Climate Dependency of Tree Growth Suppressed by Acid Deposition Effects on Soils in Northwest Russia

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Increased tree growth in temperate and boreal forests has been proposed as a direct consequence of a warming climate. Acid deposition effects on nutrient availability may influence the climate dependency of tree growth, however. This study presents an analysis of archived soil samples that has enabled changes in soil chemistry to be tracked with patterns of tree growth through the 20th century. Soil samples collected in 1926, 1964, and 2001, near St. Petersburg, Russia, showed that acid deposition was likely to have decreased root-available concentrations of Ca (an essential element) and increased root-available concentrations of Al (an inhibitor of Ca uptake). These soil changes coincided with decreased diameter growth and a suppression of climate–tree growth relationships in Norway spruce. Expected increases in tree growth from climate warming may be limited by decreased soil fertility in regions of northern and eastern Europe, and eastern North America, where Ca availability has been reduced by acidic deposition.

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Introduction

Increased tree growth in temperate and boreal forests has been proposed as a direct consequence of climate warming on the basis of longer growing seasons and satellite imagery that shows an increase in canopy greenness (*1*–*3*). However, limited availability of nutrients, such as Ca, may constrain the response of trees to a more favorable climate. The role of Ca as a control of tree growth may have increased in significance if Ca availability has been lowered by acid deposition. The importance of Ca in forest function is well-established (*4*), and the possibility of Ca depletion is increasingly being considered in investigations of forest health and productivity (*5*–*7*). Declines of both red spruce (*Picea rubens*) and sugar maple (*Acer saccharum*) in the eastern United States have been linked to Ca deficiency in soils (*5*, *8*, *9*).

Despite progress in defining relationships between Ca availability and tree growth, the effect of acid deposition on tree growth remains uncertain because of a lack of information on the magnitude and timing of Ca loss from soils. Without this information, changes in Ca availability cannot be related to tree-growth patterns. Soil Ca data that predate the onset of high acid deposition rates are available from only two studies worldwide (*10*, *11*). In the Adirondack region of New York decreases in Ca concentrations and pH, attributable to acid deposition and net forest growth, were identified in forest soils between 1930–1932 and 1984 (*10*). Decreased Ca concentrations and pH, attributed largely to acid deposition, were also identified between 1893 and 1997 through analysis of archived soil samples in a Scots pine–Norway spruce forest near Moscow, Russia (*11*). Because data were available at only two points in time in these studies, the pattern of change between the sampling dates is unknown, which make the data difficult to relate to tree-growth patterns.

Further use of archived soil samples provided by the Dokuchaev Central Soil Museum, St. Petersburg, Russia, has now enabled soil chemistry to be evaluated in forest soils in 1926, 1964, and 2001, at a site that has received high levels of acid deposition during the 20th century. Tree cores were also collected at this site for dendrochronological analysis. The objectives of this study were to (a) compare the chemical concentrations of the soils at this site on the three sampling dates to determine if long-term changes had occurred and (b) determine if relationships existed between changes in chemical concentrations of soil and patterns of tree growth during the 75-year period.

Experimental Section

Study Site. The study was conducted in a southern boreal forest of predominantly Norway Spruce (*Picea abies*) and Scots pine (*Pinus sylvestris*), 40 km southeast of St. Petersburg, Russia, within the flat, low-lying area east of the Bay of Finland. Current mean annual temperature is 6.4 °C and has increased about 1 °C over the past century. Current mean annual precipitation is 73 cm and has increased 10% over the past century. The forest has been the property of the St. Petersburg State Forest Technical Academy since 1865, although it was a state-run forest dating back into the 1700s. Details of forest history and management are published elsewhere (*12*). Soil sampling was done in the Parkovyi quadrangle, a 4 ha parcel, 70 m elevation, which served as the demonstration stand for this forest throughout the 20th century. There is no record of harvesting in the quadrangle after 1845. Shallow ditching was done in 1842–1845 to

