TIMING, MAGNITUDE, AND IMPACT OF ACIDIC DEPOSITION ON
SENSITIVE FOREST SITES

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ABSTRACT. Adverse effects of acidic deposition on forest health are most
likely to occur in forests which develop a thick raw or "mor" humus
layer in which the effective cation exchange capacity is highly
sensitive to acid input. A study of the trend of exchangeable Ca and Mg
ions in sensitive humus layers over the past six decades indicated that
a downward shift in equilibrium has occurred that is consistent with
theories of ion mobilization and coincident in time with increasing
acidic deposition in the mid-1900's. Independent records of a base
cation mobilization in wood supports the view of a change in the root
zone in sensitive forest sites and in lake water chemistry. Induced
nutrient deficiency in declining forest stands was indicated by a high
Al:Ca ratio in fine root tip tissue as a marker of altered uptake
efficiency which leads to increased vulnerability to biotic factors and
greater sensitivity to abiotic stresses. Evidence indicates that
sensitive sites were damaged by acidic deposition 20 to 40 yr ago, long
before the problem of "acid rain" was recognized.

1. Introduction

A major concern of the 1980's was the possible linkage between the
declining health of some forest types in eastern North America and
central Europe, and the rapid rise in emissions of oxides of S and N
which brought about increased acidic deposition ("acid rain"). One key
factor, especially in old growth forests of spruce, fir, and hemlock, is
the role of the raw or "mor" humus layer in nutrient storage. The
location of the fine root system in this layer tends to isolate the

nutrient cycle from the mineral soil, especially in stands with a small hardwood component (Moore, 1922; Coile, 1938; Hopkins, 1939; Donahue, 1940; Kimmins and Hawkes, 1978). The efficiency of the humus-root complex in preventing Ca from leaching is illustrated by the bleached mineral soil layer under thinner mor layers on podzols, or the very low pH and Ca concentrations which were present in the lower parts of very thick mor layers (Heimburger, 1934; Plice, 1934) before the advent of significant acidic deposition.

The substantial isolation of the root system from the mineral layer results, therefore, in a significant reliance on the atmosphere to make up bases naturally lost by leaching. In this respect the nutrient dynamics in these forests resemble those of ombrotrophic bogs, an idea first alluded to by Mattson et al. (1944). Unfortunately, this natural "sponge" stores and concentrates mineral acids. Because base adsorption capacity is pH-dependent, the accumulation of mineral acids during periods of infrequent drainage and the acidity of spring snow melt during periods of maximum drainage both reduce the effective cation exchange capacity of the humus. This reduction in exchange capacity will override the naturally high efficiency of Ca removal by the root-humus complex. Increased acidic deposition on forest stands with thicker mor-type humus will likely accelerate mobilization of extant cation reserves and limit resupply from the atmosphere and litter mineralization. The net effect is an initial and brief "fertilizer effect" brought about by increased uptake as well as leaching followed by prolonged base impoverishment.

Evidence of a Ca signal in wood formed coincident with significant increases in acidic deposition in the eastern United States from the late 1940's to the early 1960's has been reported in bolewood of spruce, fir, hemlock, and birch consistent with cation mobilization concepts (Momoshima and Bondietti, 1990; Bondietti et al., 1990). This base cation mobilization signal was observed in more than half of the more than 80 mature red spruce sampled from North Carolina and Tennessee through to New Brunswick. They reflected the mobilization of bases in the raw humus that would have occurred as regional acidic deposition rapidly increased through the mid-1900's, causing a disequilibrium between the changing acid-base status of deposition and the concentration of adsorbed bases.

The consequence of mobilization, disequilibrium, and debasification in the root zone of mature trees is the creation of nutrient imbalances that can result in dieback, decline, and mortality from many local causes. Deficient trees are more vulnerable to diseases and insects, and more sensitive to cold and drought injury and nutrient deficiencies. Calcium deficiencies can be induced in mature trees by increased interactions with Al^{3+}, H^+, and Fe^{3+} (Shortle and Smith, 1988; Schroeder et al., 1988; Stienen and Bauch, 1988). Unlike crop plants, trees have a high Ca requirement (Rennie, 1955), with deficiency resulting in the dysfunction and death of primary and secondary meristems. Magnesium deficiency is a secondary problem, with uptake being under stronger metabolic control (Williams, 1970).

The purpose of this paper was, (1) to determine the trend of exchangeable Ca^{2+} + Mg^{2+} in the sensitive humus of forest mor and

forested bogs in eastern North America reported in the literature over the past six decades for which a limited record exists, (2) to compare the Ca mobilization signal in wood samples to changes in soil and lake chemistry at sensitive sites in this region, and (3) to compare a marker (molar Al:Ca ratio) of Al-induced Ca deficiency in fine root tip tissue obtained from sensitive sites differing in acid input.

