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Acid rain and soils of the Adirondacks. II. Evaluation of calcium and aluminum as causes of red spruce decline at Whiteface Mountain, New York

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Several recent publications suggest that acid deposition produces high anion concentrations in the soil solution of high-elevation forests, causing Al-induced Ca deficiency in red spruce (*Picea rubens* Sarg.) root tips. Some authors suggest that this results in decreased growth below and above ground, decline, and ultimately in death. Accordingly, we examined soil solution Al and Ca, soil and foliar chemistry, and their relationship to red spruce decline on Whiteface Mountain, New York, where severe red spruce mortality occurred between the mid-1960s and late 1980s. Spruce mortality and crown condition are strongly related to elevation, but there is no evidence that the occurrence or degree of spruce decline is related to exchangeable Al, Mg, Ca, or their ratios in mineral or organic horizons. Foliar Ca and Mg concentrations that we measured were in the range judged to be sufficient based on field fertilization studies. Soil solution ratios of Ca:Al in mineral and organic horizons were above the threshold (1:1 molar ratio) proposed as detrimental to Ca uptake in all samples collected during three of four growing seasons, and soil-solution Al concentrations were well below levels thought to inhibit spruce root growth. If Al toxicity is a factor in red spruce decline on this mountain, it is acting in a way that is not detectable using soil and foliar analyses.

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Plusieurs publications récentes suggèrent que les dépôts acides produisent des concentrations anioniques élevées dans la solution de sol des forêts en haute altitude, causant une déficience en Ca induite par Al dans les apex racinaires de l'épinette rouge (*Picea rubens* Sarg.). Certains auteurs suggèrent que cela se traduit par des diminutions de croissance tant dans la partie hypogée que dans la partie épigée de la plante, du dépérissement et finalement, par la mort de l'arbre. En conséquence, nous avons examiné Al et Ca dans la solution de sol, la chimie du sol et la chimie foliaire et leur interrelations avec le dépérissement de l'épinette rouge à montagne Whiteface, New York, où une mortalité sévère de l'épinette rouge est apparue entre le milieu des années 1960 et la fin des années 1980. La mortalité de l'épinette et la condition des cimes sont fortement reliées à l'altitude mais il n'y a aucun signe que l'apparition ou le degré de dépérissement soient reliés à Al, Mg et Ca échangeables ou à leurs rapports dans les horizons organiques ou minéraux. Les concentrations foliaires en Ca et Mg étaient dans la zone de suffisance basée sur des études de fertilisation au champ. Les rapports Ca:Al de la solution de sol dans les horizons minéraux et organiques étaient au-dessus du seuil (rapport molaire 1:1) proposé comme nuisible au prélèvement de Ca dans tous les sols échantillonnés durant trois des quatre saisons de croissance. Les concentrations en Al de la solution de sol étaient bien inférieures aux niveaux considérés comme inhibant la croissance racinaire de l'épinette. Si la toxicité de Al est un facteur dans le dépérissement de l'épinette rouge sur cette montagne, elle agit d'une manière qui n'est pas détectable par les analyses de sol et de feuillage.

[Traduit par la rédaction]

Introduction

For more than a decade, researchers have been seeking links between acid deposition and the decline of red spruce (*Picea rubens* Sarg.) in the montane spruce–fir forests of the Adirondacks and New England (Johnson and Siccama 1983; Eagar and Adams 1992). There is consensus that winter injury due to freezing is a preeminent factor promoting decline and mortality in the northern Appalachians and that exposure of foliage to ambient cloud water or artificial acid mist significantly increases the risk of winter injury (Friedland

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et al. 1984; A.H. Johnson et al. 1986, 1988, 1991; DeHayes et al. 1991; DeHayes 1992; Perkins et al. 1991; Vann et al. 1992; Peart et al. 1992).

In addition to the effects of weather, pathogens, and direct effects of acid deposition on foliage, there may be belowground effects that add stress and contribute to decline. Several researchers have focused on Ca as a nutrient that when in short supply, has been linked to decreased photosynthesis:respiration ratios in foliage, reduced growth below and above ground, and possibly increased mortality (Shortle and Smith 1988; McLaughlin et al. 1990, 1991; McLaughlin and Kohut 1992; Shortle and Bondietti 1992). A substantial amount of evidence suggests that Ca uptake by red spruce can be inhibited by elevated levels of Al that are released to the soil solution preferentially to Ca and Mg when anion concentrations increase (e.g., Raynal et al. 1990; McLaughlin et al. 1990, 1991; McLaughlin and Kohut 1992). In laboratory seedling studies, an inhibition of Ca uptake has been observed at Al concentrations occasionally reached in the soil solution at Whiteface Mountain , New York (100–200 μmol/L; Raynal et al. 1990; Miller et al. 1992).

Several studies have provided indirect evidence of an adverse effect of Al on Ca uptake. Bondietti et al. (1989) measured Ca and Al in red spruce bolewood from the northern Appalachians and found radial patterns that suggested to them that Ca was mobilized in the 1940–1960 period, then depleted after 1960–1970 with concomitant increases in Al. They suggested that this represents an effect of increasing sulfate deposition with attendant mobilization and leaching of Ca followed by preferential mobilization of Al. Shortle and Smith (1988) observed low Ca/Al ratios in root tips of declining spruce in Vermont compared with spruce roots sampled from healthy spruce in Maine that experienced lower rates of acid deposition. They suggested that root tissue composition was affected by strong-acid anion deposition in acid rain and that this Al-induced Ca deficiency led to reduced growth, increased susceptibility to natural stresses, and ultimately red spruce mortality in high-elevation stands. Schlegel et al. (1992) measured Al, Ca, and Mg in cross sections of red spruce roots in an area of declining spruce on Whiteface Mountain and concluded that Al toxicity (e.g., blocking of Ca and Mg uptake) was a factor contributing to red spruce decline at Whiteface Mountain. They asserted that Ca/Al ratios in the soil solution at Whiteface Mountain are below values considered to be critical for adequate Ca uptake (<1.0) and that foliar Ca values are low.

