



Half a century of Scots pine forest ecosystem monitoring reveals long-term effects of atmospheric deposition and climate change

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Abstract

At two forest sites in Germany (*Pfaffenwinkel*, *Pustert*) stocked with mature Scots pine (*Pinus sylvestris* L.), we investigated changes of topsoil chemistry during the recent 40 years by soil inventories conducted on replicated control plots of fertilization experiments, allowing a statistical analysis. Additionally, we monitored the nutritional status of both stands from 1964 until 2019 and quantified stand growth during the monitoring period by repeated stand inventories. Moreover, we monitored climate variables (air temperature and precipitation) and calculated annual climatic water balances from 1991 to 2019. Atmospheric nitrogen (N) and sulfur (S) deposition between 1964 and 2019 was estimated for the period 1969–2019 by combining annual deposition measurements conducted in 1985–1987 and 2004 with long-term deposition records from long-term forest monitoring stations. We investigated interrelations between topsoil chemistry, stand nutrition, stand growth, deposition, and climate trends. At both sites, the onset of the new millennium was a turning point of important biogeochemical processes. Topsoil acidification turned into re-alkalinization, soil organic matter (SOM) accumulation stopped, and likely turned into SOM depletion. In the new millennium, topsoil stocks of S and plant-available phosphorus (P) as well as S and P concentrations in Scots pine foliage decreased substantially; yet, age-referenced stand growth remained at levels far above those expected from yield table data. Tree P and S nutrition as well as climate change (increased temperature and drought stress) have replaced soil acidification as major future challenges for both forests. Understanding of P and S cycling and water fluxes in forest ecosystems, and consideration of these issues in forest management is important for successfully tackling the new challenges. Our study illustrates the importance of long-term forest monitoring to identify slow, but substantial changes of forest biogeochemistry driven by natural and anthropogenic global change.

KEYWORDS

carbon sequestration, N eutrophication, *Pinus sylvestris*, soil acidity, stand growth, stand nutrition, topsoil chemistry

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1 | INTRODUCTION

European forest ecosystems have been subject to substantial nutrient losses for centuries. In many forests, timber and fuelwood export, forest pasture, and litter raking resulted in considerable loss of soil organic matter (SOM) and nutrients such as nitrogen (N), phosphorus (P), potassium (K), magnesium (Mg), and calcium (Ca) from soils, accompanied by soil acidification. Forest exploitation in Germany and most European countries peaked in the 19th century, decreased in the early 20th century, and ceased after World War II. Since then, increased fossil fuel use and agriculture intensification resulted in elevated atmospheric deposition of sulfur (SO_4^{2-}), N (NH_4^+ , NO_3^-), and protons to forests (Meesenburg et al., 2016). Atmospheric S and N input culminated in the 1980s, followed by a rapid decrease in S deposition (Göttlein & Mellert, 2019; Oulehle et al., 2011), whereas N inputs remained high (Meesenburg et al., 2016; Schmitz et al., 2019).

Termination of soil-degrading forest overutilization and elevated atmospheric S and N deposition resulted in marked topsoil chemistry changes in German forests (Meesenburg et al., 2016; Prietzel, Kolb, & Rehfuss, 1997; Prietzel & Stetter, 2010; Prietzel, Stetter, Klemmt, & Rehfuss, 2006). Stocks of soil organic carbon (SOC) increased and topsoil SOM became enriched in N, whereas pH values, base saturation (BS), and stocks of plant-available nutrient “base” cations (Ca^{2+} , Mg^{2+} , K^+) and P decreased. Forests often showed an unprecedented growth acceleration (Mellert, Prietzel, Straussberger, & Rehfuss, 2004; Pretzsch, Biber, Schütze, Uhl, & Rötzer, 2014) but also developed an increasingly unbalanced nutritional status, characterized by excessive N supply and insufficient Ca, Mg, K, and P nutrition (Mellert et al., 2004; Prietzel, Rehfuss, Stetter, & Pretzsch, 2008). Another challenge for European forests is the recent climate change (Lindner et al., 2014), including a general trend of increasing air temperature and prolonged growing seasons (Menzel & Fabian, 1999), which results in increased water demand of trees and forests. Published results about recent climate warming effects on SOC stocks are scarce, and completely absent concerning other chemical forest soil properties. Recent climate warming has resulted in SOC losses of forest soils in Great Britain (Bellamy, Loveland, Bradley, Lark, & Kirk, 2005) and the German Alps (Prietzel, Zimmermann, Schubert, & Christophel, 2016), while other studies reported no SOC change (Callesen et al., 2015) or SOC increases (Grüneberg, Ziche, & Wellbrock, 2014; Jonard et al., 2017).

At present, information about long-term effects of acid deposition and climate warming on topsoil chemistry, stand nutrition, and growth of European forests is scarce. It is unknown to which extent recent decreases in atmospheric S and N input to European forests were associated with reversal of topsoil acidification and slowdown or halt of topsoil N eutrophication. This lack of knowledge is caused by the scarcity of long-term forest monitoring studies. Increases in pH and BS in forest floor and in some cases also in mineral topsoil horizons were reported after decreased atmospheric S, N, and acid deposition in forests of North America (Fraser, Bailey, & Ducey, 2019; Lawrence et al., 2015) and Austria

(Berger, Türtscher, Berger, & Lindebner, 2016). The Solling dataset (Meesenburg et al., 2016) comprises a long-term (1968–2019) record of climate, deposition, and soil chemistry data obtained for a Norway spruce (*Picea abies*) and a European beech (*Fagus sylvatica*) stand in Northern Germany. Scots pine (*Pinus sylvestris*) is the third key tree species in European forestry. Here, we present a comprehensive evaluation of data from two Scots pine sites in Southern Germany, covering a period from the early 1960s (stand nutrition, growth, and climate) and 1970s (topsoil chemistry) until 2019. More specifically, we address long-term effects of elevated S and N deposition in the 20th century after World War II, S deposition decrease after 1990, and recent climate warming on topsoil chemistry, stand nutrition, and stand growth.

2 | MATERIALS AND METHODS

2.1 | Study sites

The study was conducted in the stands *Pfaffenwinkel* and *Pustert* (S Germany). At the beginning of monitoring in 1963, both stands already were mature (age > 80 years) and characterized by poor quality and growth. Both sites had been degraded by long-term litter raking, forest pasture, and forest overexploitation. Important site and stand properties are presented in Table S1. In both stands, amelioration experiments were established in 1963 to identify feasible methods to increase stand productivity (Prietzel et al., 2008; Rehfuss & Schmidt, 1971). Every experiment included three randomly distributed control plots, 30 m × 40 m in size, which were separated from the fertilized plots by buffer areas. Fertilized and control plots were investigated intensively during the last 50 years. The investigation included repeated assessments of the chemical topsoil (forest floor + uppermost 30 cm mineral soil) status in 10-year intervals, (bi)annual Scots pine nutrition assessments by foliar analysis, and repeated stand inventories between 1963 and 2016.

The stand *Pfaffenwinkel* (50°00′26″N, 12°20′12″E; 535 m a.s.l.) has a cool, subcontinental climate. Parent material is intensively weathered phyllite covered by Pleistocene periglacial solifluidal debris. From this material acidic, nutrient-poor Dystric Cambisols with silt loam texture have developed. The soil (Table S2) is acidic down to >1 m depth; in the uppermost 70 cm BS is <25%. In 1964, the site was densely stocked (Table S1) with 86-year-old Scots pines of poor growth (site index IV.6 according to Wiedemann, 1943). The stand *Pustert* (49°19′44″N, 12°00′19″E; 480 m a.s.l.) has a warmer (Table S1) and less continental climate than *Pfaffenwinkel*. Parent material is Pleistocene solifluidal silt loam covering intensively weathered cretaceous sediments. The soil is a Stagnic Albeluviol with silt loam topsoil and clayey subsoil (Table S2); it shows seasonally impeded drainage and water logging. It is less deeply acidified than the soil at *Pfaffenwinkel*; subsoil (44–80 cm depth) BS is 50%. In 1963, *Pustert* was densely stocked (Table S1) with 81-year-old Scots pine of poor growth; however, with a site

index of III.8 according to Wiedemann (1943), stand growth was better than at *Pfaffenwinkel*.

2.2 | Soil inventories

On the three control plots at each site, we conducted soil inventories in 2014 and 2019, with the methods being identical with those used in earlier inventories performed in 1974, 1984, 1994, 2004 at *Pustert*, and in 1982, 1994, and 2004 at *Pfaffenwinkel* (Prietz et al., 1997, 2006). At each plot, we identified 20 sampling points located on the knot positions of an orthogonal grid net. At each point, we collected the entire forest floor material within a metal frame (size 20 cm × 20 cm). Then we sampled the mineral topsoil down to 30 cm depth, divided into 10 cm increments, with a stainless cylinder auger (inner diameter 5 cm). We pooled the sample material of five random points at each plot by depth increment. Thus, for each study site, 12 samples per depth increment originating from three replicate plots were available. We dried all samples to constant weight at 65°C and passed them through a 2 mm mesh sieve. We weighed the fine earth fraction <2 mm diameter for calculation of hectare-based soil masses and element stocks. From sieved samples, we measured the pH value with a glass electrode in 0.01 M CaCl₂ and in deionized water (soil:solution ratio 1:10 for forest floor, 1:2.5 for mineral soil samples). The measurements were performed under constant stirring after an equilibration time of 2 hr. Exchangeable cations and exchangeable H⁺ were analyzed after 2 hr batch extraction with 0.5 M NH₄Cl and subsequent filtration with 0.45 μm cellulose acetate filters (Schleicher & Schuell Comp.). Concentrations of Ca²⁺, Mg²⁺, K⁺, Na⁺, Al³⁺, Mn²⁺, and Fe²⁺ in the extracts were analyzed by ICP-OES (Varian Vista Pro). Exchangeable H⁺ was calculated from the pH value measured with a glass electrode, taking into consideration H⁺ produced by Al³⁺ hydrolysis. On ground subsamples, we determined concentrations of total C and N with a LECO analyzer CHN-2000. All samples were free of carbonate; therefore, total C equals organic C. Total P and S concentrations were analyzed by ICP-OES after digestion of ground subsamples with HF/HNO₃/HClO₄. Additionally, we determined the concentration of citric acid-soluble orthophosphate (P_{cit}), which is a good proxy for plant-available P (Fäth, Kohlpaintner, Blum, Göttlein, & Mellert, 2019; Prietz et al., 2010). This was done by extraction of sieved subsamples with 1% citric acid (soil:solution mass ratio 1:10; extraction time 23 hr) and analysis of orthophosphate concentrations in the extracts by colorimetry, using the method of Murphy and Riley (1962) as modified by John (1970). All samples were analyzed in duplicate.

