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Influence of excess nitrogen deposition on a white spruce (*Picea glauca*) stand in southern Alaska

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Abstract. Excess N delivered to forest ecosystems has been shown to alter internal ecosystem biogeochemical cycles, contribute to forest decline, and negatively affect the health of receiving waters. In the vicinity of the Nikiski Industrial Complex, Kenai Peninsula, Alaska, there has been recent concern about the influence of NH_3 emissions that have occurred for over two decades on local soils and vegetation. The study site represented an opportunity to examine the influence of elevated N deposition on a northern coniferous ecosystem in an area with a low background of N deposition. Overstory vegetation in the area is dominated by white spruce (*Picea glauca* Moench. Voss) and paper birch (*Betula papyrifera* Marsh.). Mortality of both species has occurred adjacent (<2 km) to the industrial complex. Average annual N deposition rates ranged from 0.7 to 21.0 kg $ha^{-1}y^{-1}$ in the area, with the highest rates closest to the complex. Sulfate deposition at the site was low. Due to the high NH_3 deposition, precipitation near the complex was less acidic than precipitation in general; bulk precipitation pH ranged from 5.51 to 7.06. Within 1.80 km of the facility there was an increase in KCl- and resin- extractable soil NH_4^+ and NO_3^- in the O horizon, and a decrease in soil pH compared to soils further from the facility. Spruce near the facility had chlorotic foliage and thinning crowns, higher concentrations of N, but lower foliar Ca and Mg. Foliar Mg levels approached deficiency levels, but foliar Ca was well above reported deficiency levels at all sites. Both Mg:N and Ca:N ratios, however, suggest nutrient imbalances in the high N deposition zone. Canopy death and fertilization by N appear to have encouraged growth of the native bluejoint grass. The presence of elevated NO_3^- in O horizon soil extracts, elevated NO_3^- in resin bags placed between the O and E horizons, and nutritional imbalances in the foliage suggest that N saturation may be occurring in soils adjacent to the facility.

Introduction

Pre-industrial atmospheric N inputs are estimated to have been relatively low (1–2 kg $ha^{-1}y^{-1}$) (Hedin et al. 1995). Air pollution has increased N deposition far beyond natural levels in some areas, and excess N deposition is thought to cause nitrogen saturation and forest health problems in Europe and the northeast USA (Nihlgard 1985; Agren & Bosatta 1988; van Breemen

& Van Dijk 1988; Aber et al. 1989, 1993; Schulze et al. 1989). Nitrogen deposition rates as high as 70 kg $ha^{-1}y^{-1}$ have been reported from Europe (Grennfelt & Hultberg 1986; Nihlgard 1990; van Breemen 1990). Based on soil chemistry, Nilson (1989) suggested that critical loads for N deposition in European forests could be 3–15 kg $ha^{-1}y^{-1}$ for coniferous forests and 5–10 kg $ha^{-1}y^{-1}$ for deciduous forests. Excess N is though to cause nutrient imbalances in foliage, lower root production and mycorrhizal associations, decrease productivity, increase susceptibility to diseases and insects, and also to change successional patterns.

Johnson & Lindberg (1992) and Van Cleve & Alexander (1981) reported N inputs of 2.1 kg $ha^{-1}y^{-1}$ in central Alaska, which appear close to pre-anthropogenic levels. However, there has been recent concern about the influence of industrial atmospheric emissions on vegetation in the vicinity of the Nikiski Industrial Complex, Kenai Peninsula, Alaska, where facilities for urea processing, petroleum refinement, gas extraction and electrical power generation are located. The dominant vegetation is white spruce (*Picea glauca* Moench. Voss) and paper birch (*Betula papyrifera* March.) with some poplar (*Populus balsamifera* L. × *Populus trichocara* Torr. & Gray). In very wet low lying environments, muskeg communities (sphagnum/shrub/*Picea mariana* bog-like areas) are present. Mortality of both white spruce and birch has occurred adjacent to the industrial complex since 1972 when the fertilizer plant opened (Keith Laurie, Unocal Chemicals Division, Kenai, Alaska, pers. comm.). Emergency releases of concentrated NH_3 gas during the early years of operation (1972 to 1979) have been informally attributed as the cause of the earliest tree mortality. However, since the 1980s, NH_3 emissions have been reduced and safety features have been added at the urea fertilizer plant so that N outputs have been reduced (Denise Newbould, Unocal Chemicals Division, Kenai, Alaska, pers. comm.). In high concentrations, NH_3 is known to cause direct damage to vegetation (Temple et al. 1979; Malhotra & Blauel 1980). Sulfur dioxide is also emitted from the complex, but in relatively low amounts and so is not considered a serious threat to ecosystem health.

Because most of the forest in the Nikiski area is not under the influence of N deposition beyond natural levels, the area immediately around the Unocal facility site represented an opportunity to examine the influence of excess N deposition, without significantly elevated S deposition, on northern coniferous ecosystems. The goal of this study was to examine the effect of chronic high NH_3 addition to soils and vegetation in the area immediately adjacent to the facility, and to determine if symptoms of nitrogen saturation were present. This was done by examining: (1) the pH and total N in precipitation (snow and rain), (2) soil solution chemistry and the uptake of N species by resin

bags, (3) the chemical status of the foliage, especially that of N, Mg, K and P, and (4) results between visibly affected and visibly unaffected forests.