Direct evidence that red spruce respond to added Ca has been provided by field fertilization studies. Van Miegroet et al. (1993) fertilized red spruce saplings with Ca and Mg at two elevations in Great Smoky Mountain National Park, Tennessee, and using vector analysis of foliar Ca determined that red spruce foliage responds to Ca additions if foliar Ca concentrations are <1700 μg/g. At foliar concentrations of ca. 1900 μg/g, there was not a significant response to Ca additions. Mg fertilization did not improve either Ca or Mg levels in foliage, suggesting that even though foliar Mg levels were low (ca. 600 μg/g), they were not indicative of Mg shortage. J.D. Joslin (Tennessee Valley Authority, unpublished data) has obtained nearly identical results by fertilizing mature red spruce on Whitetop Mountain, Virginia. These fertilization studies suggest that foliar Ca levels are a reasonable measure of plant Ca status. In this regard, Friedland et al. (1988) reported mean Ca concentrations in current-year red spruce foliage at pooled high-elevation sites in northern New York and Vermont that averaged less than 1700 μg/g, suggesting the possibility of a moderate Ca deficiency in one or both regions.

Few of the studies that suggest Ca deficiency in populations of red spruce in the Northeast have reported on Ca levels in foliage or soils. Joslin et al. (1992) reviewed available data on soil and foliar properties in high-elevation red spruce stands in the northern and southern Appalachians. Foliar Ca levels they report are nearly twice as high at the northern sites, and well above the 1700 μg/g threshold for deficiency. Interestingly, mortality of red spruce in the northern Appalachians has been dramatically higher and more widespread than in the southern Appalachians (Eagar and Adams 1992). At Mount. Moosilauke, New Hampshire, Huntington et al. (1990) found that red spruce crown condition was significantly correlated with Ca in the Oi + Oe horizon (the upper forest floor) but not with Ca in the Oa horizon (lower forest floor) or in the mineral soil.

A traditional approach to forest nutrition suggests that nutrient deficiencies can be diagnosed by a combination of soil, foliar, and fertilization studies. If this approach is valid for Ca, then the following hypotheses should be supported by soil, foliar and soil-solution data if Ca deficiency is a causal factor in red spruce decline

(1) The severity of decline should be greatest on soils that have the lowest ratios of exchangeable Ca/Al. These soils will produce the lowest Ca/Al ratios in the soil solution when anion concentrations are high.

(2) (*i*) Foliar Ca levels should be lowest in declining trees and in stands where decline and mortality are greatest. (*ii*) Foliar Ca levels should be low on soils that have low Ca or low Ca/Al ratios and are subject to high levels of acidic deposition.

(3) Al concentrations and (or) Ca/Al ratios in the soil solution should be in the range associated with reduced Ca uptake.

We report on data that address these three hypotheses.

Spruce decline at Whiteface Mountain

Between 1964 and 1982, spruce basal area decreased by about 60% in the mature stands studied by Scott et al. (1984). Silver et al. (1991) resurveyed the transects sampled by Johnson and Siccama (1983) and found that the percentage of dead spruce doubled between 1982 and 1987. J.J. Battles, A.H. Johnson, T.G. Siccama, A.J. Friedland and E.K. Miller (unpublished data) found that the mortality rate between 1985 and 1990 on the permanent plots used for soil solution studies (see above) was 3% per year.

The percentage of standing dead red spruce at Whiteface Mountain (55% at elevations above 1000 m) is consistent with the data for other Adirondack and Green Mountain peaks (Leopold et al. 1988; Craig and Friedland 1991; Peart et al. 1992). Red spruce crowns and roots have been studied in detail at Whiteface Mountain by Wargo et al. (1993). There are no unusual pathogen problems and no specific symptoms that differ from those reported in other areas where declining red spruce have been studied. Thus, all evidence we are aware of suggests that decline and mortality at Whiteface Mountain are representative of the region-wide decline.

Methods

Study site

Whiteface Mountain is an isolated massif in the northeastern Adirondacks (44°22′N, 73°54′W), which rises to a maximum

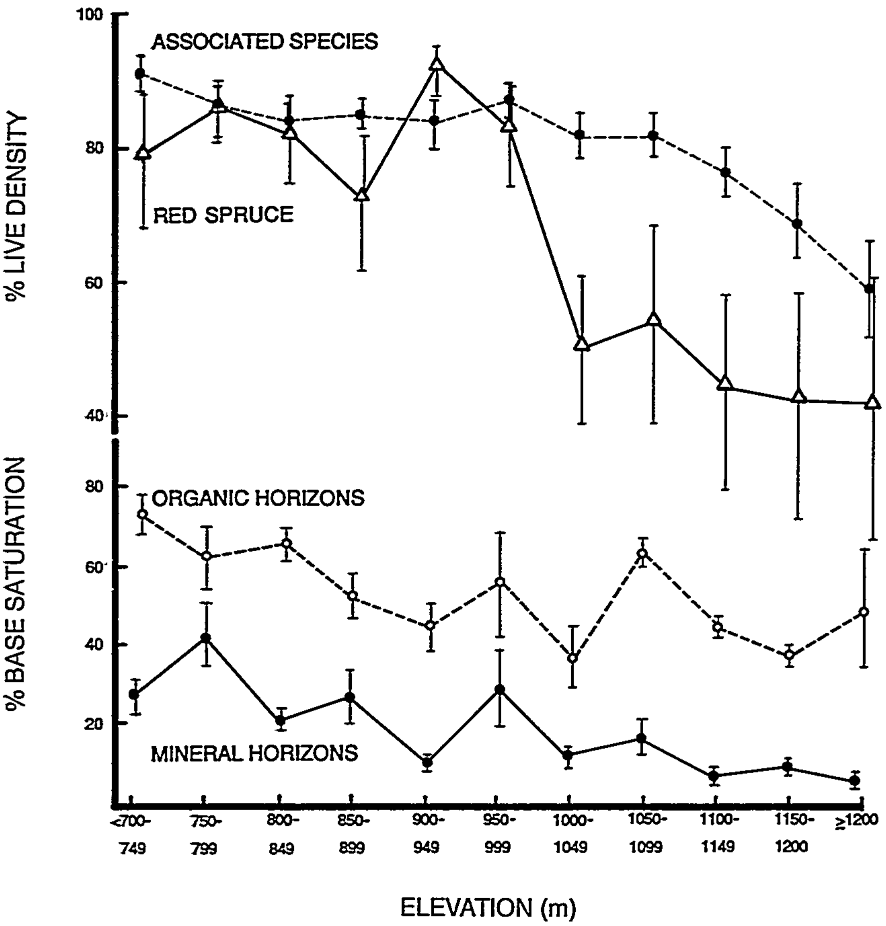

FIG. 1. Elevational gradient in percentage of the individuals alive in 1987 for red spruce (upper solid curve) and other species (upper broken line) at Whiteface Mountain, New York, and elevational gradient in base saturation of organic horizons (lower broken line) and mineral horizons (lower solid line).

