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Foliar Nutrient Analysis of Sugar Maple Decline: Retrospective Vector Diagnosis

Victor R. Timmer and Yuanixin Teng¹

Abstract

Accuracy of traditional foliar analysis of nutrient disorders in sugar maple (*Acer saccharum* Marsh) is limited by lack of validation and confounding by nutrient interactions. Vector nutrient diagnosis is relatively free of these problems. The technique is demonstrated retrospectively on four case studies. Diagnostic interpretations consistently suggest that decline incidence involves Ca and Mg deficiency induced by toxic accumulation of Mn.

Introduction

The evaluation of nutrient sufficiency in trees is complex because there are many nutrients considered essential for vigorous tree growth. These nutrients are usually required in different amounts and balanced proportions, and their availability to trees may be affected by interactions occurring between different nutrients and also between nutrients and other elements in the soil or plant system. The interactions are difficult to detect and quantify, thus confounding interpretations of simple measures of deficiency and excess of nutrient supply. These problems are particularly pertinent to diagnosing nutritional symptoms of maple decline because of the widespread belief that multiple rather than single factors of stress may be contributing to the incidence of disorders (Cote and Oimet 1996). Thus reliable diagnostic techniques must have the capacity to cope effectively with multiple nutrients and their complex interactions.

Under such circumstances, foliar analysis rather than soil chemical analyses is the preferred method of assessing tree nutrient status since elemental composition of the leaf is considered a more direct index of nutrient availability to trees compared to measures of soil nutrient supply. It is also simpler to screen multiple element status (such as both macro- and micronutrients) by foliar analyses rather than by soil analysis because fewer and less complex laboratory procedures are involved. Traditionally, three major interpretive techniques have been applied to assess leaf chemistry of sugar maple: the critical concentration concept, the nutrient ratio approach and the DRIS norm system. Each technique has inherent limitations in terms of interpretive accuracy and reliability (Timmer 1991). This paper will introduce a fourth approach to foliar analysis interpretation, called vector nutrient diagnosis that has not been previously applied to sugar maple decline disorders. The technique is more comprehensive than the others, and has the potential to improve diagnostic power.

Current Approaches

We reviewed recent studies relating sugar maple leaf chemistry to decline ratings or dieback symptoms in a range of stand conditions in eastern Canada and the United States (Bernier and Brazeau 1988; Bernier and Brazeau 1988; Cote and Camire 1995; Cote et al 1995; Cote and Ouimet 1996; Fyles et al 1994; Heisey 1995; Kolb and McCormick 1993; Liu et al 1997; Mader and Thompson 1969; McLaughlin 1992; Ouimet et al 1995; Ouimet et al 1996; Ouimet and Fortin 1992; Wilmot et al 1995; Wilmot et al 1996) to determine the most popular diagnostic technique used. We noted a clear reliance on pre-established "critical" or "threshold" concentrations of elements as indicators of nutrient sufficiency. Although simple in application, the diagnostic reliability and sensitivity of the critical level approach must be questioned. Published critical concentrations for mature sugar maple are highly variable and poorly defined (Kolb and McCormick 1993; van den Burg 1985), they are seldom verified and validated by controlled fertilization experiments (Timmer 1991), and may not account for nutrient interactions. Interpretations can be confounded by inconsistencies of leaf tissue concentration with age, season and development stage, and by possible dilution and accumulation effects when comparing plants or plant components of unequal size (Timmer 1991).

In general, diagnoses offered in these studies tended to be qualitative rather than quantitative, focusing more on nutrient deficiency rather than toxicity because of the lack of published critical toxicity levels, and the difficulty in distinguishing between luxury consumption and toxic uptake above sufficiency concentrations. All studies screened macronutrient status, only a few included micronutrients, which implies that the role of micronutrients and their interactions were not considered crucial in diagnosing this disorder. In three studies (Cote and Camire 1995; Cote et al 1995; Long et al 1997), diagnostic interpretations were supplemented by comparing leaf nutrient status of healthy and unhealthy trees, and using optimum nutrient ratios or DRIS norms to assess nutrient balance. This approach is more inclusive than that based solely on critical levels, but reliability suffers from similar problems of weak definition, calibration and validation. Vector diagnosis may avoid the interpretive problems associated with traditional foliar analyses because its application is independent on pre-established critical levels, nutrient ratios or DRIS indices, and the technique has been calibrated and validated by fertilization trials. We have applied this approach retrospectively to foliar chemistry data published in these studies to demonstrate its potential in evaluating nutritional problems associated with maple decline.

Vector Diagnosis

Graphical vector diagnosis is explained in detail (Timmer 1991), and has been reviewed by Weetman 1989 and Haase

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and Rose 1995, thus it will only be briefly described here. Initially developed by Timmer and Stone 1978, the technique has undergone periodic refinements to enhance reliability and flexibility. Interpretations of complex multi-vector responses were simplified by normalization procedures (Timmer and Morrow 1984). Identifying nutrient interactions was improved by comparing multiple nutrients in similar space (Teng and Timmer 1990a; Teng and Timmer 1990b; Timmer and Teng 1990). Concepts of steady-state nutrition were introduced by integrating dynamic or temporal parameters in the system (Imo and Timmer 1997). Recently, interspecific nutrient competition between species was identified by adopting a two-crop format of the technique (Imo and Timmer 1997).

Its application recognizes the biological principle that growth of plants is dependent on nutrient uptake, hence the nutrient concentration in the plant is a function of two fundamental processes: nutrient uptake and biomass accumulation. This relationship is examined by comparing growth and nutrient status of trees, or tree components, differing in health and/or productivity in a nomogram that plots biomass (accumulation) on the upper horizontal axis, nutrient (uptake or) content on the lower horizontal axes, and corresponding nutrient concentration (nutrient content divided by component biomass) on the vertical axis (Figure 1). When normalized to a specified reference sample (usually the control set to 100), differences are depicted as vectors because of shifts in both direction and magnitude. Diagnosis is based on vector direction of individual nutrients, identifying occurrence of deficiency (C), sufficiency (B), luxury consumption (D), toxicity (E), antagonism (F) and dilution (A). Each configuration corresponds to a specific phase in dose response curves relating changes [increasing (+), decreasing (-), or none (0)] in plant growth, nutrient content and nutrient concentration to increasing soil nutrient supply (Timmer 1991). Vector magnitude reflects the extent or severity of specific diagnoses, and facilitates relative ranking and prioritizing.

When only part of the tree is sampled (usually the case with large trees, not seedlings), the presumption is that biomass changes in plant components (such as shoots or foliage) accurately reflect growth changes of the sample trees (Timmer and Morrow 1984). In this exercise, it was assumed that routine measures of maple decline in stands, such as crown transparency, branch defoliation, canopy dieback, growth decline, etc. are closely correlated with tree productivity. Hence these measures served as surrogate estimates of foliar biomass in the nomograms. Accordingly, the upper and lower horizontal axes in Figures 2, 3 and 5 were labeled as indices of foliar biomass and nutrient content.

Case Studies

We present here data from studies that monitored both macronutrients and micronutrients in foliage in relation to a range of maple decline. The nomograms (Figure 2 and 3) show a common pattern of the largest downward, left-pointing vectors associated with Ca and Mg (Shift F, antagonism in Figure 1), and the largest, upward left-pointing vectors associated with Mn (Shift E, toxicity in

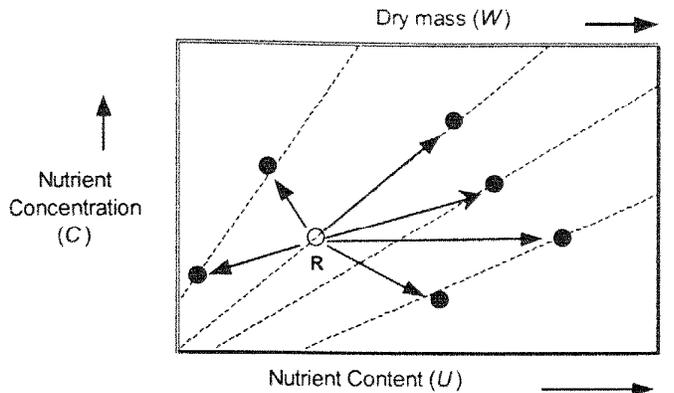
Figure 1). Vector length increased with reduced foliar biomass or the severity of decline. Since the Mn vectors were larger than corresponding Ca or Mg vectors, diagnosis suggest that the disorder involved a toxic build up of Mn that inhibited uptake of Ca and Mg uptake, i.e., a case of Mn-induced Ca and Mg deficiency. Thus, as decline ratings increased, uptake (content) of base cations (mainly Ca and Mg) in foliage was severely depressed, while uptake (content) of Mn was slightly reduced in highly affected trees, or increased in less affected trees.

The mechanism could be explained by soil acidification increasing exchangeable Mn levels in the soil, which resulted in excess or toxic build up of this ion in trees (Marschner 1995). High levels of soluble Mn may in turn displace exchangeable Ca, Mg or K on the soil exchange complex inducing deficiencies of these nutrients for the trees. It is also well known in agriculture that within the plant high supplies of Mn may inhibit transport of Ca and Mg into fast growing tissues (Graham et al 1988).

Our diagnosis was supported indirectly by growth and nutritional responses of trees in independent fertilization experiments testing soil base status and liming treatments. Response patterns of sugar maple seedlings raised on a gradient of soil base saturation and pH under greenhouse conditions (Figure 4) were similar to those of Figure 2 and 3. Biomass and uptake of Ca and Mg in foliage was markedly reduced (Shift F) as base saturation of the soil decreased (Figure 4). Since Mn uptake was little changed, and concentration increased appreciably (Shift E), interpretations suggest growth inhibition due to Mn-induced Ca and Mg deficiency. Logically, the problem could be alleviated by effective liming to increase pH and base status of the soil. Liming of mature sugar maple affected by decline raised soil pH and stimulated tree growth, while reducing crown dieback symptoms (Figure 5). The response was accompanied by increased uptake of Ca and Mg (Shift C, deficiency in Figure 1) and decreased uptake of Mn (Shift F) in foliage. Apparently, applications of dolomitic limestone corrected a deficiency of Ca and Mg, and antagonistically reduced Mn uptake thus counteracting possible Mn toxicity.

Conclusion

We have limited our demonstration of retrospective vector nutrient diagnoses to four studies. Vector nomograms (not shown here) of other studies (Cote and Camire 1995; Liu et al 1997; Ouimet et al 1995; Ouimet and Fortin 1992; Wilmot et al 1995; Wilmot et al 1996) that monitored mostly macronutrient status in foliage, revealed similar patterns of reduced uptake of Ca and Mg (or K on some Quebec sites) in declining trees, supporting the diagnosis that base cation limitation may be associated with maple dieback. However, interactions with Mn could not be confirmed because of the lack of Mn data. We surmise from our combined results that sugar maple decline may be linked to induced base cation deficiency that is caused by toxic Mn accumulation in the rooting zone of soils. This preliminary diagnosis needs to be confirmed by field trials testing controlled additions of Mn to the soil in problem stands.