Site description

The study site was located on the Kenai Peninsula, Alaska, adjacent to the Nikiski Industrial Complex containing the Unocal urea processing facility, the Tesoro oil refinery, and the Phillips 66 natural gas facility north of the city of Kenai at 60.38°N, 151.18°W (Figure 1). This area is part of a lowland outwash plain less than 30 m above sea level on flat topography that stretches from the northwest "elbow" of the peninsula down the west side of the peninsula (Reiger 1979).

The northern area of Kenai peninsula is a mixture of maritime and continental taiga climate (Critchfield 1983). Summer rains dominate the hydrologic regime with an average of 278 mm (55%) falling as rain between June 1 and September 31 and 227 mm (45%) falling as snow or a rain/snow complex the rest of the year out of 505 mm annually (Anonymous 1992). Average monthly temperatures range from –11 °C to 12 °C (Reiger 1979).

The surrounding forest is dominated by white spruce. Paper birch (*Betula papyrifera*) and cottonwoods (hybrids between *Populus balsamifera* and *Populus trichocarpa*) are also present. The average age in 1992 of the white spruce was 136 years with most trees between 50 and 150 years old, and DBH ranging from 15 to 41 cm. The uneven age distribution and presence of deciduous trees may indicate multi-species invasion after glacial retreat or stand-replacing disturbance (Van Cleve & Viereck 1981). In undisturbed areas, various crustose and foliose lichens grow on tree branches, with one genus, *Peltigera*, found on the forest floor. Understory species composition is similar to other white spruce upland sites previously described by Van Cleve & Viereck (1981). Shrub, herb, moss, grass and lichen species in undisturbed areas are typical of other white spruce ecosystems. Located to the northeast of the facility is 5–7 ha of open area vegetated primarily by bluejoint grass (*Calamagrostis canadensis* (Michx.) Beauv.), where white spruce previously grew.

Soil in the area is classified as a Typic Cryorthod (Rieger 1979). The parent material of the region is a silty loess or mixture of loess and volcanic ash varying in thickness over a substratum that ranges from outwash gravel to compact till. Forest floor thickness varies from 8–10 cm. Volcanic ash has been identified as an important part of Kenai-area soils, which lowers the bulk density of and adds exchangeable aluminum to the soils (Ping et al. 1988; Shoji et al. 1988). The cryorthods of the Kenai Peninsula are also

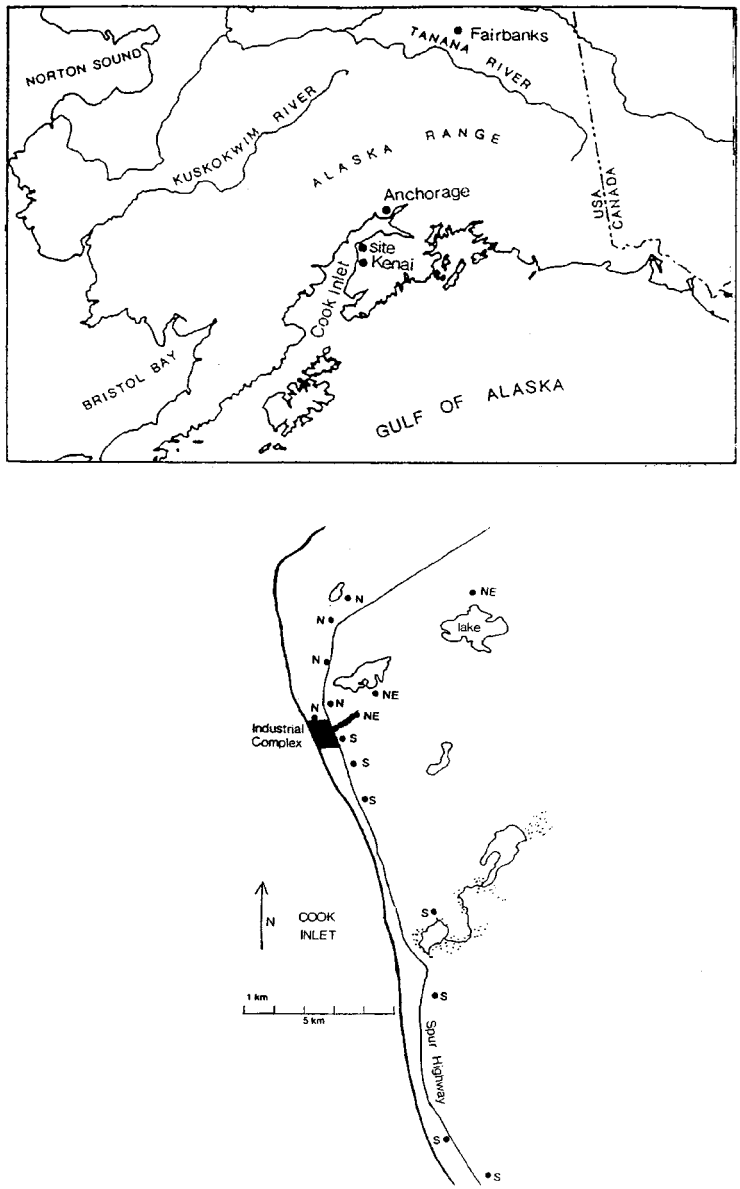

Figure 1. Map of the industrial complex and study plots along three transects on the Kenai Peninsula, Alaska.

characterized by low base saturation (2–14%) with most of the exchange sites contributed by soil organic matter (Shoji et al. 1988; Ping et al. 1989).