elevation of 1483 m. The bedrock is Precambrian anorthosite, which is overlain by locally derived glacial till. As a result of the Ca-rich anorthosite, the soils have a very high Ca content (4% on a total weight basis; Johnson and Lindberg 1991). The till is several meters deep on lower slopes and intermittent above 1100 m. Soils developed in till are mostly Cryorthods, while at the highest elevations, soils are largely Cryofolists (essentially organic mats overlying bedrock; Witty 1968).

Forest types at Whiteface Mountain differ as a function of elevation and disturbance history. Lower elevations (<800 m) are dominated by hardwoods with scattered red spruce, mid-elevations (800–1100 m) are mixed forests of balsam fir (*Abies balsamea* (L.) Mill.), red spruce, and paper birch (*Betula papyrifera* var. *cordifolia* (Marsh.) Regel), and the upper slopes are dominated by balsam fir.

The data presented below were obtained from three studies: (*i*) a study of foliar chemistry and soils at the base of those trees conducted at 700–1200 m on the northwest face of the Whiteface massif; (*ii*) a stand-level study of soil characteristics and their relationship to red spruce vigor conducted at 60 permanent plots that represented all aspects on the mountain between 700 and 1200 m; and (*iii*) soil solution studies conducted at four plots with highly acid soils in the spruce–fir zone located at 1020–1090 m on the northwest face.

Sampling plots and red spruce condition

In 1986–1987, three hundred thirty-one 10 m diameter circular plots were established between 600 and 1300 m on Whiteface Mountain on a randomly located, 21-transect grid (Battles et al. 1992). Individual trees in a subset of these plots were used to assess relationships among foliar chemistry, crown health, and soil chemistry at the base of the sampled trees. In 1987, another set of sixty 20 × 20 m intensive study plots was located in a stratified systematic manner in the spruce–fir–birch zone to study diseases and the relationship between soil characteristics (including Ca, Mg, and Al) and stand condition (Battles et al. 1992). Crown health of red spruce used in the statistical analyses reported below was rated on a five-point scale according to U.S. Forest Service Forest Response Program protocols: 1, 0–10% loss of foliage from live crown; 2, 11–50% foliage missing; 3, 51–99% foliage lost; 4, dead with relatively intact stem; 5, dead with broken stem.

Foliage

In October 1986, 69 trees in the 10 m diameter plots on the northwest aspect were selected for foliar sampling. Because of the large difference in spruce mortality above and below 1000 m we elected to sample an approximately equal number of trees in each elevation zone. Below 1000 m, each spruce >10 cm DBH nearest plot center was sampled, and above 1000 m we sampled the spruce >10 cm DBH nearest plot center, plus the second nearest tree if there was more than one in the plot. We climbed the trees to sample foliage from whorls 4–7 on the south-facing side. Current-year (1986) and 1-year-old (1985) needles were separated and dried at 60°C for 24 h upon returning from the field.

In the laboratory, 200 needles of each sample were counted, redried at 60°C, weighed, and digested in HCl–HF after dry-ashing at 500°C (Friedland et al. 1988). Analysis for Ca, Mg, and K in the extracts was performed by atomic absorption spectroscopy. National Bureau of Standards standard pine needles (reference No. 1575), internal standards, replicate samples, and procedural blanks were used to assess analytical accuracy and precision.

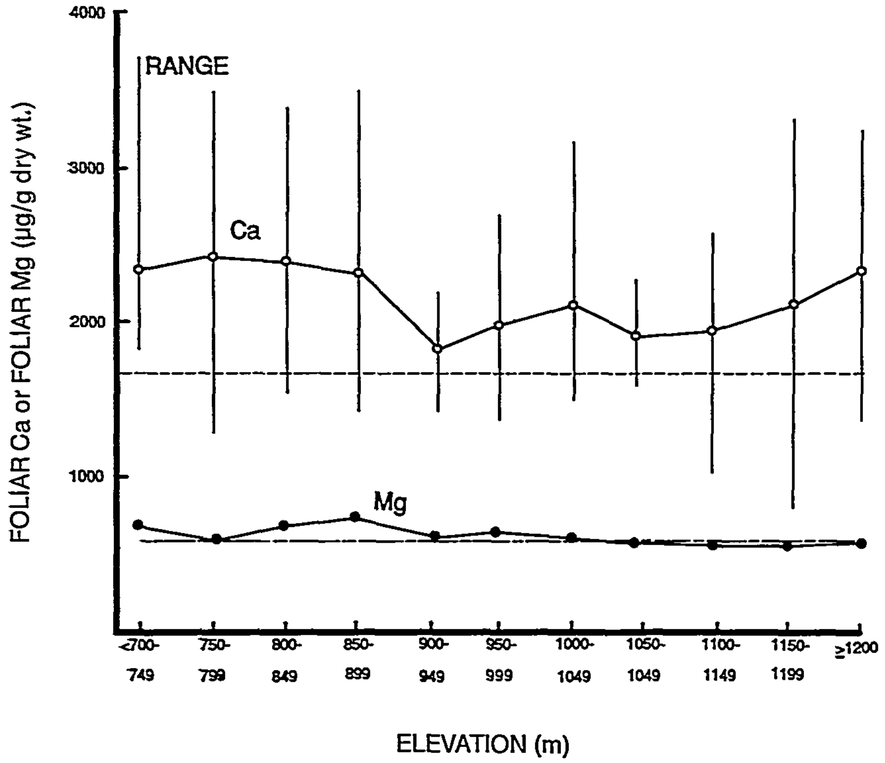

FIG. 2. Ca and Mg content of red spruce foliage in October 1986 as a function of elevation at Whiteface Mountain, New York. Bar length represents the range of Ca values observed in each elevation band ($n = 69$ trees total). Brokens lines for Ca and Mg represent incipient deficiency (Van Miegroet et al. 1993).

