

# Is Nitrogen Deposition Altering the Nitrogen Status of Northeastern Forests?

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*Concern is resurfacing in the United States over the long-term effects of excess nitrogen (N) deposition and mobility in the environment. We present here a new synthesis of existing data sets for the northeastern United States, intended to answer a single question: Is N deposition altering the N status of forest ecosystems in this region? Surface water data suggest a significant increase in nitrate losses with N deposition. Soil data show an increase in nitrification with decreasing ratio of soil carbon to nitrogen (C:N) but weaker relationships between N deposition and soil C:N ratio or nitrification. Relationships between foliar chemistry and N deposition are no stronger than with gradients of climate and elevation. The differences in patterns for these three groups of indicators are explained by the degree of spatial and temporal integration represented by each sample type. The surface water data integrate more effectively over space than the foliar or soil data and therefore allow a more comprehensive view of N saturation. We conclude from these data that N deposition is altering N status in northeastern forests.*

*Keywords: foliar N, nitrification, nitrate, soil C:N*

**L**ong-term exposure to excess nitrogen (N) from atmospheric deposition and other human activities has had a range of impacts on natural ecosystems. The five articles in this special series in *BioScience* present a general theory of N impacts as this element cascades through the global environment. They also provide case studies of coastal eutrophication and its effects on terrestrial ecosystems in the northeastern and western United States.

This article results from a meeting and data synthesis effort supported by the US Environmental Protection Agency (EPA) and designed to answer a single question: Is the N status of northeastern forests being altered by N deposition? This relatively simple question becomes complex in the context of ambient spatial and temporal variation in climate, species composition, and disturbance effects across the region. Here we provide a brief review of changes in forests with N deposition as predicted by N saturation theory, discuss factors that can complicate trends over a large region, and present the results of a new, large-scale data synthesis designed to overcome these factors using a large sample size. This approach is intended to complement model analyses at intensive study sites, experimental studies, and earlier, limited regional analyses of extensive data sets. To the extent that this kind of regional survey supports theoretical analyses and site-level experimentation, it also supports the occurrence of change in the landscape in response to N deposition.

Human activities have dramatically increased the mobility and deposition of reactive forms of N (Galloway et al. 2003). In the northeastern United States, atmospheric deposition has increased at least 5- to 10-fold over preindustrial conditions (Galloway et al. 1984), and 60% to 80% of deposited N occurs as nitrate ( $\text{NO}_3^-$ ; NADP 2001). Title IV of the 1990 Clean Air Act Amendments established the goal of reducing stationary source nitrogen oxide ( $\text{NO}_x$ ) emissions from 1980 levels by 2 million tons through a limitation on emission rates from certain categories of stationary coal-fired boilers and mobile sources. Given the predicted increases in power generation and vehicle miles traveled, it is unlikely that  $\text{NO}_x$  emissions will decrease significantly without further regulatory action.

Regional variations in the emission and atmospheric transport of N have resulted in a strong gradient of N deposition

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across the northeastern United States, from relatively high rates (10 to 12 kilograms N per hectare per year [kg N per ha per yr]) of wet and dry N deposition in southern New York and Pennsylvania to relatively low deposition (< 4 kg N per ha per yr) in eastern Maine (figure 1; Ollinger et al. 1993). Long-term averages for wet deposition show a clear west-to-east gradient, while dry deposition exhibits an equally strong trend from south to north (Ollinger et al. 1993), suggesting different source areas. Wet deposition declines with increasing distance from industrial areas to the west, while dry deposition declines with increasing distance from the coastal urban corridor of the northeastern United States. Within the Northeast, however, local storm paths and elevational effects can cause substantial variation in N deposition from region to region (e.g., the Adirondacks [Ito et al. 2002] and the Catskill Mountains [Weathers et al. 2000]).

Nihlgard (1985) was the first to propose that excessive N deposition could damage forest ecosystems. Through focused, coordinated international research efforts (e.g., Aber et al. 1998, Fenn et al. 1998), a generalized description of the continuous response of forests to continuing, chronic N additions has been developed (see Galloway et al. 2003). The primary characteristics of this process, which is termed N saturation, are (a) nonlinear changes over time and (b) a combination of initial fertilization effects followed by negative impacts on plant function and growth.

