term (<20 years) fertilization effects. The main questions to be addressed in this study are:

- Which long-term (40 years) amelioration effects can be observed regarding topsoil chemistry, stand nutrition, and stand growth?
- Is NPKMgCa fertilization or combination of PKMgCa fertilization, tillage, and lupine sowing in the long run more effective in improving soil chemistry, stand nutrition, and stand growth?
- How has the fertilization affected long-term C and N storage in different compartments (topsoil and stand biomass) of the two Scots pine forest ecosystems?

Material and methods

Study sites

The investigation was conducted in the Scots pine stands Pfaffenwinkel and Pustert in Northern Bavaria, Germany, which recently have been described in detail by Prietzel et al. (2006). Important properties of the sites, stands, and soils are presented in Tables 1 and 2. In 1964, the sites were densely stocked with 86-year-old (Pfaffenwinkel) and 77year-old (Pustert) Scots pine of poor quality with a site index (Wiedemann 1943) of IV.6 at Pfaffenwinkel and III.8

| | Pfaffenwinkel | Pustert |
|-----------------------------|--|---|
| Elevation, slope, aspect | 528–542 m a.s.l., 5% NW | 477–482 m a.s.l., 3% NW |
| Parent material | Solifluidal silt loam cover over deeply weathered phyllite residues (saprolith) | Solifluidal silt loam over clayey weathering residues of cretaceous sediments |
| Mean annual air temperature | 5.8°C | 7.2°C |
| Mean annual precipitation | 615 mm | 650 mm |
| Water regime | fairly dry | seasonal change from wet to dry |
| Soil type | Dystric Cambisol (formerly litter-raked) | Stagnic Albeluvisol (formerly litter-raked) |
| Stand | 100% Pinus sylvestris | 98% Pinus sylvestris, 2% Picea abies |

Table 2 Important propertiesof the soils at the sites Pfaffen-winkel 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) |
|-----------|---------------|-------------------------|-----------|------------------------------------|----------------------------------|---|-----------|----------------------------|
| Pfaffenwi | nkel | | | | | | | |
| 0 | 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 | | | | | | | | |
| 0 | 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 |
| Е | 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 NH₄Cl), *BS* base saturation, *NA* not analyzed

at Pustert. At Pfaffenwinkel the atmospheric S deposition (bulk deposition) in the year 1986 was 47 kg ha⁻¹ year⁻¹, and the N bulk deposition was 10 kg ha⁻¹ year⁻¹. In 2004, the atmospheric S input was only 15 kg ha⁻¹ year⁻¹, but the atmospheric N deposition had increased to 24 kg ha⁻¹ year⁻¹ (Prietzel et al. 2006). At Pustert, in 2004 the corresponding values were 10 kg S ha⁻¹ year⁻¹ and 30 kg N ha⁻¹ year⁻¹. Thus, the stand at Pustert receives 50% less S, but 30% more N by atmospheric deposition than that at Pfaffenwinkel. The decrease in atmospheric S and the increase in N deposition at Pfaffenwinkel and probably also at Pustert during the last 20 years reflect the general deposition trends in Central Europe (Alewell 2001; Wright et al. 2001).

In 1964, at both sites amelioration experiments were established to compare several techniques to increase stand productivity. The experimental setup is described in detail by Rehfuess and Schmidt (1971), Preuhsler and Rehfuess (1982), and Prietzel et al. (1997, 2006). All experimental variants, represented by three replicates per site, have been studied 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), (2) annual or biannual assessment of the nutritional status of the Scots pines by foliage analysis, and (3) forest inventories conducted in 3- to 7-year intervals. This paper presents results about the variants CON (control), FER (NPK-MgCa fertilization, including an application of 4,000 kg CaCO₃ ha⁻¹), and LUP (combination of PKMgCa fertilization—again including the application of lime, tillage, sowing of lupine) (Table 3).

Table 3 Amelioration techniques in the experimental treatments FER (superficial application of NPKMgCa fertilizer and lime, and LUP (combination of PKMgCa fertilization, liming, tillage, and lupine sowing)

| Year | Variant | Type of amelioration | | |
|---|------------------------------|---|--|--|
| 1964 | FER, LUP | Superficial application of 4,000 kg ha ⁻¹ CaCO ₃ , 1,000 kg ha ⁻¹ Thomas phosphate $(Ca_3(PO_4)_2[SiO_4]:$ 15% P), and 400 kg ha ⁻¹ K ₂ /MgSO ₄ | | |
| | LUP | Tillage (15–20 cm deep); sowing of lupine (<i>Lupinus polyphyllus</i>) seeds (20 kg ha ⁻¹) | | |
| 1964, 1966, 1972 | FER | Superficial application of 500 kg ha ⁻¹ nitrochalk (70% NH ₄ NO ₃ ; 30% CaCO ₃) | | |
| 1967 | FER, LUP | 300 kg ha ⁻¹ superphosphate [Ca(H ₂ PO ₄) ₂ + CaSO ₄ : 18% P] (Pfaffenwinkel only) | | |
| Total applied amount of | nutrients | | | |
| Pfaffenwinkel | | Pustert | | |
| N: 370 kg ha ⁻¹ (FER) | | N: 370 kg ha^{-1} (FER) | | |
| P: 95 kg ha ⁻¹ (FER, LU | IP) | P: 70 kg ha^{-1} (FER, LUP) | | |
| K: 90 kg ha ⁻¹ (FER, LU | JP) | K: 90 kg ha^{-1} (FER, LUP) | | |
| Ca: 2,095 kg ha ⁻¹ (FER (LUP) | 2)/1,915 kg ha ⁻¹ | Ca: 2,035 kg ha ⁻¹ (FER)/1,855 kg ha ⁻¹ (LUP) | | |
| Mg: 32 kg ha ⁻¹ (FER, I | LUP) | Mg: 32 kg ha ⁻¹ (FER, LUP) | | |

Assessment of topsoil chemistry and stand nutrition

On all control and ameliorated plots at both experimental sites, soil inventories were carried out in fall 2004 with the method described in detail in Prietzel et al. (1997, 2006). The forest floor and the mineral soil down to 30 cm depth were sampled at 20 places per plot; the latter in 10 cm sections (0-10, 10-20, and 20-30 cm). For each study site and experimental variant, 12 composite samples per depth increment originating from three replicate plots were available. All samples were dried to constant mass at 65°C, and sieved (<2 mm). For each sample, the pH-value in 0.01 M CaCl₂, the concentration of organic C (OC), total N, and exchangeable cations were analyzed as described in Prietzel et al. (1997, 2006). Additionally, from 1964 until 2004 the nutritional status of the stands at the different plots 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 plot. From current-year needles, representative foliage samples were taken and analyzed as described in detail in Prietzel et al. (1997, 2006) after the 100-fascicle masses had been determined.

Assessment of stand growth, calculation of biomass, and calculation of element sequestration in the pine stands

At both sites, repeated forest inventories were conducted in 3- to 7-year intervals (Pfaffenwinkel) and 5-year intervals (Pustert). At Pustert, in 1998, and 2003 additionally the trees, which had developed as natural regeneration during the experiment and had grown to a diameter at breast height (1.3 m; dbh) <6.5 cm were recorded.