Soils

At the base of each tree sampled in the foliar composition study, four small soil pits were dug along the four primary compass directions, approximately midway between the bole and edge of the crown. After excluding loose litter, samples from the four pits were composited to represent the top 15 cm of soil and the 15–30 cm depth around each tree.

At the 60 intensive study plots, soil pits were systematically located 5 m below the midpoint of the lower boundary of each plot. Soils were excavated within a 50 × 50 cm frame according to the quantitative pit method described in detail by Hamburg (1984) with slight modifications. In this procedure, all of the soil, roots, and rocks excavated from carefully measured depth increments were weighed so that accurate estimates of soil (<2 mm) and nutrient quantities could be made. The Oi and Oe horizons were collected as a single sample. The Oa (or a weighed subsample) was then collected, followed by the top 10 cm of mineral soil, the 10–20 cm mineral soil layer, and all soil from 20 cm to bedrock or the top of the C horizon. We analyzed subsamples from the Oa horizon and the 0–10 cm, ≥10–20 cm, and ≥20 cm depths separately.

All soil samples were air dried to constant weight and then seived through either a 5-mm stainless steel screen (organic horizons) or a 2-mm screen (mineral horizons). Seived soil (2.5 g, weighed to ±1 mg) was extracted in a vacuum extractor with 50 mL of 1.0 M NH_4Cl and extracts analyzed for Ca, Mg, Na, and K by AAS. Exchangeable H^+ and Al were measured on KCl extracts according to the procedure of Thomas (1982). Internal standards (U.S. Forest Service Forest Response Program standards WRO and WRM) and procedural blanks and a cross-laboratory check of 21 samples (W.P. Robarge, North Carolina State University, Durham) were used to ensure analytical accuracy.

Organic matter content of samples from the quantitative pits was estimated by loss on ignition of ovendried soil at 500°C overnight. Soil pH was determined by glass electrode in a 1:1 (v/v) suspension.

Base saturation was calculated as the ratio of exchangeable base cations to $H^+ + Al +$ exchangeable bases.

To facilitate the statistical analyses, we used average values for organic and mineral horizons. In many cases (particularly above 1000 m), the soil underlying the Oa horizon had high enough organic matter content to qualify as an organic horizon (loss on ignition ≥40%), and in these cases the values obtained from the organic layers were averaged as an estimate of organic horizon chemistry at each site. To estimate the chemical characteristics of the mineral soil at each site, the values obtained from the layers that had <40% organic matter were averaged.

Soil solution studies

In 1985, four 0.1-ha vegetation plots were established for biogeochemistry studies as part of the Integrated Forest Study (Johnson and Lindberg 1991). These plots were located at 1020–1090 m on the northwest aspect. In these plots >53% of the red spruce basal area was accounted for by dead or severely declining trees. Miller et al. (1992) give the range of composition for these soil pits as well as ranges for the whole mountain. Soils in the lysimeter plots are very acid relative to those elsewhere on the mountain (Miller et al. 1992). Organic horizons have a pH of 3.4 and mineral horizons pH 4.1 (A–Bhs) to 4.7 (Bs), and base saturation is 9–11% (Miller et al. 1992). At each plot, three fritted glass plate (60 mm diameter, 2- to 4-µm pore size, Corning 60 VF) tension (10-kPa) lysimeters were installed in each of three horizons (O, A–Bh, and Bs). Collections of soil solution were made weekly and pooled for monthly analysis. Total dissolved Al and dissolved Ca were determined by atomic absorption spectroscopy (AAS) at Oak Ridge National Laboratory, Oak Ridge, Tennessee. In a related study (Miller et al. 1992) we fractionated soil-solution Al using the method of Bartlett et al. (1987) and found that 62 and 77% of the Al measured by AAS in O and mineral horizons, respectively, was kinetically labile (the most phytotoxic form).

Statistical analyses

Owing to the variety of studies using various combinations of class and continuous variables with varying distributions, we

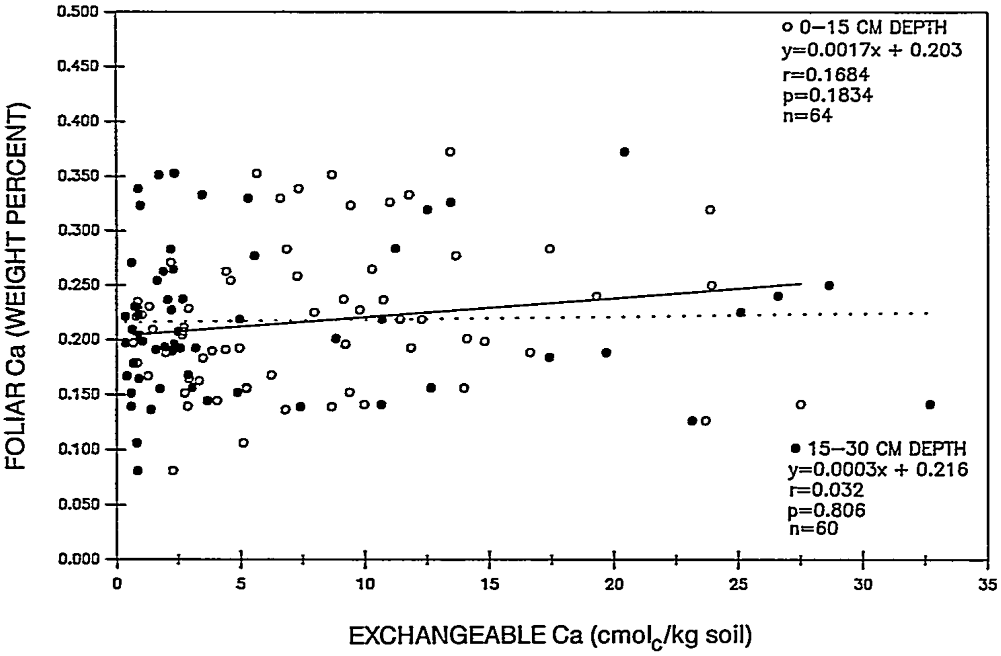

FIG. 3. Foliar Ca concentration as a function of exchangeable Ca in surface soil (0–15 cm) and subsoil (15–30 cm) at Whiteface Mountain, New York. Soils were collected at the base of each tree sampled for foliar analysis (see text).

used a variety of approaches available from SAS Institute Inc. (1985) as noted below. We considered results to be statistically significant if $p \leq 0.10$.