Two sets of N saturation hypotheses have been presented, one based on forest processes (Aber et al. 1989, 1998) and the other focused on seasonal changes in  $\text{NO}_3^-$  concentrations in surface waters (Stoddard 1994). Both hypotheses include four stages of response, and both focus on  $\text{NO}_3^-$  dynamics as a key characteristic of N status, because the mobility and loss of  $\text{NO}_3^-$  drive the primary negative effects of N saturation. These effects include soil and water acidification, in-

creased aluminum mobility and movement to streams, and nutrient imbalances in trees (reviewed in Vitousek et al. 1997, Aber et al. 1998, Fenn et al. 1998). Contributions of stream-water N to coastal eutrophication are described by Driscoll and colleagues (2003). In forests, the onset of  $\text{NO}_3^-$  production in soils through net nitrification is a critical process. Increasing N concentrations in foliage and changes in ratios of carbon to nitrogen (C:N) in soils are other primary indicators of forest N status (Aber et al. 1989). In surface waters, stages of N saturation are defined by the quantity and seasonality of  $\text{NO}_3^-$  concentrations, ranging from low and seasonal to high and relatively constant (Stoddard 1994).

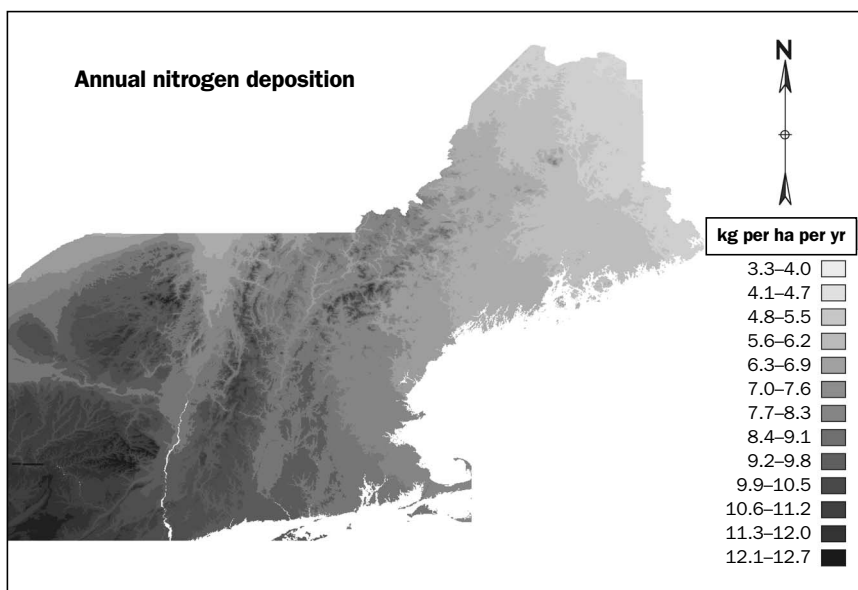
### Detecting changes in nitrogen status of forests of the northeastern United States

Both the terrestrial- and the aquatic-based N saturation theories suggest that  $\text{NO}_3^-$  concentration in streamwater should be the primary indicator of N status and should increase over time with chronic N deposition. Is this happening in the northeastern United States? Long-term data sets give conflicting results. At the Hubbard Brook Experimental Forest in the White Mountains of New Hampshire, where N deposition has remained at 6 to 8 kg N per ha per yr for at least the last three decades, stream  $\text{NO}_3^-$  has declined erratically from peak losses in the 1970s (Likens et al. 1996). In contrast, the Fernow Experimental Forest in West Virginia, which receives about 10 to 12 kg N per ha per yr (Adams et al. 1997), shows a long-term increase in stream  $\text{NO}_3^-$  (Peterjohn et al. 1996). A longer-term data set for Schoharie Creek in the Catskill Mountains of New York shows increased stream  $\text{NO}_3^-$  over the last 100 years but with cyclical increases and decreases, including the recent decline evident at Hubbard Brook (Stoddard 1991). Lakes and streams across the Adirondack and Catskill Mountains in New York have also shown increases in

surface water  $\text{NO}_3^-$  concentrations in the 1980s and declining values in the 1990s, whereas New England surface waters showed few trends in either decade. These conflicting results suggest that factors other than N deposition can affect temporal patterns of  $\text{NO}_3^-$  loss from forest ecosystems.

