Eleven-year response of foliar chemistry to chronic nitrogen and sulfur additions at the Bear Brook Watershed in Maine

Jose Alexander Elvir, Lindsey Rustad, G. Bruce Wiersma, Ivan Fernandez, Alan S. White, and Gregory J. White

Abstract: The foliar chemistry of sugar maple (*Acer saccharum* Marsh.), American beech (*Fagus grandifolia* Ehrh.), and red spruce (*Picea rubens* Sarg.) was studied from 1993 to 2003 at the Bear Brook Watershed in Maine (BBWM). The BBWM is a paired-watershed forest ecosystem study, with one watershed treated bimonthly since 1989 with ammonium sulfate ($(NH_4)_2SO_4$) at a rate of 25.2 kg N·ha^{-1}·year^{-1}. Foliar N concentrations were higher in all tree species within the treated watershed compared with trees within the reference watershed. Foliar Ca and Mg concentrations were lower in American beech and red spruce within the treated watershed. There were no significant differences in foliar K concentrations between watersheds. Foliar P and Mn concentration differences between watersheds were inconsistent among years. Differences in foliar N concentrations between watersheds declined over time in sugar maple but not in red spruce or American beech. Differences in foliar Ca and Mg concentrations between the treated and reference watersheds increased over time for American beech and red spruce, primarily because of a consistent decline in concentrations of these nutrients in trees within the treated watershed. No temporal trends in foliar Ca and Mg concentration differences between watersheds were observed for sugar maple.

Résumé : Les caractéristiques chimiques des feuilles d'érable à sucre (*Acer saccharum* Marsh.), de hêtre à grandes feuilles (*Fagus grandifolia* Ehrh.) et d'épinette rouge (*Picea rubens* Sarg.) ont été étudiées de 1993 à 2003 dans le bassin de Bear Brook, dans le Maine, où se déroule une étude sur l'écosystème forestier à l'échelle du bassin. Cette étude est menée dans deux bassins jumelés dont un est traité sur une base bimensuelle depuis 1989 avec du sulfate d'ammonium ($(NH_4)_2SO_4$) au taux de 25,2 kg N·ha^{-1}·an^{-1}. La concentration de N dans les feuilles de toutes les espèces d'arbres dans le bassin traité était plus élevée comparativement à celle des arbres du bassin témoin. La concentration de Ca et Mg dans les feuilles était plus faible chez le hêtre à grandes feuilles et l'épinette rouge dans le bassin traité. Il n'y avait pas de différence significative entre les bassins dans la concentration foliaire de K. Dans le cas de la concentration foliaire de P et Mn, la différence entre les bassins n'était pas consistante d'une année à l'autre. La différence dans la concentration foliaire de N entre les bassins a diminué avec le temps chez l'érable à sucre mais pas chez l'épinette rouge ni le hêtre à grandes feuilles. La différence dans la concentration foliaire de Ca et Mg entre les bassins traité et témoin a augmenté avec le temps chez le hêtre à grandes feuilles et l'épinette rouge, surtout à cause d'une diminution constante de la concentration de ces nutriments dans les arbres du bassin traité. Dans le cas de l'érable à sucre, aucune tendance dans le temps n'a été observée dans la différence de concentration de Ca et Mg entre les bassins.

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Introduction

Elevated deposition of atmospheric nitrogen (N) increases soil N availability, which may in turn result in increased plant N uptake in N-limited ecosystems (Aber et al. 1989; 1998; Driscoll et al. 2001). Foliar N concentrations, for example, have been shown to increase as a result of both enhanced atmospheric N deposition and N fertilization (Magill et al. 1997; Bertills and Nasholm 2000). Given the reported positive relationship between foliar N concentrations and CO_2 uptake (Evans 1989; Lambers et al. 1998), increased soil N availability could increase forest growth, resulting in

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J.A. Elvir.[1] Escuela Nacional de Ciencias Forestales, Apartado Postal 2, Siguatepeque, Comayagua, Honduras.
L. Rustad. USDA Forest Service Northeastern Research Station, 271 Mast Road, Durham, NH 0324, USA.
G.B. Wiersma. Department of Forest Ecosystem Science, The University of Maine, 105 Winslow, Orono ME 04469, USA.
I. Fernandez. Department of Plant, Soil, and Environmental Sciences, The University of Maine, 1 Deering Hall, Orono ME 04469, USA.
A.S. White. Department of Forest Ecosystem Science, The University of Maine, 217.Nutting Hall, Orono ME 04469, USA.
G.J. White. Idaho National Engineering and Environmental Laboratory, P.O. Box 1625-2213 Idaho Falls, ID 83415–2213, USA.

[1]Corresponding author (e-mail: alexit98@yahoo.com).

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greater C storage in biomass (Lloyd 1999) and greater inputs of C to the soil via detritus and root exudates (Hagedorn et al. 2003). Alternatively, elevated N deposition has also been shown to accelerate the leaching of soil base cations, leading to imbalances of foliar nutrients and ultimately to forest decline (Oren et al. 1993).

A decline in the availability of nutrients such as calcium (Ca) and magnesium (Mg) and a concurrent increase in N availability and acidity in forest soils have been attributed to prolonged N deposition in some areas in the northeastern United States and central Europe (Graveland et al. 1994; Johnson et al. 1994; Driscoll et al. 2001). As N accumulates in the soil, ecosystem N retention capacity can be surpassed, resulting in increased NO_3^- leaching and concomitant base cation leaching (Norton el al. 1999). Nasholm et al. (2000) discussed how forest trees growing under high N deposition could be exposed to two forms of excess N: an absolute excess of N, when there is more N available than the tree is able to use at its maximum growth rate; or a relative N excess, when there is a deficiency of other nutrients that limit growth, resulting in a surplus of N over time.

Foliar nutrient imbalances have been reported in trees growing in controlled N-fertilization experiments over areas with different N deposition rates and along N deposition gradients (Friedland et al. 1991; McNulty et al. 1996; Magill et al. 1997; Boxman et al. 1998). However, long-term foliar nutrient concentration data are not usually available because of a lack of long-term N studies and systematic measurements.