In the inventories, the height and the diameter at breast height of all trees at the various plots was measured. For all experimental variants the annual increment of merchantable wood (stem volume over 7 cm minimum diameter at the smaller end) was calculated by standard procedures published in Pretzsch (2002). The amount of C, N, P, K, Mg, and Ca accumulated per annum in the growing merchantable wood was calculated as described in Prietzel et al. (2006), accepting a wood density of 440, 430, 660, and 600 kg m⁻³, for the wood of pine, spruce, oak, and other broadleaf tree species, respectively (Rademacher et al. 1999; Jandl et al. 2007). 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 calculations. 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 elements tied up in the annual increment of merchantable wood by the factor 1.3.

Statistics

The differences between the cumulative volume increments of merchantable wood and the mean foliar nutrient concentrations in the stands of the studied experimental variants as calculated for the 3- to 7-year periods between the growth measurements, which proved to be normally distributed according to Kolmogorov–Smirnov tests, were tested for statistical significance by one-factorial analysis of variance (ANOVA) followed by a post-hoc least significant difference (LSD) test. Because the soil chemical data were not normally distributed, the Kruskal–Wallis H test, followed by a post-hoc Nemenyi test was used for testing the significance of differences between soil chemical features of a given horizon for the various treatments.

Relationships between the mean nutrient concentration of current-year foliage of dominant pines and the mean annual volume increment of the respective stands for different 3- to 7-year periods between stand inventories were tested by non-parametric correlation (Spearman's ρ), and with simple and multiple linear regression models with stepwise inclusion of significant (P < 0.05) independent variables. According to Durbin–Watson tests, autocorrelation did not constitute a significant problem in our time series. In 72% of all cases, (first-order) autocorrelation could be strictly rejected on a 0.05 level. Not in a single case autocorrelation was identified; 28% of all cases were inconclusive. All statistics was performed using SPSS 12.1 for Windows.

Results

Acidification status of the topsoil

In 2004, 40 years after application of 4 t ha^{-1} CaCO₃ as part of the fertilization treatment (and 32 years after the last additional application of 150 kg ha^{-1} CaCO₃ at the plots

Fig. 1 pH value (0.01 M CaCl₂) in the forest floor and the mineral topsoil for different variants of the experiments **a** Pfaffenwinkel and **b** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*

with NPKMgCa fertilization—Table 3), the pH value in the topsoil of both study sites was still significantly higher at the ameliorated compared to the respective control plots (Fig. 1). In the entire topsoil of both study sites except the forest floor at Pustert, the base saturation also was significantly larger at the ameliorated plots compared to the control plots (Fig. 2). No significant differences between the plots with NPKMgCa fertilization + lime application (FER) and the plots with tillage, PKMgCa fertilization, liming, and lupine introduction (LUP) could be noticed. The stocks of exchangeable Ca in the topsoil were significantly increased (Fig. 3) at both sites and for both amelioration techniques. At Pfaffenwinkel, the topsoil of the ameliorated plots in 2004 contained about 800 kg ha⁻¹ more exchangeable Ca than the topsoil of the control plots (Fig. 3a), which is about 40% of the Ca amount applied in the 1960s. At Pustert, the difference is 60% of the applied Ca amount for the LUP plots, and 70% for the FER plots (Fig. 3b). With $15-30 \text{ kg ha}^{-1}$, the amount of fertilizer-Ca sequestered by the additional growth of merchantable wood on FER and LUP plots is negligible at both study sites compared to the amount of fertilizer-Ca retained in the soil. The stocks of exchangeable Mg in the topsoil were increased significantly only for the LUP variants at Pfaffenwinkel and Pustert. Due to a large spatial variation on the FER plots, the increases of the exchangeable Mg stocks for the latter variant were not statistically significant, but nevertheless considerable. Forty years after Mg application, the difference between the exchangeable Mg stocks in the topsoil of the ameliorated and the control plots at Pfaffenwinkel (Fig. 3c) corresponds to 38 (FER) and 44% (LUP) of the applied Mg amount. At





Fig. 2 Base saturation in the forest floor and the mineral topsoil for different variants of the experiments **a** Pfaffenwinkel and **b** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*



Fig. 3 Stocks of exchangeable Ca and Mg in the forest floor and the mineral topsoil (uppermost 30 cm) for different variants of the experiments **a**, **c** Pfaffenwinkel and **b**, **d** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*

Pustert, the respective differences (Fig. 3d) even correspond to 197 (FER) and 337% (LUP) of the applied Mg. In contrast to Ca, the amount of fertilizer-Mg sequestered in the additional merchantable wood of the ameliorated stands considerably contributes to the retention of fertilizer-Mg in the system. For both amelioration variants at both sites, the amount of fertilizer-Mg sequestered in this compartment ranged between 4 and 7 kg ha⁻¹ (13–22% of the applied Mg amount).

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

At both sites, in 2004 the concentration of OC in the forest floor was significantly decreased on the ameliorated compared to the control plots (Fig. 4a, b); in the uppermost mineral topsoil, however, an increase could be noticed (Fig. 4c, d). At Pustert, but not at Pfaffenwinkel, the humus form has changed from mor at CON to moder and mull at the ameliorated plots. The NPKMgCa fertilization + lime application and particularly the combination of PKMgCa fertilization, liming, tillage, and introduction of lupine resulted in a significant decrease of the C/N ratio in the forest floor and in the mineral topsoil (Fig. 5). Both amelioration treatments resulted in considerable losses of OC in the topsoil (forest floor + uppermost 30 cm mineral soil; Fig. 6a, b), corresponding to 12-16% of the topsoil OC stocks at the control plots. The topsoil OC losses were caused by significant decreases of the forest floor OC amounts, whereas the mineral topsoil at both sites generally

Fig. 4 Concentration of organic C in the forest floor and the mineral topsoil (0–10 cm depth) for different variants of the experiments **a**, **c** Pfaffenwinkel and **b**, **d** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*

Fig. 5 C/N ratio in the forest floor and the mineral topsoil (0– 10 cm depth) for different variants of the experiments **a**, **c** Pfaffenwinkel and **b**, **d** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*



gained OC. However, these increases in the mineral topsoil could not compensate for the OC losses of the O layer.

At both sites, the N stocks in the topsoil of the ameliorated plots were slightly—but due to the large spatial variation not significantly—increased relative to the control plots (Fig. 6c, d), with the increase generally being larger for the plots with NPKMgCa fertilization than for the plots where lupines had been introduced. As with OC, the forest floor of the ameliorated plots also lost a considerable amount of N, whereas the mineral topsoil gained N. At Pustert the N losses of the forest floor and the N gains of the mineral topsoil were significant for both amelioration variants, whereas at Pfaffenwinkel only LUP showed significant changes. Thirty-two years after the last nitrochalk amendment in 1972, the topsoil of the FER plots in Pfaffenwinkel contained 234 kg ha⁻¹ more N than the topsoil of the control plots, corresponding to 63% of the applied N amount. At Pustert this difference between FER and CON



Fig. 6 Organic carbon and nitrogen stocks in the forest floor and the mineral topsoil (uppermost 30 cm) for different variants of the experiments **a**, **c** Pfaffenwinkel and **b**, **d** Pustert in 2004. Significant (P < 0.05) differences between treatments are indicated by different *letters*

was 291 kg ha⁻¹ (78% of the applied N amount). The respective differences between the topsoil N stocks of the plots with lupine introduction and CON were 147 kg N ha⁻¹ for Pfaffenwinkel and 32 kg N ha⁻¹ for Pustert.