Results and discussion

Spatial patterns in soil characteristics, foliar characteristics, and spruce condition

In evaluating hypotheses 1 and 2, we looked for evidence that severity of decline was greatest in stands where soils had the lowest base saturation, that crown condition was worst in trees with the lowest base saturation in the root zone (hypothesis 1), and that declining trees had the lowest levels of Ca in foliage (hypothesis 2).

Figure 1 summarizes soil base saturation and the percentage of red spruce and associated species alive in 1986–1987. Trends in base saturation follow expected patterns, with organic horizons showing higher base saturation than mineral horizons, and base saturation tending to decrease with increasing elevation. As observed in other spruce–fir forests (e.g., Robarge and Johnson 1992) the base saturation of organic soil horizons can be high in spite of low pH. At Whiteface Mountain organic horizons generally have pH values between 3.5 and 4.5 with base saturation generally exceeding 40% at any pH value, while mineral horizons have base-saturation values of about 8–30% with a trend toward decreased base status at higher elevations. Whiteface Mountain has relatively high base status soils relative to those summarized by Joslin et al. (1992), probably due to the high Ca content of the till and the abundance of soil organic matter.

Elevational trends in foliar Ca and Mg (newest year class) are shown in Fig. 2. There are not significant elevational trends in the foliar concentrations of either element, and mean values are typical for healthy high-elevation spruce in the northern Appalachians (Joslin et al. 1992). As shown in Fig. 2, a relatively small percentage of the samples had Ca <1700 µg/g, the threshold proposed by Van Miegroet et al. (1993) and J.D. Joslin (unpublished data) for incipient Ca deficiency, and the low values were spread across nearly all elevations. The foliar Mg values at Whiteface Mountain are similar to those found elsewhere (Friedland et al. 1988; Joslin et al. 1992), and in the range judged to be sufficient by van Miegroet et al. (1993) on the basis of field fertilization experiments conducted with spruce saplings.

In comparing soil exchangeable cations (Ca, Mg, Ca/Al, and Mg/Al) with foliar concentrations, we found no significant correlations, as illustrated by Fig. 3. This finding is similar to that of Huntington et al. (1990) and suggests that foliar concentrations of Ca and Mg are regulated by factors other than limited availability in the soil. Evaluating foliar chemistry using traditional concepts of forest nutrition does not suggest deficiency of either Ca or Mg on Whiteface Mountain. Some investigators have raised the point that total foliar Ca is not an appropriate measure of Ca status, though no alternative measures have been proposed.

Table 1 shows the results of stepwise regression analysis (SAS Institute Inc. 1985) using percent dead and severely declining red spruce (angular transformation of percentage in crown condition 3 + 4 + 5) in the intensive study plots as the dependent variable and elevation and exchangeable cation concentrations as candidate predictors. Crown condition is related to elevation, but no soil variables were significantly correlated with decline severity. We also explored the possibility that quantities of Ca and Mg might be limiting, even though their concentrations on exchange sites were rather high. Some soils at high elevation are shallow to bedrock, and the total quantity of nutrients available in thin soils can be low (<200 kg/ha). As illustrated by Fig. 4, most soils contain relatively large pools of available cations (>400 kg Ca/ha), and there are not significant relationships between quantities of exchangeable Ca or Mg and decline severity in the study plots.

Table 2 shows a summary of the relationships between crown condition and foliar and soil chemistry for the 69 red spruce trees sampled in the 10 m diameter circular plots. In this multiple range analysis (SAS Institute Inc. 1985), there was a significant difference in crown condition with ele-

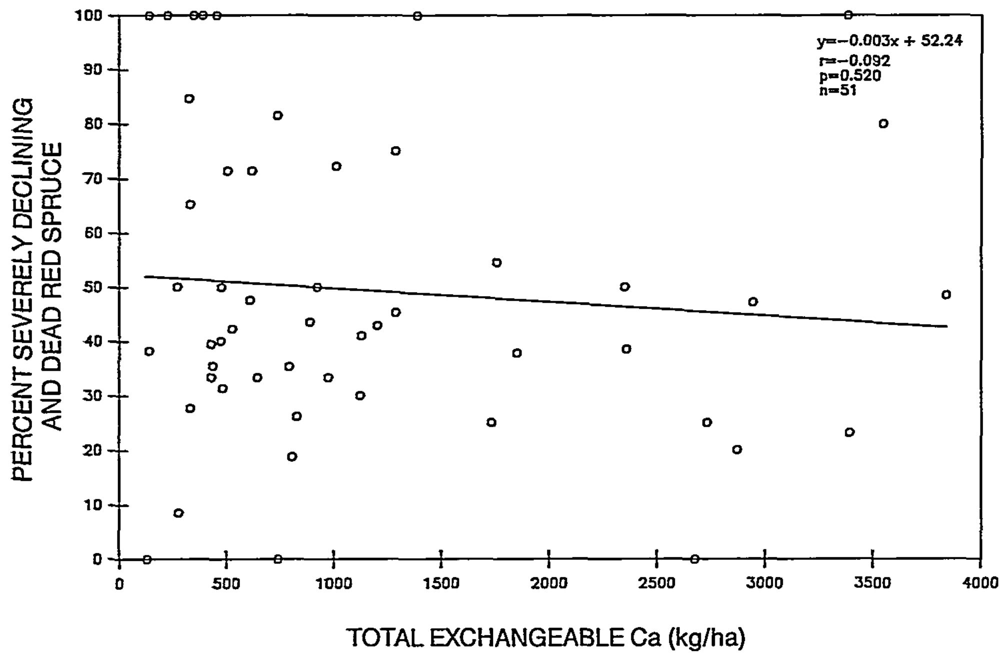

FIG. 4. Red spruce decline plotted as a function of the quantity of exchangeable Ca in the root zone (O, A, and B horizons) at Whiteface Mountain, New York. One calcareous profile with > 20 000 kg Ca/ha is not shown.