Although less is known about temporal trends in soil and foliar indicators, several factors are known to change the availability and cycling of N in ecosystems and would be expected to alter foliar, soil, and stream indexes in ways contrary to those expected from N deposition alone.

**Climate gradients and variation.** Gradients in foliar N concentration have been measured across gradients in latitude and elevation and attributed to expected physiological responses to lower temperatures and shorter growing seasons (Körner 1989, Yin 1992, Haxeltine and Prentice 1996).



**Figure 1. Estimated annual nitrogen deposition across the eastern United States (Ollinger et al. 1993).**

Interannual variability in climate may also result in as much as a 20% difference in foliar N concentration across several years (Magill et al. 2000). Variation in stream  $\text{NO}_3^-$  has been attributed to variation in several climate variables. Mitchell and colleagues (1996) attributed a 1-year spike in  $\text{NO}_3^-$  losses in northeastern forests to an excessively cold December with low snow cover, possibly accompanied by significant soil frost. Various studies including experimental manipulation have shown that soil freezing can result in substantial increases in soil  $\text{NO}_3^-$  (e.g., Groffman et al. 2001). Murdoch and colleagues (1998) presented a 12-year data set for a Catskill site showing a significant relationship between mean annual temperature and stream  $\text{NO}_3^-$  concentration. In a modeling analysis, Aber and colleagues (2002) demonstrated that high  $\text{NO}_3^-$  losses at Hubbard Brook in the 1970s resulted from an anomalous combination of climatic variation and biotic disturbance.

**Species effects.** Tree species common to the northeastern United States often have very different patterns of N cycling (Pastor et al. 1984, Finzi et al. 1998) and may respond differently to increased N deposition. Across a gradient of N cycling rates in New Hampshire, different hardwood species with the same rate of N mineralization had significantly different foliar N concentrations (Ollinger et al. 2002). Mitchell and colleagues (1992) attributed the much greater  $\text{NO}_3^-$  loss at Turkey Lakes in Ontario than at Huntington Forest in New York to the predominance of sugar maple at the former site. Similarly, Lovett and Rueth (1999) found that soils sampled along an N deposition gradient showed increased nitrification under sugar maple but not under American beech. Lovett and colleagues (2002a) have argued that most of the differences in the spatial patterns in surface water  $\text{NO}_3^-$  concentrations in the Catskill Mountains result from differences in tree species composition that drive differences in soil C:N ratios.

**Hydrologic pathways.** In some catchments  $\text{NO}_3^-$  can be consumed in streams, riparian areas, groundwater (Groffman et al. 1996), hyporheic zones (Triska et al. 1993), and wetlands (mostly due to denitrification), while in other catchments these zones may be  $\text{NO}_3^-$  sources (Mitchell 2001). The relative importance, spatial distribution, and hydrologic connectivity of uplands, wetlands, streams, and lakes results in varying spatial and temporal patterns in the relative importance of biotic and abiotic processes affecting N retention (Inamdar et al. 2000). Creed and Band (1998) showed how topographic features can be used to explain much of the variability in  $\text{NO}_3^-$  export among subcatchments for the Turkey Lakes watersheds in Ontario. Burns and colleagues (1998) have suggested that deep groundwater with high concentrations of  $\text{NO}_3^-$  may be an important source of N in surface waters in the Catskill Mountains, especially during periods of base flow when groundwater contributions would be most important.

**Disturbance.** Stream  $\text{NO}_3^-$  losses generally increase during the first few years following disturbances such as forest harvest (e.g., Aber et al. 2002) or fire (Knoepp and Swank 1993) and then are maintained at low levels during periods of regrowth (Vitousek and Reiners 1975, Bormann and Likens 1979). Insect defoliation has been shown to cause pulses of  $\text{NO}_3^-$  loss in some streams (Eshleman et al. 1998, Aber et al. 2002; compare Lovett et al. 2002a), and ice damage may have similar effects in northern New England (Nielson et al. 2001). Clearing land for agricultural use may reduce (or augment) forest N pools and N cycling, and it can also alter species composition.

Disturbance can leave a lasting signal in forest ecosystems. Plowing decreases the soil C:N ratio and enhances net nitrification for decades after agricultural abandonment and forest regrowth (Compton and Boone 2000). Century-old harvests and fires can still be detected in reduced net nitrification rates (Goodale and Aber 2001) and losses of  $\text{NO}_3^-$  in stream water (Goodale et al. 2000). The duration and diversity of human uses of the land in the northeastern United States may underlie spatial complexity in patterns of  $\text{NO}_3^-$  concentrations in surface waters.