2. Materials and Methods

An extensive literature search was conducted by Bondietti to determine all pertinent forest mor values of exchangeable Ca^{2+} + Mg^{2+} for humus in eastern North American before 1950 and after 1970. Values outside this region in areas in which anthropogenic acid inputs were not likely were also included. Although many profiles of forest mor and forested bogs are presented in the literature, chemical measurements on the raw humus layers are usually not reported, making the available data base quite small. Furthermore, to assure a high sensitivity to mineral acid input, we restricted the literature analysis to sites meeting a specific set of criteria. These criteria required that the sites be older forests generally dominated by spruce, that humus have a high loss-on-ignition (LOI) indicating an organic matter (OM) content of >70% by weight, that the raw humus have a pH of < 4.5 (most sites contained humus with a pH < 4.2), and that the combined F + H soil layers be thicker than 5 cm. Base saturation was not used as a criteria because it is not a standardized measurment. LOI is strongly correlated with relative cation exchange capacity (Chandler, 1939a) and thickness of the humus layer (Chandler, 1942; Gorham, 1954). Thick mor layers commonly develop under spruce, fir, and hemlock forests; somewhat thinner layers can develop under pine, oak, or northern hardwoods. Red spruce litter in forests over thick mor layers has the lowest Ca content of any common species in eastern North America (Chandler, 1939b).

Nonmenclature for humus horizons differ. Generally the fermentation layer is considered to contain partially decomposed, but recognizable, organic debris immediately under the litter layer and is designated F, O_1, O_e, or A_o. The humified layer occurs below the fermentation layer and is often referred to as "greasy" because of its high H^+ saturation when relatively mineral-free, and is designated H, O_2, O_a, or A_{oo}. The term F-layer and H-layer are used in this paper; however, the two were sometimes mixed into one sample in the various studies reported.

To facilitate comparison of one report with another all values of exchangeable essential bases were converted to Ca^{2+} + Mg^{2+} in $mmol(+) \cdot kg^{-1}$ OM. All necessary values for this conversion were seldom available and must be derived by a series of conversion factors derived from the entire data set. If LOI was not reported to convert to OM basis, it was readily derived from % ash or by multiplying % C by the conversion factors of 1.85 (F-layer) or 1.80 (H-layer) (Lunt, 1932). If none of these was available, conversion factors of 1.25, 1.67, and 1.43 were used for F-layers (80% OM), H-layers (60% OM), and mixtures of F-

and H-layers (70% OM), respectively. If only Ca^{2+} values were given, increasing that value by 30% gives a reasonable estimate of Ca^{2+} + Mg^{2+} because most molar Mg:Ca ratios ranged from 0.1 to 0.6 with the exception of some low Ca sites or those strongly affected by marine salts in which case Mg:Ca was somewhat higher. In two cases Ca^{2+} + Mg^{2+} values were derived from "total exchangeable bases" by multiplying by 0.8 (80% of exchangeable base cations). If only total values were given, they were converted to exchangeable Ca^{2+} + Mg^{2+} by multiplying by 0.7 (exchangeable:total values ranged from 50 to 90% in high LOI humus with an average value of about 70%). By using these conversions we believe the resulting rough estimates are reasonable over data from many different investigators that used different methods at different times to make determinations in a highly conserved biological system, the forest floor.

Soil samples used for analysis were taken from small 30 x 15 cm pits dug down to mineral soil, 6 to 10 pits per site. These pits were associated with the rooting zone of canopy red spruce trees and were kept as small as possible and refilled after taking samples of 100 to 200 cc to produce as little site disturbance as possible. Samples of humus from F-layers and H-layers were taken at depths of 0 to 5, 5 to 10, and 10 to 20 cm (F usually in upper 5 cm, H of variable depth down to mineral soil). The lower 1/4 of H-layers was avoided where humus was thick. Samples from Whiteface Mountain, NY, and Isle au Haut, ME, were less well defined coming from F and H layers in the upper 10 cm from which absorbing fine roots were collected for tip tissue analysis.

Humus samples used for the determination of total Ca^{2+} + Mg^{2+} for Whiteface Mountain, NY, Big Moose Lake, NY, and Isle au Haut, ME, were air-dried, sieved through a 2-mm sieve, dry ashed at $450^{\circ}C$, dissolved in 0.2 M HNO_3, and analyzed by plasma emission spectrometry (DCP) at the USDA Forest Service Plant and Water Analysis Laboratory, Berea, KY. Humus samples of the H-layer under a virgin red spruce stand in Waterville, NH, first sampled by Lunt (1932) 60 yr ago, were used to determine the base exchange capacity using a Ba^{2+} titration method (Belkevich and Chistova, 1963) as well as for Ca^{2+} + Mg^{2+} determinations. Humus samples collected from Mt. Abraham, VT, Crawford Notch, NH, Cherry Mountain, NH, Howland, ME, Mt.Mitchell, NC, and Roan Mountain, NC, used for the determination of exchangeable Ca^{2+} + Mg^{2+} were oven-dried at $60^{\circ}C$, sieved through a 1-mm sieve, extracted 4 times with 0.5 N ammonium acetate, and analyzed by inductively coupled plasma spectrometry (ICP) (Stark, 1990).