Methods

Experimental design

Twenty one plots were laid out on three transects radiating from the facility in the N, NE and S directions with plot distances ranging from 0.35 km to 13.23 km from the facility (Figure 1). Regular spacing was not possible due to problems with land ownership and access. Plot centers were chosen from Bureau of Kenai aerial photos. Each plot was 12×12 m square with the sides in the cardinal directions.

Deposition

Each site had one bulk deposition collector that was placed in the nearest canopy gap to ensure that throughfall was not sampled. Collectors were constructed from a 10-cm diameter Nalgene funnel inserted through a rubber stopper which was inserted into a 1 L Nalgene container. Washed polyester fiber was placed in the bottom of funnels to restrict large particle collection. Collectors were then elevated 1 m above the ground by wooden poles.

Precipitation collectors were left in the field and collected biweekly between July and October 1992, to allow enough precipitation to collect (Leonardi & Fluckiger 1987), but also to collect the precipitation as often as possible to avoid algal growth in the collectors and loss of NH_4^+. Snow samples were collected in April of 1992, roughly at the beginning and end of each transect for a total of six samples. After collection, precipitation samples were refrigerated at 4 °C. pH was determined with a Extech model 671 pH meter within 4 days of collection. Samples were then refrigerated (4 °C) or frozen, depending upon available facilities. All water samples were filtered through Whatman GF/A filters in acid-washed glassware prior to laboratory analysis.

Soil sampling

Soils were randomly sampled by horizon in 188 cm^3 cores at three randomly chosen sites within 20 m of each plot center. The three samples from each horizon were composited in the field. Soils were stored between 10 to 20 °C in paper bags until passed through a 2 mm mesh sieve. Two lab samples were analyzed for major ions from the O horizon. A portion of each sample was

oven dried at 105 °C for 24 hours to obtain an oven dry weight conversion factor. Soil pH was measured with a 1:1 (by volume) soil: water slurry on air dried soil using a Corning (model 220) pH meter. Soil extracts were analyzed for NH_4^+ and NO_3^-.

Resin bags

Two resin bags per plot were placed along the north and south directions and close to each precipitation collector. Resin bags were made of approximately 5 g (wet weight) of mixed resign (Baker HOH ion exchange mixed resin), held in nylon bags, and were placed between the O and E horizons and left for 10 weeks (7/14/92–9/23/92). Bags were removed and extracts were analyzed for NH_4^+ and NO_3^-.

Foliage sampling

At 13 of the forested plots, five foliage samples per plot were collected from five healthy spruce trees, not more than 30 m from the center of the plot. Trees were chosen from a predetermined DBH (21–34 cm) and height (13–23m) range in order to reduce variability. From each tree a pole pruner was used to sever a sun-exposed branch from the mid third of the canopy. A combination of current-and first-year foliage was collected and stored for 30 days at room temperature prior to laboratory analysis.

Laboratory analysis

Chemical analyses were conducted in the Analytical Laboratory in the College of Forest Resources, University of Washington. Internal lab standards were used for quality control. All soil, foliage, and bulk precipitation samples were digested using a modified Kjeldahl digest (Parkinson & Allen 1975). Analysis of Ca, K, Mg for precipitation, soil, and P for foliage samples were then determined by ICP (Jarrell Ash model 61e). Soil and resin were extracted with 2M KC1 at the ratios of 1:10 and 1:5 by volume, respectively. The extracts were then analyzed for NO_3^- and NH_4^+ using a Technicon II Autoanalyzer. Total N in foliage and soil was determined using a Perkin-Elmer 2400 CHN analyzer.

Calculations

The Systat program was used to calculate non-linear regression statistics and to quantify trends in the data sets to determine the effects of transect and distance. The GLM procedure of SAS was used to perform all analysis

of variance (ANOVA) on data sets to determine the effects of transect and distance. When the GLM indicated statistical significance, a least squares mean was calculated to separate differences among transects and distance. Significance for this study was established at $\alpha = 0.05$.

Concentrations of chemical constituents from precipitation were multiplied by the precipitation received from May to September, 1992 as determined by the National Weather Service (Anonymous 1992). Snow samples were analyzed and the average value for each snow plot was used as a snow measurement for the closest transect plot where only precipitation was collected.

Results

Bulk deposition

Calamagrostis canadensis (native bluejoint grass) dominated the understory vegetation close to the facility creating a natural and visually obvious separation between the plots within 1.80 km of the facility and those farther away. We used this natural break for statistical purposes. The plots within 1.80 km of the facility had the highest measured total N in bulk precipitation ($P = 0.0277$) compared to precipitation collected at farther sites (Figure 2A), with total N deposition in bulk precipitation decreasing exponentially with distance from the facility ($y = 9.852x^{-0.480}$).