Foliar nutrient concentration of the mature pine stand

At the beginning of the experiments, the Scots pine stands at Pfaffenwinkel (Fig. 7) and Pustert (Fig. 8) were characterized by an insufficient supply with N and P. The pine trees on the control plots of *Pfaffenwinkel* showed an increasing foliar N concentration trend from 1965, when this value according to Arbeitskreis Standortskartierung (2003) indicated N deficiency, up to a status of luxurious N nutrition in 1991. From 1992 to 2004, however, a decreasing trend of foliar N concentrations was noticed (Fig. 7a, Table 4). The foliar concentrations of P (Fig. 7b) and K (Fig. 7c) at CON exhibited a decreasing trend, which was significant for K. Whereas the foliar K concentrations indicated a good K supply of the pines throughout the entire monitoring period, the foliar P concentration decreased to values characteristic for a low P supply according to Arbeitskreis Standortskartierung (2003). The foliar Ca concentrations of the pines (Fig. 7e) at the control plots of Pfaffenwinkel decreased significantly to concentrations indicating Ca deficiency ($< 2 \text{ mg g}^{-1} \text{ dw}$) at the end of the monitoring period. The pine stands on the ameliorated plots showed a sustainable and statistically significant improvement in Ca nutrition relative to those on the control plots (Fig. 7e, Table 4). Also the mean foliar concentrations of N and P (Table 4) as well as the 100-fascicle masses (Table 6) were increased relative to CON throughout the entire monitoring period. The positive effect of amelioration was particularly strong at the beginning (1965–1980) and at the end of the 40-year monitoring period (1993–2004), when the pines at CON showed the smallest foliar N and P concentrations and 100-fascicle masses (Fig. 7).

In contrast to Pfaffenwinkel, the pines at the control plots of Pustert showed a systematic increase of the foliar N concentrations throughout the entire 40-year period of monitoring (Fig. 8a; Table 5) from an initial status of insufficient N supply to a status of optimal to luxurious N nutrition. Concomitantly the foliar concentrations of P, K, and Ca declined (Fig. 8b, c, e). As with Pfaffenwinkel, also at Pustert the amelioration resulted in a significant increase of the foliar N and P concentrations of the pines relative to those on CON in the first two decades of the monitoring period (Table 5), and also in a significant increase of the 100-fascicle mass during the first decade of the experiment (Table 6). The foliar N and P concentrations as well as the 100-fascicle masses remained-statistically not significant-elevated in the Scots pines on the ameliorated plots throughout the entire 40 years of experimentation (100-fascicle mass: 30 years). Additionally, the foliar concentrations of Ca and Mg were increased for the pines after amelioration in the second half of the monitoring periodhowever, the differences were not significant.

Stand growth

Important growth data of the pine stands at the different treatments of the experiments at Pfaffenwinkel and Pustert are presented in Table 7. The annual volume increment of merchantable wood in the stands on the control plots at both sites (Fig. 9a, c) showed an increasing trend from 1965 until 1992, followed by slightly reduced values. The temporal growth pattern of the pines at the control plots has been described and discussed in detail recently by Mellert et al. (2004b). At both sites, the volume increment response of the stands to the amelioration can be distinguished into four phases (Fig. 9a, c). In phase I (Pfaffenwinkel: 1964-1969; Pustert: 1965–1973), the increment was particularly enhanced by NPKMgCa fertilization + liming. In phase II, (Pfaffenwinkel: 1970–1978; Pustert: 1974–1983), the pines at both amelioration treatments grew much better than those at the control plots. In phase III (Pfaffenwinkel: 1979–1992; Pustert: 1984–1988), the stand growth at CON accelerated considerably, with the annual volume increment of merchantable wood approaching that of the ameliorated



Fig. 7 Foliar concentrations of N, P, K, Mg, and Ca as well as 100-fascicle mass of current-year needles of the pine stands for different variants of the experiment Pfaffenwinkel in the period 1964 through 2004

Fig. 8 Foliar concentrations of N, P, K, Mg, and Ca as well as 100-fascicle mass of current-year needles of the pine stands for different variants of the experiment Pustert in the period 1964 through 2004



plots. In phase IV (Pfaffenwinkel: 1993–1999; Pustert: 1989–2003), volume increments remained at a high level (Pfaffenwinkel) or showed a slightly decreasing trend (Pustert), without any significant differences between the experimental variants. In Pfaffenwinkel the mean annual increment of merchantable wood during the whole monitoring period was 9.6 m³ for the NPKMgCa-fertilized pines, 9.4 m³ for the LUP, and 8.2 m³ for the CON variant. NPK-MgCa fertilization (FER; total volume increase in the

period 1964–1999: $344 \pm 9 \text{ m}^3$; Fig. 9b) thus resulted in a statistically significant increment increase by 16% relative to the control (total volume increase: $296 \pm 31 \text{ m}^3$); the combination of PKMgCa fertilization, tillage, and lupine introduction (LUP; total volume increase in the period 1964–1999: $338 \pm 15 \text{ m}^3$) resulted in a statistically significant acceleration by 14%.

In Pustert, the mean annual increment of merchantable wood was 9.2 m^3 for the NPKMgCa-fertilized pines

Table 4Foliar concentration(arithmetic mean

value \pm standard deviation in mg g⁻¹ dry matter) of N, P, K, Ca, and Mg in current-year needles of the Scots pines at the variants CON (control), FER (superficial application of NPK-MgCa fertilizer and lime, and LUP (combination of application of PKMgCa fertilizer and lime, tillage, and lupine sowing) of the amelioration experiment Pfaffenwinkel for the periods 1965–1974, 1975–1984, 1985– 1994, and 1995–2004

Significant differences between variants are indicated by different letters

Table 5 Foliar concentrat (arithmetic mean value \pm standard deviation mg g^{-1} dry matter) of N, P Ca, and Mg in current-year dles of the Scots pines at th variants CON (control), FE (superficial application of M MgCa fertilizer and lime, a LUP (combination of appli tion of PKMgCa fertilizer a lime, tillage, and lupine sow of the amelioration experin Pustert for the periods 1965 1974, 1975-1984, 1985-19 and 1995-2004