Cores were taken from 3 to 10 canopy red spruce using a 12-mm diameter increment borer. Core surfaces were scraped with a stainless steel scalpel and segmented into 5- or 10-yr increments, which were dry ashed and dissolved as described elsewhere (Bondietti et al., 1990). Elemental analysis was done by DCP at Berea, KY, as stated above.

Samples of absorbing fine root tips of 57 mature red spruce trees were taken in late September and October from sites with relatively high acid input (mountain or coastal sites strongly affected by cloud water; fogs of pH 3 occur) (Kimball et al., 1988) and relatively low acid input (mid-elevation sites lacking strong cloud influence). High and low

input sites today probably differ by 2x in acid input, but both are
receiving 3 to 10x the estimated acid input occurring before the 1940s.

Absorbing fine root tips (0.1 to 0.3 mm diam x 1 to 5 mm length) were
most often found in the raw humus layer with rapidly decreasing
frequency down toward the upper mineral soil. Sufficient material was
collected from the humus to yield 20 to 50 mg dried tissue per tree (60
to 100 root tips per mg; 1200 to 5000 tips per tree). Samples from the
upper mineral soil were also taken for comparison, but yields were often
less than 20 mg. Field collections of fine root clusters were kept
frozen until tips were individually removed in distilled-deionized water
and oven-dried for analysis of divalent and trivalent metal ions.

Analysis of oven-dried (55°C) root tip tissue was done initially by
atomic absorption spectroscopy and ion chromatography (Shortle and
Smith, 1988; Shortle and Stienen, 1988), then by plasma emission
spectrometry at the USDA Forest Service, Plant and Water Analysis
Laboratory, Berea, Kentucky (details of quality control/assurance
available upon request). The molar ratio of Al:Ca in the root tip
tissue is based on triplicate determinations per tree, 2 to 8 trees per
location. The estimated mean molar ratio of Al:Ca in root tip tissue
and the 95% confidence limits are reported for high and low input sites,
and the mean and 95% confidence limits for each site are given when n $\geq$
6 trees.

3. Results

Estimates of exchangeable Ca^{2+} + Mg^{2+}, mmol(+) $\cdot$ kg^{-1}OM, in the
raw humus of the forest floor was derived form 24 references reporting
results from 1 to 26 profiles each that met the criteria specified above
(Table I). Estimates before 1950 were limited to the northeastern
United States (NY and NH in particular, plus two sites from Quebec) and
consistently contained 150 to 300 mmol(+) $\cdot$ kg^{-1}OM as predicted by
the concept of Ponomareva (1969) for northern forest mor (and peat).
Exceptions to this rule was humus from deep in thick organic layers.
Six samples of H-layer > 20 cm from Romell and Heiberg (1931) averaged
47 mmol(+) $\cdot$ kg^{-1} OM with a broad range of 4 to 233. Twelve samples
of H-layer from the lower portion of the profile at unspecified depths
from Heimburger (1934) averaged 143 mmol(+) $\cdot$ kg^{-1} OM with a range
of 71 to 214. Thus, the mixing of humus from deeper layers in thick
profiles can tend to depress the results.

Most soil profiles described from NH and NY were located in the
western Adirondacks, NY, and the White Mountains, NH, and had developed
under old growth spruce, fir, and hemlock. Six additional profiles
listed separately (Table I) developed under balsam fir in Quebec (2),
under black spruce (2), and under white pine (1) or northern hardwoods
(1). The three separate sets of values listed for PA (Table I) were
from the same area of Pennsylvania sampled by three different
investigators over a twelve-year period with equivalent results.

TABLE I. Estimates of exchangeable essential base cations in humus of the forest floor over the past six decades.

Reference; Location	Exchangeable $Ca^{2+} + Mg^{2+}$ $mmol(+) \cdot kg^{-1} OM^a$	
	F-layer	H-layer
Eastern region before 1950:		
1931 Romell and Heiburg; NY, NH	233(16)	188(15)
1934 Heimburger; NY, NH	295(26)	236(25)
NY, Quebec	231(6)	214(6)
1931 Romell and Heiberg; PA	219(1)	228(3)
1934 Plice; PA	226(3)	143(2)
1943 Hough; PA	180(2)	
Outside region 1940 to 1975:		
1942 Chandler; AK	233(1)	--
1953 Mader; Quebec	208(1)	
1954 Gorham; Ireland	203(4)	
1955 Sirèn; Finland	183(1)	
1957 Bonnevie-Svendsen and Gjems; Norway	246(2)	--
1958 Zaboyeva; USSR	293(1)	--
1958 Ovington; Britain	213(3)	
1959 Krause et al.; AK	278(1)	--
1960 Soil Survey Staff; Norway	271(2)	
1968 Heilman; AK	208(2)	--
1975 Soil Survey Staff; AK	233(1)	--
Eastern region after 1970:		
1971 Damman; Newfoundland	106(2)	73(2)
1971 Page; Newfoundland	--	138(10)
1973 Hoyle; NH	--	115(1)
1977 Pilgrim and Harter; NH	--	126(3)
1980 Hanson; NH, ME	117(2)	52(1)
1981 Lang et al.; NH	94(1)	--
1982 Molliter and Raynal; NH	39(2)	21(1)
1985 Cronan; NY	120(2)	
1988 Arp and Manasc; New Brunswick	135(2)	

[a]   Mean of estimates from each reference; number of estimates in parenthesis; estimates derived from F(fermentation)-layer, H(humified)-layer, or mixed FH-layer.