We calculated stocks of organic C, total N, P, and S as well as of citric acid-extractable orthophosphate and exchangeable nutrient cations by multiplying the respective concentrations analyzed in the different inventories for each depth increment with the soil mass of the respective increment. Assuming mineral soil mass not to change between inventories, we used the arithmetic mean of dry soil masses retrieved in different inventories for the respective mineral soil depth increments. For calculation of O layer OC and nutrient stocks,

we used the respective O layer masses as determined specifically for each sample. Comparability of data obtained with different analytical methods in earlier phases of the 45-year monitoring period had been assured by Prietz et al. (1997). Additionally, archived soil samples were re-analyzed to confirm the consistency of analytical methods. Since most data were not normally distributed, we used the non-parametric Kruskal-Wallis H test, followed by a Nemenyi test for identification of statistical significances ($p < .05$) of differences between soil samples taken from a given horizon in different inventories. We performed all statistical computations with the software package SPSS 12.1. for Windows.

2.3 | Assessment of stand nutrition

From 1964 until 2019, we annually (*Pfaffenwinkel*) or bi-annually (*Pustert*) assessed the nutritional status of the pine stands by foliar analysis. We sampled current-year needles during winter dormancy from the uppermost crown of 12 dominant pine trees at each plot. All foliage samples were dried at 65°C. After determination of 100-needle pair masses, we ground all samples and determined the concentrations of N, S, P, K, Mg, and Ca. Until 1986, we analyzed foliar N concentrations using the Kjeldahl method, later with a Heraeus Macro N analyzer (1987–1990) and a LECO analyzer CHN-2000 (since 1991). Until 1991, we analyzed foliar P, Ca, Mg, and K concentrations after combustion at 450°C in a muffle oven and digestion of the residue in HCl by colorimetry (P: molybdenum-blue-method), with a flame emission spectrophotometer (Ca, K: Eppendorf ELEX 6361), or AAS (Mg; Perkin Elmer 380). From 1992 on, we subjected the samples to a 6 hr pressure digestion with concentrated HNO₃ at 160–180°C and determined concentrations of Ca, Mg, K, P, and S in the digests by ICP-OES (Perkin Elmer Optima 3000). We assured the comparability of different methods used during monitoring by analysis of reference samples.

2.4 | Assessment of stand growth

During the monitoring period, we conducted forest inventories in 3-year to 10-year intervals (in the following termed growth periods) at *Pfaffenwinkel* and 5-year to 10-year intervals at *Pustert* (Table S3). In each survey, diameter at breast height (DBH) was fully inventoried for all trees with a DBH > 7cm. Due to the general strong correlation between tree diameter and tree height, heights were sampled from approximately 30 trees evenly spread over the diameter distribution. Stand height curves (e.g., Petterson equation) were fitted to these data and used for estimating the missing tree heights. Tree volume was calculated using the volume equations for merchantable wood (stem volume <7 cm minimum diameter) by Franz et al. (1973) which require tree species, height, and diameter. Stand level volume (m³/ha) was scaled up from the tree volumes per plot. Annual volume increment at stand level was calculated from two subsequent surveys. We referenced all growth increment data to the expected growth

increment according to the yield table of Wiedemann (1943; moderate thinning variant) for Scots pine. Relationships between stand growth and climate, stand nutrition, and topsoil chemistry were investigated by correlation analysis after appropriate assignment of the growth data obtained in different stand inventories to the respective soil inventories (Table S4).

2.5 | Climate data

For both study sites, air temperature and precipitation data were acquired for the monitoring period from 1 km² grid cells provided by the German Meteorological Service (DWD) which were regionalized from appropriate data of nearby meteorological stations. The regionalization followed the routine described earlier (Mayer, Kudlinski, & Müller-Westermeier, 2003; Mayer & Müller Westermeier, 2010; Müller-Westermeier, 1995): Data from stations were reduced to a common reference level using spatially variable regression functions. The reduced values were interpolated by inverse distance weighting and subsequently re-conformed to values in the actual topography. For precipitation, instead of fields of regression coefficients, a background field consisting of interpolated ratios of actual monthly station precipitation to 30-year long-term reference precipitation was used to derive grid cell values of precipitation. Monthly data were aggregated to mean values for each calendar year. Growing season length at both sites in different years was calculated from mean annual air temperature (MAT) according to Chmielewski and Rötzer (2001).

2.6 | Atmospheric deposition estimation

Continuous atmospheric deposition monitoring in European forests with few exceptions (Germany: Solling sites; Meeseburg et al., 2016) started not earlier than in the 1990s, 30 years after commencement of our study, and our study sites were not part of the German long-term forest deposition monitoring program. However, atmospheric deposition was monitored 1985–1987 and in the entire year 2004 at *Pfaffenwinkel*, and in the entire year 2004 at *Pustert* in the course of short-term research programs. Moreover, deposition has been monitored continuously since 1991–1995 at two nearby long-term forest monitoring stations (*Flossenbuerg*, distance to *Pfaffenwinkel*: 27 km; *Altdorf*, distance to *Pustert*: 49 km). Detailed information on the sites and stands *Flossenbuerg* and *Altdorf* is presented in the Supporting Information (text; Table S5). We estimated the long-term development of atmospheric N and S deposition at our study sites from 1991–1995 to 2019 by assuming identical inter-annual deposition changes for the stands *Pfaffenwinkel* and *Flossenbuerg* and for *Pustert* and *Altdorf*, respectively. For estimation of atmospheric S deposition between 1970 and 1990, the long-term deposition dataset of the *Solling* Norway spruce stand (Meeseburg et al., 2016; Tiktak, Bredemeier, & van Heerden, 1995) was used. Deposition estimation details are presented in the Supporting Information (text; Table S5; Figure S1).

3 | RESULTS

3.1 | Topsoil chemistry

Topsoil pH values at both sites decreased in the period between the first soil inventory and the end of the 20th century (Figure 1a–p), but a reversal of this trend occurred afterwards. The reversal started in the O layer after 1994, and was delayed until 2004 with increasing soil depth. Similar to the pH, at both sites mineral soil BS decreased significantly between 1974–1982 and 2004, followed by a systematic, significant increase thereafter (Figure 2a–p). O layer BS at *Pfaffenwinkel* showed the same pattern, but differences between inventories were not statistically significant. In contrast, O layer BS at *Pustert* increased systematically between 1974 and 2019. At both sites, effective cation exchange capacity in the O layer decreased significantly during the monitoring period. No systematic change was present in the mineral topsoil, except for the 0–10 cm increment at *Pfaffenwinkel* (Figure 2d; Figure S2). O layer stocks of exchangeable Ca²⁺ (Ca_{ex}) at *Pfaffenwinkel* by trend decreased during the monitoring period, Mg_{ex} stocks showed no trend, and K_{ex} stocks increased significantly (Table 1). Mineral topsoil Ca_{ex} stocks also decreased between 1982 and 2019, Mg_{ex} and K_{ex} stocks decreased between 1982 and 2004, but increased later. In contrast to *Pfaffenwinkel*, O layer Ca_{ex}, Mg_{ex}, and K_{ex} stocks at *Pustert* increased during the monitoring period (Table 2). Mineral topsoil Ca_{ex} stocks showed no trend; Mg_{ex} and K_{ex} stocks decreased between 1984 and 2004 and increased thereafter.

At both study sites, SOC stocks in the O layer and in the mineral topsoil by trend increased in the period between the first soil inventory and 2004. However, the increases were not statistically significant due to considerable spatial SOC stock variability. Maximum SOC stocks in 2004 were followed by marked (but also statistically insignificant) SOC stock decreases during the most recent 15 years. Forest floor N stocks at both sites increased significantly between 1974–1982 and 2004, followed by an insignificant decrease in later years (Tables 1 and 2). Mineral topsoil N stocks showed no trend at *Pfaffenwinkel*, but were significantly elevated in the period 1984–2004 at *Pustert*. The significant forest floor C/N ratio decrease from 35 to 29 between 1982 and 2004 at *Pfaffenwinkel* and from 36 to 23 between 1974 and 2004 at *Pustert* did not continue during the most recent 15 years. Instead, O layer C/N ratios remained constant between 2004 and 2019 (29–30 at *Pfaffenwinkel*; Figure 3a; 23 at *Pustert*; Figure 3b), while both C and N concentrations decreased concomitantly (Figure S3).

At *Pfaffenwinkel*, topsoil S concentrations (Figure 4a,d,g,j) and stocks (Table 1) increased significantly (concentrations) between 1982 and 1994, but declined strongly during the last 25 years. In 2019, topsoil S concentrations were lower than in 1982, and topsoil S stocks were <20% of those in 1994. This trend was associated with significant S depletion of forest floor SOM; C/S ratios increased from <200 to almost 300 (Figure 3c). The same pattern of significant S concentration and stock increase until 1994, followed by significant decreases in subsequent years, was present

FIGURE 1 pH values measured in soil suspensions with deionized H₂O and with 0.01 M CaCl₂ for O layers and mineral topsoil (0–10, 10–20, 20–30 cm) at *Pfaffenwinkel* (a–h) and *Pustert* (i–p) in the period 1974–1982 to 2019. Shown are arithmetic mean values and standard errors of 12 replicate samples for each year. Significantly different ($p < .05$) mean values among years are indicated by different letters

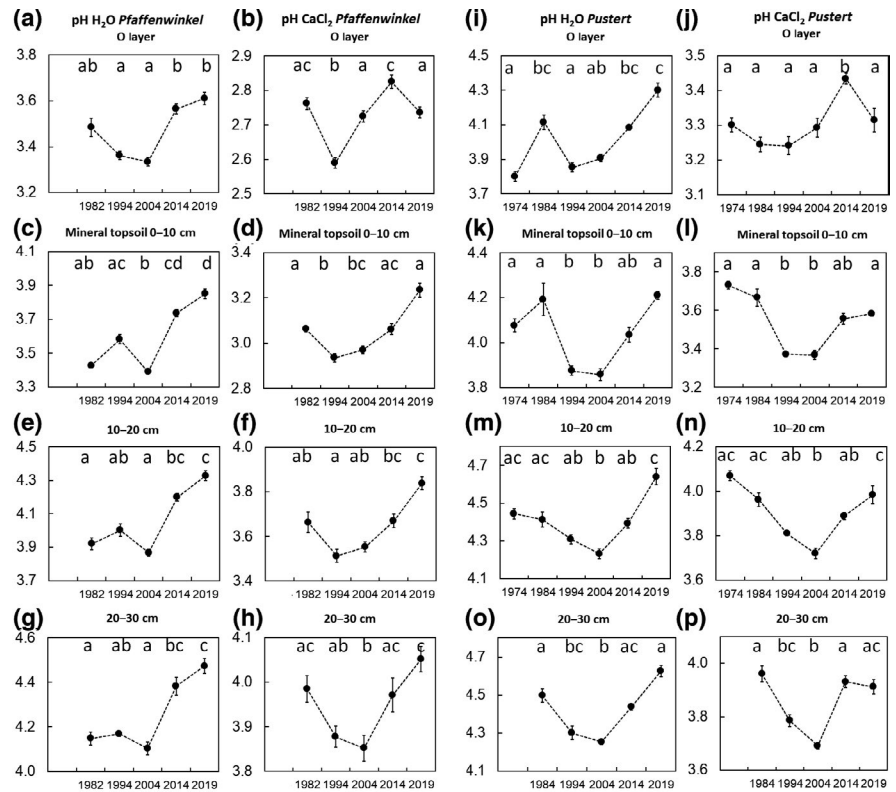
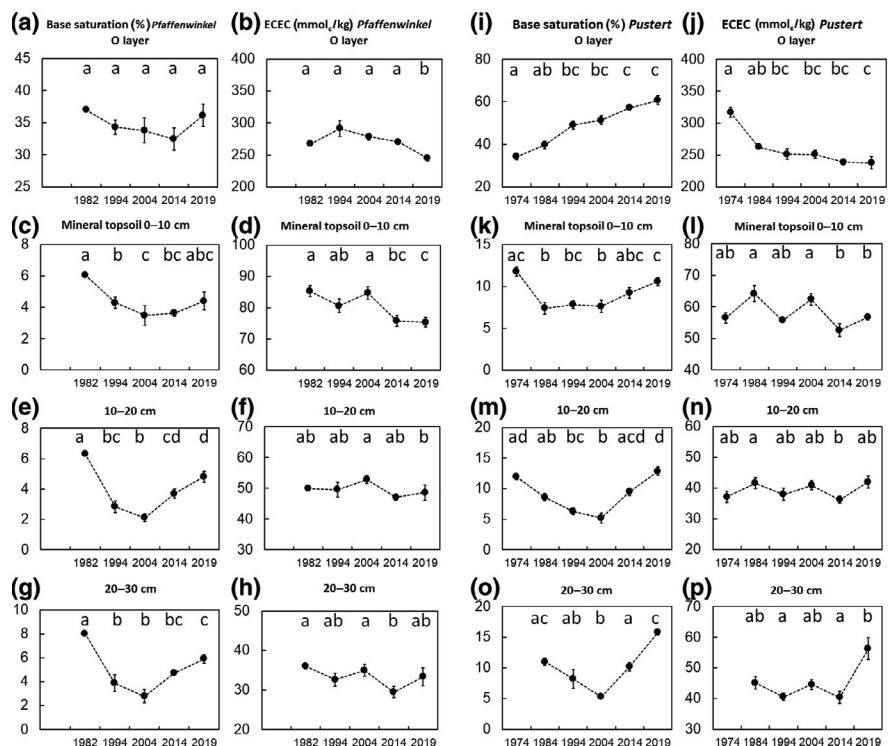