Our study focused on temporal changes in the foliar nutrient concentrations of three tree species, sugar maple (*Acer saccharum* Marsh.), American beech (*Fagus grandifolia* Ehrh.), and red spruce (*Picea rubens* Sarg.), at the Bear Brook Watershed in Maine (BBWM). The BBWM is a paired-watershed study in which one watershed has been treated with bimonthly additions of ammonium sulfate ($(NH_4)_2SO_4$) from 1989 through 2003. Previous whole-ecosystem mass balance and soil chemical studies at the BBWM have indicated that although 80% of the added N has been retained (Kahl et al. 1999), the watershed is evolving through the stages of N saturation (sensu Aber et al. 1989, 1998), as characterized by increased rates of net N mineralization and net nitrification (Jefts et al. 2004), increased export of N in surface waters (Kahl et al. 1999; Fernandez et al. 2003; Szillery 2003), and an initial increase followed by a subsequent reduction in exports of Ca, potassium (K), and Mg in surface waters, reflecting an initial mobilization followed by depletion of these elements in the soil exchange complex (Fernandez et al. 1999; Fernandez et al. 2003). We therefore hypothesized that this decadal-scale N addition would also induce changes in foliar chemistry, including increases in foliar N concentrations and decreases in foliar base cation concentrations. Our objective was to test this hypothesis using foliar nutrient concentration data collected over 11 years.

Materials and methods

Study site

The BBWM is located in eastern Maine (44°52′15″N, 68°06′25″W) on the southwest slope of Lead Mountain and is formed by two contiguous small, forested watersheds, West Bear (WB, 11.42 ha) and East Bear (EB, 10.77 ha). The BBWM was established in 1985 as a major component of the Watershed Manipulation Project, part of the National Acid Precipitation Assessment Program designed by the US Environmental Protection Agency (Church 1999). The objective of the BBWM project was to study the effects of increasing N and sulfur (S) deposition on whole watersheds (Church 1999). During a calibration period between 1986 and 1989, it was determined that the hydrology, water chemistry, soils, vegetation, topography, relief, aspect, and exposure of the two watersheds were similar (Uddameri et al. 1995; Kahl et al. 1999; Norton et al. 1999). Five dominant and codominant species (American beech, sugar maple, red spruce, red maple (*Acer rubrum* L.), and yellow birch (*Betula alleghaniensis* Britt.)) distributed in three cover types (hardwood, softwood, and mixedwood) comprised about 98% of all trees in each watershed (Eckhoff 2000; Elvir 2001).

Wet plus dry deposition at BBWM was estimated at 8.4 kg $N\cdot ha^{-1}\cdot year^{-1}$ and 14.4 kg $S\cdot ha^{-1}\cdot year^{-1}$ (Norton et al. 1999; Wang and Fernandez 1999). The treatment of WB started in 1989 and consisted of bimonthly ammonium sulfate ($(NH_4)_2SO_4$) additions at the rate of 25.2 kg $N\cdot ha^{-1}\cdot year^{-1}$ (Norton et al. 1999), while the EB watershed served as an untreated reference. Additional information on the BBWM and effects of the chemical treatment on water and soil chemistry and forest growth is available elsewhere (Kahl et al. 1999; Church 1999; Chen and Beschta 1999; Norton et al. 1999, 2004; Elvir et al. 2003).

Foliar sampling and analysis

The three most dominant tree species at the BBWM (American beech, sugar maple, and red spruce) were sampled for foliar chemical analysis in independent studies in 1993 (White et al. 1999), 1998–2000, and 2002–2003 (current study). Time of sample collection during the growing season varied somewhat between studies. White et al. (1999) collected samples during the first week of September 1993 for hardwood species and in mid-October for red spruce. The 1998–2000 collections for both hardwood and softwood species were made during the first 2 weeks of August. In 2002–2003, samples were collected during the last week of July and first week of August for sugar maple, in mid-August for American beech, and during the first week of September for red spruce.

Foliar samples were collected following similar procedures in all years. Dominant or codominant healthy trees of each species were randomly selected in each watershed. Individual trees sampled were different for each of the three studied periods. In 1993, sampling included 36 American beech, 31 sugar maple, and 21 red spruce from the treated WB watershed and 23 American beech, 11 sugar maple, and 29 red spruce on the reference EB watershed (White et al. 1999). In 1998–2000, foliar samples from 20 trees of each species in each watershed were collected, while for 2002 and 2003 foliar samples from 9 trees of each species in each watershed were collected. For all three data sets, foliar samples were collected from the south side of the upper crown of selected trees.

Samples were stored in paper bags, and sample handling in the field and laboratory was done using powder-free latex gloves. Foliar samples were not washed after collection and were oven dried to a constant mass at 70 °C. Dried samples were ground in a Wiley mill (40 mesh). Chemical analyses were performed by the Analytical Laboratory of the Maine Agricultural and Forest Experiment Station at The University of Maine for 1993, 2002, and 2003 and by the USDA Forest Service chemistry laboratory at the Northeastern Research Station for the 1998–2000 period. In all studies, total N was determined by combustion analysis or the Dumas method. For 1993, 1998–2000, and 2002, Ca, Mg, K, phosphorus (P), and manganese (Mn) were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) after samples had been dry ashed at 550 °C for 5 h in a muffle furnace and digested in 50% HCl (Kalra and Maynard 1991). In 2003, nutrients were analyzed by ICP-AES following nitric acid (HNO_3) microwave digestion. For quality control, field and laboratory split samples were included with the foliar samples, as were samples of standard reference materials from the National Institute of Standards and Testing. The ICP-AES was set to run analyses within a ±10% control error for concentrations of the certified reference materials.