No record of exchangeable $Ca^{2+} + Mg^{2+}$ was found for the southeastern United States before 1958. The first record (McGinnis, 1958) of exchangeable cations under a spruce-fir stand appeared to be
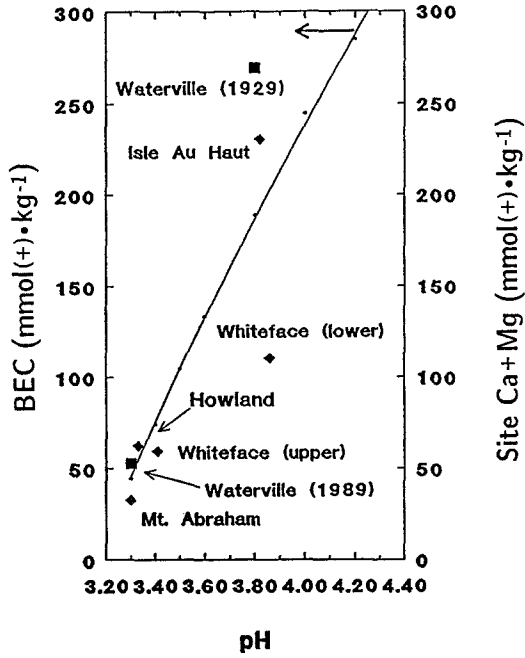
Figure 1. Base Exchange Capacity (BEC) of Oa- or H-layer humus from a virgin red spruce stand in Waterville, New Hampshire, and the total $Ca^{2+} + Mg^{2+}$ concentrations and pH of the same type of humus collected at Whiteface Mountain (2 elevations), New York; Mount Abraham, Vermont, and Isle au Haut and Howland, Maine. Measurements made in 1929 and 1989 of exchangeable $Ca^{2+} + Mg^{2+}$ at the pH at the Waterville site are also shown.

generally comparable to a virgin spruce stand in NH (Lunt, 1932, Figure 1). Estimated exchangeable $Ca^{2+} + Mg^{2+}$ for humus of the F-layer was 178 (Lunt, 1932), 109 (McGinnis, 1958); H-layer 277 (Lunt, 1932), 251 (McGinnis, 1958). McGinnis (1958) reported the difference between the upper 3/4 and the lower 1/4 of the deep H-layer as 251 and 30, respectively. By 1967, four F-layers and nine H-layers had an average of 161 and 46 mmol(+) · kg^{-1} OM, respectively (Wolfe, 1967). More recently reports of F-layer 60 and 212, H-layer 52 and 97 have occurred (McGuire, 1983). Thus, the very sparse record of forest floor data in spruce-fir stands of the southern Appalachians appeared to be following trends similar to the Northeast (Table I).

    Estimates of exchangeable $Ca^{2+} + Mg^{2+}$ in the forest floor after 1970 were consistently below 150 mmol(+) · kg^{-1} OM and often well below that value. This is in contrast to estimates in the same region

before 1950, to estimates from areas unlikely to be strongly influenced by acidic deposition, and to the generalized concepts of Ponomereva (1969) for northern forest mor (Table I).   Samples of humus from the F-layer and H-layer of the upper 10 cm of forest floor in the northeastern and southeastern United States, 1988 to 1990, confirmed this general trend derived from published data (Table II).   Two notable exceptions occurred at Crawford Notch, NH, and Isle au Haut, ME.   Old growth red spruce at Crawford Notch, NH, has been dead for a decade longer than at other high mortality sites observed across the region and the huge amount of dead branches, roots, and boles are undergoing mineralization, which is likely to restore a more normal equilibrium in the forest floor.   Isle au Haut is under strong marine influence where base resupply is likely to result from sea salt as indicated by a high Mg:Ca ratio and Na in the humus.

TABLE II.   Estimates of exchangeable essential base cations in the upper 10 cm of humus of the forest floor sampled in 1988 to 1990.

Reference; Location	Exchangeable $Ca^{2+} + Mg^{2+}$ $mmol(+) \cdot kg^{-1}OM$[a]	
	F-layer	H-layer
Whiteface Mountain, NY	119(8)	
Big Moose Lake, NY	--	55(3)
Mount Abraham, VT	43(10)	26(10)
Crawford Notch, NH	145(10)	228(10)
Cherry Mountain, NH	123(6)	93(6)
Howland, ME	130(10)	81(10)
Isle au Haut, ME	205(4)	
Mount Mitchell, NC	--	30(17)
Roan Mountain, NC	--	69(17)

[a]See footnote for Table I.