TABLE 1. Stepwise regression relating the severity of spruce decline (arcsine-transformed percentages in crown condition 3 + 4 + 5) to elevation and soil characteristics at the 20 × 20 m permanent vegetation plots (df = 42)

	F	*p*
Variables in model		
Elevation	5.773	0.348
Variables not in model		
Organic horizon K	0.0063	0.012
Organic horizon Ca	0.0066	0.013
Organic horizon Mg	0.168	0.064
Organic horizon Ca/Al	0.362	0.096
Organic horizon Mg/Al	0.263	0.080
Mineral soil Al	0.887	0.146
Mineral soil K	0.115	0.053
Mineral soil Ca	0.165	0.063
Mineral soil Mg	0.075	0.043
Mineral soil Ca/Al	0.379	0.096
Mineral soil Mg/Al	0.333	0.090

vation, but no significant differences in foliar elemental concentrations among the three crown classes (class 1, 0–10% loss of foliage from the crown; class 2, 11–50% loss; class 3, 51–99% loss). Other statistical procedures and the addition of base cation concentrations in 1-year-old foliage do not alter the relationships shown in Table 2 with one exception. The K concentrations in 1-year-old foliage was significantly related ($p < 0.01$) to crown condition, with K in class 3 tree foliage < class 2 < class 1.

The exchangeable Ca, Mg, and Al concentrations in soils from the 0–15 and ≥15–30 cm depths at the base of the trees whose foliage was analyzed do not suggest that trees with poorer crowns were associated with soils with low levels of exchangeable Ca or Mg or low base cation / Al ratios. Table 2 shows that the most severely declining trees (class 3) tend to grow on the soils with the highest available Ca, though the differences in soils under trees of different crown classes are not statistically significant.

Soil solution studies

As suggested by hypothesis 3, we looked for evidence that soil solution levels of Al and Ca/Al ratios in soil solutions were in the range found to be unfavorable in laboratory experiments. Miller et al. (1992) report on a number of studies of dissolved Al in soil solutions collected at Whiteface Mountain. They show that labile (i.e., toxic) Al is higher in the forest floor soil solutions than in solutions obtained from the mineral soil (mean 76 vs. 46 μmol/L). Concentrations of toxic forms of Al found to inhibit root growth in solution culture experiments (180–250 μmol/L) were reached in only one winter sample over a 4-year period. From controlled field acid irrigation experiments, Miller et al. (1992) concluded that anion concentrations needed to be unrealistically high before Al reached phytotoxic levels because Al balanced only one-third or less of the anion charge. By comparison, D.W. Johnson et al. (1991) have shown soil-solution concentrations for dissolved Al collected in spruce stands in Great Smoky Mountain National Park. In that study, Al concentrations in solutions from each of the soil horizons averaged approximately 50–60 μmol, similar to the values measured at Whiteface Mountain. Peak values at the Great Smoky Mountain site appear to be greater than at Whiteface Mountain, approaching 200 μmol/L with pulses of nitrate and sulfate. At the Great Smoky Mountain site, there is no unusual mortality of red spruce nor signs of decline.

Figure 5 shows the Ca/Al ratio measured in soil solutions collected from highly acidic soils in declining spruce–fir stands on Whiteface Mountain. In three of the four growing seasons sampled, Ca/Al ratios in samples from all horizons were >1.0, a ratio for spruce root growth that has been cited as critical (e.g., Schlegel et al. 1992). In the summer of 1989, Ca/Al ratios declined to <1.0 in both mineral and organic horizons. In winter Al becomes dominant. Soil temperature measurements (E.K. Miller, unpublished data) indi-

TABLE 2. Relationship between crown condition and soil chemistry or foliar composition for 69 red spruce trees on Whiteface Mountain, New York

Crown condition*	Elevation	0–15 cm Ca‡	0–15 cm Ca/Al	0–15 cm Mg‡	0–15 cm Mg/Al	15–30 cm Ca‡	15–30 cm Ca/Al	15–30 cm Mg‡	15–30 cm Mg/Al	% dry wt. in needles† Ca	% dry wt. in needles† Mg	% dry wt. in needles† K
1	913^a	8.5^a	1.7^a	1.3^a	0.26^a	6.0^a	1.2^a	0.71^a	0.14^a	0.22^a	0.06^a	0.61^a
2	1062^b	6.5^a	0.84^a	0.77^a	0.09^a	5.4^a	2.3^a	0.50^a	0.14^a	0.21^a	0.06^a	0.57^a
3	1103^b	11.4^a	2.5^a	1.4^a	0.31^a	9.9^a	1.6^a	1.3^a	0.21^a	0.22^a	0.06^a	0.52^a

NOTE: Values in each column followed by the same letter are not significantly different by multiple range test (SAS Institute Inc. 1985).

*Crown class 1 = 0–10% foliage lost from live crown 2 = 11–50% lost; 3, >50% loss.

†Values are for 1986 needles.

‡Charge measured in cmol$_c$/kg soil (dry wt.)