Significant differences between variants are indicated by different letters

| | Period 1965-1974 | Period 1975-1984 | Period 1985–1994 | Period 1995-2004 |
|-----|---------------------------|---------------------------|--------------------------|--------------------------|
| N | | | | |
| CON | 13.72 ± 0.71 a | 14.32 ± 1.07 a | 16.22 ± 1.95 a | 14.07 ± 1.27 a |
| FER | $16.53\pm0.96~\mathrm{b}$ | $15.75\pm1.03~\mathrm{b}$ | 16.88 ± 1.73 a | 14.86 ± 1.21 a |
| LUP | $14.56\pm0.64~\mathrm{c}$ | $16.37\pm0.84~\mathrm{b}$ | 17.06 ± 1.94 a | 15.10 ± 1.24 a |
| Р | | | | |
| CON | 1.56 ± 0.11 a | 1.61 ± 0.09 a | 1.66 ± 0.21 a | 1.55 ± 0.20 a |
| FER | $1.79\pm0.12~\mathrm{b}$ | $1.73\pm0.09~\mathrm{b}$ | 1.67 ± 0.19 a | 1.61 ± 0.19 a |
| LUP | $1.67\pm0.18~\mathrm{ab}$ | $1.81\pm0.12~\mathrm{b}$ | 1.74 ± 0.20 a | 1.63 ± 0.25 a |
| K | | | | |
| CON | 5.89 ± 0.41 a | 5.99 ± 0.40 a | 5.97 ± 0.41 a | 5.51 ± 0.87 a |
| FER | $5.98\pm0.32~\mathrm{a}$ | 5.70 ± 0.43 a | $5.52\pm0.21~\mathrm{b}$ | 5.11 ± 0.72 a |
| LUP | 5.70 ± 0.31 a | 5.74 ± 0.52 a | $5.55\pm0.28~\mathrm{b}$ | 4.92 ± 0.77 a |
| Ca | | | | |
| CON | $2.24\pm0.14~\mathrm{a}$ | 2.07 ± 0.28 a | $1.89\pm0.33~\mathrm{a}$ | 1.75 ± 0.25 a |
| FER | $2.79\pm0.25~\mathrm{b}$ | $2.63\pm0.34~\mathrm{b}$ | $2.53\pm0.27~\mathrm{b}$ | $2.46\pm0.29~\mathrm{b}$ |
| LUP | $3.22\pm0.32~\mathrm{c}$ | $2.70\pm0.33~\mathrm{b}$ | $2.30\pm0.19~\mathrm{b}$ | $2.37\pm0.27~\mathrm{b}$ |
| Mg | | | | |
| CON | $0.69\pm0.15~\mathrm{a}$ | $0.66\pm0.14~\mathrm{a}$ | 0.79 ± 0.08 a | 0.77 ± 0.11 a |
| FER | 0.72 ± 0.17 a | $0.72\pm0.14~\mathrm{a}$ | 0.81 ± 0.06 a | 0.80 ± 0.10 a |
| LUP | $0.89\pm0.17~\mathrm{b}$ | 0.74 ± 0.12 a | 0.85 ± 0.06 a | 0.79 ± 0.09 a |
| | | | | |

| | Period 1965–1974 | Period 1975-1984 | Period 1985-1994 | Period 1995-2004 |
|-----|---------------------------|---------------------------|--------------------------|---------------------------|
| N | | | | |
| CON | 13.48 ± 0.57 a | 14.44 ± 0.84 a | 16.39 ± 0.77 a | 17.33 ± 1.55 a |
| FER | $15.36\pm0.90~\mathrm{b}$ | $15.62\pm0.59~\mathrm{b}$ | 16.46 ± 1.08 a | $17.94\pm0.82~\mathrm{a}$ |
| LUP | $14.80\pm0.58~\mathrm{b}$ | $15.88\pm0.65~\mathrm{b}$ | 16.55 ± 0.70 a | 17.50 ± 1.49 a |
| Р | | | | |
| CON | $1.43\pm0.05~\mathrm{a}$ | 1.42 ± 0.06 a | 1.34 ± 0.07 a | $1.32\pm0.07~\mathrm{a}$ |
| FER | $1.67\pm0.08~\mathrm{b}$ | $1.63\pm0.08~\mathrm{b}$ | 1.45 ± 0.14 a | $1.33\pm0.13~\mathrm{a}$ |
| LUP | $1.63\pm0.06~\mathrm{b}$ | $1.65\pm0.11~\mathrm{b}$ | 1.38 ± 0.06 a | $1.30\pm0.09~\mathrm{a}$ |
| Κ | | | | |
| CON | 5.14 ± 0.32 a | 5.10 ± 0.24 a | 5.08 ± 0.16 a | $4.97\pm0.40~\mathrm{a}$ |
| FER | 5.43 ± 0.27 a | 5.31 ± 0.37 a | 4.77 ± 0.44 a | $4.70\pm0.52~\mathrm{a}$ |
| LUP | 5.34 ± 0.22 a | 5.42 ± 0.26 a | $4.93\pm0.26~\mathrm{a}$ | $4.79\pm0.27~\mathrm{a}$ |
| Ca | | | | |
| CON | $3.62\pm0.52~\mathrm{a}$ | $3.21\pm0.24~\mathrm{a}$ | $2.63\pm0.16~\mathrm{a}$ | 2.73 ± 0.40 a |
| FER | 3.17 ± 0.28 a | $3.28\pm0.26~a$ | 2.90 ± 0.34 a | 2.96 ± 0.27 a |
| LUP | $3.53\pm0.31~\mathrm{a}$ | $3.18\pm0.50~a$ | 2.91 ± 0.15 a | $3.16\pm0.22~a$ |
| Mg | | | | |
| CON | 0.94 ± 0.14 a | 0.84 ± 0.11 a | 0.81 ± 0.04 a | $0.88\pm0.11~\mathrm{a}$ |
| FER | 0.89 ± 0.19 a | 0.87 ± 0.12 a | 0.86 ± 0.08 a | $0.91\pm0.10~\mathrm{a}$ |
| LUP | 0.97 ± 0.19 a | 0.87 ± 0.12 a | 0.90 ± 0.05 a | 0.97 ± 0.11 a |

(+28%), 8.2 m³ for the pines with lupine introduction (+15%), and 7.2 m³ for the pines on the control plots. The total volume increase in the period 1965–2003 was $287 \pm 26 \text{ m}^3$; at CON, significantly larger at LUP

 $(330 \pm 9 \text{ m}^3)$, and largest at FER $(368 \pm 18 \text{ m}^3; \text{ Fig. 9d})$. Thus, FER had a larger long-term effect on volume increment at Pustert than at Pfaffenwinkel, whereas the effects of LUP were similar at both sites.

Table 6 One-hundred-fascicle masses (arithmetic mean value \pm standard deviation in g dry matter) of current-year needles of the Scots pines at the variants CON (control), FER (superficial application of NPKMgCa fertilizer and lime, and LUP (combination of application of PKMgCa fertilizer and lime, tillage, and lupine sowing) of the amelioration experiments Pfaffenwinkel and Pustert for the periods 1965–1974, 1975–1984, 1985–1994, and 1995–2004

| | Period 1965–1974 | Period 1975–1984 | Period 1985–1994 | Period 1995–2004 |
|---------|--------------------------|--------------------------|--------------------------|---------------------|
| Pfaffer | nwinkel | | | |
| CON | $4.18\pm1.01~\mathrm{a}$ | $4.05\pm0.50~a$ | $5.21\pm0.61~a$ | 3.73 ± 1.18 a |
| FER | 5.28 ± 0.93 a | $4.59\pm0.74~\mathrm{a}$ | $5.80\pm0.60~a$ | 4.13 ± 1.09 a |
| LUP | 4.56 ± 0.99 a | 4.50 ± 0.48 a | 5.64 ± 0.48 a | 4.06 ± 1.19 a |
| Puster | t | | | |
| CON | $4.10\pm0.72~\mathrm{a}$ | $4.43\pm0.55~a$ | $5.47\pm0.94~a$ | 4.62 ± 0.72 a |
| FER | $5.23\pm0.79~\text{b}$ | 4.55 ± 0.77 a | 5.56 ± 0.75 a | 4.14 ± 0.83 a |
| LUP | $4.96\pm0.43~b$ | 5.04 ± 0.77 a | $5.73\pm0.95~\mathrm{a}$ | 4.41 ± 0.95 a |