FIGURE 2 Base saturation and effective cation exchange capacity (ECEC) of O layers and mineral topsoil (0–10, 10–20, 20–30 cm) at *Pfaffenwinkel* (a–h) and *Pustert* (i–p) in the period 1974–1982 to 2019. Shown are arithmetic mean values and standard errors of 12 replicate samples for each year. Significantly different ($p < .05$) mean values among years are indicated by different letters



in the mineral topsoil of *Pustert* (Figure 5a,d,g,j; Table 2); however, forest floor S concentrations, stocks, and C/S ratios (Figure 3d) at *Pustert* did not change systematically. Topsoil P concentrations and stocks as well as the P content of forest floor SOM neither changed systematically at *Pfaffenwinkel* (Figures 3e and 4b,e,h,k; Table 1) nor at *Pustert* (Figures 3f and 5b,e,h,k; Table 2) during

the monitoring period. In contrast, concentrations and stocks of citric acid-extractable orthophosphate, which is a proxy variable for plant-available P (Prietzel & Stetter, 2010) at both sites (Figures 4c,f,i,l and 5c,f,i,l) decreased in the mineral topsoil and particularly in the forest floor. In 2019, topsoil stocks of citric acid-extractable orthophosphate were significantly decreased by

TABLE 1 Stocks (kg/ha), arithmetic mean values and standard errors (in brackets) of organic C, total N, exchangeable Ca, Mg, and K, as well as of total S, P, and citric acid-extractable orthophosphate P at Pfaffenwinkel

	Organic C	N	Ca _{ex}	Mg _{ex}	K _{ex}	S	P	P _{citric}
Forest floor								
1982	47,207 (973) a	1,331 (22) a	160 (27) ab	23.3 (0.3) ab	3.7 (3.4) a	262 (18) a	62 (2) ab	13.9 (0.3) a
1994	47,710 (2,200) a	1,438 (48) ab	184 (15) a	26.1 (1.5) a	17.3 (1.6) b	395 (50) a	53 (3) ac	9.6 (0.3) b
2004	48,565 (2,221) a	1,613 (81) b	155 (12) ab	26.0 (1.9) ab	19.3 (1.1) b	244 (13) a	66 (3) b	9.8 (0.5) b
2014	42,135 (2,081) a	1,445 (62) ab	120 (8) b	19.6 (1.2) b	28.0 (1.2) c	238 (11) a	50 (1) c	8.0 (0.3) b
2019	43,480 (1,159) a	1,453 (42) ab	126 (8) b	27.8 (1.6) a	36.4 (3.4) c	151 (3) b	67 (1) b	9.3 (0.5) b
Mineral topsoil (uppermost 30 cm)								
1982	44,757 (1,531) ab	2,504 (50) a	93 (14) a	36.3 (14.5) a	58.2 (4.9) a	1,010 (60) ac	1,941 (100) a	44.6 (5.6) a
1994	43,502 (1,943) ab	2,353 (66) ab	58 (7) b	15.3 (4.2) b	38.9 (3.5) ab	3,459 (392) b	1,505 (75) b	35.6 (5.3) ab
2004	51,637 (1,998) a	2,195 (67) b	67 (12) b	10.0 (1.4) b	24.3 (1.4) b	892 (44) ad	1,897 (80) a	40.1 (6.0) ab
2014	41,151 (1,545) b	2,391 (60) ab	31 (3) b	15.8 (0.7) b	58.6 (2.3) a	1,525 (68) bc	1,394 (49) b	28.8 (8.4) ab
2019	40,693 (1,102) b	2,366 (75) ab	29 (4) b	20.6 (2.1) ab	70.8 (4.4) a	519 (20) d	1,845 (16) a	23.6 (4.2) b
Forest floor + mineral topsoil								
1982	91,965 (597) a	3,835 (63) a	253 (30) ab	59.5 (4.2) a	61.9 (5.9) ab	1,273 (57) ac	2,003 (98) a	58.5 (5.7) a
1994	91,212 (3,589) a	3,790 (98) a	241 (17) a	41.4 (1.8) ab	56.1 (3.9) ab	3,853 (459) b	1,557 (74) b	45.2 (5.4) ab
2004	100,202 (3,815) a	3,850 (120) a	222 (16) ab	36.0 (2.5) ab	43.6 (1.8) a	1,136 (45) ad	1,964 (80) a	49.9 (6.1) ab
2014	90,286 (2,834) a	3,837 (86) a	151 (9) b	35.4 (1.4) b	86.5 (2.5) bc	1,763 (75) bc	1,444 (49) b	46.8 (8.3) ab
2019	81,827 (1,772) a	3,819 (91) a	155 (7) b	48.4 (2.7) a	107.2 (7.6) c	670 (20) d	1,912 (20) a	32.9 (4.3) b

Note: Stocks with different letters among years are statistically different ($p < .05$).

TABLE 2 Stocks (kg/ha); arithmetic mean values and standard errors (in brackets) of organic C, total N, exchangeable Ca, Mg, and K, total S, P, and citric acid-extractable orthophosphate P at Pustert

	Organic C	N	Ca _{ex}	Mg _{ex}	K _{ex}	S	P	P _{cit}
Forest floor								
1974	20,155 (925) ab	552 (25) a	76 (4) a	13.7 (2.0) a	10.2 (0.5) a	ND	ND	ND
1984	20,400 (1,262) ab	783 (43) ab	102 (8) ab	11.6 (0.9) a	12.3 (1.0) ab	71 (8) ab	37 (4) ac	7.4 (0.4) a
1994	22,070 (1,100) a	842 (39) bc	110 (6) bd	15.8 (0.9) ab	13.1 (0.5) ab	107 (17) ab	41 (2) ac	6.6 (1.3) a
2004	26,342 (1,441) a	1,143 (53) c	151 (9) c	23.2 (0.8) bc	14.7 (0.5) bc	106 (5) b	59 (3) b	7.9 (0.5) a
2014	16,429 (1,034) b	697 (39) ac	110 (8) bd	17.7 (4.3) bc	15.5 (1.2) bc	106 (8) b	34 (2) a	4.4 (0.3) b
2019	20,065 (1,502) ab	858 (63) bc	148 (15) cd	27.9 (1.2) c	21.1 (1.3) c	82 (6) ab	48 (3) bc	5.3 (0.6) ab
Mineral topsoil (uppermost 30 cm)								
1974	30,114 (1,086) a	1,197 (49) a	ND	ND	ND	ND	ND	ND
1984	29,150 (2,399) b	2,294 (179) bc	137 (10) a	43.0 (3.2) ac	56.8 (4.0) a	567 (96) a	755 (25) a	11.8 (1.2) a
1994	32,828 (1,158) a	1,990 (152) b	102 (10) ab	31.9 (1.7) ab	57.3 (1.0) ab	2,053 (178) b	698 (51) a	11.4 (1.2) a
2004	37,976 (2,059) a	1,992 (156) bc	116 (15) ab	22.3 (2.9) b	46.7 (1.8) a	529 (87) a	864 (86) a	9.8 (1.3) a
2014	33,456 (2,527) ab	1,680 (81) a	89 (12) b	38.5 (2.9) a	90.9 (5.3) bc	838 (130) ab	678 (67) a	12.5 (1.3) a
2019	27,401 (809) a	1,518 (60) b	112 (8) ab	59.0 (3.8) c	107.0 (4.3) c	423 (43) a	761 (17) a	7.5 (1.0) a
Forest floor + mineral topsoil								
1974	50,269 (2,466) ab	1,750 (72) ac	ND	ND	ND	ND	ND	ND
1984	49,550 (2,042) a	3,077 (156) bc	239 (14) ab	54.6 (3.2) a	69.1 (3.8) a	638 (99) a	792 (24) a	19.2 (1.2) a
1994	54,898 (1,541) b	2,832 (155) b	212 (13) ab	47.7 (2.0) a	70.4 (1.0) a	2,159 (209) b	739 (52) a	17.7 (1.4) ab
2004	64,317 (1,844) b	3,135 (135) b	267 (15) a	45.5 (2.8) a	61.4 (1.9) a	634 (85) ab	924 (85) a	17.8 (1.3) ab
2014	49,884 (2,763) ac	2,377 (93) c	199 (16) b	56.2 (3.6) a	106.4 (5.8) b	944 (130) ab	713 (68) a	16.9 (1.5) ab
2019	47,467 (2,120) bc	2,376 (94) b	261 (17) ab	86.9 (3.7) b	128.1 (4.7) b	505 (44) a	809 (17) a	12.8 (0.9) b

Note: Stocks with different letters among years are statistically different ($p < .05$).

Abbreviation: ND, no data.