Soil

KCl- extractable NH_4^+ (Figure 2B) was highest within 1.80 km of the facility ($P = 0.0006$), and showed a significant decrease with distance from the facility in all transects ($y = 0.16x^{-0.259}$). Patterns for resin NH_4^+ concentrations were similar, with significantly more NH_4^+ extracted by resins close to the facility than in plots farther away ($P = 0.0215$). KCl- extractable NO_3^- in soil was also highest within 1.80 km of the facility ($P = 0.0006$) (Figure 2C), and showed a significant trend with distance ($y = 0.736x^{-0.04}$). Total soil N was significantly higher close to the facility as well ($P = 0.0001$). The southern transect showed significantly higher NO_3^- levels compared to other transects in plots close to the facility, through south transect values were significantly lower than other transects in plots farther away from the facility. The dichotomy may have been due to the erratic spacing of the plots. A more consistent spacing of plots would have provided a larger sample size and possibly less polarized results. Resin NO_3^- values, like soil NO_3^-, were significantly higher within 1.80 km of the facility compared with plots farther away ($P = 0.0027$), and were significantly lower throughout the southern transect ($P < 0.0005$).

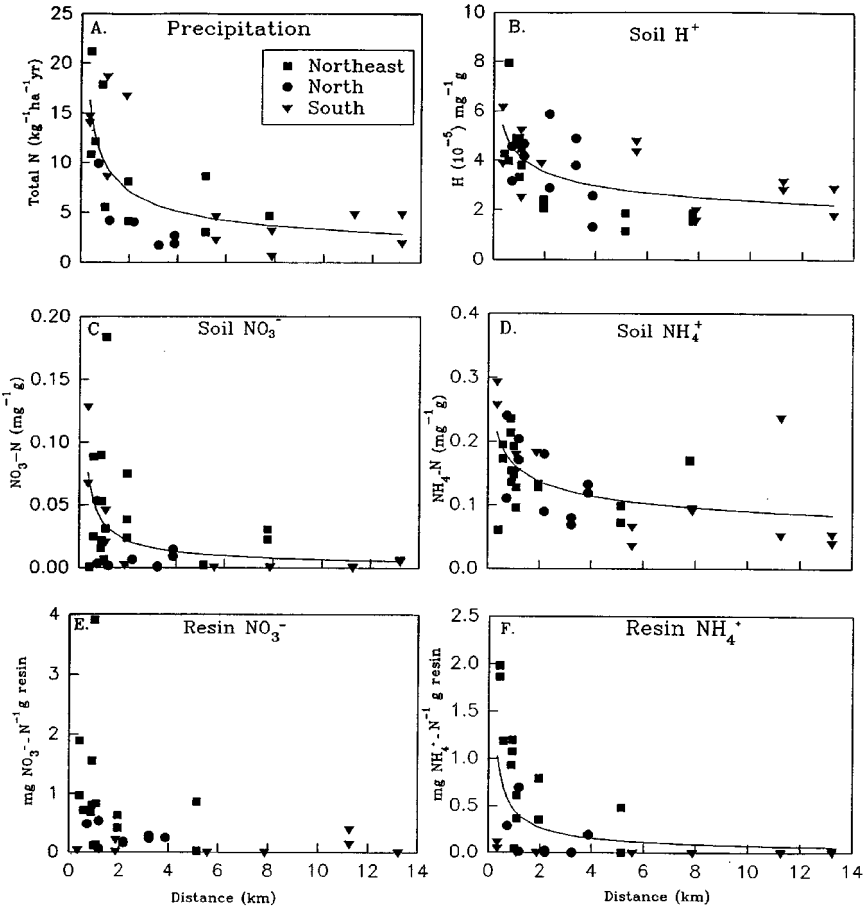

Figure 2. A) Total N in bulk precipitation with distance from the facility, B) Extractable NH_4^+-N of soils with distance from the facility, C) Extractable NO_3^--N of soils with distance from the facility, and D) Hydrogen ion concentration in soils with distance from the facility. Different transects are denoted by the three different symbols.

pH values were expressed as $[H^+]$ for statistical analysis (Figure 2D). Plots within 1.80 km of the facility had higher $[H^+]$, and therefore lower pH, than plots farther away ($P < 0.0001$), and there was a significant decrease in $[H^+]$ with distance from the facility ($y = 4.12(10^{-5})\ x^{-0.248}$). Beyond 1.80 km, the NE transect had the lowest $[H^+]$ concentration of any of the transects ($P = 0.0156$), probably due to vegetation differences near the end of this transect (Whytemare 1994). The two most acidic soils were found in the closest southern site (pH = 4.2) and a close northeastern plot (pH = 4.1), both within 1 km of the facility.

Foliage

Data on nutrient deficiency levels in foliage for *Picea glauca* are not common, therefore we have used Ingestad's (1959) optimum and deficiency levels of macronutrients for *Picea abies*. These numbers are comparable to deficiency levels in most conifers (Wilde et al. 1985). The four forested plots within 1.80 km of the facility had higher foliar N than plots farther from the facility (Figure 3A) (P = 0.0001). Foliage in the NE transect showed significantly higher levels of N within the close plots (P = 0.011) than foliage in the N and S transects. In the plots less than 1.80 km for the facility, S transects foliage had the lowest N concentrations (P = 0.0233). All N values in foliage were above deficiency levels as determined by Ingestad (1959) for *Picea abies*. Foliar Ca (Figure 3B) was significantly lower (P = 0.0001) in the 4 plots closest to the facility, but all foliar Ca levels were well above deficiency levels. Both foliar K (Figure 3C) and foliar P (Figure 3E) concentrations were significantly higher in the far sites compared to sites within 1.80 km of the facility (P = 0.006 and P = 0.0261, respectively). However, the interaction between distance and transect was significant, as only the NE transect showed the increase in foliar P and K with distance, and only in the furthest two plots. The furthest two NE inland sites may well have developed on slightly different soils, as the surrounding vegetation was dominated by different shrubs and tended to have more deciduous trees in the overstory than the north or south transect sites which were similar distances from the ocean (Whytemare 1994). S and N transect foliage did not show significant changes in foliar P and K with distance. Foliar Mg (Figure 3D) was significantly lower in plots close to the facility (P = 0.004), with one close NE plot's mean below deficiency levels.