Significant differences between variants are indicated by different letters

Biomass of different tree species at Pustert

At the beginning of the experiment in 1964, the biomass of merchantable wood at the various was 68 ± 5 Mg ha⁻¹ (dry matter). Scots pine strongly dominated (>95% of total wood biomass); the percentages of Norway spruce and broadleaf trees were marginal (Fig. 10). In 1998, 44 years later, the total wood biomass has doubled at all experimental variants. At all plots, the contribution of other tree species than pine to the total biomass of merchantable wood in 1998 was increased strongly compared to 1964. The species change was largest at the FER plots, intermediate at the LUP plots, and smallest, but still considerable at the CON plots. In 1998, at the FER plots Scots pine comprised 91%

of total merchantable wood biomass; the contribution of oak, spruce, and other broadleaf trees were 5, 4, and 1%, respectively. At the LUP plots, Scots pine comprised 94% of total merchantable wood biomass, followed by other broadleaf species than oak (birch, willow) with 4%, and spruce with 2%. At the CON site, 96% of total merchantable wood was assigned to Scots pine; the contribution of oak, spruce, and other broadleaf trees was 1, 1, and 2%, respectively. Between 1998 and 2003 a further slight increase (+1 to +6%) of total merchantable wood biomass was observed, which was entirely due to an increase of the woody biomass of other tree species than pine. Within the 5-year-period 1998 through 2003, the absolute biomass of broadleaf trees (merchantable wood) increased by 63 (LUP), 78 (FER), and 87% (CON), The contribution of broadleaf trees to the total stand biomass (merchantable wood) almost doubled within that period: it increased from 3 to 6% at the CON plots, from 4 to 7% at the LUP plots, and from 6 to 10%, at the FER plots. Additionally, at the FER plots a strong increase in spruce biomass was observed; in 2003 spruce comprised 7% of total tree biomass at the FER plots compared to 2% for the other experimental variants.

Discussion

Long-term amelioration effects on topsoil acidity

In both experiments, the amelioration procedures, which included an application of lime as well as of Ca, Mg, and K-bearing fertilizers resulted in a sustainable increase of pH and base saturation, and in a significant long-term enhancement of the stocks of exchangeable Ca, Mg, and K

Table 7 Growth and yield characteristics for the treatment variants CON, FER, and LUP of the experiments Pfaffenwinkel and Pustert

| | Stand data at the first survey | | Stand data for the treatment period | | | | Stand data 2000 and 2003, respectively | | |
|--------|--------------------------------|-----------------------|--|--|--|---|--|---|---------------------------------------|
| | Age (year) | Mean height (m) | Standing volume (m ³ ha ⁻¹) | Mean annual volume increment (m ³ ha ⁻¹ year ⁻¹) | Mean annual volume increment (%) | Cumulative volume increment $(m^3 ha^{-1})$ | Cumulative volume increment (%) | Cumulative volume increment $(m^3 ha^{-1})$ | Cumulative volume increment (%) |
| Pfaffe | nwinke | 1 | | | | | | | |
| CON | 86 | 13.2 | 121.3 | 8.0 | 100.0 | 289.3 | 100.0 | 509.3 | 100.0 |
| FER | 86 | 12.5 | 114.7 | 9.6 | 118.9 | 344.0 | 118.9 | 543.3 | 106.7 |
| LUP | 86 | 12.8 | 119.3 | 9.4 | 116.7 | 337.7 | 116.7 | 547.3 | 107.5 |
| Puster | t | | | | | | | | |
| CON | 81 | 15.3 | 143.7 | 7.2 | 100.0 | 286.0 | 100.0 | 573.7 | 100.0 |
| FER | 81 | 15.6 | 165.0 | 9.2 | 128.4 | 367.3 | 128.4 | 664.3 | 115.8 |
| LUP | 81 | 15.5 | 152.7 | 8.2 | 115.0 | 328.7 | 115.0 | 627.0 | 109.3 |

Essential information is presented for the first survey in 1963 and 1964, respectively, for the some 40 years lasting observation period, and for the last survey in 2002 and 2003, respectively



Fig. 9 Mean annual and cumulative volume increments of merchantable wood (stem volume over 7 cm diameter at the smaller end; without bark) of the pine stands for different variants of the experiments



Fig. 10 Contribution of different tree species to the total biomass of merchantable wood for different variants of the experiment Pustert in 1964, 1998, and 2003

in the topsoil despite the high atmospheric deposition of N and S at both sites. More than 30 years after the last fertilizer application, at Pfaffenwinkel still about 40% of the applied Ca and Mg are present as exchangeable Ca and Mg in the uppermost 30 cm of the soil; another 15% of the applied Mg has been taken up by the stand. At Pustert, the long-term retention of fertilized Ca and Mg was even more pronounced. Here, more than 50% of the applied Ca was recovered as exchangeable cations in the topsoil; the amount of recovered Mg even exceeded the applied amount by a factor of 2–3. These results are strongly contrasting to findings of other studies in Norway spruce stands where (1) pH increases were restricted to the forest floor and the uppermost 5 cm of the mineral topsoil (Huber





a, **b** Pfaffenwinkel and **c**, **d** Pustert in the period 1964/1965 through 1999/2003

et al. 2006) and (2) most of the base cations applied by addition of lime and/or fertilizer were lost from the topsoil by leaching and stand uptake within 10-20 years (e.g., Ulrich and Keuffel 1970; Hildebrand 1986). Huber et al. (2006) report that 20 years after superficial application of $4,000 \text{ kg ha}^{-1}$ dolomitic limestone to a loess-derived soil stocked with mature Norway spruce forest, only 20% of the applied Ca, but all of the applied Mg was lost from the uppermost 40 cm of the soil by seepage water leaching. Consequently, Hildebrand (1994) recommended a repetition interval of 25 years for the application of dolomitic lime at a rate of 2.5–3 t ha⁻¹ to compensate soil acidification and base cation losses in forests subject to elevated atmospheric acid deposition. The cation exchange capacity in the topsoils of the liming experiments described by Ulrich and Keuffel (1970) and Hildebrand (1986) was larger than that of the soils in our study; thus the prolonged retention of applied base cations in our experiment cannot be due to a larger cation storage capacity in the topsoil. However, the annual precipitation at the sites studied by Ulrich and Keuffel (1970) (745 mm) and Hildebrand (1986) (950–1,000 mm) is considerably larger than at our sites (600-615 mm), and the soils are sandy and thus more permeable than our soils. Also the larger fertilizer retention in the topsoil of Pustert compared to Waldsassen can be at least partly explained (another crucial process is the vigorous establishment of broadleaf tree regeneration at Pustert, see below) explained by the strongly retarded water movement in the clayey, compacted subsoil at Pustert.