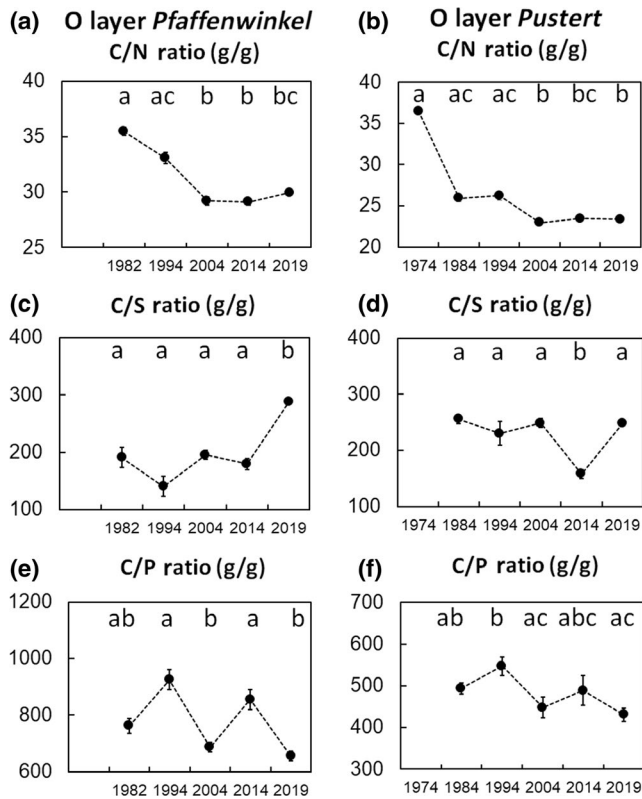


FIGURE 3 Forest floor C/N (a, b), C/S (c, d), and C/P ratios (e, f) at *Pfaffenwinkel* and *Pustert* in the period 1974–1982 to 2019. Shown are arithmetic mean value and standard error of 12 replicate samples for each year. Significantly different ($p < .05$) mean values among years are indicated by different letters

33% at *Pustert* and 44% at *Pfaffenwinkel* compared to 1982–1984 (Tables 1 and 2).

3.2 | Stand nutrition

At both sites, Ca concentrations in half-year-old Scots pine foliage decreased between 1964 and 2004, followed by an increasing trend in later years (Figures 6a and 7a). Whereas the Ca nutritional status of the pines was good throughout the monitoring period at *Pustert*, the initially good Ca supply of the pines at *Pfaffenwinkel* changed into a status of latent insufficiency according to the foliar concentration threshold values of Göttelein (2015) in the late 1970s, and remained insufficient until the end of monitoring. Similar to Ca, also pine Mg nutrition was sufficient throughout monitoring at *Pustert*, even though we observed a trend of deterioration (Figure 7c). In contrast, at *Pfaffenwinkel* foliar Mg concentrations indicated a general status of latent Mg insufficiency without marked long-term trend (Figure 6c). Foliar K concentrations indicated a good K supply during the first 30 years of monitoring at both sites (Figures 6e and 7e), but decreased markedly after 1995, resulting in latent K deficiency of the pines at *Pustert* after 2006.

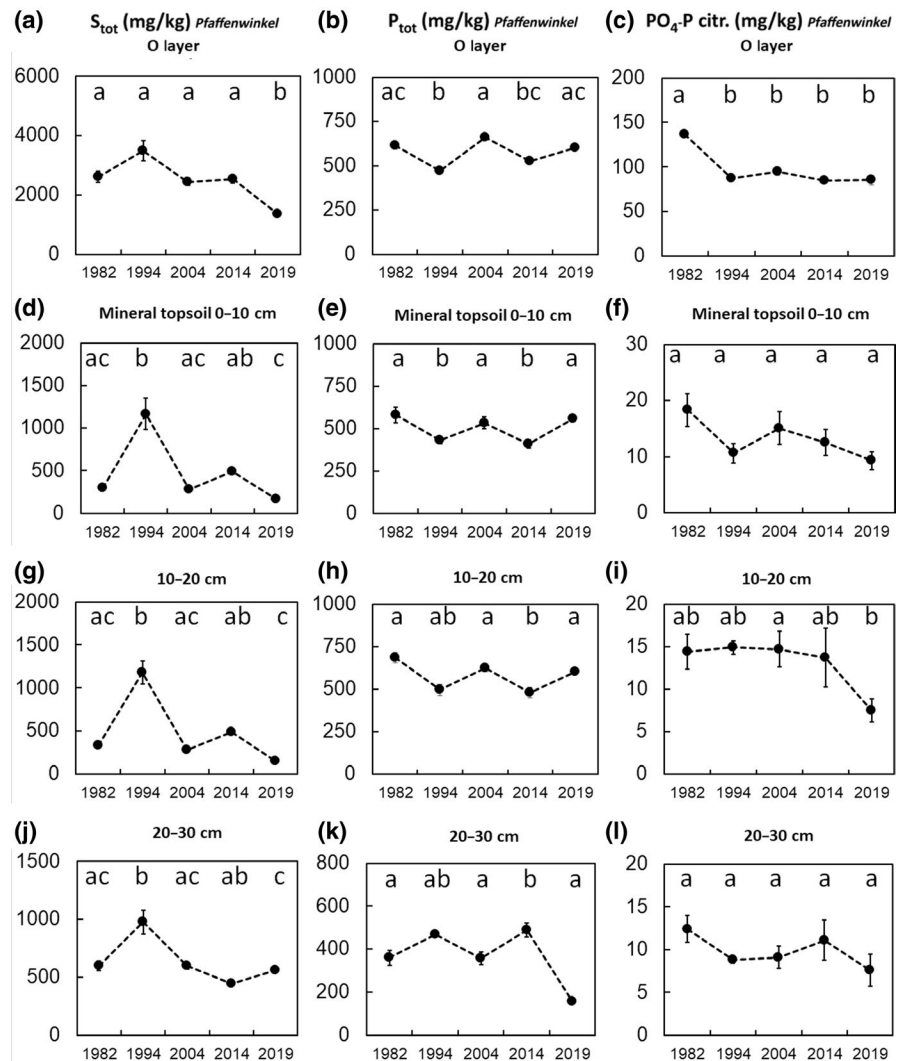
At the beginning of monitoring, the pines at both study sites showed latent N deficiency (Figures 6b and 7b). Starting in the

1970s, pine foliar N concentrations at both sites increased considerably and N deficiency disappeared at the beginning of the 1980s. At *Pfaffenwinkel*, a peak foliar N concentration of 20 mg/g was measured in 1990, followed by a concentration decrease and re-establishment of an insufficient N nutrition status 10 years later. At *Pustert*, N concentrations in pine foliage showed an increasing trend throughout the late 20th century; yet, as at *Pfaffenwinkel*, a trend of decreasing foliar N concentrations was present during the last 20 years. The development of pine foliar P concentrations (Figures 6d and 7d) differed between both sites during the first part of the monitoring period, but was similar during the second part. In the 1960s, both stands showed a sufficient P supply. However, foliar P concentrations of the pines at *Pustert* developed an accelerating trend of P decrease, indicated latent P deficiency after the year 2000, and a status close to acute P deficiency during the most recent 10 years of monitoring. At *Pfaffenwinkel*, foliar P concentrations increased until 1990, followed by a marked trend of decrease thereafter. Similar to *Pustert*, also the pines at *Pfaffenwinkel* showed insufficient P nutrition during the last years of monitoring. Sulfur concentration analysis in pine foliage started in 1994 at *Pfaffenwinkel* and in 2000 at *Pustert*. Foliar S concentrations of the pines at both sites (Figures 6f and 7f) decreased markedly during the recent two decades, indicating a status of insufficient S nutrition according to Göttelein and Mellert (2019) after 2000 and 2015 at *Pfaffenwinkel* and *Pustert*, respectively. According to their foliar nutrient ratios (Figure 7g–k), the pines at *Pustert* systematically approached or reached a status of imbalanced nutrition, characterized by excess supply with N relative to S, Mg, K, and, most pronouncedly, P. During the most recent 10–15 years, the imbalance decreased for the base cations, but not for P and S. At *Pfaffenwinkel* (Figure 6g–k), N/base cation ratios in pine foliage showed different trends for Ca (increase until 1990; decrease thereafter), Mg (no trend), and K (modest long-term trend of increase). However, in contrast to *Pustert*, the range of balanced nutrition was never or seldom (N/Mg during early monitoring) exceeded. Similar to *Pustert*, foliar N/P and N/S ratios of the pines at *Pfaffenwinkel* showed a long-term trend of increase, yet at the end of monitoring, N/P and N/S nutrition still were balanced according to the threshold values of Göttelein (2015). At both sites, average mass of current-year pine needles increased by 100% between 1964 and 1990 and decreased again thereafter (Figures 6e and 7e). In 2004, foliage masses were similar to those in the early 1960s.

3.3 | Stand growth

At *Pfaffenwinkel*, annual Scots pine growth at the beginning of monitoring was low ($<4 \text{ m}^3 \text{ merchantable wood ha}^{-1} \text{ year}^{-1}$ in 1964–1966; Figure 8a). Growth doubled to $8 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ in 1967–1969, and remained in that range until 1983. Growth further accelerated to a maximum rate of $10 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ in 1984–1988 and continuously decreased thereafter to $8 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ in the 21st century. However, consideration of the typical growth decrease in Scots pine trees with increasing age (black lines in Figure 8a,b) in the analysis

FIGURE 4 Concentrations of total S (a, d, g, j), total P (b, e, h, k), and citric acid-extractable orthophosphate-P (c, f, i, l) in O layers and mineral topsoil (0–10, 10–20, 20–30 cm) at *Pfaffenwinkel* in the period 1982–1984 to 2019. Shown are arithmetic mean value and standard error of 12 replicate samples for each year. Significantly different ($p < .05$) mean values among years are indicated by different letters



(“age-referenced growth”; Figure 8c) indicates that site conditions at *Pfaffenwinkel* have become increasingly favorable for pine growth during the entire monitoring period until the last stand inventory in 2016. At *Pustert* (Figure 8b), pine growth increased from 6 m^3 merchantable wood $\text{ha}^{-1} \text{ year}^{-1}$ at the beginning of monitoring to almost $10 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ in the period 1984–1988. The increase was followed by a continuous growth decrease in later years, and pine growth at *Pustert* in the 21st century was as low as before 1980. However, age-referenced growth suggests that site conditions at *Pustert* have remained favorable until the last stand inventory in 2013 (Figure 8d).

3.4 | Climate variables

At both sites, MAT increased during the monitoring period (Figure 9a,b; Table 3). The increase rate was small ($<0.1 \text{ K/decade}$) in the period 1960–1985, larger ($0.3\text{--}0.4 \text{ K/decade}$) in the period 1985–2010, and very large ($2.1\text{--}2.2 \text{ K/decade}$) in the period 2011–2019. Mean growing season length increased by 10 days (Table 3), in line with results reported by Menzel and Fabian (1999). Mean annual

precipitation (MAP) between 1960 and 2010 showed a slightly increasing trend (20 mm/decade), which was not significant due to large inter-annual variation (Figure 9c,d). However, it decreased considerably after 2010, and on average was 10% smaller in the period 2011–2019 compared to the period 1960–2010. Simultaneous MAT increase and MAP decrease at our sites resulted in deteriorated water availability to forest trees and decreased climatic water balance (CWB; Table 3; Figure 9e,f), which in contrast to the period 1991–2010 (single drought year 2003) often were negative in most recent years (2015, 2018, and 2019).