Statistical analysis

Mean concentrations as well as N:Ca and N:Mg ratios were calculated. All nutrient concentration data were subjected to analyses of variance in a 2 × 3 factorial using a combination of fertilization (no addition and $(NH_4)_2SO_4$ addition) and species (American beech, sugar maple, and red spruce) with repeated measures (1998–2000, 2002–2003) using SAS® software (SAS Institute Inc. 1999), except for 1993, where results of statistical analysis were already published (White et al. 1999). Main or interactive effects of treatments and time were considered significant at $\alpha = 0.05$. Correlations between foliar nutrient concentration differences (treated minus reference) and time (1993–2003) were evaluated using two-tailed Pearson correlation tests.

Results and discussion

The effects of a treatment are typically studied either by comparing data before and after treatment or by applying the same treatment to treatment units in different replicates. As pretreatment foliar chemistry data are not available for BBWM and there are no replications of other watersheds with identical treatment histories available, our discussion is based on the assumption that foliar chemistry of selected trees in both watersheds was similar before the treatment. As described earlier, the BBWM watersheds were calibrated over a 3-year pretreatment period, and water chemistry and other factors were found to be similar. Norton et al. (1999, 2004) showed that the treatment clearly affected water chemistry, with an increase in base cation and NO_3^- export in stream water in WB compared to pretreatment levels. They also showed that, as N accumulated in the system and the soil became depleted of base cations, export of N increased while export of base cations declined. The effects of the treatment on water chemistry, soil chemistry, and ecosystem N retention compared to pretreatment levels (Fernandez et al. 1999, 2003; Norton et al. 1999, 2004; Jefts et al. 2004) support our assumption that changes in foliar chemistry in trees within the treated WB watershed likely result from the fertilizer treatment.

Nitrogen

Foliar N concentrations were usually significantly greater in trees within the treated WB watershed compared to those in trees within the reference EB watershed for all tree species in all years (Fig. 1). Exceptions were red spruce in 1998, 1999, and 2000, where no significant differences between watersheds were observed. The factorial analysis showed that American beech had significantly higher foliar N concentrations than the other two species, and sugar maple had significantly higher foliar N concentrations than red spruce. These results are consistent with studies showing that hardwoods generally have higher foliar N concentrations than conifers (Magill et al. 1997; Persson et al. 2000; Scarascia-Mugnozza et al. 2000).

Although differences between watersheds were always significant, the magnitude of the differences in foliar N concentration between the treated and reference watersheds declined over time for sugar maple (Fig. 1), and this difference was negatively correlated with time (Table 1). This reduction over time in foliar N concentration differences between watersheds was a result of a decline in foliar N concentrations in trees within the treated WB watershed and an increase in foliar N concentrations in trees within the reference EB watershed.

Differences in foliar N concentrations between treated and reference watersheds for American beech showed no temporal trends (Table 1), indicating that the differences observed in 1993 have been maintained (Fig. 1). Other studies have shown similar results in foliar N concentration in response to elevated N deposition. Magill et al. (2000), for example, reported an increase in foliar N concentrations in black oak (*Quercus velutina* Lam.), black birch (*Betula lenta* L.), and red maple at the Harvard forest in central Massachusetts for the first 5 years under elevated N additions (113 kg $N \cdot ha^{-1} \cdot year^{-1}$), after which foliar N concentrations either remained stable or showed a slight decline. They also reported that foliar N concentrations in red pine (*Pinus resinosa* Ait.) consistently increased during the first 9 years under the same treatment.

Studies in central Europe also reported increased foliar N concentrations between 1960 and 1980 in coniferous species under conditions of elevated N deposition (Nohrstedt et al. 1993; Huttl and Schaaf 1995). Recent studies, however, have failed to find further temporal changes in foliar N concentrations between the late 1980s and early 1990s in the same forest (Thelin et al. 1998). In a Swedish forest treated with 100 kg $N \cdot ha^{-1} \cdot year^{-1}$, foliar N concentrations of Norway spruce (*Picea abies* (L.) Karst) increased after the first 3 years of treatment, and differences between treated and reference trees were constant after 5 years (Rosengren-Brinck and Nihlgård 1995). Taken collectively, these studies suggest that foliar N concentrations of trees under chronic N deposition reach a maximum in a relatively short time, after which concentrations are maintained or decline slightly. Stable N concentrations may indicate a maximum physiological limit to foliar N concentration for a particular species under enhanced N deposition. Stable or declining foliar N concen-

Fig. 1. Foliar N concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at $\alpha = 0.05$.

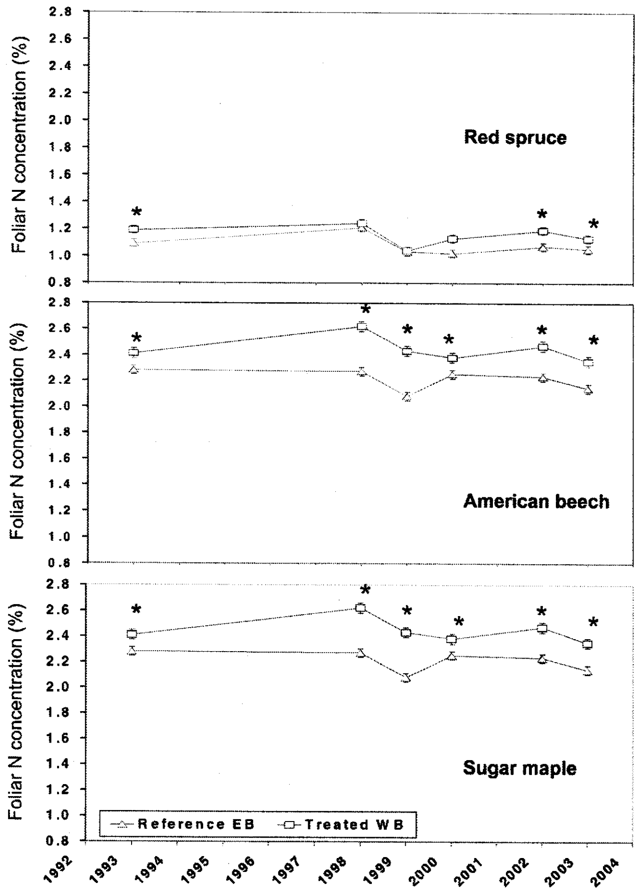


Table 1. Correlation coefficients (two-tailed Pearson correlation tests) between foliar nutrient concentration differences (treated minus reference) and time (1993–2003) for sugar maple, American beech, and red spruce at the Bear Brook Watershed in Maine.