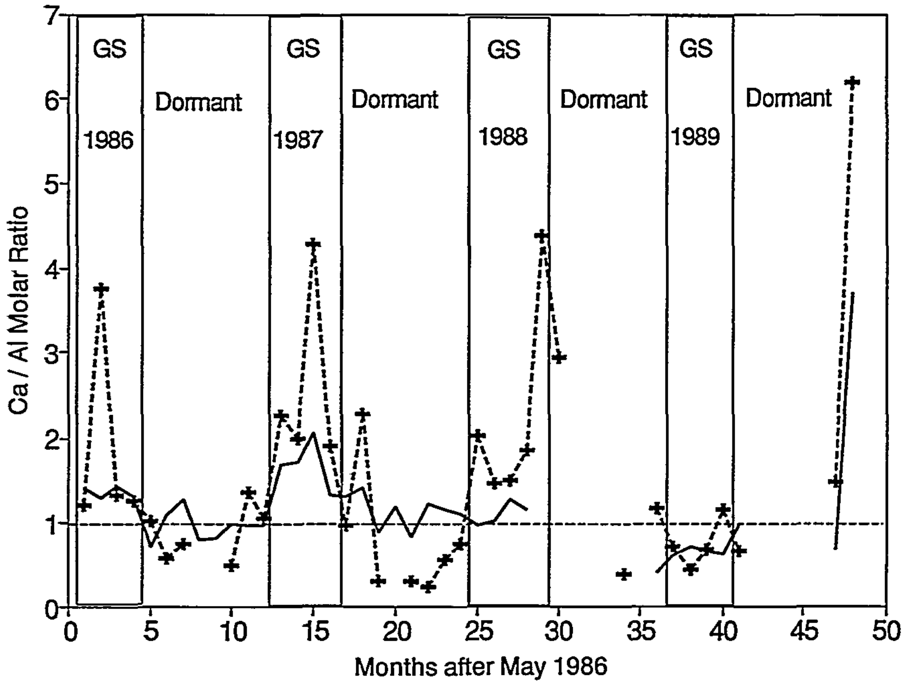

FIG. 5. Molar ratio of Ca/Al in soil solutions from organic (broken line) and mineral (solid line) horizons at 1020–1090 m on Whiteface Mountain. New York. Ratios are calculated on the basis of labile Al (0.62× total Al in organic horizons, and 0.77× total Al in mineral horizons). Soil solution data are from Miller et al. (1992). GS, growing season.

cate that the high Al/Ca values occur when most of the soil water is frozen, concentrating the remaining solution and, by necessity, raising the ratio of dissolved Al/Ca (e.g., Reuss 1983). Raynal et al. (1990) determined that soil solution Al at 100–200 μmol/L reduced base cation uptake in solution culture experiments. In our lysimeter studies, 100 μmol/L (total dissolved Al) was exceeded in approximately 5, 10, and 20% of the samples from the Bs, A/Bhs, and O horizons, respectively, with all occurrences in October–March.

Summary

Compared with other spruce–fir sites where soil, soil-solution, and foliar chemistry have been examined (e.g., D.W. Johnson et al. 1991; Joslin et al. 1992; Huntington et al. 1990), Whiteface Mountain is a relatively high base status site and it has experienced the greatest degree of spruce mortality. The hypotheses regarding spatial consistency between the severity of decline and low foliar Ca or low available Ca/Al ratios in soils are not supported by our data. Measurements of exchangeable pools of Ca, Mg, and Al, foliar concentration of base cations, and soil solution studies do not support arguments suggesting that Al is limiting Ca or Mg uptake. Based on the foliar samples and collected in 1986 and soils collected in 1986–1988, spruce trees and soils look to be well supplied with Ca. Levels of Al known to inhibit Ca uptake or reduce root growth (100–200 μmol/L) occur sometimes during the dormant season, but not during the growing season. The only indication of potential for Al interference with Ca uptake is from soil solutions collected during one of four growing seasons (1989) and inferences that this is detrimental to red spruce have been made from laboratory studies of Norway spruce (*Picea abies* (L.) Karst.), rather than from experiments with red spruce. Overall our data cannot be counted as support for the idea that Ca and Al imbalances are important stresses involved in spruce decline at Whiteface Mountain when the data are viewed in light of traditional approaches used to detect nutrient shortages in forest trees. We do acknowledge, however, that the type of soil and foliar measurements made in this study may not be adequate for detecting Ca shortages. If this is the case, then new criteria for detecting soil-induced Ca deficiency need to be developed.

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Bartlett, R.J., Ross, D.S., and Magdoff, F.R. 1987. Simple kinetic fractionation of reactive aluminum in soil solutions. Soil Sci. Soc. Am. J. **51**: 1479–1482.

Battles, J.J., Johnson, A.H., Siccama, T.G., Friedland, A.J., and Miller, E.K. 1992. Red spruce death: effects on forest composition and structure on Whiteface Mt., NY. Bull. Torrey Bot. Club, **119**: 418–430.

Bondietti, E.A., Baes, C.F., and McLaughlin, S.B. 1989. Radial trends in cation ratios in tree rings as indicators of the impact of atmospheric deposition on forests. Can. J. For. Res. **19**: 586–594.

Craig, B.W., and Friedland, A.J. 1991. Spatial patterns in forest composition and standing dead red spruce in montane forests of the Adirondacks and northern Appalachians. Environ. Monitoring Assess. **18**: 129–143.

DeHayes, D.H. 1992. Winter injury and developmental cold tolerance. *In* Ecology and decline of red spruce in the eastern United States. *Edited by* C. Eagar and M.B. Adams. Springer-Verlag, New York. pp. 295–337.

DeHayes, D.H., Thornton, F.C., Waite, C.E., and Ingle, M.A. 1991. Ambient cloud deposition reduces the cold tolerance of red spruce. Can. J. For. Res. **21**: 1292–1295.

Eagar, C., and Adams, M.B. (*Editors*). 1991. Ecology and decline of red spruce in the eastern United States. Springer-Verlag, New York.

Friedland, A.J., Gregory, R.A., Karenlampi, L., and Johnson, A.H. 1984. Winter damage to foliage as a factor in red spruce decline. Can. J. For. Res. **14**: 963–965.

Friedland, A.J., Hawley, G.J., and Gregory, R.A. 1988. Red spruce foliar chemistry in northern Vermont and New York, USA. Plant Soil, **105**: 189–195.

Hamburg, S.P. 1984. Effects of forest growth on soil nitrogen and organic matter pools following release from subsistence agriculture. *In* Forest Soils and Treatment Impacts. Proceedings of the 6th North American Forest Soils Conference, 19–23 June 1983, Knoxville, Tenn. *Edited by* E.L. Stone. pp. 145–158. University of Tennessee, Knoxville.

Huntington, T.G., Peart, D., Hornig, J., Ryan, D., and Russo-Savage, S. 1990. Relationships between soil chemistry, foliar chemistry, and condition of red spruce at Mount Moosilauke, New Hampshire. Can. J. For. Res. **20**: 1219–1227.

Johnson, A.H. 1992. The role of abiotic stresses in the decline of red spruce in high elevation forests of the eastern U.S. Annu. Rev. Phytopathol. **30**: 349–367.

Johnson, A.H., and Siccama, T.G. 1983. Acid deposition and forest decline Environ. Sci. Technol. **17**: 294A–305A.