10.1021/es048759o CCC: $30.25

Published on Web 02/16/2005

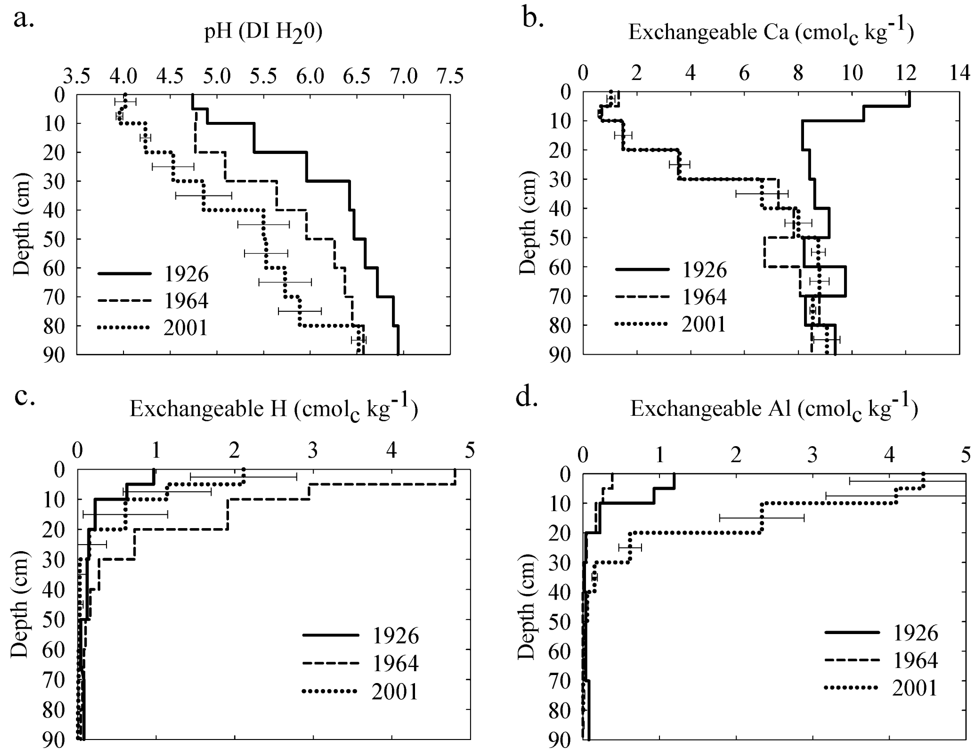

FIGURE 1. Measurements of (a) pH, (b) exchangeable Ca concentrations, (c) exchangeable H concentrations, and (d) exchangeable Al concentrations in soil profiles collected in 1926, 1964, and 2001. The lateral position of each graphed vertical line represents the measurement value. The height of the vertical line represents the thickness of the profile segments sampled. Data for 2001 are means ($n = 5$) ± 1 standard error, graphed at the midpoint of each sampled segment of the profile.

improve drainage. Forest management during the second half of the 19th century and throughout the 20th century consisted of periodic sanitation—removal of deadwood, and cutting of saplings to prevent overcrowding. Tree cores were collected from this quadrangle plus the Suitti quadrangle, approximately 5 km away. Selective cutting before 1940 resulted in a younger stand structure in the Suitti quadrangle than in the Parkovyi quadrangle, although trees predating 1875 were available for coring. Canopy condition in both quadrangles appeared normal at the time of sampling in 2001.

The site has been subject to high levels of acid deposition, which were likely to have begun shortly after World War II. Bulk deposition rates in St. Petersburg ranged from 20 kg of SO_4^{2-} ha^{-1} $year^{-1}$ in 1956 (the earliest postwar measurement) to a peak of 70 kg of SO_4^{2-} ha^{-1} $year^{-1}$ in 1978 (*11*). Deposition then declined to 20 kg of SO_4^{2-} ha^{-1} $year^{-1}$ in 1993 as the Russian economy declined but increased to 30 kg of SO_4^{2-} ha^{-1} $year^{-1}$ in 1998 as the economy redeveloped in the 1990s.

Methods

Archived soils from 1926 and 1964, collected as intact profiles of 1 m depth, 25 cm width, and 8 cm thickness, were made available by the Dokuchaev Central Soil Museum, St. Petersburg, Russia. These soils were stored horizontally in dustproof, wooden boxes that had not been treated with adhesives or resins. The intact profile enabled soil to be removed by depth in a manner that could be precisely duplicated by the modern sampling, conducted in July 2001. Records of the original sampling location and methods were made available by the Soil Museum. Locations of the original pits to within 50 m were facilitated by a grid system used by the Forest Technical Academy that predates the historic sampling. To assess local variations in soil chemistry, four replicate pits were excavated within 50 m of a centrally located pit that was considered to best represent the original sampling location. Samples were collected from these pits, and the archived profile, in 5 cm increments from the top of the mineral soil (depth 0) to a depth of 10 cm. Below 10 cm, samples were collected in 10 cm increments down to a depth of 90 cm. The profile of each pit was described in detail by an experienced pedologist from the Soil Museum. The variability among pits, expressed as standard error, was generally small relative to the differences between sampling dates discussed in this paper (Figures 1−4). The difference between the archived values and the mean of the modern sampling was often greater than twice the value of the standard error. Similarly small levels of spatial variability were also found in rocky forest soils where direct remeasurement of archived samples also enabled long-term changes to be assessed (*13*).

The stone-free soils of our study site, classified as Podzoluvisol Gleyic in the FAO system (Typic Glossaqualf in the U.S. system), have developed since the last glaciation (10 000 years ago) from calcareous till that overlies Silurian limestone. Soil texture was estimated in the field to range from silt loam in the upper profile to clay loam in the lower profile. Fine roots were densely concentrated in the O horizon but were described as few in the uppermost mineral soil of all pits. Roots were absent below 30 cm in three pits and described as few or very few in the other two pits.