Vector shift	Change in relative			Nutritional effect	Nutrient status	Diagnosis
	W	U	C			
A	+	+	-	Dilution	Non-limiting	Growth dilution
B	+	+	0	Accumulation	Non-limiting	Sufficiency, steady-state
C	+	+	+	Accumulation	Limiting	Deficiency response
D	0	+	+	Accumulation	Non-limiting	Luxury consumption
E	-	-, +	+	Concentration	Excess	Toxic accumulation
F	-	-	-	Antagonism	Limiting	Induced deficiency by E

Figure 1.—Interpretation of directional changes in relative dry mass and nutrient status of plants (or plant components) contrasting in growth and/or health. The reference condition (R) is usually normalized to 100. Diagnosis (A to F) is based on shifts (increase [+], decrease [-] or no change [0]) of individual nutrient characterized in dose response curves relating plant growth, nutrient concentration, and nutrient content to increasing soil nutrient supply. Vector magnitude reflects extent or severity of the diagnosis identified (modified from Timmer 1991). Note that results in this paper involve mostly vectors E and F, suggesting that the toxic accumulation of nutrient E antagonistically induced a deficiency of nutrient F.

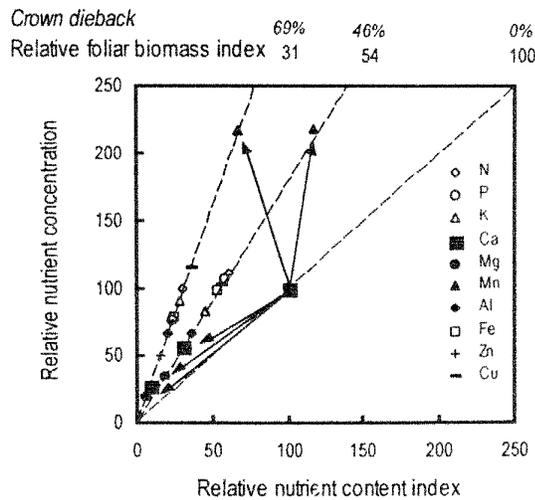


Figure 2.—Relative biomass index, nutrient concentration and nutrient content index of foliage of sugar maple stands of various degrees of crown dieback severity in Pennsylvania (Kolb and McCormick 1993). Crown dieback was defined as the proportion (percentage) of total crown volume containing dead branches with tips less than 2.5 cm in diameter. Rating was visually scored using the North American Maple Decline Project system. Relative foliar biomass index was calculated as 100 minus crown dieback rating. Foliar nutrient content index was the product of relative nutrient concentration and relative biomass index. Status of the healthy stand (with 0 to 5% crown dieback) was normalized to 100. Note that dieback severity increased with higher Mn uptake and lower Mg and Ca uptakes, suggesting a diagnosis of Ca and Mg deficiency induced by Mn toxicity.

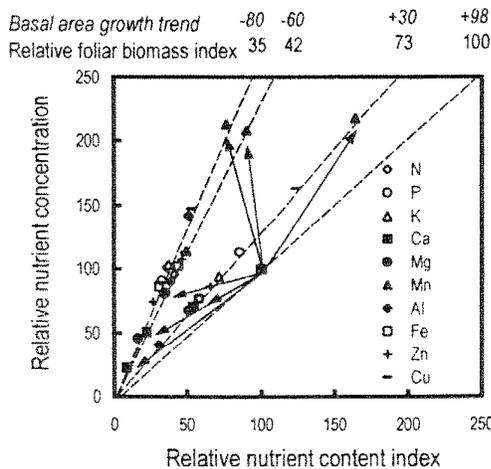


Figure 3.—Relative foliar biomass index, nutrient concentration and nutrient content index of sugar maple stands of similar age, but varying in relative basal area growth (RAG) compared to a 60-yr growth period (1928-1987) in Pennsylvania (Heisey 1995). The trend is positive if RAG increased and negative if it declined. Relative foliar biomass index was estimated as a linear function of RAG for the 1978-87 period. Foliar nutrient content index was the product of nutrient concentration and biomass index. Nutrient status of the stand with the highest RAG difference (+98) was normalized to 100. Note that basal area growth trends decreased with higher Mn uptake and lower Mg and Ca uptake, suggesting Ca and Mg deficiency induced by Mn toxicity.

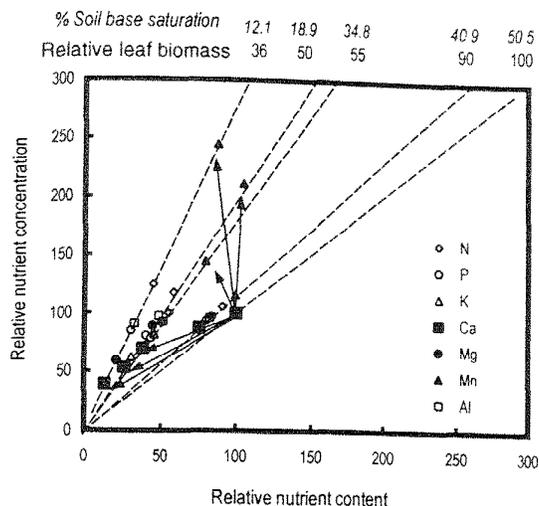


Figure 4.—Effect of soil base saturation on relative leaf biomass, nutrient concentration, and nutrient content of sugar maple seedling in a greenhouse pot trial in Quebec (Ouimet et al. 1996). The soil was a strongly acidic (pH = 4.1) sandy loam podzol with low base saturation (5%). A gradient of increasing soil base saturation was created by adding base cation solutions. The seedlings were transplanted to the pots at cotyledon stage and grown for three months. Note that low soil base saturation inhibited leaf biomass and depressed uptake of Ca and Mg most, and Mn least.

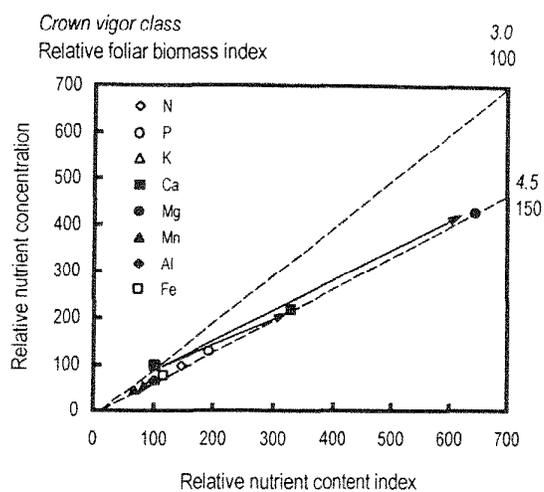


Figure 5.—Response of declining maple trees to liming in Pennsylvania (Long et al. 1997). The liming treatment was a single application of commercial pulverized dolomitic limestone (Ca = 21%, Mg = 12%, CaO equivalent = 58.8) at 22.4 Mg.ha⁻¹. Foliar biomass index was estimated as a linear function of total basal area. Nutrient status of the unlimed plots was normalized to 100. Foliar nutrient content index was the product of nutrient concentration and biomass index. Note that the positive biomass response was associated with depressed Mn uptake (-65%), and enhanced Ca (+330%) and Mg (+640%) uptake, suggesting that liming alleviated Ca and Mg deficiency, and countered Mn toxicity by reducing Mn availability.

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Root Dynamics and Nutrient Allocation in Sugar Maple

Ronald L. Hendrick¹

Abstract

My colleagues, Kurt Pregitzer and Andy Burton, and I have been studying root dynamics in sugar maple forests in Michigan for the past 10 years. We have used a combination of traditional soil cores, minirhizotron video images and physiological techniques in an effort to understand patterns of carbon and nutrient allocation to roots, particularly the ephemeral, small diameter roots.

With respect to the role of roots in ecosystem annual net primary production (NPP), roots <2.0 mm in diameter account for about 60% of ecosystem NPP. These same roots account for about 50 – 60% of the total amount of N returned through above and below ground litterfall. Foliage production and litterfall account for about 1/3 of NPP and N returns, respectively. About 85% (numbers) of the roots are less than 0.50 mm in diameter (70% of biomass and 80% of length), and less than 2% exceed 1.0 mm. The average diameter of sugar maple fine roots is about 0.35 – 0.37 mm. In addition to their overall importance to the C and N economy of sugar maple forests, we have measured a substantial degree of simultaneity in root production and mortality throughout the year. Traditional approaches that rely on measurements of temporal biomass changes to quantify root production and mortality would yield estimates about 50% too low in these forests. Combined with the tendency to use a relatively coarse (1.0 – 2.0 mm) screen through which to wash and recover roots, we believe that the importance of very fine roots has been underestimated in at least some previous root studies.

We have used minirhizotrons to follow the appearance and fate of individual roots as a measure of root longevity. In sugar maple forests, about 40% of all annual root production occurs prior to and coincident with canopy expansion. Most mortality occurs after leaf fall and over winter. Root mortality rates for any time period during the year are the same for all roots produced that year, regardless of when they first appeared. However, mortality rates drop dramatically over

winter, and remain low for roots surviving into their second growing season. However, roots produced the subsequent year die off at the same, higher mortality rates as did roots produced the previous year.

Based upon the above data, it appears that changing environmental conditions during the growing season do not affect mortality rates. However, we have measured substantial differences among sites arrayed along a north-south gradient of about 400 km. In earlier studies we observed that root mortality rates were highest at our warmest, most southern site, suggesting that higher rates of root respiration might be associated with shorter root life spans. However, we subsequently measured equally high root mortality rates at the coolest, most northern site. Root chemistry assays and soil nutrient dynamics data revealed that root nitrogen content, and ultimately root nitrogen supply potential, explain the differences. Contrary to observations from other forests, high levels of soil N increase sugar maple root lifespan, regardless of soil temperature regime. Greater tissue N concentrations were associated with higher respiration rates at all temperatures, but high rates of respiration do not appear to be shortening root lifespans.

Further studies of the relationship between root dynamics and soil temperature, matric potential and site water balance showed that there are no strong effects of the soil environment on the dynamics of fine roots at our sites. There is some tendency for periods of abundant water to be associated with lower mortality and higher production rates, but the relationship is weak. There does appear to be a temporal correspondence between the onset of periods of high, midsummer water demand and the production of deep (80 – 100 cm) roots, and between autumn leaf fall and the death of the deep roots, but it is highly variable spatially.

Our current efforts are focused upon better understanding the role that soil N, mycorrhizae, and root physiology play in regulating root dynamics, and how controls over root dynamics in sugar maple forests compare with a variety of coniferous and deciduous forests across North America.

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Mechanisms of Base-cation Depletion by Acid Deposition in Forest Soils of the Northeastern U.S.

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Abstract

Several studies have indicated a long-term decrease in exchangeable Ca in forest soils within the northeastern United States, but the regional extent of these decreases and the importance of acid deposition as a cause has not been fully resolved. Results from two recently completed studies have provided the opportunity to further investigate the role of acid deposition in depletion of base cations in northeastern forest soils. These two studies entailed 1) a survey of soils in northeastern red spruce (*Picea rubens* Sarg.) stands, which were selected to encompass the range of environmental conditions to which this species is subjected, and 2) a study of base-cation leaching in soils of a hardwood watershed in the Catskill Mountains of New York.

Results from the survey of spruce stands indicated that exchangeable Ca concentrations varied considerably in the Northeast as a result of differences in parent material mineralogy. A stronger relationship between relative weathering potential of parent material and exchangeable Ca concentrations was observed in the Oa horizon than in the B horizon of soils at these sites. Concentrations of exchangeable Ca in the B horizon may have decreased to a level at which further decreases are buffered by high concentrations of exchangeable Al.