Mg:N ratios were significantly lower in the close plots than in plots far away (P = 0.0001), and all close plots fell below the ratios indicating relative Mg deficiency established by Ingestad (1959) (Figure 3F). Within the plots close to the facility, the N transect showed significantly higher Mg:N ratios (P = 0.0471) than the other transects, through there were no differences among farther transects. Foliar Ca:N did not show differences among transects at any distance, and close plots had significantly lower ratios than those farther away (P = 0.0001) (Figure 3G).

Discussion

The pattern of N in precipitation is consistent with data on wind direction taken in 1989 by the state of Alaska (Sullivan et al. 1990), which showed that the predominant wind direction is from the south, corresponding to higher N deposition in the northern transects, downwind of the facility. The lowest

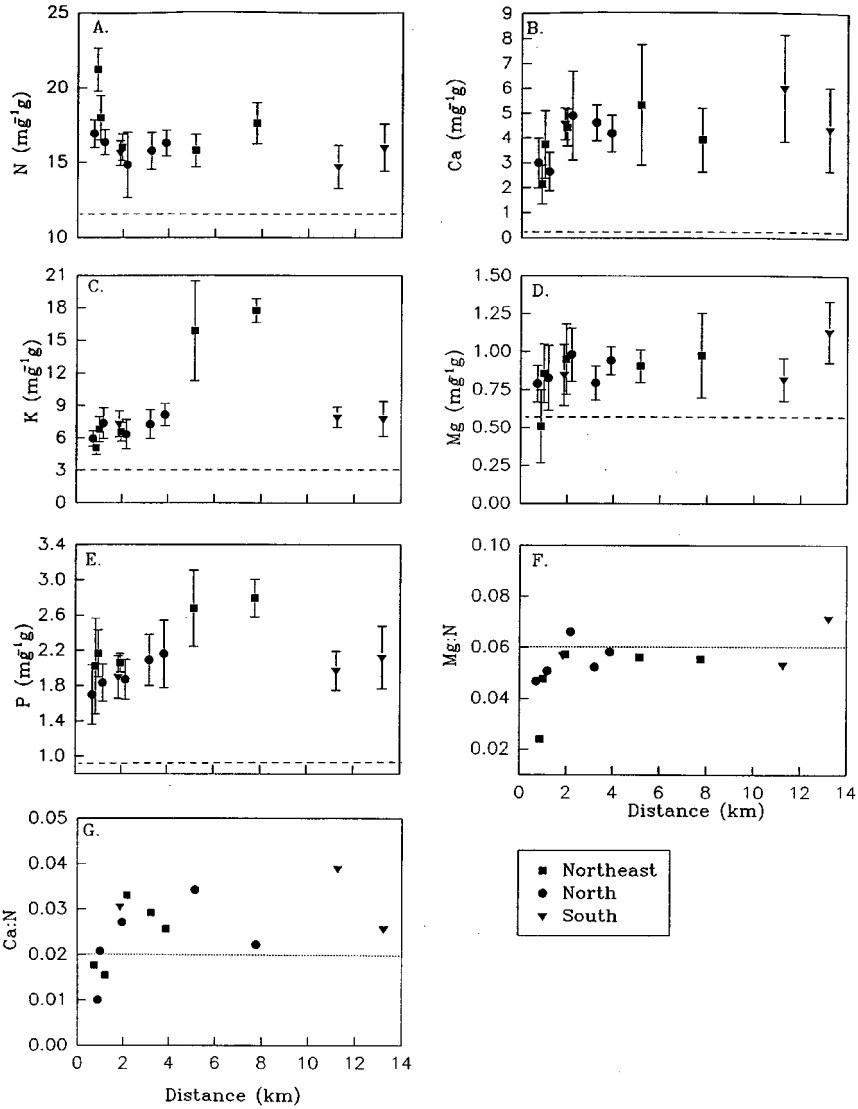

Figure 3. Concentrations of: A) N, B) Ca, C) K, D) Mg, E) P in foliage with distance from the facility. F) Mg:N, and G) Ga:N in foliage with the distance from the facility. Dashed lines (A–E) indicate the point of absolute deficiency, and dotted lines (F & G) indicate relative deficiency based on Ingestad's (1959) study of *Picea abies*.

levels of N in precipitation were similar to that of a remote coastal site in SE Alaska (Bormann et al. 1989) and another site in Central Alaska (Van Cleve & Alexander 1981). The highest levels of total N in bulk deposition observed in this study were similar to high-deposition areas of Germany (Matzner 1986;

Renk & Fischer 1988; Krzak et al. 1988; Kaupenjohann 1989; Schulze 1989, 1990; Schulze et al. 1990) while Dutch sites have shown up to twice these levels (van Breemen et al. 1987; Tietema & Verstraten 1991; Duyzer et al. 1992).