The exchangeable base cation concentration in forest mor is controlled by ambient H^+ and divalent cation concentrations.   The base exchange capacity (BEC)-pH relationship of a high LOI humus from the H-layer samples, collected under a virgin red spruce stand in Waterville, NH, 60 yr after the stand was first sampled (Lunt, 1932), illustrates this point (Figure 1).   Around pH 4, the BEC, measured by titration with $Ba(OH)_2$, is about 250 $mmol(+) \cdot kg^{-1}$.   As solution pH declines, BEC rapidly drops to about 100 $mmol(+) \cdot kg^{-1}$ at pH 3.5, for example.   Thus, increasing the ambient H^+ by a factor of 3.2 (decreasing pH from 4 to 3.5) decreased the BEC by a factor of 2.5.   This sensitivity of BEC to H^+ is the reason why mor and peat are good for storage and supply of essential base cations to plant roots despite the naturally high acidity of the root-humus

complex, and why it is so sensitive to atmospheric acidity associated
with strong acid anions.

Some present day concentration - pH relationships have been plotted
as total Ca^{2+} + Mg^{2+} and 1:1 (v:v) water pHs for several high LOI
samples from mor (Figure 1). These samples were collected in the
upper 10 cm at locations where samples of the fine root system were
collected. We also see the 80% decrease in estimated exchangeable
Ca^{2+} + Mg^{2+} at the Waterville, NH, site over the past 60 yr (Figure
1).

Cores taken from 150 to 250 year-old red spruce trees at Wateville,
NH, and Big Moose Lake, NY, have the Ca mobilization signal found in
more than half the red spruce sampled in the East (Bondietti et al.,
1990). The signal at Big Moose Lake (Fig. 2) coincides with changes
in lake water chemistry reconstructed from sediments (Charles, 1984).
From the 1950s and 1960s pH sensitive systems in the lake and the
root-humus system appeared to be altered in the way predicted by a
rapid increase in atmospheric acidity associated with strong acid
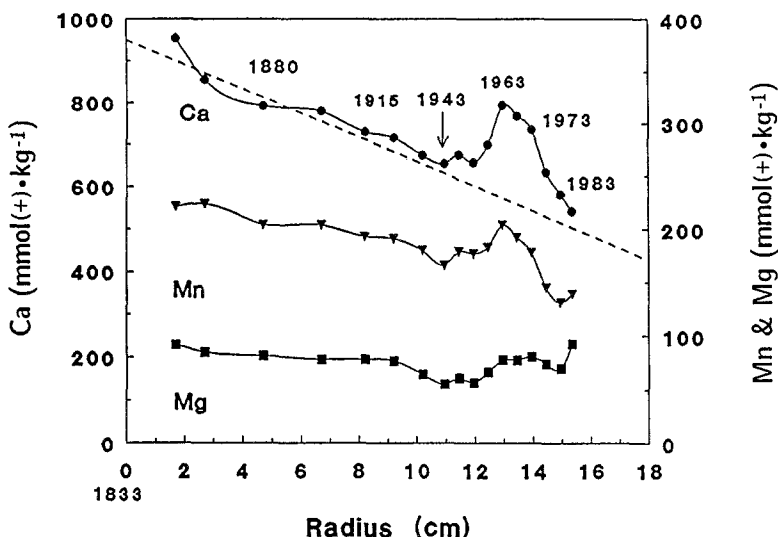anion input.

Figure 2. Concentration of major divalent cations (Ca^{2+}, Mg^{2+}, and
Mn^{2+}) in wood, expressed as milliequivalents per kilogram, of red
spruce at Big Moose Lake, New York is plotted as a function of radial
distance from the pith. The dashed line is the predicted trend in wood
divalent cation storage under constant sap conditions (Momoshima and
Bondietti, 1990). Tree ring dates are indicated along the curve to
show how the cation storage has changed over time.

TABLE III. Molar ratio of Al:Ca in red spruce fine root tip tissue from locations having relatively high acid input compared to those having relatively low acid input.

Location	Elevation	Molar Al:Ca ratio[a]		
	m	n	x	+ 95% CL
Higher acid input[b]				
Mount Mitchell, NC	1400	8	1.6	1.1
Whiteface Mountain, NY	1000	8	0.9	0.3
Mount Abraham, VT	1000	6	3.1	2.1
Crawford Notch, NH	800	6	0.9	0.9
Isle au Haut, ME	10	4	1.4	-
Over all locations		32	1.5	0.5
Lower acid input				
Hubbard Brook, NH	550	2	0.3	-
Grafton Notch, ME	700	2	0.1	-
Howland, ME	150	8	0.4	0.2
Kassuth, ME	200	6	0.3	0.1
Beddington, ME	100	7	0.2	0.1
Over all locations		25	0.3	0.1

[a] Number of observations, mean, and 95% confidence limits (n>6) for each location. Each observation was derived form a collection of 1500 to 3000 individually taken fine root tips 1-5 mm long from a single mature red spruce tree.