An upslope increase in atmospheric deposition of SO_4^{2-} corresponded to an upslope increase in the leaching of base cations, and an upslope decrease in acid-neutralizing capacity of stream water in the Catskill Mountain watershed. Upslope gradients in natural factors that could increase base-cation leaching were not detected. Decreases in concentrations of exchangeable base cations were approximately balanced by increases in concentrations of exchangeable Al. Greater Al mobilization in the mineral soil by acid deposition at upper elevations than at lower elevations was the most probable explanation for the upslope increase in leaching of base cations and stream water acidity.

Low Ca to Al ratios in mineral soil solution were found to be related to high levels of Al saturation in the forest floor in the survey of spruce stands. Mobilization of Al in the mineral soil tends to increase Al saturation in the forest floor by making Al available for upward transport by biocycling and water movement. Reversal of past decreases in exchangeable Ca concentrations will be hindered by 1) continued inputs of mobile anions that facilitate leaching, 2) high Al saturation in

the mineral soil that will interfere with uptake of Ca by roots, 3) increasing Al saturation in the forest floor, and 4) decreasing trends of atmospheric deposition of Ca.

Introduction

The possibility that acid deposition could deplete base cations in soils was acknowledged in the early stages of acid rain research in the 1970's (Cowling and Dochinger 1980). The effect of acid deposition on forest soils remained unresolved at the close of the National Acid Precipitation Program (NAPAP) in 1990, however, because (1) there was no historical soil data to show that changes in soil Ca concentrations had actually occurred, either through natural processes or acid deposition, (2) release of bases through weathering could possibly compensate for losses from leaching, and (3) the importance of natural sources of acidity relative to acid deposition was unknown. A comprehensive literature review by Shortle and Bondietti (1992) provided the first indication that exchangeable base concentrations may have decreased in the second half of the 20th century. This paper showed that, before 1950, concentrations of exchangeable Ca plus Mg in the soils of coniferous forests in the Northeast were generally about twice that of concentrations measured after 1970. The first study to directly compare Ca concentrations measured early in the century with those measured after the discovery of acid deposition in the 1970's showed that extractable Ca concentrations were significantly lower in 1984 than in 1930-32; a result attributed to forest aggradation in this study (Johnson et al. 1994a). Detailed Ca budgets developed on Whiteface Mountain, NY, indicated that the exchangeable Ca pool in the forest floor of this site declined from 1986 to 1990 (Johnson et al. 1994b) at a rate greater than that found in the study of Johnson et al. (1994a). In this study, a long-term decreasing rate of atmospheric deposition of Ca, identified by Hedin et al. (1994), was cited as a possible cause for the decrease in the 80's (Johnson et al. (1994b). The development of ecosystem Ca budgets through the use of stable isotopes of Sr (a biogeochemical analog of Ca) at Whiteface Mountain, NY (Miller et al. 1993), and Cone Pond, NH (Bailey et al. 1996) provided additional evidence of decreasing pools of exchangeable Ca in the Northeast.

The results of these studies indicate that decreases in exchangeable soil-base concentrations have occurred at several locations within the Northeast, but the regional extent of these changes, and the importance of acid deposition as their cause was not fully resolved. Results from two recently completed studies have provided the opportunity to further investigate the role of acid deposition in depletion of base cations in northeastern soils. In the first of these projects, soil and soil solution samples were collected from 12 red spruce stands (*Picea rubens* Sarg.) in New York, Vermont, New Hampshire and Maine (Figure 1), which were chosen to encompass the range of environmental conditions and stand health experienced by

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Sites sampled in this study

- 1 Sleepers River
- 2 Groton
- 3 Howland
- 4 Kossuth
- 5 Barlett
- 6 Hubbard Brook
- 7 Whiteface Mtn
- 8 Crawford Notch
- 9 Bear Brook
- 10 Big Moose Lake
- 11 Cone Pond
- 12 Mt Abraham
- 13 Neversink River

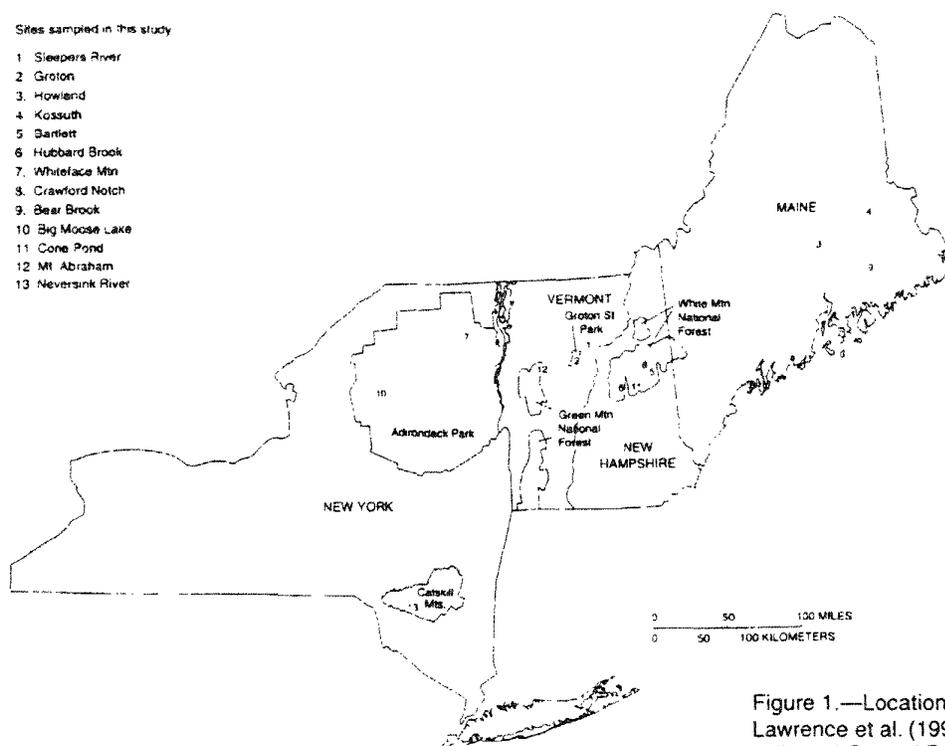


Figure 1.—Locations of red spruce stands where Lawrence et al. (1997) and Lawrence et al (in press) collected Oa and B horizon soil samples.

red spruce in this region. In the second project, soil-base leaching was studied over an elevational gradient in a hardwood forested watershed in the Catskill Mountains of southeastern New York (Figure 1). The purpose of this report is to integrate the published results of these two studies to summarize the mechanisms through which acid deposition affects the availability of base cations in the forest soils Northeast.

Methods

Survey of Soil Chemistry in Northeastern Red Spruce Stands

Red spruce was the predominant species in each of the 12 stands, although other coniferous and deciduous species were common. Increment cores indicated that the average age of canopy spruce at each site ranged from 96 to 175 years. Apparent stand health varied from good to poor where extensive dieback had occurred. Stand elevations ranged from 80 m at Howland ME to 975 m on Whiteface Mountain, NY. Wet deposition of SO_4 at six of the sites, was determined from on-site monitoring; at the remaining six sites, wet deposition was estimated from the nearest monitored site. Details of how wet deposition estimates were derived are available in Lawrence et al. (1997). Wet-only atmospheric deposition of SO_4 ranged from $12.7 \text{ mmol m}^{-2} \text{ yr}^{-1}$ at 150 m elevation at Kossuth, ME to $28.8 \text{ mmol m}^{-2} \text{ yr}^{-1}$ at 810 m elevation in Crawford Notch, NH.

A relative value of calcium weathering potential was assigned on the basis of general mineralogy, determined from bedrock maps and a till-source model that identified rock types that could be present in non-local till (Bailey and Hornbeck, 1992). Relative weathering potential varied from 1 at sites underlain by metapelite and quartzite, to 3 at sites influenced by calcareous schist. Further details about these sites including ratings of relative Ca weathering potential are available in Lawrence et al. (1997). Soils at all sites were classified as Spodosols.

Soil samples were collected from the faces of nine individual soil pits at each site (grouped in three plots, each with three pits). Eight of the 12 sites were sampled in mid-June and late September or early October of 1992 and 1993; four of the 12 sites were only sampled once (early summer or early fall) in each of the two years. In each pit, a sample was collected from the middle of the Oa horizon and from the top 10 cm of the B horizon. This sampling design was chosen to compliment assessments of red spruce tree condition conducted in conjunction with the soils investigation. Sampling in the early summer and early fall was timed to coincide with the periods when nutrient concentrations in foliage were both high and low for purpose of biochemical studies of foliage processes. The three individual soil samples from each horizon, in each plot, were combined into one sample and thoroughly mixed before chemical analysis to yield one sample from each horizon in each plot. All samples were analyzed for exchangeable cations (Ca, Mg, Na and K) by extraction with $1M \text{ NH}_4\text{Cl}$ (Blume et al.

1990) and exchangeable acidity (Al, H) by extraction with 1M KCl (Thomas 1982). Samples from Oa horizons collected on two dates at each site were analyzed for acid-extractable-Ca concentrations (Friedland et al. 1984), and Oa-horizon samples collected on one date were analyzed for total Ca concentrations by neutron activation (Parry 1991). Mineral Ca concentrations were calculated by subtracting concentrations of acid-extractable Ca from concentrations of total Ca; reactive nonexchangeable Ca concentrations were calculated by subtracting concentrations of exchangeable Ca from acid-extractable Ca concentrations. Exchangeable Ca is probably the most representative form of plant-available Ca.

The study of Johnson et al. (1994a), in which Ca concentrations in Adirondack Mountain soils in 1930-32 were compared to values in 1984, followed the methods of Heimburger (1934), which were not directly comparable to the method of Friedland et al. (1984); a commonly used method for determination of extractable Ca concentrations in the 1980's and 1990's. Methods tests by Lawrence et al. (1997) showed that the data from the studies of Johnson et al. (1994a) and Heimburger (1934) yielded concentrations about 25% higher than the Friedland et al. (1984) method. This factor was applied to the data of Johnson et al. (1994a) and Heimburger (1934) to allow comparisons with data from other studies.

Soil solutions were collected from all soil samples by expulsion (described and evaluated in Lawrence and David 1996). The procedure entails 1) manually packing field moist soil in a PVC cylinder to reduce porosity, 2) adding simulated throughfall solution to saturate the soil column, 3) discarding solution that drains off by gravity, and 4) applying air pressure (172 kPa for Oa horizon samples; 138 kPa for B horizon samples) to force out the solution that is collected for analysis of total Al, total monomeric Al and organic monomeric Al by methods in Lawrence et al. (1995). Concentrations of polymeric Al were determined by subtracting total monomeric Al concentrations from total Al concentrations; inorganic monomeric Al concentrations were determined by subtracting organic monomeric Al concentrations from total monomeric Al concentrations.