The modern soils were air-dried for storage, and moisture content was determined after drying at 100 °C. Modern and historic samples were passed through a 2 mm sieve before chemical analysis. Because the archived soils had never been analyzed, we could not directly evaluate possible storage effects. However, a recent study was able to show minimal storage effects over 30 years in an analysis of archived soils (*13*). All soil samples were analyzed for soil pH (*14*) (deionized water, 1:1 soil:water), exchangeable acidity and Al (*15*) (Al quantification limit, 0.04 $cmol_c$ kg^{-1}; H quantification limit, 0.02 $cmol_c$ kg^{-1}), and exchangeable base cations (*14*) (unbuf-

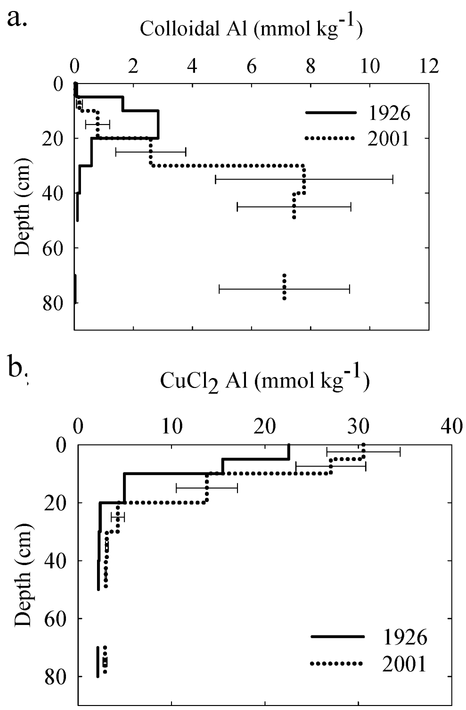

FIGURE 2. Concentrations of (a) colloidal Al (polymeric Al < 0.2 μm) and (b) $CuCl_2$ extractable (organically complexed) Al, in soil profiles collected in 1926 and 2001. The height of the vertical line represents the thickness of the profile segments sampled. Data for 2001 are means ($n = 5$) $\pm$ 1 standard error, graphed at the midpoint of each sampled segment of the profile.

fered 1 N NH_4Cl; quantification limit, 0.02 $cmol_c$ kg^{-1}), and total C (C−H−N analyzer). Organically complexed Al was determined by extraction with 0.5 M $CuCl_2$ for 2 h (*16*). Carbonate content (as percent calcite equivalent: 0.05% quantification limit) was determined by gas chromatography measurement of the CO_2 evolved from treating a soil sample with 6 M HCl. Cation exchange capacity (CEC) was calculated by summing the concentrations of exchangeable Ca, Mg, Na, K, Al, and H (once the absence of carbonate was verified). Base saturation was calculated by expressing the sum of exchangeable Ca, Mg, Na, and K as a percent of CEC.

To measure what we termed colloidal Al, 3 g of soil was equilibrated with 30 mL of 1 mM NaCl for 5 days (*17*). Filtrate that passed through a 0.2 μm membrane filter was analyzed for total Si and total Al (ICP−OES), molybdate reactive Si (*18*), and quickly reactive Al (*19*). Total Si and molybdate reactive Si (an estimate of monomeric Si) enabled concentrations of polymeric Si to be determined by their difference. Concentrations of colloidal Al (polymeric Al) were similarly determined by the difference between total Al and quickly reacting Al; however, total Al in samples from upper horizons included organically complexed monomeric forms. Total Si concentrations did not include organic monomeric Si because dissolved Si does not complex with dissolved organic matter. Therefore, we were able to develop a regression model (polymeric Al = 0.509 × polymeric Si + 0.001; $p < 0.01$; $R^2 = 0.995$) from the relationship between polymeric Si and polymeric Al for samples from the lower profile. This relation held if dissolved organic carbon (DOC) concentrations in filtrates were less than 833 μmol L^{-1}. Polymeric Al concentrations (hereafter referred to as colloidal Al) in samples with DOC greater than 833 μmol L^{-1} were then estimated from polymeric Si measurements and the Si−Al regression model.

All laboratory analyses were done at the U.S. Geological Survey, Troy, NY, except colloidal Al (Swedish University of Agricultural Sciences, Department of Soil Sciences, Uppsala, Sweden) and CO_3 content (Hubbard Brook Experimental Forest, NH).

A single tree core (12 mm diameter) was collected from each of 31 dominant and codominant Norway spruce trees in the Technical Academy Forest, within 5 km of the soil pits. Trees > 30 cm diameter at breast height (approximately 1.4 m above ground) were selected to include a range of sizes that were free of visible stem defects. Annual rings were cross-dated to validate assignment of calendar dates.

Ring-width increment data of individual trees were converted to Z scores (*20*) and then grouped into the three age classes presented in Figure 4a. The grouping was based on a break in the distribution of the two oldest classes and a split of the trees dated from 1903 to 1954 to yield similar numbers in each of the two youngest age classes. Models of ring-width increment were developed from forward stepwise regression (STATISTICA 6.0 software) that used *Z* scores of ring-width increment as the dependent variable and mean monthly values of air temperature and precipitation, which yielded 24 possible predictors. Temperature and precipitation data for November and December were grouped with the following 10 months for correlation to annual ring-width increment. Temperature data were collected at the Voiekov Meteorological Observatory, 25 km from the site, at an elevation of 78 m. These data, available back to 1891, were obtained from the Global Historical Climatology Network Temperature Database (*21*) in May 2003. Precipitation data from the same site and period were obtained from the National Climatic Data Center, Asheville, NC (*22*).

Normalized difference vegetation index (NDVI) data were derived from 10 day composite imagery collected by the National Oceanic and Atmospheric Administration (NOAA) meteorological satellites through the Pathfinder advanced very high resolution radiometers (AVHRRs) land (PAL) program, run jointly by NOAA and the National Aeronautics and Space Agency (NASA). The imagery has been processed by the Global Inventory Monitoring and Modeling Studies (GIMMS) at NASA. The NDVI record was corrected for degradation of satellite orbits and variations in atmospheric transparency. Data were analyzed as accumulated values for the growing season. Individual values of 9 pixels (8 × 8 km) were averaged to obtain a single value for a 24 × 24 km^2 that encompasses the Academy Forest and surrounding forested landscape.