3.5 | Atmospheric S and N deposition

At both sites, atmospheric S deposition (Figure 10a) culminated in the late 1970s and early 1980s at levels of $60 \text{ S kg ha}^{-1} \text{ year}^{-1}$, and strongly decreased thereafter to $<20 \text{ kg S ha}^{-1} \text{ year}^{-1}$ at the turn of the century and $<10 \text{ kg S ha}^{-1} \text{ year}^{-1}$ at the end of monitoring. Atmospheric N deposition (Figure 10b) after 1991–1995 without trend fluctuated around $30 \text{ kg ha}^{-1} \text{ year}^{-1}$ at *Pustert* and $25 \text{ kg ha}^{-1} \text{ year}^{-1}$ at *Pfaffenwinkel*. N deposition estimates before

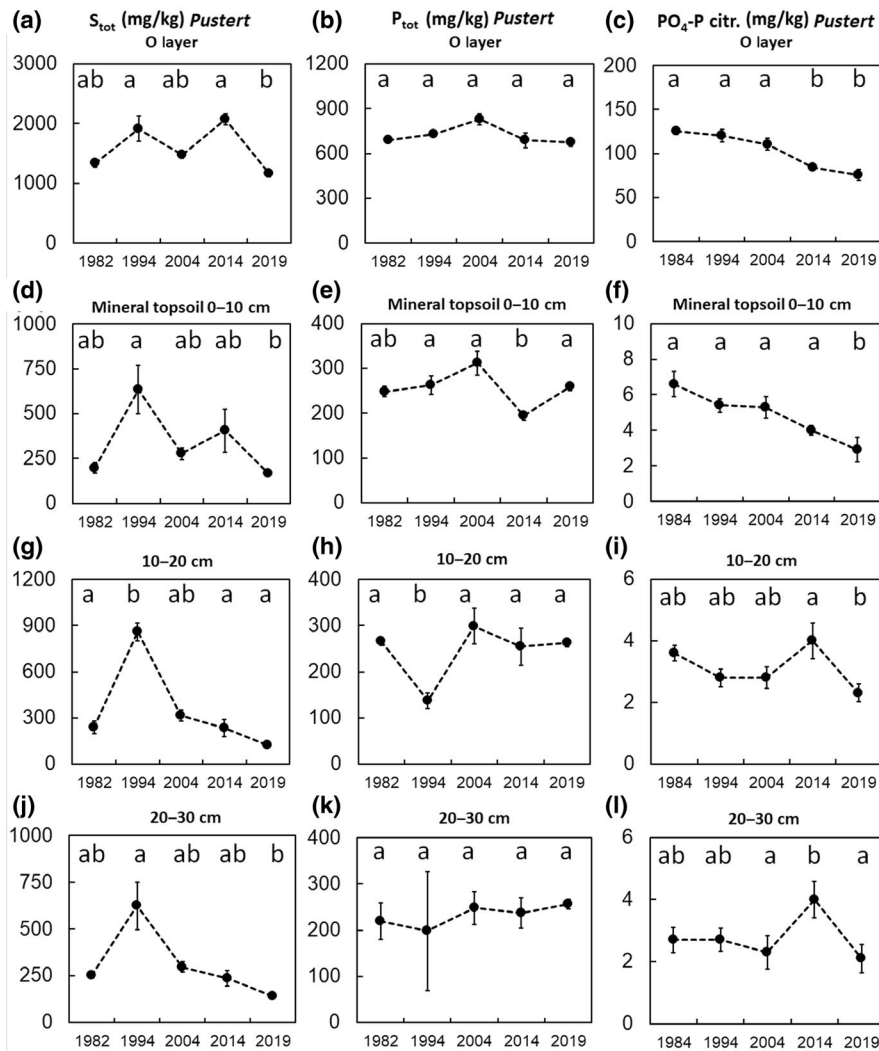


FIGURE 5 Concentrations of total S (a, d, g, j), total P (b, e, h, k), and citric acid-extractable orthophosphate-P (c, f, i, l) in O layers and mineral topsoil (0–10, 10–20, 20–30 cm) at Pustert in the period 1982–1984 to 2019. Shown are arithmetic mean value and standard error of 12 replicate samples for each year. Significantly different ($p < .05$) mean values among years are indicated by different letters

1991–1995 must be viewed with caution; yet, it can be assumed that N deposition levels at both sites in the 1960s were in the typical range of pristine European forests with low N deposition ($<10 \text{ N kg ha}^{-1} \text{ year}^{-1}$; Schmitz et al., 2019). At Pfaffenwinkel N deposition probably steadily increased in the 1980s and 1990s, whereas at Pustert a rapid increase to present levels can be assumed after establishment of a large chicken farm in 1970 as local N emitter (distance to stand: 1 km).

4 | DISCUSSION

4.1 | The beginning of the new millennium: A turning point of biogeochemical processes in Central European forests

At both study sites, the onset of the new millennium was a turning point for several topsoil parameters. Topsoil pH, BS, and stocks of exchangeable K⁺ and Mg²⁺, which had decreased significantly during the last decades of the 20th century, reached minimum values in the soil inventory 2004 (O layer pH: 1994), and re-increased thereafter.

The turnaround from topsoil acidification to re-alkalinization has been observed in several European and North American forests with decreasing atmospheric N and S deposition (Berger et al., 2016; Cools & De Vos, 2011; Fraser et al., 2019; Lawrence et al., 2015; Schmitz et al., 2019). It seems to be a straightforward effect of decreased atmospheric S and H⁺ deposition. Weathering of primary soil minerals, together with eolic base cation deposition and base cation uplift (“base-pumping”) by trees (Clarholm & Skjellberg, 2013), obviously have overcompensated atmospheric acid input after 2004, resulting in increasing topsoil pH, BS, and base cation stocks.

Topsoil C and N stocks showed an inverse pattern. They increased during the last decades of the 20th century, reached peak values in 2004, and decreased thereafter. As indicated by decreasing C/N ratios, forest floor SOM became progressively enriched in N during the last decades of the 20th century; however, this trend did not continue in the 21st century. Even though the measured SOM stock changes were not statistically significant due to their large spatial variation (Saby et al., 2008), the systematic SOM stock increase during the last quarter of the 20th century as well as the systematic decrease between 2004 and 2019 can be assumed to represent real trends (Amrhein, Greenland, & McShane, 2019). SOM dynamics at

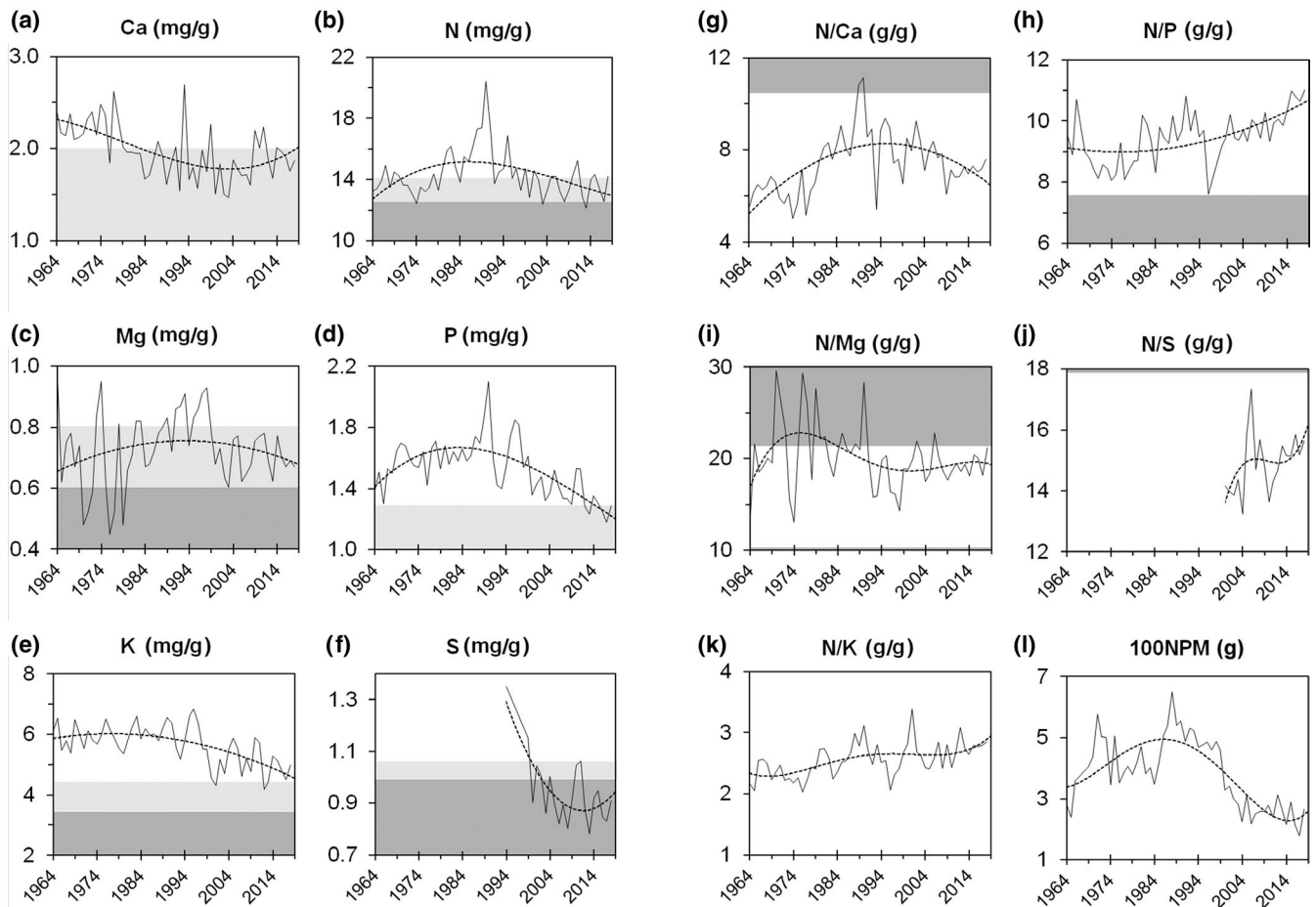


FIGURE 6 Foliar Ca, Mg, K, N, P, and S concentrations (a–f) as well as N/Ca, N/Mg, N/K, N/P, N/S ratios, and mass of 100-needle pairs (g–l) obtained for current-year foliage of dominating and co-dominating *Pinus sylvestris* trees at Pfaffenwinkel (pooled from 12 trees). Gray and dark gray areas in left panels indicate latent and manifest nutrient deficiency, respectively, according to the threshold values defined by Göttlein (2015). Dark gray areas in right panels indicate disharmonic nutrition according to the threshold values defined by Göttlein (2016) and Göttlein and Mellert (2019). Trend equations provided in Table S6

both sites during monitoring likely were driven by four major factors, whose relative influence changed between 1974 and 2019: (a) Recovery from litter raking and other historic SOM-depleting forest utilization practices (Prietzel et al., 2006). Together with elevated atmospheric N deposition and prolonged growing seasons, the recovery resulted in considerable forest growth acceleration (Mellert et al., 2004; Prietzel et al., 2008), which was larger than that observed for spruce and beech stands at more fertile sites in S Germany (Pretzsch et al., 2014), as well as in increased tree foliage masses (Prietzel et al., 2006), and probably also in (b) increased litter input to the soil. (c) Deceleration of litter and SOM decomposition due to progressive soil acidification and N fertilization (Berg & Matzner, 1997; Greaver et al., 2016; Oulehle et al., 2011) during the late 20th century. (d) Acceleration of SOM decomposition caused by topsoil re-alkalinization after 2004 similar to that reported for the nearby Czech Norway spruce forest Nacetin (50.59°N, 13.26°E; 784 m a.s.l.; soil: Dystric Cambisol) by Oulehle et al. (2011), and reinforced (Greaver et al., 2016) by markedly increasing air temperatures after 1990. Overlapping effects of site recovery from historic degradation, deposition change, and climate warming well explain the

temporal topsoil SOM stock changes at both sites, exhibiting peak values in 2004. Site-specific overlapping effects of different drivers may also explain the controversial results of SOC stock inventories performed to quantify climate change effects on SOC in European forests (Bellamy et al., 2005; Callesen et al., 2015; Grüneberg et al., 2014; Jonard et al., 2017; Prietzel et al., 2016). At our sites, the SOM loss after 2004 may have contributed to topsoil re-alkalinization in an opposite way as SOM accrual results in soil acidification (van Breemen, Driscoll, & Mulder, 1984).