As part of the regional survey of spruce stands, soil chemistry also was evaluated in spruce stands of two adjacent watersheds in eastern Maine. Through the EPA-sponsored Watershed Manipulation Project 1800 equiv ha⁻¹ yr⁻¹ of (NH₄)₂SO₄ was applied to one of these watersheds between 1989 and 1995 (Norton et al. 1994). This treatment resulted in significantly higher concentrations of NO₃ in stream water of the treated watershed than the reference watershed, in response to elevated nitrification rates in soil. The effects of increased nitrification rates and associated acidity on Al mobilization were evaluated by determining pH and concentrations of total Al, total monomeric Al and organic monomeric Al in soil solutions collected by expulsion, as described above. To further investigate possible changes in soil Al concentrations associated with changes in soil acidity, archived Oa horizon soil collected in spruce stands in the Hubbard Brook Experimental Forest, NH, in 1969-1970, 1987 and 1992, were analyzed for

concentrations of exchangeable and acid-extractable Ca and Al, by the methods given above.

Soil-Base Leaching in Hardwood Forested Watershed

The effects of acid deposition on base leaching were studied over an elevational gradient (approximately 800 m to 1230 m) in Winnisook watershed, a subbasin of the Neversink River, in the Catskill Mountains of NY. Atmospheric deposition was measured along the gradient at 14 stations from 799 m to 1234 m near the summit of the watershed. At each station, four 20-cm diameter funnels collected throughfall solutions that were combined by station for analysis. During November through April, the funnels were replaced with galvanized cylinders lined with plastic bags, to collect frozen precipitation. Throughfall measurements have been shown to be a reliable method for estimating total atmospheric S deposition (Lindberg and Garten, 1988; Lindberg and Lovett, 1992, Lawrence and Fernandez, 1993). Throughfall collections also were used to estimate water input to the watershed.

This watershed is forested primarily by *Acer saccharum*, *Betula alleghaniensis*, and *Fagus grandifolia*, but stands of *Abies balsamea* are common along upper elevation ridges. Soils are classified as Inceptisols. Soil samples were collected from the B horizon at 48 locations along the elevational gradient, by taking equal volumes of soil from each 10 cm increment of the mineral profile, which were mixed before analysis to yield one B horizon sample from each location. Exchangeable cation concentrations were determined by methods used in the survey of northeastern spruce stands. Mineral soil was also collected from a single pit downslope of Winnisook watershed at 570 m elevation where soil-base saturation was higher than within Winnisook watershed. This soil (the 0-10 cm layer below the O horizon) was thoroughly mixed and put in nylon mesh bags, which were then placed beneath the Oa horizon at 35 locations along the elevational gradient. The approach followed the buried bag method described in David et al. (1990). Bags were removed and chemically analyzed after one year by the methods used in the survey of spruce stands.

Relations among acid deposition, soil base saturation and base leaching were further evaluated by leaching solutions of H₂SO₄, HNO₃, CaCl and NaCl through B horizon soil of three different base saturation values. Ionic strength (0.32 to 0.52 mM_c L⁻¹) and pH (3.7 to 4.8) of the added solutions were similar to mean values measured in Oa horizon soil solutions in the Neversink watershed (D.A. Burns, USGS, unpublished data). Soil samples used for the buried bag experiment were also used for the leaching study; samples with the highest base saturation (14.4%) had not been placed in the soil profile in Winnisook watershed, whereas the other samples (base saturation 9.6% and 4.5%) had been in the soil profile of Winnisook watershed for one year. Solutions were extracted from the soil samples with a mechanical vacuum extractor (Blume et al. 1990). Duplicate extractions were done for each treatment. Concentrations of base cations (Ca, Mg, Na and K) and acid anions (SO₄, Cl, and NO₃) were measured in leachates, as described in Lawrence et al. (1995).

Table 1.—Exchangeable Ca concentrations as $\text{cmol}_e \text{kg}^{-1}$ and as a percentage of cation exchange capacity (Ca saturation) for the Oa and B horizons of 12 red spruce stands and 2 hardwood stands in the Northeast. Samples were collected from the upper 10 cm of the B horizon in the spruce stands, from the Bh horizon at Hubbard Brook. In Winnisook Watershed equal volume samples were collected at 10 cm depth increments through the entire B horizon, then mixed before analysis.

SITE	Oa Horizon		B Horizon	
	($\text{cmol}_e \text{kg}^{-1}$)	(($\text{cmol}_e \text{kg}^{-1}$) CEC ⁻¹)100	($\text{cmol}_e \text{kg}^{-1}$)	(($\text{cmol}_e \text{kg}^{-1}$) CEC ⁻¹)100
SPRUCE STANDS				
Sleepers River, VT ¹	22	51	0.68	6.9
Groton, VT ¹	14	41	0.32	13
Howland, ME ¹	11	34	0.19	3.8
Kossuth, ME ¹	6.3	24	0.19	2.6
Bartlett, NH ¹	11	31	0.18	3.4
Hubbard Brook, NH ¹	7.9	25	not sampled	not sampled
Whiteface Mt., NY ¹	6.3	26	0.57	7.3
Crawford Notch, NH ¹	6.8	21	0.30	3.1
Bear Brook, ME ¹	5.3	21	0.32	4.1
Big Moose Lake, NY ¹	6.4	20	0.37	3.1
Cone Pond, NH ¹	3.5	10	0.11	1.6
Mt. Abraham, NY ¹	2.1	9.1	0.41	4.0
HARDWOOD STANDS				
Hubbard Brook, NH ²	6.75	38.5	0.63	8.4
Winnisook Watershed, NY ³	2.31	13.4	0.16	2.7

¹ From Lawrence et al. (1997)

² From Johnson et al. (1991)

³ Previously unpublished data, G.B. Lawrence, U.S. Geological Survey

Stream water samples were collected weekly at the base of Winnisook watershed and approximately monthly at seven upstream sites from May 1991 through September 1994. These samples were analyzed for Ca, Mg, Na, K, SO_4 , Cl, NO_3 , total monomeric Al, and organic monomeric Al (methods described in Lawrence et al. 1995). To evaluate elevational trends in stream chemistry that were most influenced by soils (and least influenced by ground water), only samples collected on dates with the highest 20% of stream flows were included.

Results and Discussion

Current Status of Soil Calcium

Concentrations of exchangeable Ca, the predominant base cation in northeastern forest soils, varied by a factor of 10 in the Oa horizon of the 12 red spruce sites (Table 1). There were no geographical patterns other than high concentrations in eastern Vermont; the concentrations at the Big Moose Lake site were similar to those measured in Maine, and the concentration at the Bartlett, NH site, in the central White Mountains, was three times that of Cone Pond, in the southern White Mountains of New Hampshire. Exchangeable Ca concentrations expressed as Ca saturation (Ca as a percentage of total cation-exchange capacity) varied by a factor of 5. Exchangeable Ca concentrations in the upper 10 cm of the B horizon at these sites were less varied than in the Oa horizon, and 1 to 2 orders of magnitude lower (Table 1). Geographic patterns of

B horizon concentrations also differed somewhat from the Oa horizon; Whiteface Mountain, NY and Mt. Abraham, VT had relatively high concentrations in the B horizon, but these sites had concentrations in the Oa horizon that were among the lowest. Although soils in hardwood stands are generally considered less acidic than soils in conifer stands, exchangeable Ca concentrations in the Oa and B horizons in Winnisook watershed were in the lower range of the spruce stands, and the average value reported for a hardwood watershed at Hubbard Brook was slightly less than that measured in the spruce stands at Hubbard Brook (Table 1).

Exchangeable Ca was the largest Ca fraction in the Oa horizon of all spruce sites except those with a mineral-matter content greater than 30 percent (Whiteface, NY, Bear Brook, ME, and Mt. Abraham, VT), where mineral-bound Ca was the largest fraction (Figure 2). Reactive-nonexchangeable Ca was generally a minor component. Averaged for the 12 sites, acid-extractable Ca was comprised of 92 percent exchangeable Ca and 8 percent reactive-nonexchangeable Ca.

Concentrations of acid-extractable Ca measured in the spruce survey ranged from $13.9 \text{ mmol kg}^{-1}$ to 103 mmol kg^{-1} (Figure 3). This range encompassed values determined by McNulty et al. (1991) for 11 red spruce stands, also in New York, Vermont, New Hampshire and Maine. The average of samples collected in 59 red spruce stands throughout the Adirondack Mountains of New York by Johnson et al. (1994a) fell in the middle of the range of the spruce survey data of Lawrence et al. (1997), but the average value

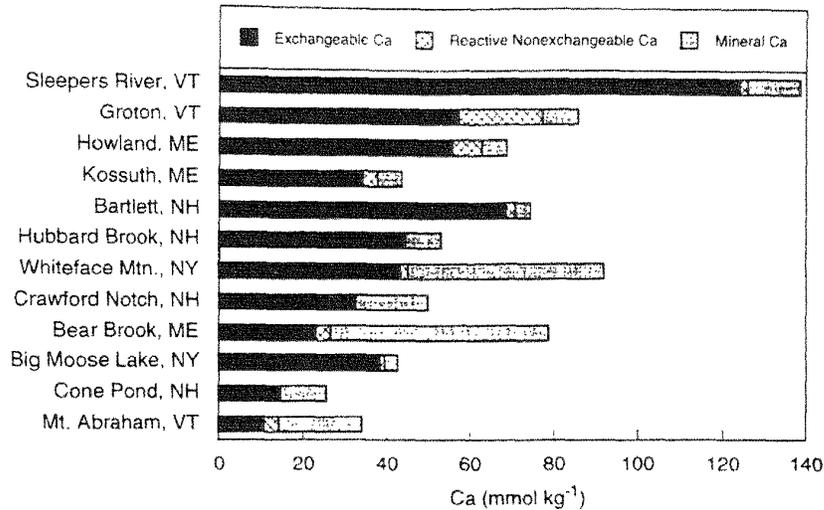


Figure 2.—Mean concentrations of exchangeable Ca, reactive nonexchangeable Ca (acid-extractable Ca concentrations minus exchangeable Ca concentrations) and mineral Ca (total Ca concentrations minus acid-extractable Ca concentrations) in the Oa horizon of 12 northeastern red spruce stands. Reprinted with kind permission from Kluwer Academic Publishers (Biogeochemistry, vol. 38, 1997, Assessment of soil calcium in red spruce forests in the northeastern United States, Lawrence, G.B., David, M.B., Bailey, S.W., and Shortle, W.C., figure 4, p. 30, copyright 1997).

obtained for the same sites by Heimburger in 1930-34 was approximately 30 mmol kg⁻¹ higher, and in the upper quarter of the range determined in the spruce survey of Lawrence et al. (1997).

Causes of Regional Variability

The considerable variability observed in exchangeable Ca concentrations in the spruce survey of Lawrence et al. (1997) could be a result of spatial variations in (1) net Ca uptake by trees, (2) atmospheric deposition of Ca, (3) acid deposition, or (4) mineralogy of parent material. Analysis of increment cores of 54-108 live trees at each site indicated that all of the red spruce stands averaged approximately 100 years in age or older and had been experiencing similar rates of declining growth since 1965. The extent of mortality was highly variable from site-to-site, however, which suggested that net uptake of Ca may also have varied from site-to-site. Variations in net Ca uptake could therefore not be ruled out as a factor in the observed spatial variability, although neither stand health nor stand age were related to exchangeable Ca concentrations. Atmospheric deposition of Ca was also unlikely to be the primary cause of variability in exchangeable Ca concentrations among the spruce sites because deposition of Ca decreases consistently from west to east (Table 2), whereas soil Ca concentrations did not show a spatial pattern.