Results and Discussion

From 1926 to 2001, soil pH decreased from 0.75 to one unit in the upper 10 cm of the mineral soil profile, and more than one unit from 10 to 80 cm (Figure 1a). Approximately half of this decrease occurred from 1926 to 1964, with the exception of the uppermost 10 cm, which did not change from 1926 to 1964 (Figure 1a). Concentrations of exchangeable Ca decreased up to 10-fold from 1926 to 1964 in the upper 30 cm of the profile and 1−2 $cmol_c$ kg^{-1} from 30 to 50 cm (Figure 1b), but showed little difference from 1964 to 2001 at any depth (Figure 1b). Exchangeable Mg concentrations (not shown) exhibited differences similar to Ca. In the upper 30 cm of the profile, concentrations in 1964 and 2001 were essentially the same, but were approximately one-half to one-fifth those in 1926. In contrast to Ca, concentrations of Mg below 40 cm were approximately 20% higher in 1964 and 2001 than in 1926. Exchangeable H concentrations in the upper 30 cm increased 2- to 5-fold from 1926 to 1964 but then decreased from 1964 to 2001, to concentrations that approached those in 1926 (Figure 1c). Little difference among sampling dates was observed for exchangeable H concentrations below 30 cm. Concentrations of exchangeable Al showed a small decrease in the upper 10 cm of the profile from 1926 to 1964, but a 10-fold or greater increase in the upper 30 cm from 1964 to 2001 (Figure 1d).

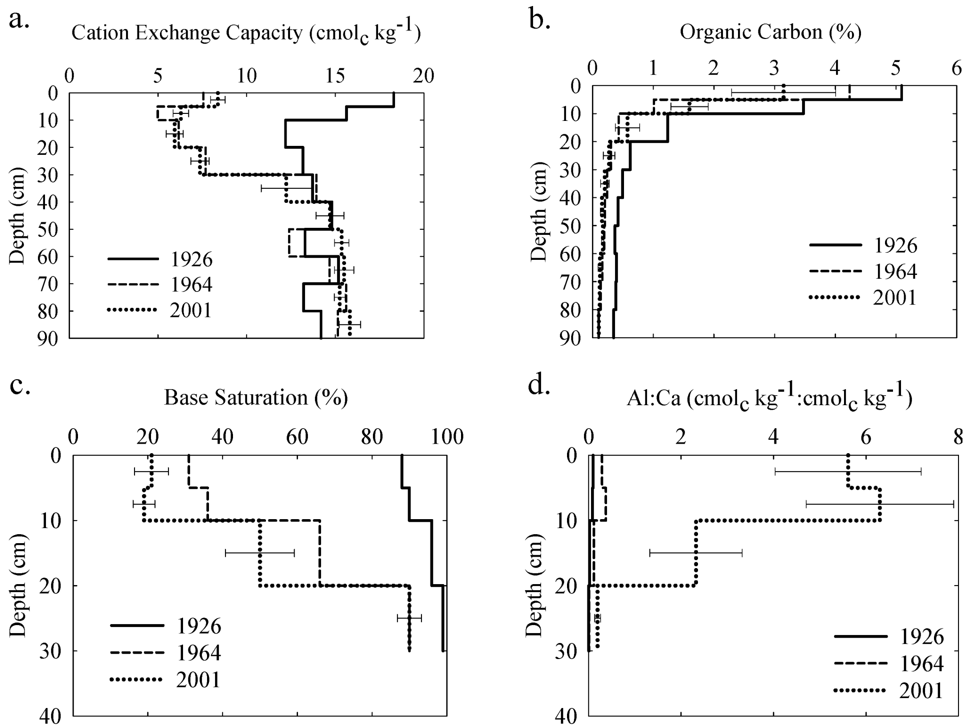

FIGURE 3. Measurements of (a) cation exchange capacity (CEC), (b) organic carbon concentrations, (c) base saturation (sum of exchangeable base cation concentrations as a percentage of cation exchange capacity), and (d) the ratio of Al to Ca, in soil profiles collected in 1926, 1964, and 2001. The height of the vertical line represents the thickness of the profile segments sampled. Data for 2001 are means (n = 5) ± 1 standard error, graphed at the midpoint of each sampled segment of the profile.

These changes in soil chemistry reflect a two-stage acidification process that was accelerated by high levels of acidic deposition. From 1926 to 1964, inputs of acidity were neutralized by replacement of exchangeable Ca by H because $CaCO_3$ was no longer available to neutralize H inputs. An absence of detectable carbonate in the upper 90 cm of the profile collected in 1926 suggests prior depletion of carbonate minerals through natural weathering. Neutralization from 1926 to 1964, accomplished largely through cation exchange, was sufficient to prevent mobilization of Al during this period.