Because precipitation inputs near the facility were less acidic from high NH_3 inputs than clean precipitation in equilibrium with CO_2, the decrease in soil pH near the facility must have been driven by dynamics other than precipitation inputs. We suggest that this relative decrease in soil pH was the result of elevated nitrification (van Breemen et al. 1982; Binkley & Richter 1987; Nilsson et al. 1988). Other investigators have shown that increased N deposition and NH_4^+ buildup leads to soil NO_3^- production, which reduces soil pH and can cause NO_3^- and cations to leach out of the soil (Johnson 1992; Klein et al. 1983; Krzak et al. 1988; Nilsson et al. 1988; Preston et al. 1983; Roelofs et al. 1985; van Breemen & Jordens 1983; van Breemen et al. 1984, 1987; Van Miegroet & Cole 1984, 1985). Typically, NO_3^- production is low in white spruce stands (Gordon 1982) due to low soil NH_4^+ concentrations, low soil pH, and the presence of Al and Cl (Roseberg et al. 1986; Ohno et al. 1988). However, the increased NH_4^+ delivered to the forest floor near the facility in this study caused an increase in NO_3^- production, as seen by soil KCl extracts and elevated NO_3^- captured by resin bags.

Only one plot close to the facility showed deficient levels of Mg in foliage, but Ca was always well above deficiency levels. However, Ingestad (1959) found that nutrient deficiencies in *Picea abies* may also occur according to ratios of nutrients to one another. If Ingestad's numbers are applied to this study, Ca:N and Mg:N ratios in foliage of trees surrounding the facility suggest nutrient imbalances. Elevated N may cause a relative deficiency of other ions such as P or base cations. Excess N deposition has been shown to increase shoot and crown growth, which in turn increases plant demand for other nutrients and decreased the tree's ability to take these ions up from the soils due to lowered root growth (Oren et al. 1988; Schneider et al. 1989). Yet acidification from acid precipitation and nitrification of NH_4^+ inputs and soil dynamics may trigger leaching of base cations from soils, exacerbating cation limitation (Schneider et al. 1989).

The decreases in foliar Mg and Ca levels close to the facility could be the result either of Mg and Ca leaching from the foliage or of the lack of Mg and Ca in the tree's rooting zone. To maintain a charge balance, the plant either takes up an amount of anion in precipitation equivalent to the protons or cations taken up, or else leaches cations equivalent to the new cation taken up (Cronan & Reiners 1983; Lovett et al. 1985; Matzner 1986). In a declining forest stand in Germany, Oren et al. (1988) found that foliar leaching was not the reason for Mg deficient foliage; rather, Mg xylem concentrations

were low. This finding suggests that root uptake is more critical to the cation balance of the tree than is foliar leaching (van Breemen & van Dijk 1988; Lovett & Schaefer 1991). Decreased root uptake of Mg and Ca might also be due to preferential uptake of NH_4^+ over nutrient cations, and both Mg and Ca are particularly susceptible to ion competition with NH_4^+ (Kirkby 1981; Schulze et al. 1989).

At the Hubbard Brook Experimental Forest in New Hampshire there is concern that acid rain has made the ecosystem susceptible to excessive depletion of soil base cations, especially Ca (Federer et al. 1989; Likens et al. 1996). Declining Ca resulting from acid deposition has also been the focus of studies on the soils of the red spruce zone in New England (Lawrence et al. 1995) and in *Picea abies* stands in the Fichtelgebirge of Germany (e.g. Meyer et al. 1988; Oren et al. 1988). However, none of these studies have made the causal link between Ca depletion in soils and Ca deficiencies in trees. Given the low foliar concentrations of Ca required by *Picea* and other conifers for healthy growth (Ingestad 1959), it may be that trees from these studies, like the trees in the current study, are well above deficiency levels for Ca. For example, Schulze (1989) showed that Mg deficiency rather than Ca deficiency caused forest decline symptoms, despite leaching of both Ca and Mg from soils, in a German forest with high acid and NH_4^+ inputs. However, given the high rates of Ca depletion from leaching and forest harvest in New England (Federer et al. 1989; Likens et al. 1996), and the suggested role for Ca to buffer Al in the soil solution (Meyer et al. 1988), it is quite possible that limitation by multiple ions will become problematic in the future.

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References

Aber JD (1992) Nitrogen cycling and nitrogen saturation in temperate forest ecosystems. TREE 7: 220–224

Aber JD, Nadelhoffer KJ, Steudler P & Melillo JM (1989) Nitrogen saturation in northern forest ecosystems. BioScience 39: 378–386

Ågren GI & Bosatta E (1988) Nitrogen saturation in terrestrial ecosystems. Environ. Poll. 54: 185–198

Alexander M (1971) Introduction to Soil Microbiology. John Wiley & Sons, New York