The delay of topsoil re-alkalinization in the mineral soil compared to the onset of S deposition decrease can be explained by remobilization (Alewell & Matzner, 1993; Prietzel & Kölling, 1999) of sulfur stored in the soils during previous times of high atmospheric S deposition (Prietzel, Mayer, Krouse, Rehfues, & Fritz, 1995; Prietzel, Mayer, & Legge, 2004). At both sites, topsoil S concentrations and stocks were highest in the 1994 inventory, and markedly decreased afterwards. Remobilized S is mostly present as sulfate, which is a mobile anion and leads so soil acidification in the same way as $\text{SO}_4\text{-S}$ entering the soil by atmospheric deposition, thus delaying soil re-alkalinization. With progressive depletion of labile soil S pools after S

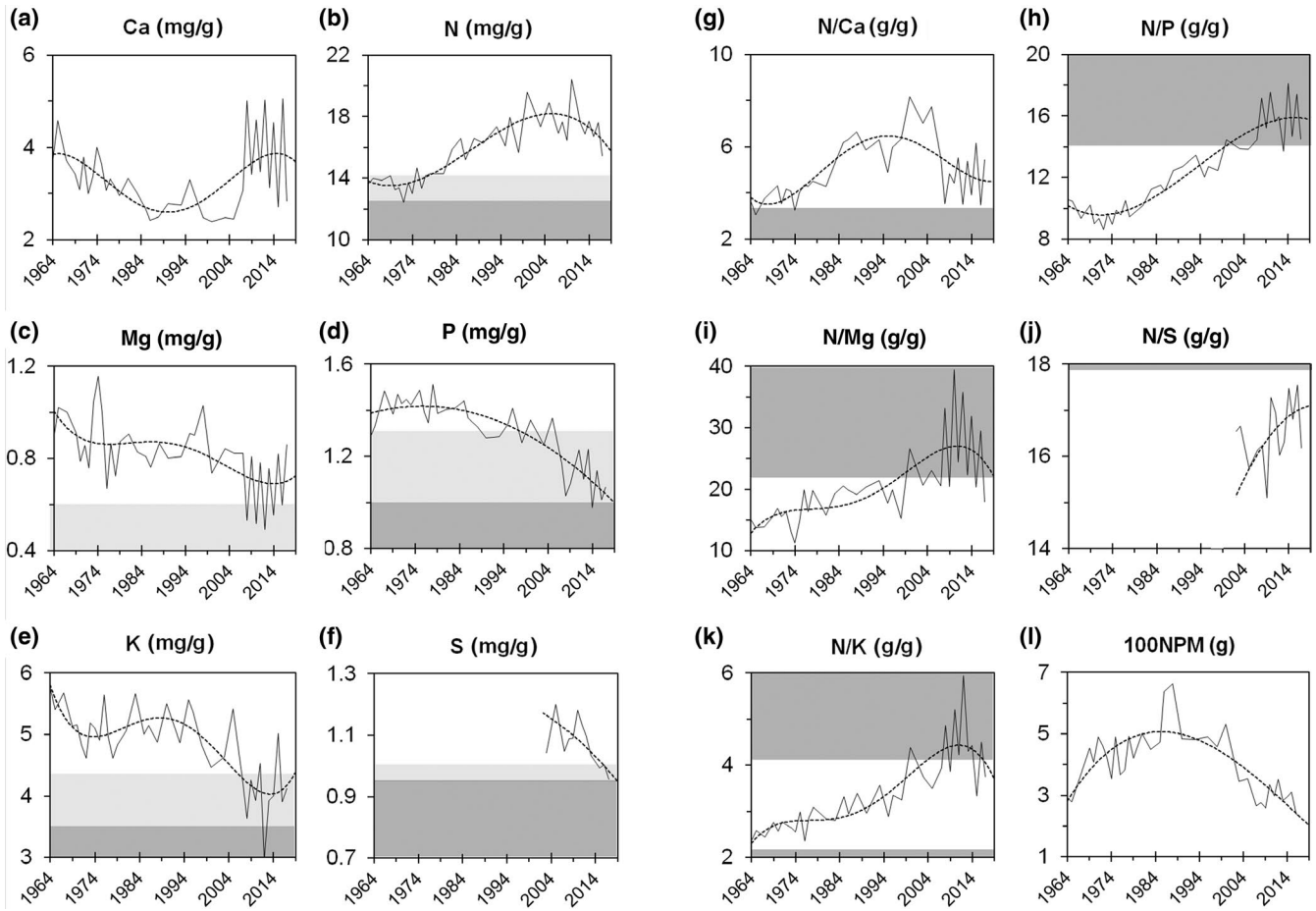


FIGURE 7 Foliar Ca, Mg, K, N, P, and S concentrations (a–f) as well as N/Ca, N/Mg, N/K, N/P, N/S ratios, and mass of 100-needle pairs (g–l) obtained for current-year foliage of dominating and co-dominating *Pinus sylvestris* trees at Pustert (pooled samples from 12 trees). For an explanation of gray and dark gray areas in panels, see Figure 6. Trend equations provided in Table S6

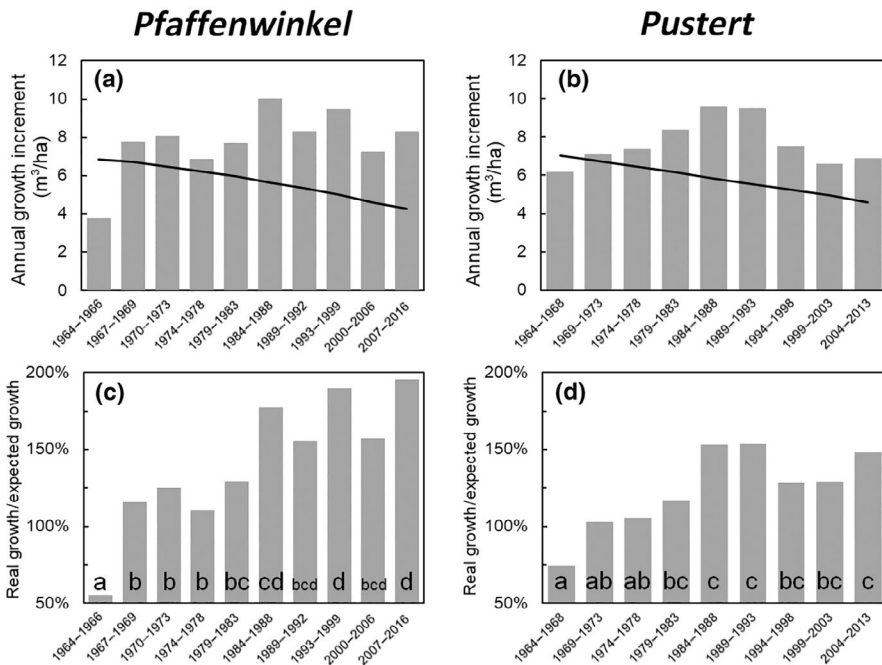


FIGURE 8 Change of mean annual increment of merchantable Scots pine wood at Pfaffenwinkel (a, c) and Pustert (b, d) during the monitoring period. Black lines represent expected age-referenced growth according to Wiedemann (1943; moderate thinning, yield class I.0). X-axis labels represent intervals between two stand inventories (growth periods). Different letters in bars indicate significant ($p < .05$) differences between mean age-referenced growth increments

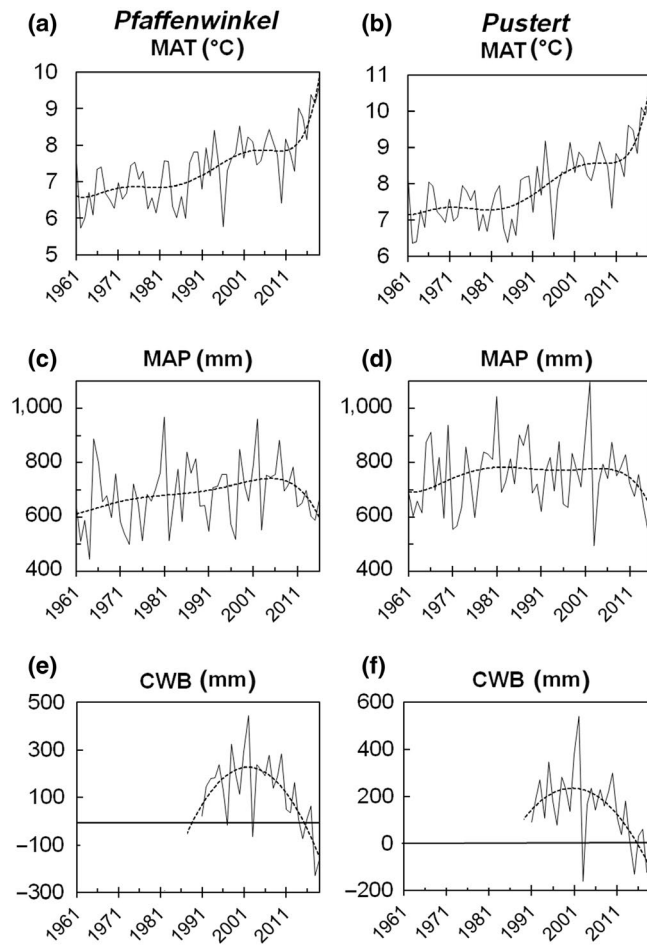


FIGURE 9 Mean annual air temperature (MAT; a, b), mean annual precipitation (MAP; c, d), and climatic water balance (CWB; e, f) at *Pfaffenwinkel* (left panels) and *Pustert* (right panels) in the period 1961–2019 (CWB: 1991–2019). Trend equations provided in Table S6

TABLE 3 Recent changes of climate parameters at the study sites

	Mean annual air temperature (°C)	Mean annual precipitation (mm/year)	Mean annual evapotranspiration (mm/year)	Mean climatic water balance (mm/year)	Growing season ^a (days)
<i>Pfaffenwinkel</i>					
1961–1970	6.63	658			175
1971–1980	6.85	631			177
1981–1990	7.00	718			177
1991–2000	7.47	680	529	151	180
2001–2010	7.77	765	544	221	181
2011–2019	8.59	618	633	–15	185
<i>Pustert</i>					
1961–1970	7.23	742			179
1971–1980	7.34	720			179
1981–1990	7.44	812			180
1991–2000	8.07	738	547	191	183
2001–2010	8.45	801	581	221	185
2011–2019	9.33	669	664	5	189

^aCalculated from MAT according to Chmielewski and Rötzer (2001). Trend equations provided in Table S6.

deposition reduction, acidifying soil S remobilization will decrease and ultimately disappear. At the end of monitoring, the pines at both sites showed dramatically reduced foliar S concentrations and were characterized by S deficiency according to Göttlein and Mellert (2019) despite high atmospheric S deposition and pronounced topsoil S accumulation just three decades before. Similar patterns were reported by Talkner et al. (2019) and Göttlein and Mellert (2019) for other German forests and by Jonard et al. (2015) for European forests, and were explained by increased tree S uptake associated with the recent forest growth acceleration (Pretzsch et al., 2014).