Nutrient	Sugar maple	American beech	Red spruce
N	–0.90*	0.20	0.14
Ca	–0.94*	0.86*	0.89*
Mg	0.51	0.85*	0.94*
K	0.46	0.71	–0.54
P	–0.22	0.03	–0.09
Mn	–0.12	–0.53	0.77

Note: *, significant correlation at $\alpha = 0.05$.

trations may be attributed to a reduction in the ability for fine roots to take up the added N as a result of a decrease in fine-root biomass, a decrease in mychorrhizal associations, or a decrease in the physiological capacity for roots to absorb the added N. For example, Jones et al. (1994) and Quist et al. (1999) both reported that the instantaneous N uptake rates of tree roots declined if roots were exposed to solutions with increased N concentrations. They suggested that an increase in N concentration in soil solution could induce a decline in the ability of roots to absorb N. Therefore, trees subjected to higher soil N availability might show an initial increase in foliar N concentration followed by stable or declining levels thereafter, which is consistent with our results.

Calcium, magnesium, and potassium

Foliar concentrations of Ca in 2000, 2002, and 2003 and Mg in 1998, 2000, 2002, and 2003 for American beech were significantly lower in trees within the treated WB watershed (Figs. 2 and 3) than in trees within the reference EB watershed. Similarly, red spruce trees within the treated WB watershed showed significantly lower foliar Ca concentrations in 2002 and 2003 and significantly lower foliar Mg concentrations in 1993, 2002, and 2003. Sugar maple, on the other hand, showed no significant differences in foliar Ca and Mg concentrations between watersheds except in 1993, when Ca concentrations were significantly lower in trees within the treated WB watershed.

Results indicated that foliar Ca and Mg concentrations of America beech and red spruce trees within the treated WB watershed have further declined in recent years compared to Ca and Mg levels in trees within the reference EB watershed (Figs. 2 and 3). This trend was supported by a significant, positive correlation between foliar Ca or Mg differences (treated minus reference) and time (Table 1). Similarly, N:Ca and N:Mg ratios for American beech and red spruce showed differences between watersheds, with trees within the treated WB watershed showing higher ratios than trees within the reference EB watershed (Table 2).

Foliar Ca and Mg concentrations of sugar maple in both treated and reference watersheds (Figs. 2 and 3) declined in 2002 and 2003 to concentrations considered below threshold levels for optimum growth of this species (Burns 1999; Horsley et al. 2000). For example, literature reviews on foliar nutrient values of healthy sugar maple trees found that K ranged from 5500 to 10 400 $mg \cdot kg^{-1}$, Ca from 5000 to 21 900 $mg \cdot kg^{-1}$, and Mg from 1100 to 4000 $mg \cdot kg^{-1}$ (Kolb and McCormick 1993). The decline in foliar Ca and Mg concentrations in sugar maple from both watersheds in 2002 and 2003 at the BBWM might be the result of the earlier collection of foliar samples in these years. At this time (last week of July and first week of august), the variability in nutrient composition and leaf mass might have affected the measurements in both watersheds. Differences in foliar Ca and Mg concentrations for sugar maple between treated and reference watersheds were significant only in 1993, with differences becoming similar in later years.

Reduced foliar Ca and Mg concentrations and increased N:Ca and N:Mg ratios in American beech and red spruce from the treated WB watershed are consistent with reports of significant reductions in soil-exchangeable base cation pools in the treated WB watershed (Fernandez et al. 2003). The $(NH_4)_2SO_4$ treatment at WB has increased base cation mobilization in soils as a result of increased soil acidification and has enhanced leaching of soil NO_3^- and SO_4^{-2}. Base cation export in stream water increased in the treated WB watershed following the first year of treatment (Norton et al. 1999). Escalating base cation export persisted in WB for approximately 6 years, followed by a decline in stream base cation export in later years (Norton et al. 2004). The in-

Fig. 2. Foliar Ca concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at α = 0.05.