Continued acid deposition did mobilize Al from 1964 to 2001, which indicates a shift from cation-exchange to weathering of solid-phase Al as the primary source of acid neutralization. This interpretation is supported by a decrease in concentrations of colloidal Al in the upper 20 cm (Figure 2a), which was concurrent with the increase in concentrations of exchangeable Al (Figure 1d) and organically complexed Al (Figure 2b). Colloidal Al, which was not detected at any depth in the 1964 profile, may have been depleted to concentrations below detection as the pH decreased in the absence of carbonate weathering from 1926 to 1964. Weathering of alumino silicates from 1964 to 2001 may have resulted in a small degree of reaccumulation. The particle size (<0.2 μm diameter) and Si-to-Al ratio of 2:1 suggest that the colloidal Al is in some form of smectite, a secondary mineral with high cation-exchange capacity. Mobilization of Al by acid deposition would be expected to increase dissolved Al concentrations in the upper profile, where weathering is most intense and organic matter is available, thereby increasing concentrations of exchangeable and organically complexed Al at the expense of colloidal Al. Depletion of secondary mineral forms of solid-phase Al in the upper profile by acid deposition has been inferred (*23*, *24*), but historical depletion has not been previously measured. Deposition of colloidal Al lower in the profile resulted from increased pH (Figures 1a and 2a).

The observed transition from cation-exchange buffering to Al buffering is fully consistent with the long-standing theory of soil acidification by acid deposition (*25*) but has not been previously demonstrated through field measurement. These changes in the weathering regime of the upper profile have degraded soil fertility through a decrease in cation-exchange capacity (CEC; Figure 3a), as well as a decrease in availability of Ca and an increase in availability of Al. A decrease in organic carbon concentrations during 1926–2001 (Figure 3b) can explain the decrease in CEC, which occurred largely by 1964, although a decrease in pH-dependent adsorption, and weathering and leaching of clay particles may also have contributed to decreased CEC in the upper 30 cm.

Net forest growth can acidify soil (*10*), but acid deposition is the most plausible explanation for the soil changes we observed. Because the stand in the Parkovyi quadrangle was mature at the time of the 1926 sampling, and no harvesting occurred during the 1900s, a large degree of aggradation is unlikely. Inventory data indicate that the growing stock was high in 1973 (350 m^3 ha^{-1}) and was only marginally higher (370 m^3 ha^{-1}) in 1994. Inventory data in the Suitti quadrangle also did not suggest strong aggradation. Growing stock increased from 180 to 210 m^3 ha^{-1} from 1953 to 1973, but then deceased to 200 m^3 ha^{-1} in 1994.

Additional information that is unsupportive of aggradation as the primary cause of acidification and Ca depletion is available in a recent study that indicated minimal soil acidification over 105 years in a strongly aggrading Russian oak forest with parent material (dolomitic limestone) similar to our study site, but with acid deposition rates that were less than half that of our site from 1970 to 1990, and 20–30% lower than our site from 1990 to 2000 (*11*). Exchangeable Ca concentrations at the two sites were similar in archived soil samples (1926 and 1893); approximately 12 to 8 $cmol_c$ kg^{-1}, from the 0–5 cm layer to the 30–40 cm layer (*11*). The site sampled in 1893 has been managed for oak production since the early 1700's and was clear-cut in 1880. The oak forest regrew to a mature stand at the time of resampling in 1998. Despite the large increase in biomass between samplings, no difference in exchangeable Ca concentrations was ap-