Thus the seepage water flux seems to be the crucial factor governing the retention of fertilizer and lime applied to forest soils. Our results show that the residence time of applied Ca and Mg in the topsoil can be considerably longer than 10-20 years even in high acid deposition environments, if vertical seepage water percolation is slow due to modest precipitation and/or fine-textured subsoil. This had already been shown in Scandinavian studies (Hallbäcken and Popovic 1985; Derome et al. 1986) and also in a previous soil inventory at our study sites conducted in 1994 (Prietzel et al. 1996). A comparison of the results of our earlier study with the data presented in this paper shows that between 1994 and 2004 the pH and the base saturation have only decreased slightly on the ameliorated plots of Pfaffenwinkel and Pustert. At the FER variant of Pfaffenwinkel, the amount of fertilizer-derived exchangeable Ca in the topsoil (defined as difference between the stocks of exchangeable Ca in the topsoil of the FER and CON plots) within the most recent 10 years has decreased from $1,133 \text{ ha}^{-1}$ (54% of the fertilized Ca amount) to 825 kg ha^{-1} (39% of fertilized Ca). The amount of fertilizer-derived exchangeable Mg decreased in the same time from 13 to 12 kg ha^{-1} (41 and 39% of fertilized Mg, respectively). At Pustert the differences between the exchangeable Ca and Mg stocks in the topsoil of the fertilized and the control plots have even increased between 1994 and 2004 from 1,072 to 1,415 kg Ca ha⁻¹, and from 43 to 63 kg Mg ha⁻¹. The difference between the exchangeable Mg stocks in the topsoil of the control and the ameliorated plots accounts for 197 (FER) and 337% (LUP) of the applied Mg amount. This increase in Ca and Mg "retention" in the topsoil of Pustert from 1994 to 2004 per se as well as its magnitude can be explained by the fact that at Pustert the initial amelioration has primed a self-rehabilitation process of the ecosystem powered by the vigorous establishment of broadleaf trees at that site (Fig. 10). Originally the site was stocked with broadleaf forest (probably a mixed oak-beech forest). Long-term human impact has resulted in a replacement of that deciduous forest by a pure Scots pine stand, and a significant depletion of the topsoil in N, P, and base cations. In contrast to Pfaffenwinkel, the subsoil of Pustert is characterized by a high base saturation, and there are still some old oak, beech, and other deciduous trees as well as Norway spruces present in the surrounding. Additionally, within a 100-m distance to the experimental site, croplands acting as refuge sites for large anecic earthworm species can be found, from which earthworms reinvaded the ameliorated plots and strongly increased in numbers (Rehfuess et al. 1984). The improvement of the chemical status of the topsoil by the initial amelioration, the activity of earthworms, and at the tilled plots also the mechanical disruption of the mor-type forest floor facilitated the successful establishment and development of a natural regeneration rich in broadleaf tree species (Quercus robur, Salix capra, Betula pendula, and Populus tremula; Fig. 10). Compared to Scots pine needles, the foliage of oak, birch, poplar, and willow is more easily decomposable and richer in N, P, and base cations (Wittich 1961; Prietzel 2004; Krauß and Heinsdorf 2005). Consequently, the introduction of deciduous trees into Scots pine stands on sites with acidic topsoils has been shown to result in an increased pH value and base saturation as well as in decreased C/N ratios of the topsoil (Prietzel 2004). Also at Pfaffenwinkel the amelioration has stimulated the establishment of broadleaf tree seedlings; however, their development was much slower than at Pustert. Moreover, small exchangeable base cation stocks in the subsoil of Pfaffenwinkel prevent a rapid improvement of the chemical status of the topsoil by the base cation pump function of broadleaf trees.

Long-term amelioration effects on soil and ecosystem carbon and nitrogen stocks

The observed losses of soil OC (8–10 t OC ha⁻¹ or 10 to 12% of the original topsoil OC stock) as well as the net transfer of OC from the forest floor into the mineral topsoil are typical features of acid soils with conifer stands after liming (e.g., Matzner 1985; Marschner and Wilczynski 1991; Kreutzer 1995; Persson et al. 1995b; Nilsson et al. 2001; Jandl et al. 2002; Huber et al. 2006). In contrast, fertilization with N, NP, or NPK alone generally results in increases of the topsoil OC pools (e.g., Mälkönen 1990; Nohrstedt 1992; Binkley and Högberg 1997), probably due to a combination of increased OC input from stand litter (Baum 1981; Mälkönen 1990) and decreased microbial humus mineralization (Nohrstedt et al. 1989; Berg and Matzner 1997; Hagedorn et al. 2003). At our study sites, aboveground stand litterfall was increased considerably after fertilization (Baum 1981). The same is probably the case for the root litter input. Nevertheless, accelerated humus mineralization probably due to a more favorable base saturation and removal of the former N and P limitation for microbial activity over-compensated the increased C input, resulting in the observed significant OC losses of the topsoils. Their magnitude $(200-250 \text{ kg ha}^{-1} \text{ year}^{-1}$ over a period of 40 years), however, is small compared to the losses reported by many other amelioration studies, where CaCO₃ or CaO had been included in the fertilizer regime (e.g., Hetsch and Ulrich 1979; Jandl et al. 2002). The comparably small OC losses from the soils at Pfaffenwinkel and Pustert may be caused by the fact that both study sites had been previously depleted in easily mineralizable OC by long-term historic litter raking. Considering the entire ecosystem, the OC losses of the topsoil at the ameliorated plots have been more than compensated by an

increased sequestration of OC in merchantable wood and coarse roots as a consequence of accelerated stand growth. At the study sites, the growth of the pine stands was increased by the amelioration procedures until 1992 (Pfaffenwinkel) and 1988 (Pustert); i.e., for 28 and 24 years (Fig. 9). This is much longer than the reported effects of N fertilization on the growth of Scots pine in most other studies (e.g., Zöttl and Kennel 1962; Nilsen and Abrahamsen 2003; Petterson and Högbom 2004), but not unique (e.g., Saarsalmi et al. 2006). According to our assessments, the C sequestration of the stands in the period between 1964 and 2004 was increased relative to the respective control variants by 8.9 t ha^{-1} (+12%) and 9.4 t ha^{-1} (+15%) for the variants with lupine treatment at Pfaffenwinkel and Pustert, respectively, and by 9.7 t ha^{-1} (Pfaffenwinkel; +13%) and 18.0 t ha^{-1} (Pustert; +28%) for the variants with superficial NPKCaMg fertilization. The increased C sequestration in woody biomass exceeded the topsoil OC losses, resulting in an overall OC balance of the entire ecosystem of +55 kg C ha⁻¹ year⁻¹ after NPKMgCa fertilization and +28 kg C ha⁻¹ year⁻¹ after lupine introduction for Pfaffenwinkel. The corresponding OC balance for Pustert was +250 kg C ha⁻¹ year⁻¹ on the FER and -23 kg C ha⁻¹ year⁻¹ on the LUP plots. In summary, our data support the conclusion of Jandl et al. (2007) that adequate forest fertilization can be a powerful management tool to increase carbon sequestration in forest ecosystems-even if it contains a lime component.