[b] Higher acid input sites are montane or coastal sites strongly influenced by cloud or fog water in which pH 3 can occur. Such sites may currently receive 2x the annual acid input of lower input sites, although both types are likely receiving 3-10x the estimated acid input before the 1940s.

Absorbing root tips collected from the humus of sensitive sites receiving a somewhat higher input of atmospheric acidity and strong acid anions had a 5x greater molar Al:Ca ratio in the tip tissue than tip tissue from sensitive sites receiving somewhat less (Table III). The incidence of mortality among mature canopy trees of the former was 5 to 10x greater than the later, although trees in sensitive sites at both levels of input had suppressed growth for the past several decades. The within site (tree-to-tree) variation in root tip analysis was quite high, but the overall confidence limits at both levels of input was about 30% compared to the 500% difference between input levels. Where input was high, concentrations of Al > Ca in the tissue of absorbing tips, the root density was sparse, and tree health was poor; where input

was lower (although still at several-fold higher levels then before 1950), concentrations of Al < Ca, root density was less sparse, and tree health was less poor.

At sites such as Crawford Notch, NH, surviving trees are beginning to recover as the huge amount of dead branches, roots, and boles are being mineralized to resupply the exchange sites in the humus. At sites such as Howland, ME, the health of trees is steadily declining, although spruce mortality is not yet occurring. Thus, we are likely to see changes in the composition of root tip tissue sampled over time in response to changing stand conditions as the trees at sensitive sites respond to stress, individuals die, and the stand recovers.

4. Discussion

The trend based on the generalization of diverse studies over many years does not allow for direct statistical comparisons because of the problem of properly weighting each data set. However, the estimated concentrations of exchangeable essential base cations (Ca^{2+} and Mg^{2+}) in the root-humus complex of well-developed forest mor appeared to fit the generalized concept of an equilibrium of 150 to 300 mmol $\cdot$ kg^{-1} OM (Ponomareva, 1969) in eastern North America before 1950 and in remote areas from 1940 to 1975, but not in eastern North America after 1970. Between 1950 and 1970 in eastern North America, atmospheric acidity increased and tree growth peaked in sensitive forest sites (McLaughlin et al., 1987). The apparent lack of fit is consistent with the theory of mobilization and sensitivity to increasing acidity which caused a shift in the equilibrium of the root-humus complex on sensitive sites and produced a cation mobilization signal in wood of mature trees on those sites (Momoshima and Bondietti, 1990; Bondietti et al., 1990).

The fact that the cation mobilization signal in mature spruce observed at Big Moose Lake, NY (Figure 2), is coincident with the period of lake acidification (Charles, 1984) supports the postulated effect of acidic deposition on sensitive forest sites. Resampling the few areas in which detailed humus chemistry data were taken before acidic deposition increased significantly across the region further supports this view (Figure 1). Additional work on such means to further validate the postulated shift in equilibrium has been delayed by the tragic death of Dr. Ernest A. Bondietti on February 17, 1990. This paper contains his final contributions to our understanding of the mechanisms by which acidic deposition can perturb the forest ecosystem.

The possible relationship of a significant perturbation in the root-humus complex of sensitive forest sites to tree health has long been known for localized damage caused by S emissions (Hartig, 1897). A deficiency of essential base cations, especially Ca, has its effects on plant meristems responsible for root, shoot, and cambial growth. Prolonged suppression of growth, especially to the root system, leads to loss of vital tissue needed for adequate plant defense. Base deficient mature trees become more vulnerable to biotic agents such as insects and rootrot fungi, and more sensitive to abiotic agents such as cold and

drought. Deterioration of health is eventually seen as dieback and decline, followed by death. High mortality brings about an abrupt decrease in nutrient demand, and initiates several decades of resupply as the dead roots, branches, and bole are mineralized into the root-humus complex.

True deficiencies of exchangeable essential cations are likely to be extremely rare due to the small amounts that need to be drawn from the exchangeable storage system. However, deficiencies induced by ion interactions are likely to occur during mobilization at the low pH of forest mor. Induced Ca deficiency may be brought about by Al^{3+} (Schroeder et al., 1988; Shortle and Smith, 1988). Similar effects at higher acidities are brought about by H^+ and Fe^{3+} (Stienen and Bauch, 1988). Over the range of pH 3.5 to 4.2 the interference of Al^{3+} with the uptake of Ca^{2+} and Mg^{2+} appears to be quite high given how common Al^{3+} is in the soil. A useful marker for an induced deficiency of this type seems to be root tip tissue in which Al^{3+} is in equal or greater molar concentration than Ca^{2+} (Shortle and Smith, 1988; Shortle and Stienen, 1988).