Johnson, A.H., Friedland, A.J., and Dushoff, J.G. 1986. Recent and historic red spruce mortality: evidence of climatic influence. Water Air Soil Pollut. **30**: 319–330.

Johnson, A.H., Cook, E.R., and Siccama, T.G. 1988. Climate and red spruce growth and decline in the northern Appalachians. Proc. Natl. Acad. Sci. U.S.A. **85**: 5369–5373.

Johnson, A.H., McLaughlin, S.B., Adams, M.B., Cook, E.R., DeHayes, D.H., Eagar, C., Fernandez, I.J., Johnson, D.W., Kohut, R.J., Mohnen, V.A., Nichols, N.S., Peart, D.R., Schiver, G.A., and White, P.S. 1991. Synthesis and conclusions from epidemiological and mechanistic studies of red spruce decline. *In* Ecology and decline of red spruce in the eastern United States. *Edited by* C. Eagar and M.B. Adams. Springer-Verlag, New York. pp. 385–411.

Johnson, D.W., and Lindberg, S.E. (*Editors*). 1991. Atmospheric deposition and forest nutrient cycling. Springer-Verlag, New York.

Johnson, D.W., Van Miegroet, H., Lindberg, S.E., Harrison, R.B., and Todd, D.E. 1991. Nutrient cycling in red spruce forests of the Great Smoky Mountains. Can. J. For. Res. **21**: 769–787.

Joslin, J.D., Kelly, J.M., and Van Miegroet, H. 1992. Soil chemistry and nutrition of North American spruce–fir stands: evidence for recent change. J. Environ. Qual. **21**: 12–30.

Leopold, D.J., Reschke, C., and Smith, D.S. 1988. Old-growth forests of Adirondack Park, NY. Nat. Areas J. **8**: 166–189.

McLaughlin, S.B., and Kohut, R. 1991. The effects of atmospheric deposition and ozone on carbon allocation and associated physiological processes in red spruce. *In* Ecology and decline of red spruce in the eastern United States. *Edited by* C. Eagar and M.B. Adams. Springer-Verlag, New York.

McLaughlin, S.B., Andersen, C.P., Edwards, N.T., Roy, W.K., and Layton, P.A. 1990. Seasonal patterns of photosynthesis and respiration of red spruce saplings from two elevations in declining southern Appalachian stands. Can. J. For. Res. **20**: 485–495.

McLaughlin, S.B., Andersen, C.P., Edwards, N.T., Hanson, P.J., Tjoelker, M.G., and Roy, W.K. 1991. Increased dark respiration and calcium deficiency in red spruce in relation to acidic deposition at high-elevation southern Appalachian Mountain sites. Can. J. For. Res. **21**: 1234–1244.

Miller, E.K., Huntington, T.G., Johnson, A.H., and Friedland, A.J. 1992. Aluminum in soil solutions from a subalpine spruce–fir forest at Whiteface Mt., NY. J. Environ. Qual. **21**: 345–352.

Peart, D.R., Nicholas, N.S., Zedaker, S.M., Miller-Weeks, M.M. 1992. Condition and recent trends in high elevation red spruce populations. *In* Ecology and decline of red spruce in the eastern United States. *Edited by* C. Eagar and M.B. Adams. Springer-Verlag, New York. pp. 125–192.

Perkins, T.D., Adams, G.T., and Klein, R.M. 1991. Desiccation or freezing? Mechanisms of winter injury to red spruce foliage. Am. J. Bot. **78**: 1207–1217.

Raynal, D.J., Joslin, J.D., Thornton, F.C., Schaedle, M., and Henderson, G.S. 1990. Sensitivity of deciduous and coniferous tree species to Al. III. Red spruce and loblolly pine. J. Environ. Qual. **19**: 163–171.

Reuss, J.O. 1983. Implications of the Ca–Al exchange system for the effect of acid precipitation on soils. J. Environ. Qual. **12**: 591–595.

Robarge, W.P., and Johnson, D.W. 1992. The effects of acidic deposition on forested soils. Adv. Agron. **47**: 1–83.

SAS Institute Inc. 1985. SAS user's guide: statistics, version 5 edition. SAS Institute Inc., Cary, N.C.

Schlegel, H., Amundson, R.G., and Huttermann, A. 1992. Element distribution in red spruce (*Picea rubens*) fine roots: evidence for Al toxicity at Whiteface Mountain. Can. J. For. Res. **22**: 1132–1138.

Scott, J.T., Siccama, T.G., Johnson, A.H., and Breisch, A.R. 1984. Decline of red spruce in the Adirondacks, New York. Bull. Torrey Bot. Club, **111**: 438–444.

Shortle, W.S., and Bondietti, E.A. 1992. Timing, magnitude, and impact of acidic deposition on sensitive forest sites. Water Air Soil Pollut. **61**: 253–267.

Shortle, W.C., and Smith, K.T. 1988. Aluminum-induced calcium deficiency syndrome in declining red spruce. Science (Washington, D.C.), **240**: 239–240.

Silver, W.L., Siccama, T.G., Johnson, C., and Johnson, A.H. 1991. Changes in red spruce populations in montane forests of the Appalachians. Am. Midl. Nat. **125**: 340–347.

Thomas, G.W. 1982. Exchangeable cations. *In* Methods of soil analysis. Part 2. *Edited by* A.L. Page, R.H. Miller, and D.R. Keeney. Agronomy, 9(2): 161–164.

Van Miegroet, H., Johnson, D.W., and Todd, D.E. 1993. Foliar response of red spruce to fertilization with Ca and Mg in Great Smoky Mountain National Park. Can. J. For. Res. 23: 89–95.

Vann, D.R., Strimbeck, G.R., and Johnson, A.H. 1991. Effects of ambient levels of airborne chemicals on freezing resistance of red spruce foliage. For. Ecol. Manage. **51**: 69–79.

Wargo, P.M., Bergdahl, D.R., Tobi, D.R., and Olson, C.W. 1993. Root vitality and decline of red spruce. Biol. Arbor. **4**: 1–134.

Witty, J.E. 1968. Classification and distribution of soils on Whiteface Mt., Essex Co., NY. Ph.D. thesis, Cornell University, Ithaca, N.Y.

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