Table 2.—Atmospheric wet deposition of Ca (kg ha⁻¹ yr⁻¹) in the northeastern United States, measured by the NADP/NTN network (<http://nadp.sws.uiuc.edu/>).

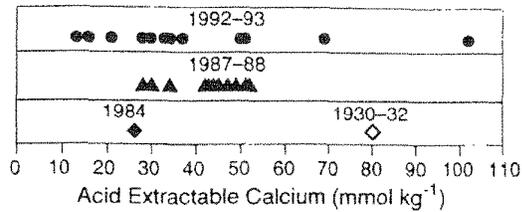
	1985	1995
Bennett Bridge, NY	1.95	1.60
Huntington Forest, NY	1.03	0.68
Underhill, VT	1.09	0.70
Hubbard Brook, NH	0.69	0.53
Greenville, ME	0.63	0.42

Calcium saturation was weakly related to wet deposition of SO₄ in the Oa horizon and unrelated to wet deposition of SO₄ in the B horizon (Figure 4), but was highly correlated with relative weathering potential in the Oa horizon. A weaker relation between Ca saturation and relative weathering potential was observed in the B horizon than the Oa horizon, despite the greater abundance of parent material in the mineral soil (Figure 4).

This relation suggests that acid deposition was unlikely to be the primary cause of variation in exchangeable Ca concentrations among sites, however enhanced leaching by acid deposition in the mineral soil can explain why Ca saturation is more strongly related to relative weathering potential in the Oa horizon than the B horizon. If base saturation is less than 20%, solute concentrations, and therefore cation leaching, are strongly controlled by exchangeable Al, which results in high rates of Al leaching and low rates of Ca leaching (Reuss, 1983). For example, in soil with a base saturation of 4.5%, only 0.11 meq L⁻¹ of Ca are released per meq L⁻¹ of added acid anion (Figure 5). The base saturation of the B horizon at all 12 spruce stands was less than 14% (Figure 6), whereas base saturation in the Oa horizon was greater than 20% at 10 stands, and greater than 16% at all 12 stands. Acidic deposition may have lowered base saturation in the B horizon to levels where further changes in base saturation were buffered by Al, which obscured the effect of differences in parent material. Parent material was more clearly expressed in the Oa horizon where vegetative recycling reduces loss of Ca to leaching.

Acid Deposition and Ca Leaching

Additional information on leaching of Ca from soil was obtained in Winnisook watershed in the recent study by Lawrence et al. (in press). Within this watershed, atmospheric deposition of SO₄²⁻ increased by a factor of 2 from the lowest to the highest elevations in the watershed



EXPLANATION

- Mean concentrations of calcium in soils of individual sites in New York, Vermont, New Hampshire and Maine sampled by Lawrence and others (1997) in 1992-3.
- ▲ Mean concentrations of calcium in soils of individual sites in New York, Vermont, New Hampshire and Maine sampled by McNulty and others (1991) in 1987-88.
- ◇ Mean concentrations of calcium in soil of 38 samples collected by Heimbürger (1934) throughout the Adirondack Mountains of New York in 1930-32.
- ◆ Mean concentrations of calcium in soil of 59 samples collected by Johnson and others (1994a) throughout the Adirondack Mountains of New York in 1984.

Figure 3.—Acid-extractable Ca concentrations in Oa horizons of red spruce stands in the northeastern US measured in this study and the studies of McNulty et al (1991), Johnson et al (1994a), and Heimbürger (1934). Values from this study and the study of McNulty et al (1991) represent means of individual sites; the values from Johnson et al (1994a) represents the mean of 59 samples collected throughout the Adirondack Mountains of New York; the values from Heimbürger (1934) represent the mean of 38 samples collected throughout the Adirondack Mountains of New York. Values reported by Johnson et al (1994a) and Heimbürger (1934) have been adjusted for method bias. To express Ca concentrations on a soil mass basis loss-on ignition values of 0.90 were assumed for the data from the McNulty et al study and the Johnson et al study.

(fig. 7a), whereas the concentrations of exchangeable base cations in both undisturbed soil and soil bags, and the ratio of base cations to acid anions in stream water, decreased from the lowest to the highest elevations (fig. 7b,c,d). The decrease in exchangeable base cation concentrations in the soil bags was approximately balanced by an increase in exchangeable Al concentrations (Lawrence et al. in press).

Effects of the spatial trend in SO_4^{2-} deposition may be analogous to the effects of past temporal trends in acidic deposition through a space-for-time substitution. This approach assumes that the elevational gradient of SO_4^{2-} deposition in Winnisook watershed has been maintained during past decades when acidic deposition rates both increased and decreased. Natural acidification processes may cause elevational gradients in the chemistry of soils and stream water, however (Lawrence et al., 1986). An elevational increase in net uptake of Ca by vegetation could contribute to an elevational trend in exchangeable base-cation concentrations in soil, as could decreased temperatures and increased abundance of conifers at upper elevations that would lead to a greater accumulation of organic acids and higher leaching rates than at lower

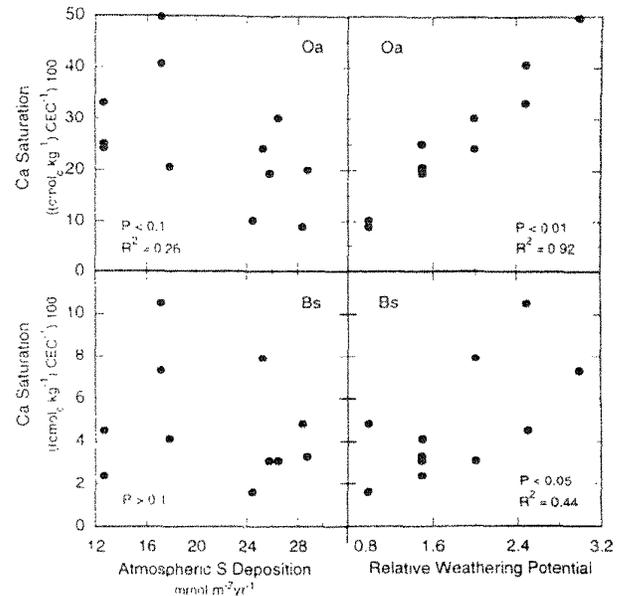


Figure 4.—Ca saturation as a function of atmospheric deposition and relative weathering potential in Oa and Bs horizons. Reprinted with kind permission from Kluwer Academic Publishers (Biogeochemistry, vol. 38, 1997, Assessment of soil calcium in red spruce forests in the northeastern United States, Lawrence, G.B., David, M.B., Bailey, S.W., and Shortle, W.C., figure 5, p. 31, copyright 1997).

elevations. Upslope increases in precipitation, decreases in evapotranspiration (ET), or decreases in soil depth would cause an elevational gradient in soil-water flux per volume of soil, which could also result in an elevational gradient in soil chemistry. Lastly, spatial variations in the mineral composition of parent material could result in an elevational gradient in base-cation concentrations in soil.

Winnisook watershed had been selectively cut up to about 900 m elevation before 1950, but we found no physical or historical evidence that the watershed was ever cleared or burned (Kudish, 1985). The cutting that had been done would most likely cause a decrease rather than an increase in exchangeable base-cation concentrations at low elevations by increasing aggradation rates. Spatial variations in tree species could not explain these spatial trends because all soil investigations (sampling and soil bag incubations) were done in similar stands of yellow birch (*Betula alleghaniensis* Britton), beech (*Fagus grandifolia* Ehrh.), and sugar maple (*Acer saccharum* Marsh). Vegetation uptake also did not explain the upslope decrease in exchangeable base-cation concentrations because decreased temperatures would tend to shorten the growing

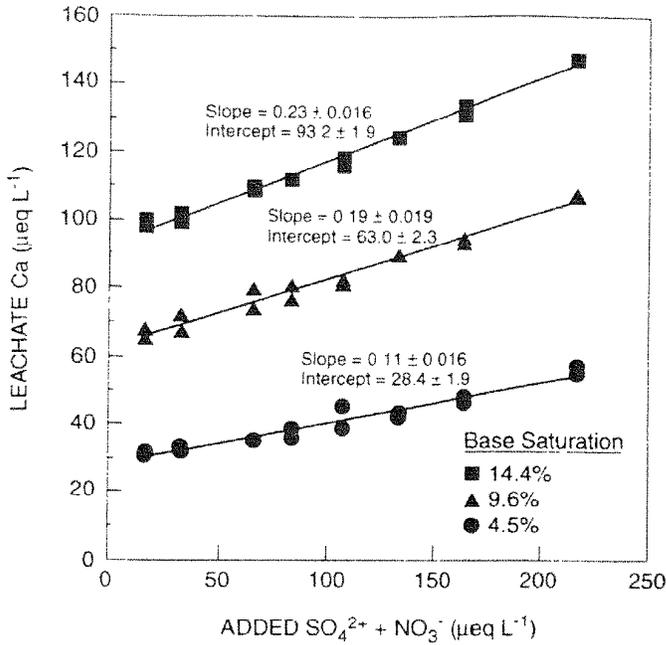


Figure 5. Concentrations of Ca in solutions leached through mineral soil with a vacuum extractor, as a function of SO_4 plus NO_3 concentrations in the added solution. Results of duplicate leachings are shown. Slope and intercepts of linear best-fit lines are given \pm 95% confidence intervals (two-sided test). Soils with initial base saturations of 14.4%, 9.6% and 4.5 % were used in the experiment. Reprinted with permission from the Ecological Society of America (Ecological Applications, in press, Soil calcium status and the response of stream chemistry to changing acidic deposition rates in the Catskill Mountains, New York, Lawrence, G.B., David, M.B., Lovett, G. M., et al., figure 8, copyright 1999).

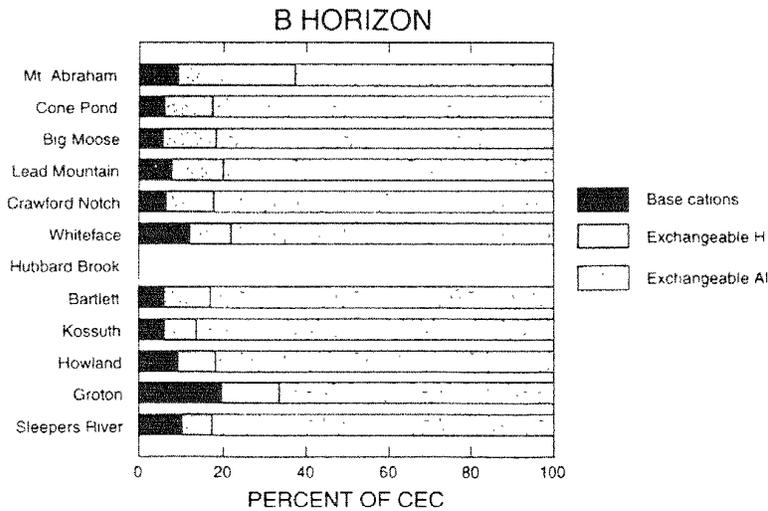


Figure 6.—Concentrations of exchangeable base cations, H and Al expressed as a percent of cation-exchange capacity for B horizons of the 12 northeastern red spruce stands. Reprinted with permission from Williams & Wilkins, (Soil Science, vol. 161, 1996, Soil and soil solution chemistry under red spruce stands across the northeastern United States, David, M.B., Lawrence, G.B., modified from figure 2, p. 320), copyright 1996).

season and decrease net uptake of Ca at upper elevations. A spatial trend in the intensity of podzolization from greater accumulation of organic acids at upper elevations than lower elevations was ruled out as a cause of the gradient in exchangeable base-cation concentrations because no spatial trends were observed in 1) organic carbon concentration in mineral soil, 2) cation exchange capacity of the mineral soil, or 3) pH of the Oa horizon.