We looked for this marker in the root tip tissue at montane and coastal sites in which high rates of acidic deposition occur and found Al:Ca > 1 and 5X the mean at mid-elevation interior sites (Table III). The data were for tips in the upper humus where Al^{3+} is least likely to be a problem, not in the lower humus in which Al:Ca $\geq$ 1 is common (Shortle and Stienen, 1988).

The loss of Ca^{2+} from forest ecosystems throughout the eastern United States is occurring at higher than postulated historic rates (Federer et al., 1989). At some sensitive sites of the type that we have been studying, Al^{3+} now exceeds Ca^{2+} in water passing from these ecosystems (Cronan and Schofield, 1979). This is due to the way in which divalent and trivalent cations mobilize at high acidity and ionic strength (Reuss and Johnson, 1986). The loss of Mg^{2+} is less dramatic due to the high degree of metabolic control governing its uptake (Williams, 1970). Although the induced deficiency syndrome has been observed most readily in red spruce, the damage to the root-humus complex of sensitive species growing on sensitive sites is not likely to be limited to this small fraction of the eastern forest ecosystem.

5. Conclusions

The evidence presented supports the view that from the late 1940's into the 1960's the mor layer of acid-sensitive forest sites in eastern North America underwent a significant change that resulted in the loss of exchangeable essential base cations and interupted the critical base nutrient cycles between mature trees and the root-humus complex. This view stands in marked contrast to those of the National Acid Precipitation Assessment Program Report (NAPAP 1990) that states: "Soil process models indicate that deposition of S and N compounds at approximately present levels could have an adverse effect on sensitive soils in certain areas of the eastern United States in 50 to 100 yr,

which is within a single rotation for managed eastern forests". The most
sensitive sites apppear to have been adversely affected 20 to 40 yr in
the past, and not 50 to 100 yr in the future. The "soil process models"
used to predict future problems are based on data gathered in the 1970's
and 1980's after significant change had already taken place. Modern
studies of effects of spatial gradients over a factor of 2X may have
been superimposed over temporal gradients of 3-5X.

   One of the most important impacts of acidic deposition on forests in
eastern North America apparently occurred before anybody was looking. As
deposition acidity increased in impact on the raw humus layers under
spruce-fir-hemlock forests, divalent bases mobilized as the effective
base retention capacity of the organic exchange sites in the forest
floor decreased. The timing of the impact, the mid-1900's, appears to
have been during the period when SO_x and NO_x emissions in eastern
North America subject to long-range transport were increasing the most
rapidly. The rapidity of the mobilization response was interpreted to be
due to a non-biological source of H^+ amplifying the natural
sensitivity of the raw humus to H^+ produced by roots.

   The base cation losses from spruce stands are, in principle,
applicable to natural stands of other species under certain specific
conditions. These conditions include: stands in which the forest floor
dominates base cation storage, stands in which ombrotrophic rather
minerotrophic resupply of bases is important, those in which Ca-poor
litter are produced, and those stands in which the available bases are
cycling rapidly. Examples of tree species that often produce mor-type
humus under Ca-poor nutritional conditions have been mentioned in the
literature in the context of productivity management. It may not be
coincidental, therefore, that some of these species: red spruce, balsam
fir, loblolly pine, shortleaf pine, and white oak, are species that also
have been reported to have experienced apparently unusual growth
patterns in recent decades. If the relationships discussed here are
valid, variables such as the extent of the stand hardwood component, mor
thickness, mor base status. fine root distribution, and ombrotrophy need
to be factored into studies on the causes of unusual stand-level growth
anomalies that started in the 1950's and 1960's.

Acknowledgments

References

Arp, P.A. and Manasc, J.: 1988, Can. J. For. Res. 18, 251.

Belkevich, P.I. and Chistova, L.R.: 1963, In Robertson, R.A. (ed.),
   Proc. 2nd Int. Peat Congress, Leningrad, pp. 904-918.

Bondietti, E.A., Momoshima, N., Shortle, W.C., and Smith, K.T.: 1990,
   Can. J. For. Res. 20, 1850.

Bonnevie-Svendsen, C. and Gjems, O.: 1957, Medd. Norsk. Skogforsk.
   48, 115.

Chandler, R.F., Jr.: 1939a, J. Agr. Res. 59, 491.

Chandler, R.F., Jr.: 1939b, Cornell (Ithaca) Univ. Agr. Exp. Sta. Mem.
   228. 15 pp.

Chandler, R.F., Jr.: 1942, Soil Sci. Soc. Am. Proc. 5, 454.

Charles, D.F.: 1984, Verh. Internat. Verein. Limnol. 22, 559.

Coile, T.S.: 1938, Soil Sci. Soc. Am. Proc. 3, 274.

Cronan, C.S.: 1985, Water Air Soil Pollut. 26, 355.

Cronan, C.S. and Schofield, C.L.: 1979, Science 204, 304.