Anonymous (1992) Climatological data, Alaska. National Climatic Data Center, NOAA
Binkley D & Richter D (1987) Nutrient cycles and H^+ budgets of forest ecosystems. Adv. Ecol. Res. 16: 1–51
Bormann BT, Tarrant RF, McClellan MH & Savage T (1989) Chemistry of rainwater and cloud water at remote sites in Alaska and Oregon. J. Environ. Qual. 18: 149–152
Cronan CS (1991) Differential adsorption of Al, Ca, and Mg by roots of red spruce (*Picea subens* Sarg.). Tree Physiol. 8: 227–237
Duyzer JH, Verhagen HLM, Westrate JH & Bosveld FC (1992) Measurement of the dry deposition of NH_3 on to a coniferous forest. Environ. Poll. 75: 3–13
Flanagan PW & Van Cleve K (1983) Nutrient cycling in relation to decomposition and organic-matter quality in taiga ecosystems. Can. J. For. Res. 13: 795–817
Gordon AM (1982) Seasonal pattern of nitrogen mineralization following logging in the white spruce (*Picea glauca* [Moench.] Voss.) forests of Interior Alaska. Interim progress report to the Vice Chancellor of Research and Advanced Study and the Institute of Northern Forestry, University of Alaska. 30 p
Grennfelt P & Hultbert H (1986) Effects of nitrogen deposition on the acidification of terrestrial and aquatic ecosystems. Water Air Soil Poll. 30: 945–963
Hedin LO, Armesto JJ & Johnson AH (1995) Pattern of nutrients loss from unpolluted, old-growth temperate forests: evaluation of biogeochemical theory. Ecology 76: 493–509
Ingestad T (1959) Studies on the nutrition of forest tree seedlings II. Mineral nutrition of spruce. Physiol. Plant. 12: 568–593
Johnson DW (1992) Nitrogen retention in forest soils. J. Environ. Qual. 21: 1–12
Kaupenjohann M (1989) Effects of acid rain on soil chemistry and nutrient availability in the soil. In: Schulze E-D, Lange OL & Oren R (Eds) Forest Decline and Air Pollution (pp 297–337). Springer-Verlag, New York
Kenk G & Fischer H (1988) Evidence from nitrogen fertilization in the forests of Germany. Environ. Poll. 54: 199–218
Kirkby EA (1981) Plant growth in relation to nitrogen supply. In: Clark FE & Rosswall T (Eds) Terrestrial Nitrogen Cycles (pp 249–267). Ecological Bulletin 33, Stockholm
Klein TM, Kreitinger JP & Alexander M (1983) Nitrate formation in acid soils from the Adirondacks. Soil Sci. Soc. Am. J. 47: 506–508
Krzak J, Hoang Dong P, Buttner G, Huttermann A, Kramer H & Ulrich B (1988) Photosynthesis, nutrient, growth and soil investigation of a declining Norway Spruce (*Picea abies*) stand in the coastal region of Northern Germany. For. Ecol. Manage. 24: 263–281
Lawrence GB, David MB & Shortle WC (1995) A new mechanism for calcium loss in forest floor soils. Nature 378: 162–165
Leonardi S & Fluckinger W (1987) Short-term canopy interactions of beech trees: mineral ion leaching and absorption during rainfall. Tree Physiol. 3: 137–145
Likens GE, Driscoll CT & Buso DC (1996) Long-term effects of acid rain: response and recovery of a forest ecosystem. Science 272: 244–246
Lovett GM, Lindberg SE, Richter DD & Johnson DW (1985) The effects of acidic deposition on cation leaching from three deciduous forest canopies. Can J. For. Res. 15: 1055–1060
Lovett GM & Schaefer DA (1991) Canopy interactions of Ca, Mg, and K. In: Johnson DW & Lindberg SE (Eds) Atmospheric Deposition and Forest Nutrient Cycling (pp 253–274). Springer-Verlag, New York
Malhotra SS & Blauel RA (1980) Diagnosis of air pollutant and natural stress symptoms on forest vegetation in western Canada. Northern Forest Research Centre, Edmonton, Alberta. Information Report NOR-X-228
Matzner E (1986) Deposition/canopy-interactions in two forest ecosystems of Northwest Germany. In: Georgi H-W (Ed) Atmospheric Pollutants in Forest Areas: Their Deposition and Interception (pp 247–262). D. Reidel Pub. Co., Dordrecht
Meyer J, Schneider BU, Werk K, Oren R & Schulze E-D (1988) Performance of two *Picea abies* (L.) Karst. stands at different stages of decline. V. Root tip and ectomycorrhiza development and their relations to above ground and soil nutrients. Oecologia 77: 7–13