Evidence of substantially higher soil water flux at upper elevations was also not apparent. Two years of throughfall measurements did not indicate an increase in precipitation

with increasing elevation within the watershed, and no trend in soil depth was detected from the 48 soil pits. Annual ET, estimated by application of the adiabatic lapse rate (Anthes et al. 1975), resulted in an increase in soil-water flux of 14.5% from base to summit. The upslope increase in soil-water flux over the elevation range where soil bags were incubated was less than half this value, but nevertheless, a distinct gradient in base-cation concentrations developed within 1 year in soil of uniform parent material. Further details of the gradient analysis in this watershed are given in Lawrence et al. (in press).

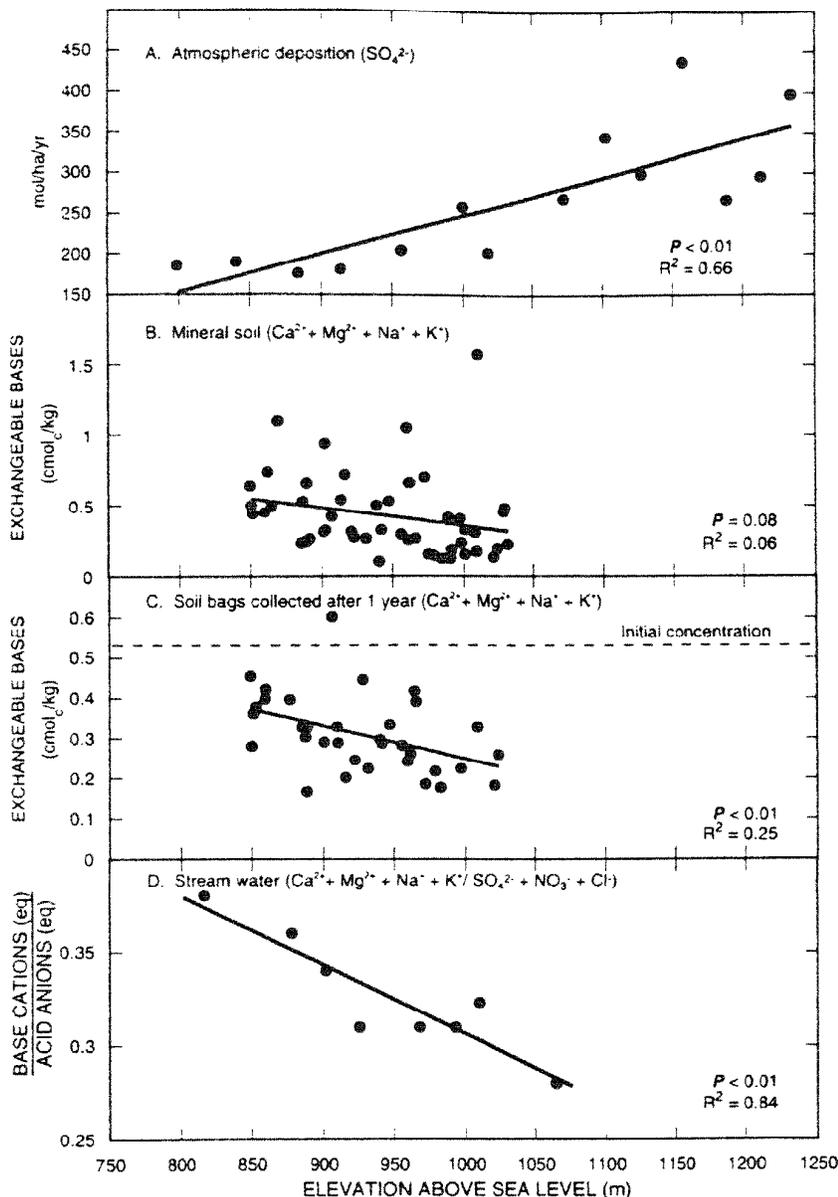


Figure 7.—Elevational gradients of selected chemical constituents in Winnisook watershed: A) annual mean SO_4 deposition measured in throughfall, June 1993 through May 1995, B) exchangeable base-cation concentrations (Ca, Mg, Na and K) in mineral soil samples collected in 1994, C) exchangeable base-cation concentrations in mineral soil samples buried in the mineral profile in mesh bags for one year, and D) the ratio of base-cation concentrations to acid-anion concentrations (SO_4 , NO_3 and Cl) in stream-water samples collected during the highest 20% of stream flows from during May 1991 through April 1997. Reprinted with permission from EOS, Transactions, American Geophysical Union, v. 79, No. 16, 1998, p. 200.

In the absence of acidic deposition it is possible that natural acidification processes would cause elevational gradients in soil-base saturation and stream chemistry in Winnisook watershed, but an inability to detect significant gradients in any acidifying factors other than acidic deposition suggests that the effects of these factors were subtle relative to the gradient in atmospheric deposition of SO_4^{2-} , which doubled from the base to the summit of the watershed.

Aluminum Mobilization

B Horizon

Further indication that acid deposition has altered soil chemistry is evident from investigations of episodic

acidification of streams in western Pennsylvania, in the Adirondack Mountains, and in Biscuit Brook, a tributary of the Neversink River, approximately 10 km from Winnisook watershed (Wigington et al., 1996). As streamflow increases during episodic periods of precipitation or melting snow, concentrations of inorganic monomeric Al can increase to levels that are toxic to fish (Baker et al. 1996). Concentrations of Al increase with increasing flow because a greater amount of water discharges directly from the soil into the stream channel than during baseflow (Chen et al. 1984; Lawrence et al., 1988; Potter, 1988). Water that has passed through the subsoil before discharging to the stream channel has had a greater opportunity for neutralization and immobilization of Al. Analysis of ionic controls of inorganic

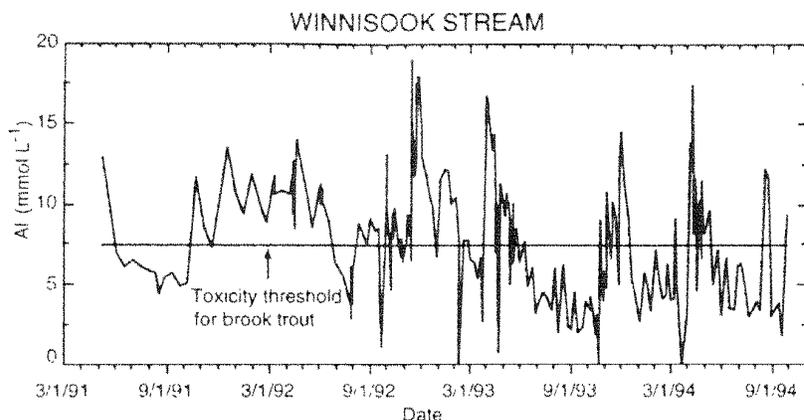


Figure 8.—Concentrations of inorganic monomeric Al in stream water at the base of Winnisook watershed from May 1991 through September 1994.

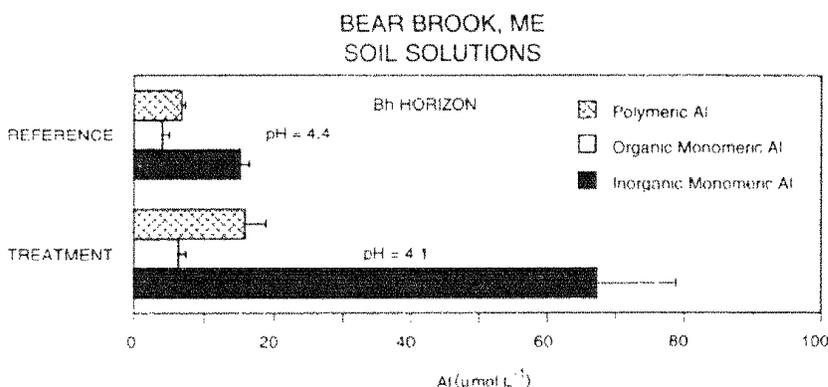


Figure 9.—Concentrations of inorganic monomeric Al (total monomeric Al minus organic monomeric Al), organic monomeric Al and polymeric Al (total Al minus total monomeric Al), in soil solutions of B horizons in treated (additions of $(\text{NH}_4)_2\text{SO}_4$) and reference stands of Bear Brook watershed, Maine. Each bar represents the mean of 45 samples that were combined into 15 samples for analysis, except for polymeric Al, which represents 33 samples combined into 11 samples. Standard errors are indicated by horizontal lines. Mean pH for reference and treatment stands is also given. Reprinted with permission from the American Chemical Society (Environmental Science and Technology, vol. 31, 1997). Response of aluminum solubility to elevated nitrification in soil of a red spruce stand in eastern Maine, Lawrence, G.B., David, M. B., modified from figure 1, p. 827, copyright 1997).

monomeric Al concentrations in stream water showed that increased concentrations during high flows were associated with increased concentrations of SO_4^{2-} and NO_3^- (Wigington et al., 1996). Naturally occurring organic acids can also mobilize Al and increase concentrations in stream water, but this process forms organically complexed Al, which is not toxic to fish (Driscoll et al., 1980). Concentrations of inorganic monomeric Al measured in the stream of Winnisook watershed from 1991-93 frequently exceeded the toxicity threshold for brook trout (Figure 8), and recent fish surveys indicated that the stream in Winnisook watershed does not support fish because of toxic concentrations of inorganic monomeric Al (Barry Baldigo and Lawrence, in press). Landowners of Winnisook watershed have reported, however, that a population of brook trout (*Salvelinus fontinalis*) previously existed in the stream (Royal Whiting, personal communication). High inorganic Al concentrations

and the loss of a previously existing fish population suggests that an increase in inorganic monomeric Al concentrations occurred in this stream sometime in the past. An increase such as this would not occur without an increase of inorganic acidity in the mineral soil. An example of the effect of increased inorganic acidity on Al mobility is shown in Figure 9. Addition of $(\text{NH}_4)_2\text{SO}_4$ to the treated watershed at Bear Brook Maine increased nitrification rates, lowered pH, and increased concentrations of inorganic monomeric Al in the mineral soil solution (Lawrence and David 1997).