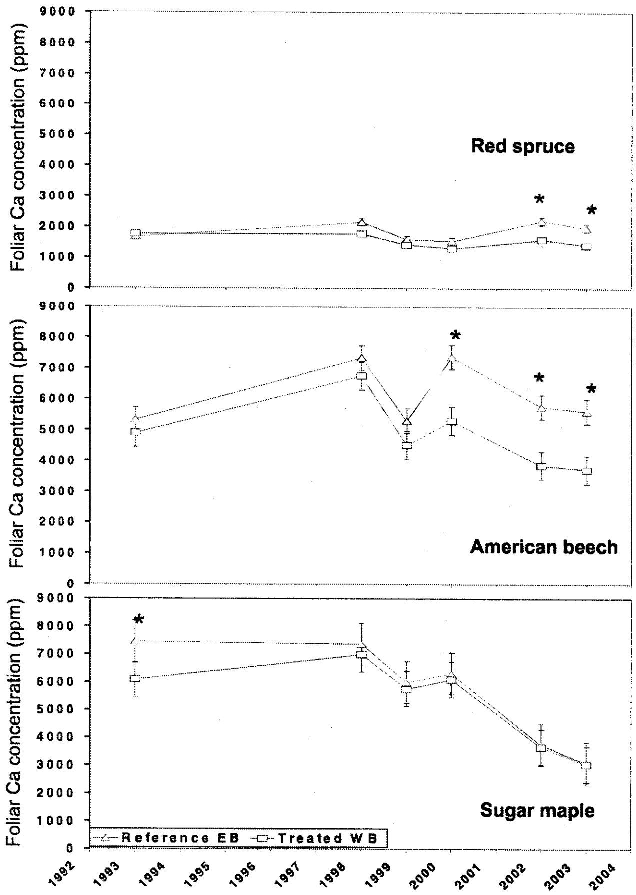
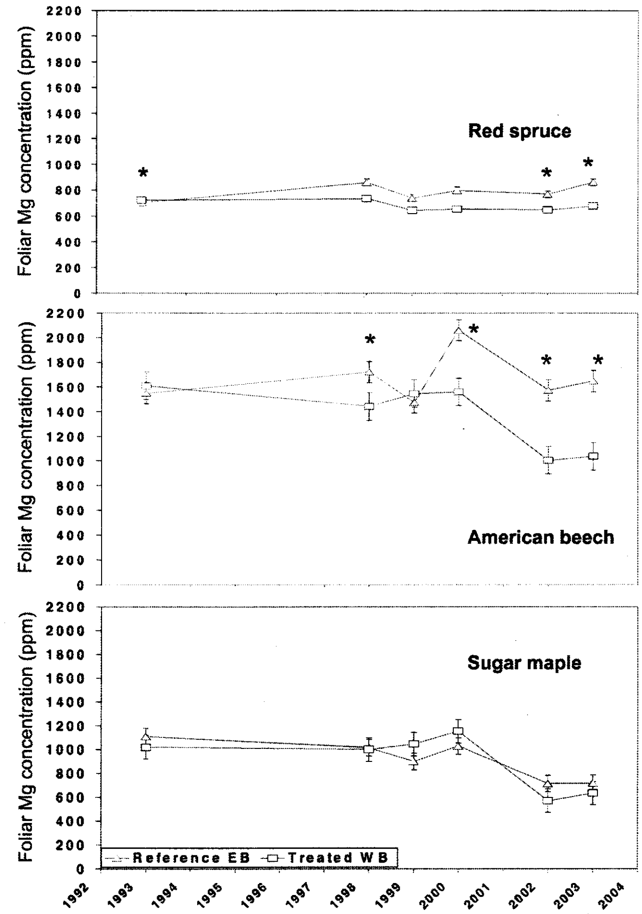


Fig. 3. Foliar Mg concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at α = 0.05.



creased rate of depletion of exchangeable base cations in soils made these nutrients less available for plant uptake, which is consistent with the sustained increase in differences in foliar Ca and Mg concentrations of American beech and red spruce trees between the treated and reference watershed observed in this study.

The significantly higher N:Ca ratio for 1993 and N:Mg ratios for 1993, 1998, 2002, and 2003 in sugar maple trees within the treated WB watershed (Table 2) were primarily the result of the significantly higher N concentrations of trees from the treated WB watershed in comparison with trees from the reference EB watershed. The nonsignificant differences in foliar Ca and Mg concentrations between watersheds in sugar maple suggest that trees within the treated WB watershed may not have experienced greater shortages in Ca and Mg supply than those experienced by trees within the reference EB watershed.

No significant differences between watersheds in foliar K concentration were observed for any species with the exception of K in 1993 and 1999, which was found to be significantly lower in American beech and red spruce trees from the treated WB watershed (Fig. 4). Similarly, no temporal trends between differences (treated minus reference) and time were observed (Table 1). Fernandez et al. (2003) reported a similar lack of notable differences between treated and reference watershed on soil K. The nonsignificant differences between treated and reference watersheds on soil and foliar K concentrations suggest a tight biogeochemical cycle of K at the ecosystem level, where available soil K is quickly taken up and recycled by the vegetation.

Phosphorus and manganese

Red spruce showed no significant differences between watersheds in foliar P and Mn concentrations except in 1998 when P and in 2003 when Mn were significantly lower in red spruce samples from the treated WB watershed (Figs. 5 and 6). American beech from the treated WB watershed showed significantly higher P concentrations in 1998 and 1999 but significantly lower foliar P concentrations in 2002 and 2003. American beech trees within the treated WB watershed showed significantly higher foliar Mn concentrations in 1993, 1998, 1999, and 2000. Sugar maple from the treated WB watershed showed significantly higher foliar P concentrations in 1993, 1998, and 2000 and significantly

Table 2. Foliar nutrient concentration ratios for sugar maple, American beech, and red spruce at East Bear (EB) or West Bear (WB) in the Bear Brook Watershed in Maine.

	Sugar maple		American beech		Red spruce	
Year	EB	WB	EB	WB	EB	WB
N:Ca						
1993[a]	2.2 (0.5)	3.8 (1.4)*	4.7 (1.5)	5.2 (1.3)	7.0 (2.1)	7.6 (3.6)
1998	2.5 (0.7)	3.5 (1.2)	3.4 (1.2)	4.2 (1.4)	6.4 (2.8)	7.8 (3.0)*
1999	3.0 (0.9)	3.7 (1.4)	4.3 (1.2)	5.6 (1.2)*	7.5 (3.6)	8.1 (3.0)
2000	3.1 (0.9)	3.5 (0.8)	3.2 (0.9)	4.9 (1.8)	7.1 (2.3)	10.6 (6.9)*
2002	4.7 (1.1)	6.0 (2.0)	4.0 (0.9)	7.5 (3.3)*	4.9 (0.7)	8.3 (3.0)*
2003	5.9 (1.5)	7.0 (2.1)	3.9 (0.8)	7.1 (2.5)*	5.5 (1.2)	9.8 (4.6)*
N:Mg						
1993[a]	15.3 (5.1)	21.9 (7.2)*	15.2 (3.2)	15.6 (3.6)	15.8 (3.1)	17.1 (3.6)
1998	18.2 (5.8)	22.8 (5.1)*	15.4 (6.5)	19.6 (6.1)*	14.4 (2.8)	17.5 (4.3)
1999	20.1 (7.3)	20.7 (8.0)	15.6 (5.6)	16.4 (4.0)	14.1 (2.6)	16.8 (4.3)
2000	19.0 (4.7)	18.0 (2.6)	11.8 (3.8)	16.0 (3.3)*	13.0 (2.1)	18.5 (7.4)*
2002	25.5 (5.6)	38.1(13.7)*	14.6 (2.8)	25.8 (6.3)*	14.6 (2.9)	19.5 (5.4)
2003	25.5 (5.4)	32.7 (8.3)*	13.3 (2.7)	23.6 (5.1)*	12.3 (1.4)	17.5 (4.4)*

Note: Standard deviations are in parentheses. *, watersheds significantly different at $\alpha = 0.05$.
[a]Data for 1993 adapted from White et al. (1999).