4.2 | Topsoil chemistry is less important for forest base cation compared to S and P nutrition

Similar to S, also the P nutritional status of the pines at our study sites deteriorated markedly during the monitoring period and ultimately reached a status of insufficient P supply. While topsoil concentrations and stocks of total P did not change, and forest floor C/P ratios rather narrowed than widened during the monitoring period, topsoil concentrations and stocks of citric acid-extractable orthophosphate decreased significantly at both sites. They reached low values particularly at *Pustert*, where pine P deficiency was more pronounced than at *Pfaffenwinkel*. Similar to results reported by Mellert, Grüneberg, Weis, and Göttlein (2017) and Fäth et al. (2019) for key forest tree species in Central Europe, foliar P concentrations of the pines at our sites were highly correlated with the stocks of citric acid-extractable orthophosphate in the uppermost 10 cm of the soil (Table S7). Temporal patterns of stand S and P nutrition thus well reflected temporal patterns of topsoil S and citric acid-extractable P concentrations and stocks. Tree P and S nutrition obviously are governed by uptake of these elements from topsoil rather than

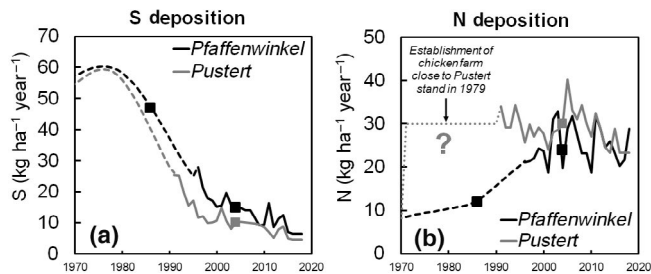


FIGURE 10 Estimated and measured (square symbols) atmospheric deposition of S (a) and N (b) at Pfaffenwinkel and Pustert in the period 1970–2019

subsoil horizons. Concerning P, this is a typical feature of forests with P-poor soils (“P-recycling systems”; Lang et al., 2016).

Despite increasing topsoil BS and exchangeable Mg^{2+} and K^+ stocks at both sites, the Mg and K supply to the pines according to their foliage concentrations has not improved during the most recent 15 years of monitoring. Obviously, topsoil concentrations and stocks of exchangeable K^+ and Mg^{2+} are poor predictors of tree K and Mg nutrition (Table S7), if not supplemented by additional information, for example, concerning subsoil K^+ and Mg^{2+} . At our sites, recent mobilization and seepage water export of sulfate that had accumulated in the subsoil during high S deposition periods (Alewell & Matzner, 1993) as well as tree K and Mg uptake (Jonard et al., 2015; van Breemen et al., 1984) likely have resulted in subsoil acidification and depletion of plant-available K^+ and Mg^{2+} during recent years (Cools & De Vos, 2011; Lawrence et al., 2015). According to our foliage nutrient data, which similar to other studies (e.g., Jonard et al., 2012, 2015) indicate a decreasing tree supply of K and Mg, recent subsoil exchangeable K^+ and Mg^{2+} depletion obviously has exceeded topsoil gains.

4.3 | Phosphorus and sulfur insufficiency as well as climate warming have replaced atmospheric S deposition as major challenge for Central European forests in the new millennium

Our results indicate an emergence (S) or aggravation (P) of insufficient forest P and S nutrition, which have become the most deficient nutrients for the pines at both study sites. Particularly at Pustert, foliar N/P and N/S ratios have left or are leaving the range of balanced nutrition according to Göttlein (2016) and Göttlein and Mellert (2019). Furthermore, foliar N/P ratios >7.8 indicate increased tree susceptibility to defoliation by insect infestation or drought (Veresoglou et al., 2014). Similar recent S and P nutrition changes were observed for other forests in Central Europe (Göttlein & Mellert, 2019; Jonard et al., 2015). The deteriorating P nutrition—a “progressive P limitation” comparable to the “progressive N limitation” hypothesized by Luo et al. (2004)—may be attributed to several factors: (a) Increased tree P uptake associated with recent forest growth acceleration due to an increased availability of N and atmospheric CO_2 (Peñuelas et al., 2013; Peñuelas, Sardans, & Rivas-Ubach, 2012). (b) Increased P export

with wood harvesting in faster growing stands (Jonard et al., 2015). (c) Decreased soil P availability due to soil acidification (Mohren, van den Burg, & Burger, 1986). (d) Decreased mass and/or P uptake activity of roots and mycorrhiza fungi (Arnebrant & Söderström, 1992; Wallander, 2000). The interaction of these factors at present still is poorly understood and just recently (Lang et al., 2016) came into the focus of forest research. Recent foliar C/N and C/P ratio increases and N and P concentration decreases for the pines at our sites may also have been caused by a positive CO_2 fertilization effect on tree N and P use efficiency (Myers et al., 2014). Indeed, high atmospheric N deposition and constant or decreasing forest floor C/N ratios at our sites during recent decades argue against a decreased soil N supply to the pines, even though negative climate warming effects on soil N mineralization, which has not been investigated in our study, may compromise tree N supply. In contrast, decreasing topsoil concentrations and stocks of plant-available P during the monitoring period suggest that depletion of plant-available soil P is a major reason for the observed changes of foliar P concentrations and C/P ratios at both sites, even though increased pine P use efficiency due to CO_2 fertilization may be an additional factor. Jonard et al. (2015) hypothesized a deterioration of the S (P, base cation) nutritional status in European forests with sustained productivity increases due to recent global changes. Nevertheless, the observation that forests on soils with large amounts of accumulated deposition S may become S-deficient within three only decades after the S deposition peak (Göttlein & Mellert, 2019)—even though S deficiency levels are hard to define—at this moment is far from being understood. Previous studies (e.g., Prietzel & Kölling, 1999) predicted sustained remobilization of S accumulated in forest soils during times of elevated S deposition after deposition decrease, which should prevent insufficient tree S nutrition for many decades. The unexpected rapid emergence of S deficiency in European forests according to current threshold values requires scientific action to understand these phenomena. Moreover, established foliar nutrient concentration threshold values may have to be re-appraised in times of changing tree nutrient use efficiencies associated with global change (e.g., of atmospheric CO_2 concentration; Myers et al., 2014).

Compared to the first part of the monitoring period, the first and second decade of the 21st century were characterized by a progressively increasing MAT at both study sites, resulting in decreased CWB, which may negatively affect tree vitality. While Scots pine is considered one of the tree species in Europe with least water demand and traditionally was characterized as well adapted to drought, it is—among the Pinaceae—particularly vulnerable to xylem embolism under drought conditions (Taeger, Zang, Liesebach, Schneck, & Menzel, 2013). Recent studies (Bigler, Bräker, Bugmann, Dobbertin, & Rigling, 2006; Galiano, Martinez-Vilalta, & Lloret, 2011) showed that increasing air temperatures and multi-year droughts during recent years were associated with increased Scots pine defoliation and mortality in various regions of Europe. According to Taeger et al. (2013), Scots pine sensitivity toward drought differs among provenances, and until now, we have not noticed increased pine defoliation or dieback at our study sites. However, recent Scots pine dieback has been observed in Switzerland (Hartmann, 2011) and also in Germany at

distances <80 km from *Pustert* (Buras et al., 2018). Scots pine needle masses at our sites in different sampling years correlated negatively with CWB; however, CWB explained only 3% and 14% of total needle mass variance at *Pfaffenwinkel* and *Pustert*, respectively. Moreover, age-referenced pine growth at both sites remained at a high level until the last growth inventories. This indicates that water shortage caused by combination of increased MAT and prolonged draughts does not yet seriously compromise Scots pine vitality at our sites. Decreasing stand growth/evapotranspiration ratios between 1991

and 2019 at both sites argue against an increased pine water use efficiency caused by elevated atmospheric CO₂ concentration (Keenan et al., 2013). Yet, the concomitant marked decline of CWB and pine needle mass at both sites is intriguing, and the poor correlation of CWB with needle mass in the same year might be caused by lagged effects of tree water supply on tree physiology, as needles are produced in spring, whereas draughts affecting the annual CWB often occur during later seasons. A detailed evaluation of soil moisture regimes at our sites based on retrospective hydrological modeling

TABLE 4 Correlation (Pearson coefficients) between age-referenced Scots pine growth (mean annual growth increments of merchantable wood compared to respective values denoted for yield class 1.0 in yield table of Wiedemann, 1943; moderate thinning) in the Scots pine stands *Pfaffenwinkel* (period 1979–2016) and *Pustert* (period 1969–2013) and climate, stand nutrition, and soil parameters

	Age-referenced Scots pine growth			
	<i>Pfaffenwinkel</i>	<i>Pfaffenwinkel</i> after 1983	<i>Pustert</i>	<i>Pustert</i> after 1983
<i>Growth periods with data</i>	4	3	5	4
Calendar year	0.858	0.577	0.759	-0.333
Mean annual air temperature	0.775	0.165	0.442	-0.745
Mean annual precipitation	-0.500	-0.593	0.413	0.206
Foliar nutrient concentrations				
N	-0.538	-0.006	0.738	-0.900
P	-0.724	-0.483	-0.740	0.037
K	-0.806	-0.518	-0.361	-0.070
Ca	0.024	0.429	-0.305	0.650
Mg	-0.347	-0.168	-0.936	-0.405
Foliar nutrient amounts				
100-needle pair mass	-0.364	-0.147	-0.208	0.229
N	-0.402	-0.125	0.165	0.051
P	-0.440	-0.199	-0.406	0.221
K	-0.476	-0.200	-0.224	0.180
Ca	-0.283	-0.059	-0.424	0.526
Mg	-0.171	-0.145	-0.615	0.090
C/N ratio O layer	-0.716	-0.144	-0.862	0.532
Base saturation				
O layer	0.946	0.818	0.797	-0.365
Mineral soil 0–10 cm	-0.776	0.078	-0.700	0.231
Mineral soil 10–20 cm	-0.577	0.999	-0.502	0.892
Mineral soil 20–30 cm	-0.580	0.986	0.623	0.998
Topsoil stocks ^a				
N	-0.217	-0.129	0.575	-0.385
S	0.390	0.132	0.409	0.013
P	-0.915	-0.919	-0.945	-0.741
Citric acid extractable PO ₄ -P	-0.888	-0.576	-0.490	0.328
Exchangeable Ca ²⁺	-0.820	-0.801	-0.987	-0.598
Exchangeable Mg ²⁺	-0.827	-0.174	0.650	0.895
Exchangeable K ⁺	0.560	0.989	0.883	0.481

Note: Numbers printed in bold type represent statistically significant ($p < .05$) correlations. Growth periods represent intervals between two stand inventories.