Results of the soil survey of spruce stands showed that Al dominated exchange chemistry in all samples of mineral soil; a factor that is also reflected in Ca to Al ratios in these soils that are well below 1.0 in both soil solution and on exchange sites (Figure 10). Inorganic acids (H_2SO_4 and HNO_3) introduced by acid deposition are the most probable

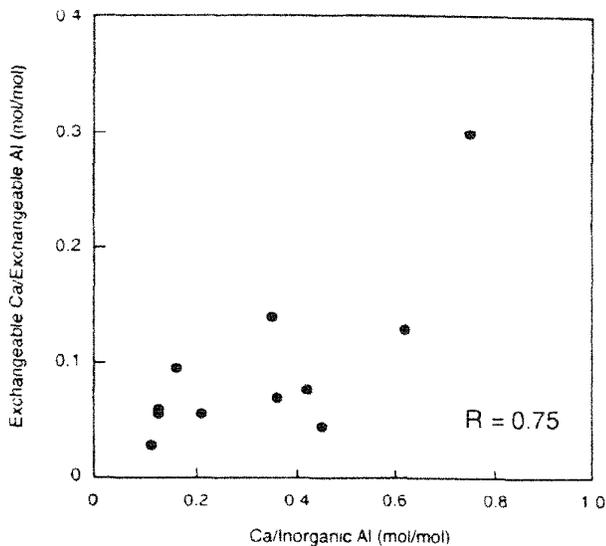


Figure 10.—Ratios of exchangeable Ca to exchangeable Al as a function of ratios of Ca to inorganic Al in soil solution of B horizons in the 12 northeastern red spruce stands. Insufficient mineral soil was available for sampling in the spruce stand at Hubbard Brook. Reprinted with permission from Williams & Wilkins, (*Soil Science*, vol. 161, 1996, Soil and soil solution chemistry under red spruce stands across the northeastern United States, David, M.B., Lawrence, G.B., modified from figure 6, p. 326), copyright 1996).

cause of high concentrations of inorganic Al in solution and on exchange sites in the mineral soil. Organic acids that are leached from the forest floor also acidify the mineral soil and mobilize Al, but this process is limited by the tendency of organic acids to be removed from solution by adsorption to mineral particle surfaces (DeConinck, 1980). As a result, the mineral soil generally has a higher pH than the forest floor where the organic acidity originates. The high mobility of H_2SO_4 and HNO_3 in the mineral soil, however, makes these acids more effective than organic acids at decreasing the solution pH, leaching cations and increasing inorganic monomeric Al concentrations in soil solutions and stream waters.

Oa Horizon

Although acid deposition is likely to have decreased solution pH (and increased Al mobility) in the mineral soil, acid deposition is unlikely to have caused a decrease in pH in the naturally acidic forest floor. Production of organic acids through decomposition caused soil solution pH values to range from 3.3 to 3.8 in the Oa horizon of the 12 spruce stands (Lawrence et al. 1995). These values are below the pH of precipitation in this region, which averages about 4.4 and is seldom less than 4.0 (NADP/NTN, 1993). There is no established mechanism to explain how addition of

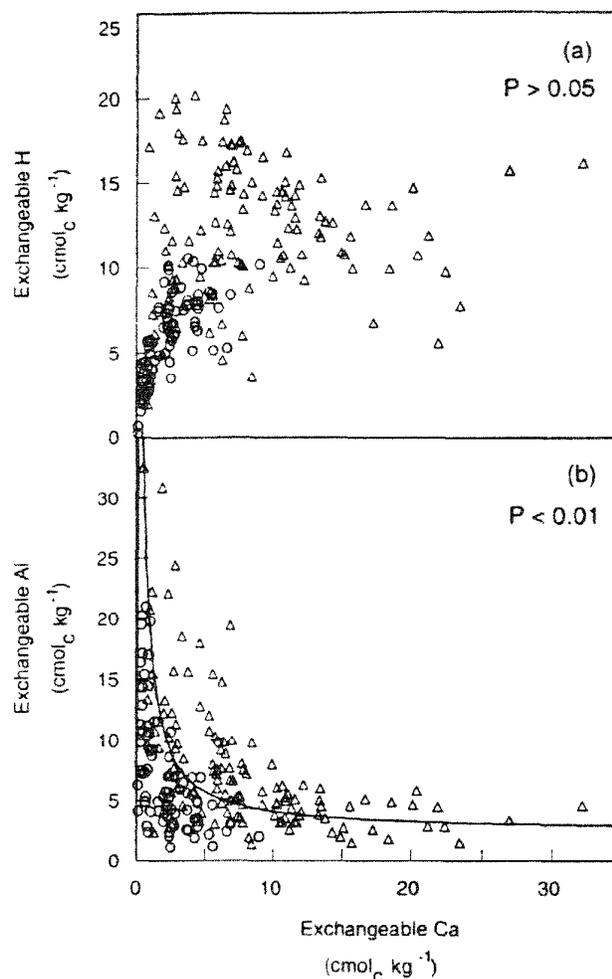


Figure 11.—Concentrations of (a) exchangeable H, and (b) exchangeable Al, as a function of exchangeable-Ca concentrations in the Oa horizon of stands of the 12 northeastern red spruce sites (triangles) and mixed hardwoods in Winnisook watershed (circles). Each triangle represents the value of three samples that were combined before analysis. Hardwood data are based on 68 individual samples. Reprinted by permission from *Nature* (vol. 378, 9 November 1995, p.163), copyright 1995, MacMillan Magazines Ltd.

precipitation above pH 4.0 could increase mobilization of Al within forest floors that have solution pH values normally less than 4.0. Although the low pH of these Oa horizons suggests that Ca availability is controlled by H, concentrations of exchangeable Ca in the Oa horizon were unrelated to exchangeable H concentrations in the 12 spruce stands and in Winnisook watershed, but inversely related to exchangeable Al concentrations (Figure 11).

The lack of a mechanism to explain mobilization of Al within the forest floor suggests that there is an outside source of Al

to this horizon. Mixing with the mineral soil when trees become uprooted provides a mechanism for introducing mineral forms of Al into the forest floor. Analysis of Al chemistry in the forest floor indicate that mineral content was not a strong controlling factor of exchangeable Al concentrations, however; dissolved Al concentrations in the Oa horizon were highly undersaturated with respect to Al bearing minerals, exchangeable Al concentrations were uncorrelated with mineral Al concentrations, and dissolved Al concentrations were unrelated to pH (Lawrence et al., 1995).

Alternatively, inorganic Al mobilized in the mineral soil by acidic deposition may be transported into the forest floor, where it outcompetes Ca (and other base cations) for exchange sites due to the high affinity of Al for the functional groups of organic matter (DeConinck 1980). Rustad and Cronan (1995) found that biocycling (uptake of Al by roots that is transported upward through the tree then returned to the soil in leaves and branches) plus atmospheric deposition of Al, was roughly 60% of forest-floor leaching losses in a red spruce stand in Maine. Biocycling of Al in the mineral soil is likely at all of the northeastern red spruce stands because ratios of Ca to inorganic Al in soil solutions are considerably less than 1.0, the value defined by Cronan and Grigal (1995) below which Al competes effectively with Ca for uptake by roots. Ratios of exchangeable Ca to Al in Winnisook watershed (0.04 mol mol⁻¹) suggest that ratios in soil solution would also be less than 1.0 (Figure 10).

A rising water table provides an additional way to increase the reactive Al concentrations in organic surface horizons, as does upward movement of water through capillary action from drying of the forest floor by evapotranspiration (Lawrence et al., 1995). When mineral-soil solution, enriched in Al relative to Ca, moves into the forest floor, Al is more effectively retained so that the Ca to Al ratio in solution draining back into the mineral soil has increased, but the ratio of exchangeable Ca to exchangeable Al in the Oa horizon has decreased. A positive correlation between exchangeable Al content of the forest floor and the ratio of inorganic Al concentrations to Ca concentrations in the mineral soil solution supports this interpretation (Figure 12). Analysis of a limited number of archived soil samples collected in 1969-1970, 1987 and 1992, also show an increase in exchangeable and extractable forms of Al and a decrease in these forms of Ca (Table 3).

Potential decreases in the availability of Ca in the forest floor are particularly significant in light of the low Ca to Al ratios measured in the mineral soil. Although the forest floor is the primary horizon for nutrient uptake, the original source of mineral nutrients has generally been considered to be weathering in the mineral soil. Two recent studies have indicated, however, that 30 to 50% of available Ca in the forest floor originates from atmospheric deposition (Miller et

TABLE 3.—Concentrations in the Oa horizon of spruce-fir stands at the Hubbard Brook Experimental Forest, New Hampshire, USA. Samples collected in 1969 and 1970 were averaged together, as were those collected in 1987 and 1992. Values are means based on 9-14 samples. Statistically significant differences ($P < 0.05$) between sampling periods, determined by the Wilcoxon nonparametric test, are indicated by superscripts with different letters. Standard deviations are given in parentheses.

Sampling period	Exchangeable (cmol _c kg ⁻¹)		Acid-extractable (cmol _c kg ⁻¹)	
	Al	Ca	Al	Ca
1969-70	2.5 ^a (1.1)	8.3 ^a (4.4)	19.3 ^a (10.2)	9.9 ^a (6.4)
1987, 1992	3.7 ^a (2.9)	3.5 ^b (2.1)	37.0 ^b (21.6)	4.6 ^b (2.9)

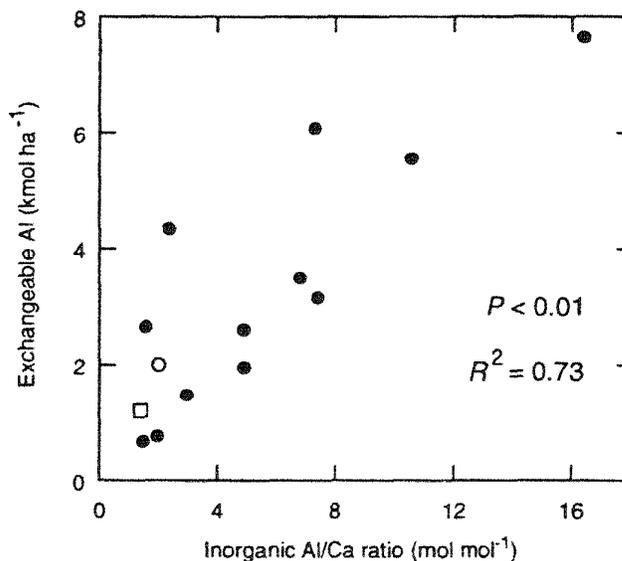


Figure 12.—Exchangeable Al content of Oa horizons of the 12 red spruce stands as a function of the molar concentration ratio of inorganic Al to Ca in B-horizon soil solution. Exchangeable Al was expressed as content to normalize the data for varying forest-floor thicknesses. Each filled circle represents the mean of 18-36 soil and soil solution samples (combined into 6-12 samples for analysis) collected at each of the 12 sites. Data from both the reference and treatment watersheds at Bear Brook Maine are included. The open circle represents the mean of 68 soil samples and 31 seep-water samples, collected in Winnisook watershed. The open square represents soil and seep-water concentrations measured at Tunk Mountain, ME, by Rustad (1988). Reprinted by permission from Nature (vol. 378, 9 November 1995, p.163), copyright 1995, MacMillan Magazines Ltd.

al. 1993, Bailey et al. 1996), a source of Ca which has been steadily declining in recent decades (Hedin et al., 1994). Decreasing levels of atmospheric Ca deposition and low Ca to Al ratios in the mineral soil will both tend to impede the reversal of Al accumulation in the forest floor.

Summary and Conclusion

Significant progress has been made during the 1990's towards establishing the past and current status of base cation concentrations in forest soils of the Northeast. The studies of Shortle and Bondietti (1992), Johnson et al. (1994a), and Lawrence et al. (1995) presented historical data that indicated long-term decreases at sites in the Adirondack Mountains and northern New England. The survey of soils in northeastern red spruce forests by Lawrence et al. (1997) identified considerable variability in exchangeable Ca concentrations, but an analysis of the methodological bias of the previous studies showed that these data did not alter the conclusion that long-term decreases are common in this region.