Fig. 4. Foliar K concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at $\alpha = 0.05$.

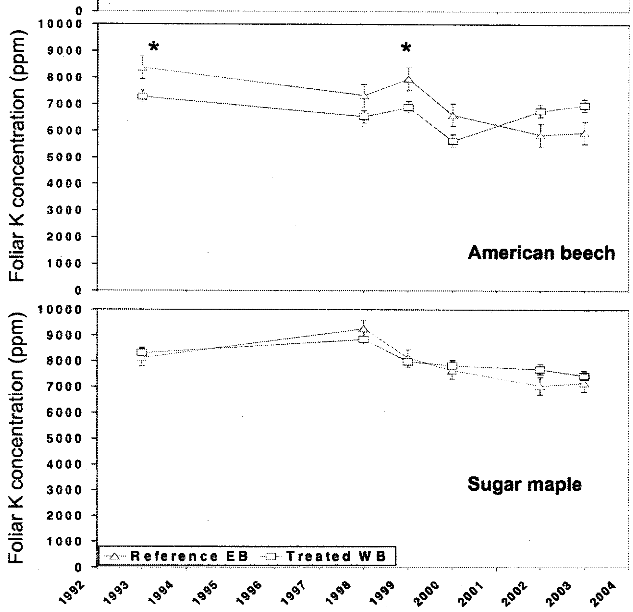


Fig. 5. Foliar P concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at $\alpha = 0.05$.

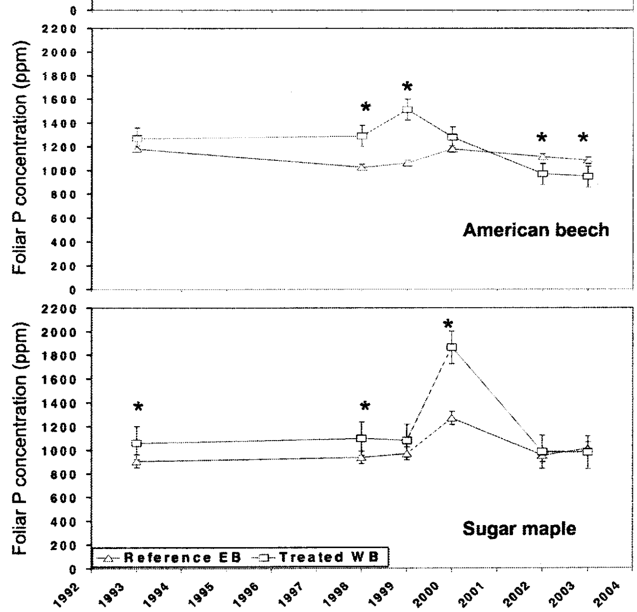
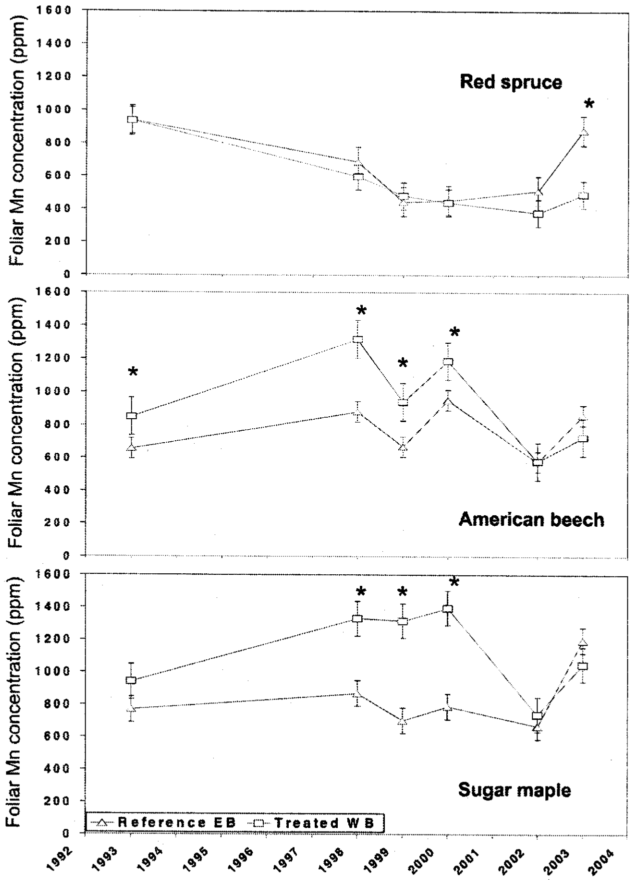


Fig. 6. Foliar Mn concentration trends for three tree species in a treated (fertilizer applied bimonthly since 1989) and reference (untreated) watershed at the BBWM. Bars indicate standard erros; an astersk indicates significance between watersheds at α = 0.05.



higher foliar Mn concentrations in 1998, 1999, and 2000 (Figs. 5 and 6). No significant temporal trends in differences between watersheds for foliar P and Mn were observed for any species (Table 1).

Foliar P and Mn data provided no evidence of differences between watersheds on soil P and soil Mn availability for plant uptake. Effects of N inputs on foliar P and Mn concentrations have not been documented in similar long-term experimental sites involving chronic applications of N in Europe (Nilsson and Wiklund 1994) and the northeastern United States (Magill et al. 1997, 2000). At the BBWM, no significant differences between treated and reference watersheds on soil Mn concentrations have been reported (Fernandez et al. 2003), and Mn exports in stream water have not been documented.