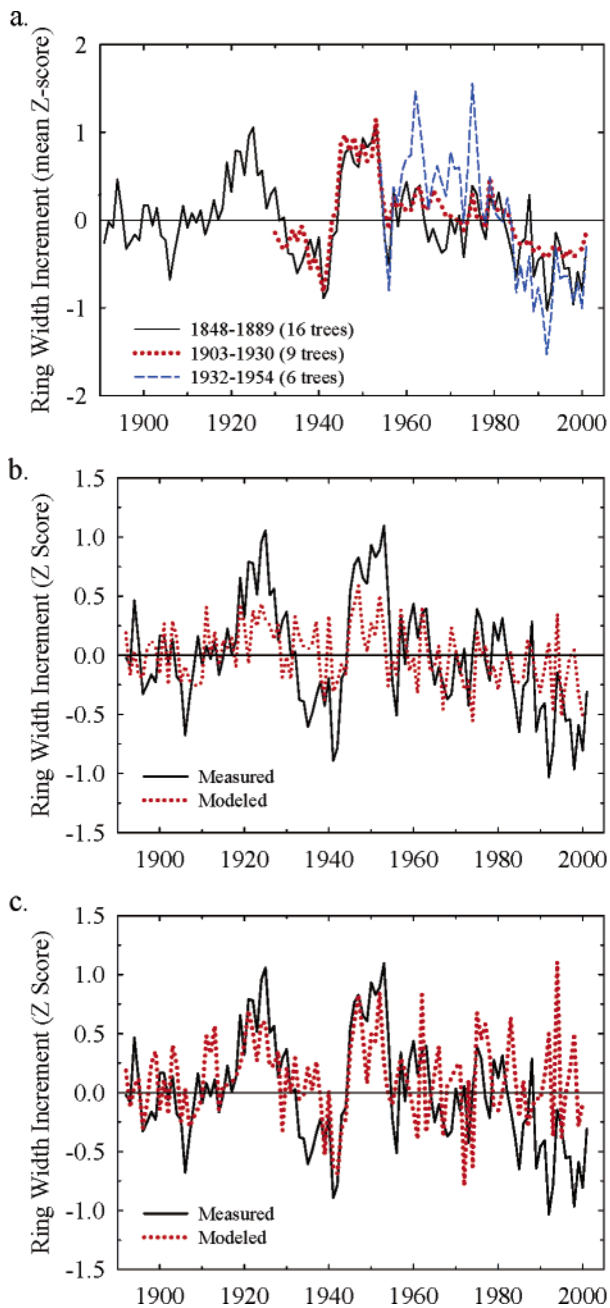

FIGURE 4. Trends of (a) ring-width increment (expressed as mean of *Z* scores) for three age classes of trees (data start at the date that includes all trees for that age group), (b) measured ring-width increment (expressed as the mean of *Z* scores) for the 16 oldest trees shown in a and ring-width increment estimated with a regression model that was developed from mean monthly temperature and precipitation for the period 1891 to 2001, and (c) measured ring-width increment (expressed as the mean of *Z* scores) for the 16 oldest trees shown in a and ring-width increment estimated with a regression model that was developed from mean monthly temperature and precipitation for the period from 1891 to 1960 and then extrapolated from 1960 to 2001.

parent in the top 40 cm of soil in the oak forest between 1893 and 1998. The soil changes observed in our study took place within 75 years, in soil also formed in calcareous parent material, which has generally been considered resistant to acidification over this time scale. Soil acidification would occur more readily than observed in this study where soils have developed from slow-weathering silicate minerals that are predominant in many regions of northern Europe and eastern North America.

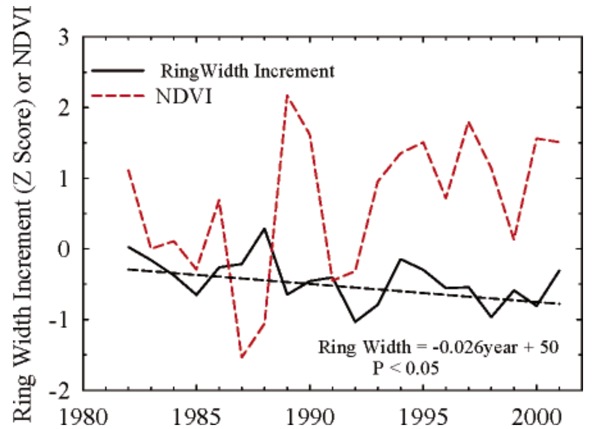

FIGURE 5. Mean of the measured ring-width increment for the 16 oldest trees shown in Figure 4a and the normalized difference vegetation index (NDVI), normalized for plotting by dividing by the 1982 value and then subtracting 1.

The decrease in exchangeable Ca concentrations resulted in a decrease in base saturation to values that approached the threshold of 15% (Figure 3c), below which forest damage may occur from an imbalance in the availability of Ca and Al (an inhibitor of Ca uptake) in the rooting zone (*26*). The ratio of exchangeable Al to Ca also increased substantially in the upper 20 cm from 1964 to 2001 (Figure 3d). Canopy damage was not evident in this forest, but mean ring-width increment (expressed as *Z* scores) of Norway spruce in three age classes showed a coincident anomaly of below average growth, which began in the early 1980s and extended through the 1990s (Figure 4a).

To evaluate what role climate may have played in this decline, stepwise forward regression was used to investigate the relationship between ring-width increment and mean monthly air temperature and precipitation for 1891 to 2001, the period for which climate data were available. Although a statistically significant model ($p < 0.01$; $R^2 = 0.28$) was obtained with the complete data set, only March temperatures (negative correlation) and October precipitation (negative correlation) were significant predictors ($p < 0.05$), and the model was not effective at capturing the magnitude of variations in the first 70 years or the below average growth in the 1980s and 1990s (Figure 4b). Negative correlations with March temperature and October precipitation are difficult to explain on the basis of physiological processes.

Variations in growth were described effectively (Figure 4c; $p < 0.01$; $R^2 = 0.44$), however, with climate data for 1891–1960, with April and January temperatures (positive correlation), April and September precipitation (positive correlation), and February temperatures (negative correlation). The negative correlation with February temperatures is also difficult to explain on the basis of physiological processes, but a similar result was observed for Norway spruce in Finland (*27*). When this model was extended to 2001, predicted growth diverged from measured growth in the late 1980s (Figure 4c). A statistically significant model could not be obtained for the last 70 years with the variables selected by stepwise regression for 1891–1960 data, or for any combination of the 24 climate variables over the past 40 years. Changes in relations of tree growth to climate variations have been previously reported for red spruce in the eastern United States (*28*, *29*) and trees near the arctic tree line (*30*), but in each study the cause was reported as unknown.

Despite the decrease in rates of diameter growth over the past 2 decades, satellite imagery indicated that canopy greenness (expressed as the normalized difference vegetation index, NDVI) at our site did not decrease and may have possibly increased ($P < 0.1$; Figure 5). A general increase in

NDVI has been observed at high northern latitudes and is considered to be an indication of increased net primary productivity that has resulted from a warming climate (*1*, *3*). The favorable climate suggested by the positive *Z* scores predicted during the 1980s and 1990s (Figure 4c) may have also contributed to good canopy growth, but the pronounced decrease in growth rings of these outwardly healthy trees suggests a decrease in net primary productivity.