Elevated concentrations of inorganic Al in surface waters, also common in this region, suggest that acid deposition has contributed to the decrease in base-cation concentrations in soils. Elevated concentrations of inorganic Al in surface waters is the result of mobilization of inorganic Al in the mineral soil, which, in turn, is the probable result of decreased pH from inorganic acids deposited from the atmosphere. Naturally derived organic acids also mobilize Al, but the high affinity of Al for organic functional groups favors the formation of dissolved organic Al over dissolved inorganic Al. The mobilization of Al in the mineral soil increases concentrations of exchangeable Al by displacing adsorbed Ca, which is then free to be leached with mobile inorganic anions.

The importance of acid deposition as a cause in the decrease of base-cation concentrations in forest soils has been further suggested by results of the survey of soils in northeastern spruce stands (Lawrence et al. 1995, Lawrence et al. 1997), and the analysis of base-cation leaching in the Catskill Mountains (Lawrence et al. in press). The strong relation between relative weathering potential and Ca saturation in the Oa horizon but not in the B horizon, observed in the survey of spruce stands, indicates that base saturation in the mineral soil has been reduced to levels at which Al dominates exchange chemistry and inorganic Al concentrations in soil solution become much greater than Ca concentrations. Data from the survey of spruce stands also indicate that exchangeable Al concentrations are likely to be increasing in the forest floor as a result of Al mobilization in the mineral soil. Lastly, an elevational gradient in base-cation leaching in a Catskill Mountain watershed that could not be explained with any factors, other than an elevational gradient in acid deposition, demonstrates the effect that acid deposition can have on base-cation leaching.

The information summarized in this paper suggests that reversal of past decreases in exchangeable Ca concentrations will be hindered by 1) continued inputs of mobile anions that facilitate leaching, 2) high Al saturation in the mineral soil that will interfere with uptake of Ca by roots, 3) increasing Al saturation in the forest floor, and 4) continued decreasing trends of atmospheric deposition of Ca.

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Changes in Base Cations in Forest Floors

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Abstract

Cation loss from forest soils may be a contributing factor in some cases of sugar maple decline. We undertook three studies examining changes in base cations in forest floors. First, we described the pattern of change in forest floor calcium and magnesium in an intensively studied site at the Hubbard Brook Experimental Forest in New Hampshire. We analyzed samples collected at eight different times between 1970 and 1997. Concentrations and contents of Ca were higher in forest floor samples collected in 1970 than in those collected from 1976 to 1997. Changes over time were not significant between 1976 and 1997 for either Ca or Mg. Second, we questioned whether losses of base cations from the forest floor were regionally important. We analyzed

samples collected from 28 mature stands of a variety of forest types in the northeastern United States. About half of the stands lost Ca and Mg from the forest floor between 1980 and 1990, but the other half showed gains. The average change in cation content or concentration during this period was not significant. Third, we hypothesized that cation loss from the forest floor might be associated with forest regrowth following logging. We sampled forest floors in a successional sequence of 13 northern hardwood stands in the White Mountains over a 15-year interval. Contrary to our prediction, young stands gained Ca, Mg, and K relative to forest floor amounts measured 15 years previously. Middle-aged stands, harvested between 1930 and 1960, were most likely to show losses of base cations from the forest floor between 1979-80 and 1994-95. None of our studies rules out the possibility of important cation losses from forest floors before the late 1970s. However, the forest floor appears to aggrade, along with living biomass, early in stand development, and may not be a good indicator of changes in mineral soil fertility.

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Response of Northern Hardwood Forests to Nutrient Perturbation

Christopher Eagar¹, Scott Bailey², and Amey Bailey²

Abstract

Substantial amounts of calcium have been depleted from the soils of northern hardwood forests in northern New England over the past 50 years. Portions of this depleted calcium have been incorporated into the biomass of the aggrading forests; however, significant amounts have been leached into drainage waters and lost from the ecosystem. The depletion of base cations from the soil exchange pool combined with the acidic conditions of most northern hardwood soils results in increased levels of exchangeable Al. Elevated levels of Al can inhibit the uptake of Ca by sugar maple (*Acer saccharum* Marsh.) and may adversely affect the health of other species. The objectives of this field experiment are to: 1) determine if sugar maple productivity is limited by the declines in calcium pools, 2) evaluate the response of sugar maple to changes in soil Ca:Al ratios, and 3) determine the effect of changes in Ca and Al availability on nutrient cycling. Twelve 45m x 45m plots were established in an 80 year old sugar maple dominated stand located at an elevation of about 750m at Hubbard Brook Experimental Forest (HBEF) in New Hampshire. Four plots received a total of 10 g m⁻² Ca applied as CaCl₂, 4 plots received a total of 4.5 g m⁻² of Al applied as AlCl₃, and the remaining 4 plots were untreated controls. Treatments began in October 1995, with additional applications in May 1996, November 1996 and May 1997. The following attributes are being measured: tree growth, tree health, sugar maple wood chemistry, sugar maple foliar nutrient content, concentration and mass, litterfall amount and chemistry, soil chemistry, soil solution chemistry, and throughfall chemistry.

Prior to treatment the soils at the site had low exchangeable base cations (A horizon Ca = 0.68 cmol × kg⁻¹ and Mg=0.17 cmol × kg⁻¹) and high exchangeable Al (A horizon Al=5.82 cmol × kg⁻¹ from 1M NH₄Cl extraction). These values are similar to other sites at HBEF and are some of the more base-poor values found in the White Mountains. Soil water is collected from three nests of 3 tension lysimeters on each plot. At each nest, lysimeters are located beneath the organic horizon, and at 25 cm and 50 cm below the top of the forest floor. A goal of the study is to increase the concentration of inorganic monomeric Al (Alⁱ) in soil solution

Table 1.—Average soil solution concentrations and the Ca:Alⁱ ratio for the 1997 growing season at three depths

Treatment	Depth	Ca	Mg	Al ⁱ	Cl	Ca:Al ⁱ
------(μmol L ⁻¹)-----						
Calcium	O	135.8	32.6	25.9	371.0	4.3
	25cm	43.0	13.1	19.4	132.3	2.3
	50cm	43.2	14.1	15.1	113.6	3.1
Aluminum	O	103.4	27.2	89.6	492.7	1.4
	25cm	48.0	16.0	40.0	218.7	1.3
	50cm	39.4	13.7	18.8	112.7	2.5
Control	O	23.0	6.8	15.0	25.1	1.5
	25cm	15.0	4.7	13.6	12.6	1.1
	50cm	17.4	5.8	8.5	11.3	2.1

in the upper soil horizons to at least 100 μmol L⁻¹, a concentration that inhibits the uptake of Ca by sugar maple. Table 1 shows the average soil solution concentrations for Ca, Mg, Alⁱ, and Cl and the average Ca:Alⁱ ratio for the 1997 growing season. The calcium treatment increased the concentration of Ca at all depths. Other cations, including Al, were elevated relative to the control plots, but not to the degree of Ca. The Ca:Alⁱ ratio was increased to a level well out of the range that would be potentially harmful to sensitive plants. The aluminum treatment increased Al concentration to near the target level in the organic horizon and to almost 50% of the target level for the B horizon (25 cm depth). The aluminum treatment also substantially increased the concentration of Ca at all depths; consequently, there was only a minimal change in the Ca:Alⁱ ratio compared to the control plots. Multi-cation response suggests that changes in soil water chemistry are due to both added cations as well as native cations contributed from soil exchange sites. Foliar samples for chemical analysis were collected in August of each year from sapling and overstory sugar maple, hobble bush (*Viburnum alnifolium* Marsh.), and wood fern (*Dryopteris spinulosa* (Muhl.) Underw.). There has not been a significant response of foliar concentrations of Ca in sugar maple saplings or overstory trees; however, the trends are in the expected directions with higher concentrations of Ca in the calcium treatment plots and lower in the aluminum treatment plots relative to the control. Wood fern showed the most dramatic response to treatments with the foliar concentration of Ca doubling in the calcium treatments compared to controls and a trend toward reduced Ca in the aluminum treatments.

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Soil Water and Xylem Chemistry in Declining Sugar Maple Stands in Pennsylvania

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Abstract

Evidence is accumulating that decline of sugar maple, *Acer saccharum* Marsh., in northern Pennsylvania may be related to overall site fertility as reflected in the chemistry of soil water and bolewood xylem. In this paper we discuss factors related to varying site fertility, including effects of soil liming, past glaciation, topographic position and soil acidification. Soil liming experiments at Cherry Springs and Black Diamond sites in northcentral PA have shown a dramatic growth response of sugar maple to improved soil Ca and Mg supplies (Long et al. 1997). Soil water chemistry at 30-cm depth at Cherry Springs and Black Diamond showed significantly higher Ca and Mg and significantly lower Al and Mn concentrations on treated than control plots in 1995-96, over ten years after liming. Ca/Al molar ratios < 1 in soil water on unlimed plots at Cherry Springs suggest that growth stress may be occurring due to nutritional disorders. Sugar maple xylem chemistry at Cherry Springs also showed significantly higher Ca and Mg and significantly lower Mn concentrations on limed than on unlimed plots. Although Al concentrations were undetectable in sugar maple bolewood, Mg/Mn or Ca/Mn molar ratios in xylem were significantly higher on limed than unlimed plots at Cherry Springs. Similar soil water and bolewood xylem chemistry differences were generally, but not always, found between stands on glaciated and unglaciated sugar maple sites in northern Pennsylvania. Generally, forest stands on

glaciated sites showed higher soil water Ca/Al ratios and bolewood xylem Mg/Mn ratios than unglaciated sites. Topographic position (upper, mid, lower slopes) did not consistently correlate with occurrence of high or low Ca/Al and Mg/Mn ratios in soil water and bolewood xylem, respectively, due to the effects of underlying geology and subsurface drainage patterns. Further sampling in 15 sugar maple stands in northern Pennsylvania also showed a positive relationship between radial growth rates and the Mg/Mn ratios in bolewood xylem, suggesting a link between site fertility and growth². Soil acidification experiments at Fernow Experimental Forest in West Virginia and Bear Brook in Maine, which were conducted to test effects of accelerated rates of atmospheric deposition simulated with additions of N and S as ammonium sulfate, have shown rapid leaching of base cations from the soil leading to reduced bolewood xylem Mg/Mn molar ratios (DeWalle et al. in press).

Overall, these studies suggest that wide variations exist in forest site fertility, as reflected by soil water and xylem chemistry, due to complex interactions of glaciation history, topography, and possible accelerated leaching of soil base cations due to atmospheric deposition. Understanding the factors causing such variations in site fertility and their effects on health of sugar maple may provide better management methods for declining sugar maple stands.

References

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²Data taken in collaboration with Stephen B. Horsley, Robert P. Long, and Scott W. Bailey, Northeastern Research Station, at Irvin, PA, Delaware, OH and Durham, NH, respectively, and Thomas J. Hall, Pennsylvania Department of Conservation and Natural Resources, Bureau of Forestry, Division of Forest Pest Management, Middletown, PA.

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