Stream water chemistry studies at the BBWM have shown that total P exports in the treated WB have increased up to four times higher than in the reference EB watershed during high-discharge events (Roy et al. 1999; Norton et al. 2004). Roy et al. (1999) indicated that the induced acidification of the treated WB resulted in increased transport of P from soil to the stream, leading to greater P export from the treated WB, compared to the reference EB. However, no differences between watersheds on soil P concentrations were reported (Fernandez et al. 2003), and in this study differences between watersheds on foliar P and Mn concentrations were not consistent.

Conclusion

Foliar chemistry from 1993 to 2003 at the BBWM suggests that the treated WB watershed might be in later stages of N saturation, where the supply of N exceeds the N demand of plants and microorganisms in the ecosystem (Aber et al. 1989). Foliar N concentrations increased during the early stages of the treatment, but either remained stable (American beech and red spruce) or declined (sugar maple) from 1993 through 2003. The continued depletion of base cations in WB compared to those in EB resulted in greater differences between watersheds in foliar Ca and Mg concentrations for American beech and red spruce, but not for sugar maple. If this reduction persists it would ultimately result in reduced growth because of nutrient imbalances in American beech and red spruce.

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References

Aber, J., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M. et al. 1998. Nitrogen saturation in temperate forest ecosystems. Hypotheses revisited. Bioscience, **48**: 921–934.

Aber, J.D., Nadelhoffer, K.J., Steudler, P., and Melillo, J.M. 1989. Nitrogen saturation in northern forest ecosystems. Bioscience, **39**: 378–386.

Bertills, U., and Näsholm, T. 2000. Effects of nitrogen deposition on forest ecosystems [online]. Report 5067. Swedish Environmental Protection Agency. Available from http://www.naturvardsverket.se/bokhandeln/pdf/620-6137-2 [cited 10 June 2004].

Boxman, A.W., Blanck, K., Brandrud, T.E., Emmet, B.A., Gundersen, P., Hogervorst, R.F. et al. 1998. Vegetation and soil biota response to experimentally-changed nitrogen inputs in coniferous forest ecosystems of the NITREX project. For. Ecol. Manage. **101**: 65–79.

Burns, B. 1999. Essential elements for sugarbush fertilization. Fact sheet. Vermont Department of Forestry, Parks and Recreation, Waterbury, Vt.

Chen, H., and Beschta, R. 1999. Dynamic hydrology simulation of the Bear Brook Watershed in Maine (BBWM). Environ. Monit. Assess. **55**: 3–96.

Church, M.R. 1999. The Bear Brook Watershed manipulation project: watershed science in a policy perspective. Environ. Monit. Assess. **55**: 1–5.

Driscoll C.T., Lawrence, G.B., Bulger, A.J., Butler, T.J., Cronan, C.S., Eager, C. et al. 2001. Acid deposition in the northeastern United States: sources and inputs, ecosystem effects, and management strategies. Bioscience, **51**: 180–198.

Eckhoff, J. 2000. Efficacy of forest health monitoring indicators to evince impacts on a chemically manipulated watershed. Ph.D. thesis, The University of Maine, Orono, Me.

Elvir, J.A. 2001. Effects of ten years of chronic ammonium sulfate treatment on the forest component at the Bear Brook watershed in Maine. M.S. thesis, The University of Maine, Orono, Me.

Elvir, J.A., Wiersma, G.B., White, A., and Fernandez, I. 2003. Effects of chronic ammonium
sulfate treatment on basal area increment in red spruce and sugar maple at the Bear Brook Watershed in Maine. Can. J. For. Res. **33**: 862–869.

Evans, J.R. 1989. Photosynthesis and nitrogen relationships in leaves of C3 plants. Oecologia, **78**: 9–19.

Fernandez, I.J., Rustad, L.E., Norton, S.A., Kahl, J.S., and Cosby, B.J. 2003. Experimental acidification causes soil base-cation depletion at the Bear Brook Watershed in Maine. Soil Sci. Soc. Am. J. **67**: 1909–1919.

Fernandez, I., Rustad, L., David, M., Nadelhoffer, K., and Mitchell, M. 1999. Mineral soil and solution responses to experimental N and S enrichment at the Bear Brook Watershed in Maine (BBWM). Environ. Monit. Assess. **55**: 165–185.

Friedland, A.J., Miller, E.K., Battles, J.J., and Thorne, J.F. 1991. Nitrogen deposition, distribution and cycling in a subalpine spruce-fir forest in the Adirondacks, New York, USA. Biogeochemistry, **14**: 31–55.

Graveland, J., Vander Wal, R., Van Balen, J.H., and Van Noordwijk, A.J. 1994. Poor reproduction in forest passerines from decline of snail abundance on acidified soils. Nature (London), **368**: 446–448.

Hagedorn, F., Spinnler, D., and Siegwolf, R. 2003. Increased N deposition retards mineralization of old soil organic matter. Soil Biol. Biochem. **35**: 1683–1692.

Horsley, S.B., Long, R.P., Bailey, S.W., Hallett, R.A., and Hall, T.J. 2000. Factors associated with the decline disease of sugar maple on the Allegheny Plateau. Can. J. For. Res. **30**: 1365–1378.

Huttl, R.F., and Schaaf, W. 1995. Nutrient supply of forest soils in relation to management and site history. Plant Soil, **168**: 31–41.

Jefts, S., Fernandez, I.J., Rustad, L.E., and Dail, D.B. 2004. Decadal responses in soil N dynamics at the Bear Brook Watershed in Maine, USA. For. Ecol. Manage. **189**: 189–205.

Johnson, A.H., Schwartzman, T.N., Battles, J.J., Miller, R., Friedland, A.J., and Vann, D.R. 1994. Acid rain and soils of the Adirondacks. II. Evaluation of calcium and aluminum as causes of red spruce decline at Whiteface Mountain, New York. Can. J. For. Res. **24**: 654–662.

Jones, H.E., Högberg, P., and Ohlsson, H. 1994. Nutritional assessment of a forest fertilization experiment in northern Sweden by root bioassays. For. Ecol. Manage. **64**: 59–69.