Damman, A.W.: 1971, Ecol. Monog. 41, 253.

Donahue, R.L.: 1940, Cornell (Ithaca) Univ. Agri. Exp. Sta. Mem.
   229. 44 p.

Federer, C.A., Hornbeck, J.W., Tritton, L.M., Martin, C.W., Pierce,
   R.S., and Smith, C.T: 1989, Env. Mgt. 13, 593.

Gorham, E.: 1954, J. Ecol. 42, 497.

Hanson, D.W.: 1980, M.S. Thesis, Univ. Maine, Orono. 91 pp.

Hartig, R.: 1897, Forstlich Naturwissenschaftliche Zeitschrift 6, 49.

Heilman, P.E.: 1968, Ecology 49, 331.

Heimburger, C.C.: 1934, Cornell (Ithaca) Univ. Agr. Exp. Sta. Mem.
   165. 122 pp.

Hopkins, H.T.: 1939, M.S. Thesis, Cornell Univ., Ithaca, NY. 66 pp.

Hough, A.F.: 1943, Soil Sci. 56, 19.

Hoyle, M.C.: 1973, USDA For. Serv. Res. Pap. NE-260. 18 pp.

Kimball, K.D., Jagels, R., Gordon, G.A., Weathers, K.C., and Carlisle,
   J.: 1988, Water Air Soil Pollut. 39, 383.

Kimmins, J.K. and Hawkes, B.C.: 1978, Can. J. For. Res. 8, 265.

Krause, H.H., Reiger, S., and Wilde, S.A.: 1959, Ecology 40, 492.

Lang, G.E., Cronan, C.S., and Reiners, W.A.: 1981, Can. J. For. Res.
   11, 388.

Lunt, H.A.: 1932, Conn. Agr. Exp. Sta. Bull. 342. pp. 743-835.

Mader, D.L.: 1953, Soil Sci. Soc. Am. Proc. 17, 155.

Mattson, S., Sandberg, G., and Turning, R.E.: 1944, Ann. Agr. Coll.
   Sweden 12, 101.

McGinnis, J.T.: 1958, M.S. Thesis, Univ. Tenn., Knoxville. 82 pp.

McGuire, G.A.: 1983, M.S. Thesis, Univ. Tenn., Knoxville. 88 pp.

McLaughlin, S.B., Downing, D.J., Blasing, T.J., Cook, E.R., and Adams,
   H.S.: 1987, Oecologia 72, 487.

Mollitor, A.V. and Raynal, D.J.: 1982, Soil Sci. Soc. Am. Proc. 46,
   137.

Momoshima, N. and Bondietti, E.A.: 1990, Can. J. For. Res. 20, 1840.

Moore, B.: 1922, J. Forestry 20, 233.

NAPAP: 1990, 1989 Annual Report, National Acid Precipitation Assessment
    Program, Washington, DC.
Ovington, J.D.:   1958, J. Ecol. 46, 391.
Page, G.:   1971, Can. J. For. Res. 1, 17.
Pilgrim, S.A.L. and Harter, R.D.:   1977, N.H. Agr. Exp. Sta. Bull.   507.
Plice, M.J.: 1934, Cornell (Ithaca) Univ. Agr. Exp. Sta. Mem. 166. 32pp.
Ponomareva, V.V.:   1969, Theory of Podzolization.   Israel Program for
    Scientific Translation.   TT 68-50442.   National Technical Information
    Service, Springfield, VA.
Rennie, P.J.:   1955, Plant Soil 7, 49.
Reuss, J.O. and Johnson, D.W.:   1986, Ecological Studies Series No. 50.
    Springer-Verlag, New York.
Romell, L.G. and Heiberg, S.O.:   1931, Ecology 12, 567.
Schroeder, W.H., Bauch, J., and Endeward, R.:   1988, Trees 2, 96.
Shortle, W.C. and Smith, K.T.:   1988, Science 240, 1017.
Shortle, W. and Stienen, H.:   1988, USDA For. Serv. Gen. Tech. Rep.
    NE-120, pp. 519-525.
Sirèn, G.:   1955, Acta Forest. Fenn. 62, 1.
Soil Survey Staff: 1960, Soil Classification.   U.S. Govt. Printing
    Off., Washington, DC.
Soil Survey Staff: 1975, Soil Taxonomy, Handbook 436, U.S. Govt.
    Printing Off., Washington, DC.
Stark, N.   1990.   Final Report Cooperative Reserach Agreement NE-23-317
    and Prelim. Report NE-23-494, USDA For. Serv. NE For. Exp. Sta.,
    Radnor, PA.
Stienen, H. and Bauch, J.:   1988, Plant Soil 106, 231.
Williams, R.J.P.:   1970, Quart. Rev. Chem. Soc. 24, 331.
Wolfe, J.A.:   1967, Ph.D. Diss., Univ. Tenn., Knoxville, 193 pp.
Zaboyeva, I.V.:   1958, Soviet Soil Sci. pp. 237-244.