At both sites, both amelioration treatments resulted in a considerable enrichment of the soil organic matter in nitrogen and an increased topsoil nitrogen stock. These findings are similar to the results of many earlier fertilization experiments with Scots pine on poor sites (e.g., Melin and Nömmik 1988; Mälkönen and Kukkola 1991). More than 30 years after the last application of N-containing fertilizer, 63 and 78% of the applied N can still be recovered in the topsoil of Pfaffenwinkel and Pustert, respectively, and another 4% and 8% in additional woody biomass of the faster-growing stands. As with OC, the N retention was larger than in other fertilization experiments conducted in Scots pine stands (e.g., Nohrstedt 1990; Tamm et al. 1995; Heinsdorf and Beck 2003), mixed stands of Scots pine and Norway spruce (e.g., Hetsch and Ulrich 1979), or Norway spruce stands (e.g., Nohrstedt et al. 2000).

Long-term amelioration effects on stand nutrition

At both sites, soil amelioration resulted in increased foliar concentrations of N, (P), and Ca as well as in larger needle masses. The effects decreased with progressing time, and were statistically significant only during the first two decades. However, they could be observed during the entire monitoring period, i.e., even more than 30 years after the last fertilizer amendment. In contrast to N and P, the amelioration effects on foliar Ca and Mg (Pustert) concentrations became more prominent with progressing time (Figs. 7e, 8d, e). Whereas the NPKMgCa fertilization + lime application was more effective than the combination of PKMgCa fertilization, liming, tillage, and lupine introduction in improving the insufficient N and P nutritional status of the stand during the first 10 years of the experiment, this effect disappeared in later years. The sustained positive amelioration effects on the nutritional status of the stands in Pfaffenwinkel and Pustert contrast to results of earlier amelioration experiments on Scots pine and other conifers at other sites, where only short-termed (<10 years) improvements had been reported (e.g., Binkley 1986; Miller 1988). However, longer-lasting fertilization effects on stand nutrition, which even included the subsequent stand generation ("carry-over effect"), have been reported for Douglas fir stands in USA (Strader and Binkley 1989; Prietzel et al. 2004) and for pine stands in Canada (Prescott et al. 1995). The long-term amelioration effects on the nutritional status of the stands Pfaffenwinkel and Pustert match well with the changes of the chemical status of the topsoil. In both ecosystems, which are characterized by a small initial supply of the soil with N, P, Ca, and Mg, and high atmospheric S and N deposition, the amelioration effects on N nutrition were particularly large at the beginning (Rehfuess and Schmidt 1971; Preuhsler and Rehfuess 1982), when the N status of the ecosystems was poor. However, this effect was gradually offset by the increasing N saturation status of the control plots (Prietzel and Kölling 1998; Prietzel et al. 2006) caused by high atmospheric N input. On the other hand, the Ca and Mg fertilization effectively and sustainably mitigated the poor and/or decreasing Ca and Mg supply of the pines at both sites.

Long-term amelioration effects on stand growth: Nutrition-growth relationships

Our single correlation analyses, which include all experimental variants and cover the whole period of 40 years of monitoring unexpectedly revealed a key relevance of the P nutrition—besides the N supply—for the growth of the stands: At both sites the mean annual increment of merchantable wood in the periods between the stand inventories showed a stronger correlation to the mean foliar P concentration during the respective period (Pfaffenwinkel: r = 0.60; P < 0.001; Pustert: r = 0.60; P < 0.01) than to the foliar N concentration (Pfaffenwinkel: r = 0.39; P < 0.05; Pustert: r = -0.08; n.s.), and no significant correlation to the foliar Concentrations of K, Mg, or Ca. Also the mean foliar P contents (mg P needle⁻¹) were better correlated with the increment of merchantable wood (Pfaffenwinkel: r = 0.73; P < 0.001; Pustert: r = 0.87; P < 0.001) than the

foliar N contents (Pfaffenwinkel: r = 0.66; P < 0.001; Pustert: r = 0.56; P < 0.01).

In multiple linear regression models (Table 8) covering again the entire monitoring period from 1964 through 1999 (Pfaffenwinkel) and from 1965 through 2003 (Pustert), the foliar P concentration explained 50 (Pfaffenwinkel) and 44% (Pustert) of the annual increment of merchantable wood in the differently treated pine stands, whereas the foliar N concentration explained only 20% of the growth variance at Pfaffenwinkel and was not even a significant predictor for stand growth at Pustert. The prominent role of foliar P for the statistical explanation of pine growth during the entire monitoring period does not necessarily mean that the stands originally suffered from P deficiency. In fact, foliar P concentrations in the Scots pines at the control plots of both sites always exceeded the threshold value of P limitation (1.2 mg P g^{-1} ; Arbeitskreis Standortskartierung 2003); up to now no indication exists that P fertilization alone would stimulate pine growth at our experimental sites. All fertilization trials conducted hitherto show that at least during the 1960s and 1970s the addition of N alone stimulated the wood production of pine.

On the other hand, the superior predictor value of foliar P concentrations for the growth of pine at both study sites must be seen also in the context of a general recovery of soil fertility at these sites from historic N (P, base cations) depletion, associated with high atmospheric N deposition. In the long run, this development shifts both ecosystems from N limitation at the start of our experiments into sys-

tems in which stand growth will be mainly determined by other growth factors than N, such as water, P, Mg, K, or even Ca. There is already some evidence available supporting that hypothesis, mainly the significant increase of foliar N/P and N/Ca ratios in the early 1980s (Prietzel et al. 1997; Prietzel and Kölling 1998). Therefore, we additionally conducted splitted regression analyses comprising either the first 20 years (1964–1983) or the second 20 years (1984-1999/2003) of the monitoring period (Table 9). At Pfaffenwinkel, in the second half of the monitoring period the foliar P and N concentrations completely lost their predictor value for the increment of merchantable wood and were replaced by foliar K (negative beta value; Table 9) and Mg. At Pustert, the coefficients of determination of foliar P and N concentrations with regard to growth were reduced considerably in the second half of the monitoring period; and-most interestinglyfoliar N turned from a positive into a negative predictor, i.e., the growth of the pines now was reduced instead of enhanced in years with large foliar N concentrations. At the moment, however, the question whether and when Mg, P, or Ca deficiency will occur and fertilization with these elements might result in additional growth increases at our experimental sites remains open. Particularly at the Pustert site, the accelerating increase of broadleaf tree biomass, which is characterized by foliage much richer in base cations and phosphorus compared to Scots pine foliage (Wittich 1961; Prietzel 2004; Krauß and Heinsdorf 2005) has counteracted or mitigated topsoil acidification and will increasingly do so in the future.