Nihlgard B (1985) The ammonium hypothesis – an additional explanation to the forest dieback in Europe. Ambio 14: 2–8
Nilsson J (1989) Critical loads for sulphur and nitrogen. Acidification Research in Sweden, Nat. Swed. Envir. Program. 8: 1–2
Nilsson SI, Berden M & Popovic B (1988) Experimental work related to nitrogen deposition, nitrification and soil acidification – a case study. Environ. Poll. 54: 233–248
Ohno T, Sucoff EI, Erich MS, Bloom PR, Buschena CA & Dixon RK (1988) Growth and nutrient content of red spruce seedlings in soil amended with aluminum. J. Environ. Qual. 17: 666–672
Oren R, Schulze E-D, Werk KS & Meyer J (1988) Performance of two *Picea abies* (L.) Karst. stands at different stages of decline. VII. Nutrient relations and growth. Oecologia 77: 163–173
Parkinson JA & Allen SE (1975) A wet oxidation procedure suitable for the determination of nitrogen and mineral nutrients in biological material. Commun. Soil Sci. Plant Anal. 6: 1–11
Ping CL, Shoji S & Ito T (1988) Properties and classification of three volcanic ash-derived pedons from Aleutian Islands and Alaska Peninsula, Alaska. Soil Sci. Soc. Am. J. 52: 455–462
Ping CL, Shoji S, Ito T, Takahashi T & Moore JP (1989) Characteristics and classification of volcanic-ash-derived soils in Alaska. Soil Sci. 148: 8–28
Preston CM, Marshall VG & McCullough K (1990) Fate of ^{15}N-labelled fertilizer applied on snow at two forest sites in British Columbia. Can. J. For. Res. 20: 1583–1592
Reiger S (1979) Exploratory soil survey of Alaska. Washington, DC, USDA-SCS
Roelofs JG, Kempers AJ, Houdijk ALFM & Jansen J (1985) The effect of airborne ammonium sulfate on *Pinus nigra* var. *maritima* in the Netherlands. Plant Soil 84: 45–56
Roseberg RJ, Christensen NW & Jackson TL (1986) Chloride, soil solution osmotic potential, and soil pH effects on nitrification. Soil Sci. Soc. Am. J. 50: 941–945
Schulze E-D (1989) Air pollution and forest decline in a spruce (*Picea abies*) forest. Science 244: 776–783
Schulze E-D (1990) What can be done to aid our ailing forests? German Research 13–15
Schulze E-D, McCracken I, Zimmerman R & Benecke U (1990) Performance of two *Picea abies* (L.) Karst. stands at different stages of decline. VIII. Photosynthesis and nutrition under polluted and clean air conditions of Germany and New Zealand. Oecologia 82: 158–161
Schulze E-D, Oren R & Lange OL (1989) Nutrient relations of trees in healthy and declining Norway spruce stands. In: Schulze E-D, Lange OL & Oren R (Eds) Forest Decline and Air Pollution: A Study of Spruce (*Picea abies*) on Acid Soils (pp 392–411). Springer-Verlag, New York
Schneider BU, Meyer J, Schulze E-D & Zech W (1989) Root and mycorrhizal development in healthy and declining Norway spruce stands. In: Schulze E-D, Lange OL & Oren R (Eds) Forest Decline and Air Pollution: A Study of Spruce (*Picea abies*) on Acid Soils (pp 370–390). Springer-Verlag, New York
Shoji S, Takahashi T, Ito T & Ping CL (1988) Properties and classification of selected volcanic ash soils from Kenai Peninsula, Alaska. Soil Sci. 145: 395–413
Sullivan TJ, Rose CL, Gilfilian RE, Eilers JM, van Breemen N, Bernert JA, Hanson D & Queitzsch BE (1990) Nikishi vegetation impact assessment. State of Alaska, DEC
Temple PJ, Harper DS, Pearson RG & Linzon SN (1979) Toxic effects of ammonia on vegetation in Ontario. Environ. Poll 20: 297–302
Tietema A & Verstraten JM (1991) Nitrogen cycling in an acid forest ecosystem in the Netherlands under increased atmospheric nitrogen input. Biogeochemistry 15: 21–46
Tukey HB Jr. (1970) The leaching of substances from plants. Ann. Rev. Plant Physiol. 21: 305–324
van Breemen N (1990) Deterioration of forest land as a result of atmospheric deposition in Europe: a review. In: Gessel SP, Lacta DS, Weetman GF & Powers RF (Eds) 7th North

American Forest Soils Conference (pp 40–48). Faculty of Forestry Publication, University of British Colombia, Vancouver

van Breemen N, Burrough PA, Belthorst EJ, van Dobben HF, de Wit T, Ridder TB & Reijinders HFR (1982) Soil acidification from atmospheric ammonium sulfate in forest canopy throughfall. Nature 299: 548–550

van Breemen N, Driscoll CT & Mulder J (1984) Acidic deposition and internal proton sources in acidification of soils and waters. Nature 307: 599–604

van Breemen N & Jordens ER (1983) Effects of atmospheric ammonium sulfate on calcareous and non-calcareous soils of woodlands in the Netherlands. In: Ulrich B & Pankrath J (Eds) Effects of Accumulation of Air Pollutants in Forest Ecosystems (pp 171–182) D. Reidel Publishing Company, Boston

van Breemen N, Mulder J & van Grinsven JJM (1987) Impacts of acid atmospheric deposition on woodland soils in the Netherlands: II. Nitrogen transformations. Soil Sci. Soc. Am. J. 51: 1634–1640

van Breemen N & van Dijk HGF (1988) Ecosystem effects of atmospheric deposition of nitrogen in The Netherlands. Environ. Poll. 54: 249–274

Van Cleve K & Alexander V (1981) Nitrogen cycling in tundra and boreal ecosystems. In: Clark FE & Rosswall T (Eds) Terrestrial Nitrogen Cycles (pp 375–404). Ecol. Bull. 33, Stockholm

Van Cleve K & Viereck LA (1981) Forest succession in relation to nutrient cycling in the boreal forest of Alaska. In: West DC, Shugart HH & Botkin DB (Eds) Forest Succession: Concepts and Application (pp 185–211). Springer-Verlag, New York

Van Miegroet H & Cole DW (1984) The impact of nitrification on soil acidification and cation leaching in a red alder ecosystem. J. Environ. Qual. 13: 586–59

Van Miegroet H & Cole DW (1985) Acidification sources in red alder and Douglas-fir fir soils – importance of nitrification. Soil Sci. Soc. Am. J. 49: 1274–1279

Wilde SA, Corey RB, Iyer GG & Voigt GK (1985) Soil & Plant Analysis for Tree Culture. 5th Revised edition. Oxford & IBH Publishing Co

Whytemare AB (1994) Industrial nitrogen inputs to an Alaskan white spruce forest ecosystem. M.S. thesis. University of Washington, Seattle, Washington