Kahl, J., Norton, S., Fernandez, I., Rustad, L., and Handley, M. 1999. Nitrogen and sulfur input–output budgets in the experimental and reference watershed, Bear Brook Watershed in Maine (BBWM). Environ. Monit. Assess. **55**: 113–131.

Kalra, Y.P., and Maynard D.G. 1991. Methods manual for forest soil and plant analysis. For. Can. North. For. Res. Cent. Inf. Rep. NOR-X-319.

Kolb, T.E., and McCormick, L.H. 1993. Etiology of sugar maple decline in four Pennsylvania stands. Can. J. For. Res. **23**: 2395–2402.

Lambers, H., Chapin, F.S., III, and Pons, T.L. 1998. Plant physiological ecology. Springer-Verlag, New York.

Lloyd, J. 1999. The CO_2 dependence of photosynthesis, plant growth responses to elevated CO_2 concentrations and their interaction with soil nutrient status, II. Temperate and boreal forest productivity and the combined effects of increasing CO_2 concentrations and increased nitrogen deposition at a global scale. Funct. Ecol. **13**: 439–459.

Magill, A., Aber, J., Hendricks, J., Bowden, R., Melillo, J., and Steudler, P. 1997. Biogeochemical responses of forest ecosystems to simulated chronic nitrogen deposition. Ecol. Appl. **7**: 402–415.

Magill, A., Aber, J., Berntson, G., McDowell, W., Nadelhoffer, K., Melillo, J., and Steudler, P. 2000. Long-term nitrogen additions and nitrogen saturation in two temperate forests. Ecosystems, **3**: 238–253.

McNulty, S.G., Aber, J.D., and Newman, S.D. 1996. Nitrogen saturation in a high elevation New England spruce–fir stand. For. Ecol. Manage. **84**: 109–121.

Nasholm, T., Nohrstedt, H., Karen, O., Kyto, M., and Bjorkman, C. 2000. How are forest trees affected? *In* Effects of nitrogen deposition on forest ecosystems [online]. *Edited by* U. Bertills and T. Näsholm. Report 5067. Swedish Environmental Protection Agency. Available from http://www.naturvardsverket.se/bokhandeln/pdf/620-6137-2 [cited 12 May 2004].

Nilsson, L.O., and Wiklund, K. 1994. Nitrogen uptake in a Norway spruce stand following ammonium sulfate application, fertilization, irrigation, drought and nitrogen-free fertilization. Plant Soil, **164**: 221–229.

Nohrstedt, H.O., Sikstrom, U., and Ring, E. 1993. Experiments with vitality fertilization in Norway spruce stands in southern Sweden. SkogForsk (Forestry Research Institute of Sweden) Report No. 2.

Norton, S., Kahl, J., Fernandez, I., Haines, T., Rustad, L., Nodvin, S. et al. 1999. The Bear Brook Watershed, Maine (BBWM), USA. Environ. Monit. Assess. **55**: 7–51.

Norton, S., Fernandez, I., Kahl, J.S., and Reinhardt, R. 2004. Acidification trends and the evolution of neutralization mechanisms through time at the Bear Brook Watershed, in Maine (BBWM), USA. Water Air Soil Pollut. Focus, **4**: 289–310.

Oren, R., Werk, K.S., Buchmann, N., and Zimmermann, R. 1993. Chlorophyll-nutrient relationships identify nutritionally caused decline in *Picea abies* stands. Can. J. For. Res. **23**: 1187–1195.

Persson, T., Van Oene, H., Harrison, A., Karlsson, P., Bauer, G., Cerny, J. et al. 2000. Experimental sites in the NIPHYS/CANIF Project. *In* Carbon and nitrogen cycling in European forest ecosystems. Ecol. Stud. **142**: 14–46.

Quist, M.E., Näsholm, T., Lindeberg, J., Johannisson, C., Högbom, L., and Högberg, P. 1999. Responses of a nitrogen saturated forest to a sharp decrease in nitrogen input. J. Environ. Qual. **28**: 1970–1977.

Rosengren-Brinck, U., and Nihlgård, B. 1995. Nutritional status in needles of Norway spruce in relation to water and nutrient supply. Ecol. Bull. **44**: 168–177.

Roy, S., Norton, S., Fernandez, I., Kahl, J. 1999. Linkages of P and Al export at Bear Brook Watershed in Maine. Environ. Monit. Assess. **55**: 133–147.

SAS Institute Inc. 1999. SAS Statistic, 8th ed. [computer program]. SAS Institute Inc., Cary, N.C.

Scarascia-Mugnozza, G., Bauer, G., Persson, H., Mateucci, G., and Maci, A. 2000. Tree biomass, growth and nutrient pools. *In* Carbon and nitrogen cycling in European forest ecosystems. Ecol. Stud. **142**: 49–62.

Szillery, Johanna. 2003. Soil solution dynamics in response to elevated nitrogen and sulfur treatments at the Bear Brook Watershed in Maine. M.S. thesis, The University of Maine, Orono, Me.

Thelin, G., Rosengren-Brinck, U., Nihlgård, B., and Barkman, A. 1998. Trends in needle and soil chemistry of Norway spruce and Scots pine stands in south Sweden 1985–1994. Environ. Poll. 99: 149–158.

Uddameri, V., Norton, S.A., Kahl, J.S., and Scofield, J.P. 1995. Randomized intervention analysis of the response to the West Bear Brook watershed, Maine to chemical manipulation. Water Air Soil Pollut. **79**: 193–199.

Wang, Z., and Fernandez, I. 1999. Soil type and forest vegetation influences on forest floor nitrogen dynamics at the Bear Brook Watershed in Maine (BBWM). Environ. Monit. Assess. **55**: 221–234.

White, G., Fernandez, I., and Wiersma, G. 1999. Impact of ammonium sulfate treatment on the foliar chemistry of forest trees at the Bear Brook Watershed in Maine. Environ. Monit. Assess. **55**: 235–250.