 Table 8
 Comparison of different linear regression models including the mean annual volume increment of merchantable wood as dependent factor and the mean 100-fascicle masses as well as the mean foliar

concentrations of current-year needles of the Scots pine stands of Pfaffenwinkel and Pustert for the periods between the stand inventories (eight inventories between 1964/1965 and 1999/2003, respectively)

| | Coefficient of determination R^2 | Standardized beta | Significance (β) |
|----------------------------------|--|-------------------|--------------------------|
| Pfaffenwinkel: independent var | iables: foliar nutrient concentrations, 100-fascicle | mass | |
| Model 1: only one independent | variable | | |
| 100-fascicle mass | 0.62 | 0.790 | *** |
| Foliar P concentration | 0.50 | 0.738 | *** |
| Foliar N concentration | 0.20 | 0.450 | * |
| Model 2: all significant indepen | dent variables (stepwise inclusion) | | |
| 100-fascicle mass | 0.62 | 0.790 | *** |
| Pustert: independent variables: | foliar nutrient concentrations, 100-fascicle mass | | |
| Model 1: only one independent | variable | | |
| 100-fascicle mass | 0.55 | 0.738 | *** |
| Foliar P concentration | 0.44 | 0.661 | ** |
| Model 2: all significant indepen | dent variables (stepwise inclusion) | | |
| 100-fascicle mass | 0.83 | 0.529 | *** |
| Foliar P concentration | | 0.641 | *** |
| Foliar N concentration | | 0.272 | * |

Number of asterisks (*, *, and ***) indicates that the independent variable is a significant predictor at P < 0.05, 0.01, and 0.001, respectively

| | 1964/1965 through 1983 | | | 1984 through 199 | 1984 through 1999/2003 | | |
|-----------------------------|------------------------------|-----------------------|---------------------|------------------------------|------------------------|------------------------|--|
| | Coefficient of determination | Standardized beta | Significance (β) | Coefficient of determination | Standardized beta | Significance (β) | |
| Pfaffenwinkel: independen | t variables: foliar nut | rient concentrations | , 100-fascicle mass | | | | |
| Foliar P concentration | 0.82 | 0.903 | *** | | | ns | |
| 100-fascicle mass | 0.74 | 0.860 | *** | | | ns | |
| Foliar N concentration | 0.33 | 0.571 | * | | | ns | |
| Foliar K concentration | | | ns | 0.72 | -0.849 | *** | |
| Foliar Mg concentration | | | ns | 0.51 | 0.715 | ** | |
| Pustert: independent varial | oles: foliar nutrient co | oncentrations, 100-fa | ascicle mass | | | | |
| Foliar P concentration | 0.84 | 0.917 | *** | 0.65 | 0.805 | ** | |
| Foliar N concentration | 0.73 | 0.854 | ** | 0.54 | -0.736 | ** | |
| 100-fascicle mass | 0.61 | 0.771 | ** | 0.70 | 0.835 | ** | |

 Table 9 Comparison of different linear regression models including the mean annual volume increment of merchantable wood as dependent factor and the mean one-hundred-fascicle masses as well as the
 mean foliar concentrations of current-year needles of the Scots pine stands of Pfaffenwinkel and Pustert for the periods between the growth inventories in the first and the second part of the monitoring period

Number of asterisks (*, *, and ***) indicates that the independent variable is a significant predictor at P < 0.05, 0.01, and 0.001, respectively

Comparison of long-term effects of NPKMgCa fertilization + lime application and combination of tillage, lupine sowing, liming, and PKMgCa fertilization

Forty years after the different amelioration treatments, the large differences between the topsoil chemistry at the plots with application of NPKMgCa fertilizer plus lime and those with combination of tillage, lupine sowing, liming, and PKMgCa fertilization, which had been reported in earlier studies (significantly smaller topsoil C, N, and Ca stocks at the LUP plots compared to the FER plots of Pfaffenwinkel 18 years after amelioration; Makeschin et al. 1985; Rehfuess et al. 1991) have leveled off. In 2004, no significant differences between the pH values, the base saturation, and the stocks of base cations, OC, and total N in the topsoil of the FER and the LUP plots could be noticed any more (Figs. 1, 2, 3, and 6). However, even after 40 years at both sites the topsoils with application of NPKMgCa fertilizer and lime still contain-even though not statistically significant-larger stocks of N and exchangeable Ca than the plots with tillage, lupine sowing, liming, and PKMgCa fertilization. At Pustert, also the topsoil OC stock is stillagain not significantly-larger at the FER than at the LUP plots, where significant humus mineralization occurred during the first years after tillage (Makeschin et al. 1985; Rehfuess et al. 1991). Moreover, after 40 years the cumulative production of merchantable wood and thus also the cumulative C sequestration in the Scots pine stands at both sites is still larger after NPKMgCa fertilization than after tillage, lupine sowing, and PKMgCa fertilization (Fig. 9).

Retrospectively, not only in the short and intermediate term (6-year retrospective: Rehfuess and Schmidt 1971;

18-year retrospective: Makeschin et al. 1985, Rehfuess et al. 1991), but also in the long term (40-year retrospective: this study), the application of NPKMgCa fertilizer plus lime can be regarded as the better alternative to the combination of tillage, lupine sowing, liming, and PKMgCa fertilization to improve soil fertility and stand growth in N-limited Scots pine ecosystems. Also in the long run, superficial application of NPKMgCa fertilizer is associated with smaller soil humus losses than the introduction of lupine, which had to be combined with tillage to create a seedbed for the lupines. Superficial application of NPKMgCa fertilizer plus lime did also better than the combination of tillage, lupine sowing, liming, and PKMgCa fertilization with respect to the general silvicultural aim of a conversion of Scots pine monocultures into broadleaf-rich mixed stands, because it resulted in a larger percentage of regeneration of desired tree species (e.g., oak) and a smaller percentage of regeneration of undesired broadleaf tree species (e.g., birch and willow) (Fig. 10). In Scots pine stands at sites with increased N saturation and/or high atmospheric N deposition and acidic soils, which are much more frequent in present-day Central Europe than they were 40 years ago, the combination of tillage, lupine introduction, liming, and PKMgCa fertilization is even less viable, because it adds additional N to the ecosystems and is probably associated with significant nitrate mobilization after tillage. At such sites application of PKMg(Ca) fertilizer or wood ash in small doses (Lundström et al. 2003; Jacobson et al. 2004; Saarsalmi et al. 2006) may help to accelerate the natural or artificial conversion of Scots pine monocultures on into mixed broadleaf-rich stands.

Conclusions

In Central European Scots pine ecosystems on sites with acidic topsoils and historic N and P depletion due to longterm over-exploitation, NPKMgCa fertilization + lime application and also combination of PKMgCa fertilization, liming, tillage, and introduction of lupine results in longterm stand growth acceleration, topsoil de-acidification, and N accumulation, as well as in a transfer of OC and N from the forest floor into the mineral topsoil, but also in a considerable OC loss of the soil.

A close relationship between stand growth and the N and P nutritional status of the stands as well as the simultaneous increase of foliar N and P concentrations and volume increments of the pines at the ameliorated plots particularly during the first 20 years of the monitoring period demonstrate the key relevance of N and P nutrition of the stands for their growth. In contrast to a sole application of lime, after multielement fertilization including a lime component the additional OC sequestration in merchantable wood and coarse roots over-compensates the soil OC losses, resulting in increased C accumulation in the ecosystem.

On N-limited sites with acid soils, atmospheric N deposition in the long-run results in the replacement of nitrogen as primary growth-regulating factor by other nutrients such as P, Mg, Ca or water. With regard to the high atmospheric N deposition in many Central European forests, the application of PMg(K,Ca) fertilizer or wood ash in small doses seems to be an adequate mean (1) to compensate soil base cation losses caused by leaching and increased nutrient uptake by faster-growing stands. Moreover, it (2) facilitates and accelerates the natural or artificial conversion of manmade Scots pine monocultures into mixed broadleaf-rich stands, which is a generally accepted silvicultural aim in Central Europe and which counteracts topsoil acidification and base cation depletion.

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