The onset of growth declines during the period of Al mobilization, suggests that the trees were responding to a decline in soil fertility. Direct effects of air pollution on the canopy are less likely because (1) canopy damage was not apparent and (2) deposition of SO_4^{2-}, which would be expected to positively correlate with emissions of SO_2 and O_3 (*32*), decreased from 70 to 30 kg ha^{-1} $year^{-1}$ from 1978 to 2000, the period of pronounced growth declines. Field studies of red spruce have shown that low concentrations of foliar Ca were associated with low concentrations of Ca in the soil, elevated rates of dark respiration, and lower rates of net photosynthesis (*4*). Furthermore, increased respiration of red spruce (*Picea rubens*) saplings was found to decrease retention of photosynthate and increase allocation of photosynthate to the maintenance of current year foliage (*31*). Regardless of canopy condition, the decline in diameter growth at our site suggests that the trees are becoming increasingly vulnerable to environmental stresses and could experience canopy dieback in the future, as was observed in red spruce trees in the northeastern U.S., where canopy dieback and mortality were attributed to possible increases in Al-to-Ca ratios in soils from acidic deposition (*4*, *5*).

Our results support the interpretation that decreased soil fertility from acid deposition can suppress the growth response of trees to favorable climate. Continued acid deposition may worsen soil Ca availability and result in further growth limitation and vulnerability to stress in the future. Because acid deposition has fallen on large areas of the northern temperate and southern boreal forest, the availability of soil Ca (and Mg) may be an important control of current and future tree growth in upper northern latitudes.

Acknowledgments

We are greatly indebted to the staff of the Dokuchaev Central Soil Museum, St. Petersburg, Russia, and the Field Station of the St. Petersburg State Forest Technical Academy. We thank Vsevolod Aparin, Ania Rozanova, and Rebecca Waysek for help collecting the samples and Mark David and Douglas Burns for helpful reviews of the manuscript. This work was funded in part by the NSF Ecosystem Studies Program under the Grant 0089939 and the U.S. Geological Survey National Atmospheric Deposition Program.

Literature Cited

(1) Myneni, R. B.; Keeling, C. D.; Tucker, C. J.; Asrar, G.; Nemani, R. R. Increased plant growth in the northern latitudes from 1981 to 1991. *Nature* **1997**, *386*, 698−702.

(2) Menzel, A.; Fabian, P. Growing season extended in Europe. *Nature* **1999**, *397*, 659.

(3) Lucht, W.; Prentice, I. C.; Myneni, R. B.; Sitch, S.; Friedlingstein, P.; Cramer, W.; Bousquet, P.; Buermann, W.; Smith, B. Climatic control of the high-latitude greening trend and the Pinatubo effect. *Science* **2002**, *296*, 1687−1689.

(4) McLaughlin, S. B.; Wimmer, R. Tansley Review No. 104, Calcium physiology and terrestrial ecosystem processes. *New Phytol.* **1999**, *142*, 373−417.

(5) Shortle, S. C.; Smith, K. T.; Minocha, R.; Lawrence, G. B.; David, M. B. Acidic deposition, cation mobilization, and biochemical indicators of stress in healthy red spruce. *J. Environ. Qual.* **1997**, *26*, 871−876.

(6) Schaberg, P. G.; DeHayes, D. H.; Hawley, G. J.; Murakami, P. F.; Strimbeck, G. R.; McNulty, S. G. Effects of chronic N fertilization on foliar membranes, cold tolerance, and carbon storage in montane red spruce. *Can. J. For. Res.* **2002**, *32*, 1351−1359.

(7) Bailey, S. W.; Horsley, S. B.; Long, R. P.; Hallett, R. A. Influence of edaphic factors on sugar maple nutrition and health on the Allegheny Plateau. *Soil Sci. Soc. Am. J.* **2004**, *68*, 243−252.

(8) McLaughlin, S. B.; Joslin, J. D.; Robarge, W.; Stone, A.; Wimmer, R.; Wullschleger, S. D. Effects of acidic deposition on nutrient uptake, nutrient cycling and growth processes of vegetation in the spruce-fir ecosystem. In *The productivity and sustainability of Southern Forest ecosystems in a changing environment*; Mickler, R. A., Fox, S., Eds.; Springer-Verlag: New York, 1997; pp 255−277.

(9) Horsley, S. B.; Long, R. P.; Bailey, S. W.; Hallet, R. A.; Hall, T. J. Factors associated with decline disease of sugar maple on the Allegheny Plateau. *Can. J. For. Res.* **2000**, *30*, 1365−1378.

(10) Johnson, A. H.; Anderson, S. B.; Siccama, T. G. Acid rain and soils of the Adirondacks. I. Changes in pH and available calcium 1930−1984. *Can. J. For. Res.* **1994**, *24*, 39−45.

(11) Lapenis, A. G.; Lawrence, G. B.; Andreev, A. A.; Bobrov, A. A.; Torn, M. S.; Harden, J. W. Acidification of forest soil in Russia: From 1893 to present. *Global Biogeochem. Cycles* **2004**, *18* (1), GB1037. doi: 10.1029/2003GB002107, GB1037.