^aO layer + Mineral topsoil 0–30 cm.

might clarify this issue. Anyway, combined recent development of pine nutrient deficiencies/imbbalances, increased water demand for tree transpiration, but limited soil water availability at our study sites is alarming. More frequent and longer-lasting droughts in the future (Beniston & Diaz, 2014) likely also will decrease soil nutrient availability by reducing SOM mineralization rates and ion mobility. Moreover, they will affect tree nutrient uptake by modifying carbon allocation to roots, root-absorbing surface, and maximum absorption (Jonard et al., 2015; Kreuzwieser & Gessler, 2010). Reciprocally, trees with nutrient deficiencies are less resistant against stress. According to our foliage data, the supply of both stands with K, a crucial element for regulating stomatal conductance and alleviating environmental stress such as drought and frost (Cakmak, 2005), showed the most pronounced decrease of all nutrient cations during recent decades, approaching the level of insufficiency at the end of monitoring.

An earlier study of Prietzel et al. (2008), covering the first (20th century) part of our monitoring period, identified needle mass, followed by foliar N and P concentrations as factors most related to pine growth at both study sites. Our current study adds valuable information on growth-determining site factors by (a) investigating an extended monitoring time, as well as including (b) additional soil and stand nutrition (S) data, and (c) climate data. The entire monitoring period considered, at both sites age-referenced stand growth was positively correlated with MAT (Table 4), which is associated with a prolonged growing season (Chmielewski & Rötzer, 2001) and—similar to most regions in Germany (Spekat, Enke, & Kreienkamp, 2007)—has increased at both sites particularly during the most recent 20 years (Table 3; Figure 9a,b). However, after 1983 the correlation leveled off at the cooler site *Pfaffenwinkel*, and at the warmer site *Pustert* age-referenced stand growth even was reduced in recent times with large MAT (Table 4). At the same time, it was positively correlated with foliar Ca concentrations and amounts, whereas for the other nutrients no positive correlation was observed. Ca storage and enrichment in tree foliage are strongly affected by tree water uptake and transpiration. At sites with excellent tree Ca nutrition (e.g., *Pustert*; Figure 7a), Ca concentrations and amounts in tree foliage indicate the amount of water that has been available for photosynthesis during the past growing season (Clarholm & Skjyllberg, 2013). Following this line of argument, the tendency of decreasing age-referenced stand growth at *Pustert* after 1983 together with its negative correlation with MAT and its positive correlation with foliar Ca concentrations and contents (Table 4) suggest that pine growth at *Pustert* is increasingly becoming limited by water shortage. In this context, it must be emphasized that the most recent six monitoring years at *Pustert*, when MAT has increased most strongly and MAP as well as CWB have decreased most strongly (Figure 9b,d,f), were not included in our correlation analysis, because stand growth data are not yet available for the time after 2013. In contrast, the P supply of the pines at *Pustert*, despite having become insufficient in most recent decades according to foliar P concentrations, showed no correlation with age-referenced stand growth after 1983. Another nutrient, whose availability may be increasingly influencing stand productivity at both sites, is S, which was not included in the correlation analysis, because foliar S concentrations

had not been measured before the year 2000 at *Pustert* and before 1994 at *Pfaffenwinkel*, and thus were available only for one complete growth period. Yet, available foliar S concentrations indicate insufficient S supply to the pines at *Pfaffenwinkel* and the onset of S insufficiency for those at *Pustert* at the end of monitoring. Age-referenced stand growth correlates positively with topsoil S stocks at both sites, but the correlation is weak and absent after 1983, when soil S stocks started to decrease (Table 4). Thus, the role of S as growth-limiting nutrient at our sites is questionable and deserves further investigation.

4.4 | Effects of stand aging versus N and S deposition and site recovery from litter raking

Forest aging is associated with nutrient accumulation in tree biomass, resulting in soil acidification and soil nutrient depletion (Binkley & Richter, 1987; Brais, Camiré, Bergeron, & Paré, 1995; Van Cleve, Oliver, Schlentner, Viereck, & Dyrness, 1983; Van Cleve & Viereck, 1981). It is difficult to disentangle environmental change effects on soil chemistry from those of stand aging; particularly because the latter are dependent on forest growth rate and thus site quality, and therefore are supposed to change in a changing environment. Yet, results from stand aging studies conducted under most possible constant environmental boundary conditions, for example, in pristine boreal forests (Brais et al., 1995; Van Cleve et al., 1983; Van Cleve & Viereck, 1981) may help separating stand aging from environmental change effects in our study. Thus, soil nutrient depletion in undisturbed forests with progressively increasing biomass is proceeding slowly and unidirectional, because nutrients are continuously sequestered in woody biomass, foliage, and roots. The relevance of net nutrient sequestration in foliage rapidly decreases with increasing tree age and is negligible in mature Scots pine forests. Foliage mass and nutrient content (Armolaitis et al., 2013; Lehtonen, Mäkipää, Heikkinen, Sievänen, & Liski, 2004) as well as litterfall amounts (Kouki & Hokkanen, 1992) in Scots pine stands after canopy closure under constant environmental boundary conditions do not change systematically with age any more. In our study, stand ages at beginning of monitoring were >80 years and the stands were well beyond canopy closure and growth culmination. At this stand age, any systematic change of stand growth and foliar nutrient concentrations can be expected to proceed slowly under constant environmental boundary conditions. Yet, it must be considered that in our study at both sites pine growth and thus net nutrient sequestration in woody biomass remained high until the end of monitoring. Topsoil acidification and base cation depletion at our sites between 1974 and 2004 therefore probably are a combined effect of elevated atmospheric S and N deposition and stand aging, and the systematic depletion of plant-available topsoil P stocks during the entire monitoring period is probably exclusively caused by stand aging. However, it must be emphasized that “natural” stand aging effects on chemistry have been strongly intensified by recent anthropogenic N deposition and CO₂ concentration increases as well as by climate warming, which very likely all have contributed to the accelerated

pine growth and nutrient sequestration at our study sites (Mellert et al., 2004; Prietzel et al., 2008). In contrast, the most recent topsoil pH and BS increases as well as the strongly declining S concentrations and amounts in pine foliage and topsoil after 1994 cannot be explained by forest aging, but by the recent strong decrease in atmospheric S deposition. In summary, most observations in our study have been explicitly caused (forest floor C/N decrease; recent foliar and topsoil S decrease, recent topsoil pH and base cation increase; sustained stand growth acceleration) or intensified (soil P depletion during stand development) by anthropogenic environmental change.

At *Pustert*, but not at *Pfaffenwinkel*, stand aging together with environmental change including site recovery from historic degradation resulted in development of regeneration from naturally imported seeds consisting of other tree species than Scots pine (Norway spruce, oak, birch; Figure S4). Between the first stand inventory in 1963 and the last inventory in 2013, the contribution of non-pine tree species to total stand biomass at *Pustert* increased from 0% to 5% (spruce), 3% (oak), and 2% (birch). Broadleaf tree development at *Pustert* to some extent may have contributed to recent topsoil pH, BS, and base cation stock increases (Prietzel, 2004). However, effects of oak introduction into Scots pine stands on topsoil chemistry are marginal even at larger oak tree regeneration contributions (Prietzel, 2004). Moreover, topsoil pH, BS, and base cation contents have increased in a similar way at *Pfaffenwinkel*, which due to the absence of nearby broadleaf seed trees has remained a pure pine stand throughout the monitoring period. This underpins the existence of a direct effect of recent environmental change on topsoil chemistry even without any mediation by tree species change.

5 | SUMMARY AND CONCLUSIONS

Stand growth at site *Pfaffenwinkel* in the early 1960s was limited by insufficient N and Mg supply due to historic site degradation. Until the mid-1980s, N and Mg limitation disappeared due to ecosystem recovery from degradation, supported by high atmospheric N deposition, and stand growth increased strongly. Later, increased stand nutrient uptake together with strongly decreased S deposition and probably also climate warming resulted in reduced Mg, N, and S nutrition and substantial needle mass reduction of the pines at *Pfaffenwinkel*, but age-referenced stand growth remained far above growth rates expected from yield tables until the end of monitoring. Stand growth at *Pustert* in the early 1960s was limited by insufficient N supply. Similar to *Pfaffenwinkel*, N limitation disappeared until the mid-1980s, and stand growth increased. Topsoil P stocks at *Pustert* were only 40% of those at *Pfaffenwinkel*, and increased tree P uptake associated with accelerated stand growth resulted in depletion of plant-available topsoil P stocks and deterioration of tree P nutrition. Here, age-referenced stand growth decreased after 1993, most likely due to increased water stress. Any increase in pine water use efficiency due to CO₂ fertilization (Keenan et al., 2013) at our sites could not compensate the decrease

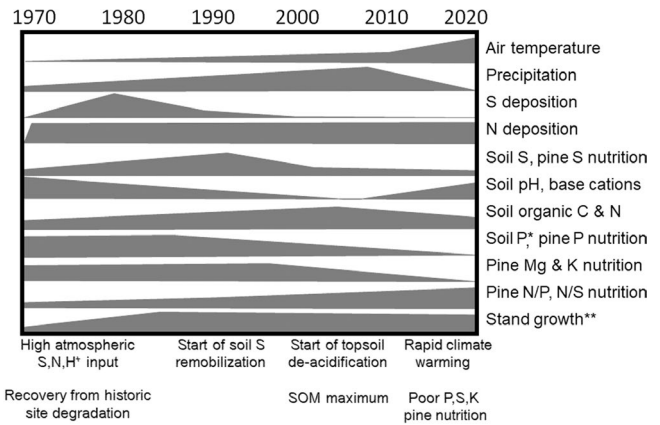


FIGURE 11 Conceptual synthesis of environmental and ecosystem changes at the studied Scots pine forest sites from 1970 to 2020. Investigated soil compartments: Forest floor + uppermost 30 cm mineral soil. *Citric acid-extractable, plant-available PO₄-P. **Age-referenced stand growth

in plant available water, and/or was overcompensated by growth-reducing effects of other limiting factors (e.g., P, S availability). Large differences of stand nutrition observed during the last two decades of the 20th century on the one hand and during the first decades of the 21st century on the other most likely are caused by a marked change of environmental conditions (Figure 11). Our study is representative for many European lowland forests with intensive utilization until World War II, elevated atmospheric N S, and acid deposition in the second half of the 20th century, and strongly reduced S deposition, but continuously high N deposition, and climate warming in the 21st century.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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