(12) Red'ko, G. I., Ed. *The 200th anniversary of the Lisino Experimental Forest Station* (in Russian); St. Petersburg Academy of Forestry: St. Petersburg, Russia, 1997.

(13) Bailey, S. W.; Horsley, S. B.; Long, R. P. Thirty years of change in forest soils of the Allegheny Plateau, Pennsylvania. *Soil Sci. Soc. Am. J.*, in press.

(14) Blume, L. J.; Schumacher, B. A.; Schaffer, P. W.; Cappo, K. A.; Papp, M. L.; Van Remortel, R. D.; Coffey, D. S.; Johnson, M. G.; Chaloud, D. J. Handbook of methods for acid deposition studies: Laboratory analyses for soil chemistry; EPA/600/4−90/023; Environmental Monitoring Systems Laboratory, U.S. Environmental Protection Agency: Las Vegas, NV, 1990.

(15) Thomas, G. W. Exchangeable cations. In *Methods of soil analysis*, 2nd ed.; Page, A. L., Miller, R. H., Reeney, D. R., Eds.; American Society of Agronomy: Madison, WI.; 1982; Agronomy 9, part 2, pp 159−166.

(16) Juo, A. S. R.; Kamprath, E. J. Copper chloride as an extractant for estimating the potentially reactive aluminium pool in acid soils. *Soil Sci. Soc. Am. J.* **1979**, *43*, 35−38.

(17) Simonsson, M.; Berggren, D. Aluminum solubility related to secondary solid phases in upper B horizons with spodic characteristics. *Eur. J. Soil Sci.* **1998**, *49*, 317−326.

(18) Weaver, R. M.; Syers, J. K.; Jackson, M. L. Determination of silica in citrate−bicarbonate−dithionite extracts of soils. *Soil Sci. Soc. Am. J.* **1968**, *32*, 497−501.

(19) Clarke, N.; Danielsson, L.-G.; Sparen, A. The determination of quickly reacting aluminium in natural waters by kinetic discrimination in a flow system. *Int. J. Environ. Anal. Chem.* **1992**, *48*, 77−100.

(20) Fritts, H. C. *Reconstructing large-scale climatic patterns from tree-ring data: A diagnostic analysis;* University of Arizona Press: Tucson, 1991.

(21) Peterson, T.; Vose, R. An overview of the global historical climatology network temperature data base. *Bull. Am. Meteorol. Soc.* **1999**, *78*, 2837−2849.

(22) Gruza, G. V.; Rankova, E. Y.; Razuvaev, V. N.; Bulygina, O. A. Indicators of climatic change for the Russian Federation. *Clim. Change* **1999**, *42*, 219−242.

(23) Driscoll, C. T.; van Breemen, N.; Mulder, J. Aluminum chemistry in a forested Spodosol. *Soil Sci. Soc. Am. J.* **1985**, *49*, 437−444.

(24) Mulder, J.; van Breemen, N.; Eijck, H. C. Depletion of soil aluminum by acid deposition and implications for acid neutralization. *Nature* **1989**, *337*, 247−249.

(25) Ulrich, B. Effects of acidic precipitation on forest ecosystems in Europe. In *Acidic precipitation, Vol. 2: Biological and ecological effects*; Adriano, D. C., Johnson, A. H., Eds.; Springer-Verlag: New York; 1988; pp 189−272.

(26) Cronan, C. S.; Grigal, D. F. Use of calcium aluminium ratios as indicators of stress in forest ecosystems. *J. Environ. Qual.* **1995**, *24*, 209−226.

(27) Makinen, H.; Nojd, P.; Mielikainen, K. Climatic signal in annual growth variation of Norway spruce (Picea abies) along a transect from central Finland to the Arctic timberline. *Can. J. For. Res.* **2000**, *30*, 769.

(28) Cook, E. R.; Zedaker, S. M. The dendroecology of red spruce decline. In *Ecology and decline of red spruce in the Eastern United States*; Eager, C., Adams, M. B., Eds.; Springer-Verlag: New York; 1992; pp 192−234.

(29) Shortle, W. C.; Smith, K.; Minocha, R.; Minocha, S.; Wargo, P. M.; Vogt, K. Tree health and physiology in a changing environment. In *Responses of northern U. S. forests to environmental*

change; Mickler, R. A., Birdsey, R. A., Hom, J., Eds.; Springer: New York; 2000; pp 229−274.
(30) Briffa, K. R.; Schweingruber, F. H.; Jones, P. D.; Osborn, T. J.; Shiyatov, S. G.; Vaganov, E. A. *Nature* **1998**, *391*, 678−682.
(31) Anderson, C. P.; McLaughlin, S. B.; Roy, W. K. A comparison of seasonal patterns of photosynthate production and use in branches of red spruce saplings at two elevations. *Can. J. For. Res.* **1991**, *21*, 455−461.
(32) Driscoll, C. T.; Lawrence, G. B.; Bulger, A. J.; Butler, T. J.; Cronan, C. S.; Eager, C.; Fallon-Lambert, K.; Likens, G. E.; Stoddard, J. L.; Weathers, K. C. Acidic deposition in the northeastern U.S.: Sources and inputs, ecosystem effects and management strategies. *Bioscience* **2001**, *51*, 180−198.

Received for review August 9, 2004. Revised manuscript received December 9, 2004. Accepted December 22, 2004.

ES048759O