## Approach

In this article we synthesize many existing data sets on stream, soil, and foliar chemistry in an attempt to overcome the confounding factors described above. While we would expect any direct relationship between N deposition and forest or surface water characteristics to be noisy because of these factors, significant relationships may emerge as a result of large sample size.

**The synthesis of regional data sets.** In April 2001, under the auspices of the Northeastern Ecosystem Research Cooperative (NERC; 29 January 2003, [www.ecostudies.org/nerc](http://www.ecostudies.org/nerc)), the EPA Clean Air Markets Division sponsored a workshop at the University of New Hampshire with the intent of assessing the status of N saturation in the northeastern United States. The effort brought together individuals from across the northeastern United States who have measured three key indicators of N status: foliar, soil, and surface water chemistry. A great many published and unpublished data sets were collated and used in the analyses presented here. These data sets were then examined for spatial patterns in foliar, soil, or stream chemistry reflecting changes in N deposition across the region (figure 1). For this analysis, the Northeast includes the region from West Virginia to Maine. We tested whether foliar, soil, and surface water chemistry responded in ways expected by theories of N saturation (Aber et al. 1989, Stoddard 1994, Aber et al. 1998) and observed in experimental fertilization studies (e.g., Gundersen et al. 1998, Magill et al. 2000). For foliage, we expected that foliar N concentration would increase and the ratio of foliar lignin to N would decrease with increasing N deposition. Similarly, for soils, we expected that nitrification would increase and the soil C:N ratio would decrease with increasing N deposition. Finally, for surface waters, we expected

that  $\text{NO}_3^-$  concentrations would increase with N deposition, particularly during the dormant season. The absence of significant trends would argue against any real effect of N deposition on N status, suggesting that N deposition effects are small compared with variation resulting from disturbance, climate variation, species effects, and other factors.

**Estimated nitrogen deposition.** For all three data sets, we estimated N deposition for each site using a statistical model developed by Ollinger and colleagues (1993), modified slightly by using dry deposition coefficients from Lovett and Rueth (1999). We estimated N deposition for each plot where foliage and soil chemistry were measured and for the elevation where each lake or stream sample was collected. Because N deposition generally increases with elevation, the use of the minimum rather than the mean elevation of each catchment underestimates N deposition to the whole watershed. Mean watershed elevations were available for 94 watersheds; for these watersheds, the underestimation of N deposition resulting from the use of minimum rather than mean elevation was 2.1 to 6.1 kg N per ha per yr (mean 4.5 kg N per ha per yr).

Sites in western Maryland and northern West Virginia fall outside the range in which Ollinger and colleagues' (1993) model applies, so N deposition to these sites was taken from published values (Peterjohn et al. 1996). None of the estimates of N deposition used here include cloud deposition, which can contribute 5 to 35 kg N per ha per yr to high-elevation sites in the northeastern United States (Lovett and Kinsman 1990). In the Adirondacks, cloud deposition increases dramatically above about 1000 meters (m; Miller et al. 1993), and so we probably markedly underestimated N deposition to sites above 1000 m.

**The calculation of mean annual temperature.** Mean annual temperature (MAT) was calculated for each study site using algorithms developed during the same regional climate modeling project described above (Ollinger et al. 1995). Monthly average maximum and minimum temperatures were

predicted using multiple linear regression equations on latitude, longitude, and elevation. Mean temperatures for each month were the average of the maximum and minimum, and the 12 monthly values were averaged to obtain the MAT.

## Results

**Foliage.** A foliar chemistry data set from 362 forested plots, concentrated in New Hampshire and New York, was available for this study (table 1). Although samples were available for a large number of species, the analyses presented here were limited to red spruce (*Picea rubens* Sarg.), a needle-leaved evergreen, and sugar maple (*Acer saccharum* Marsh.), a broad-leaved deciduous species. These two species are often dominant in two major forest types common across the region (spruce–fir and northern hardwood) and were well represented in the combined data set. For both species, the majority of samples are from the White Mountains of New Hampshire, but sample locations span the length and breadth of the region from eastern Maine to western Pennsylvania and eastern West Virginia (figure 2).

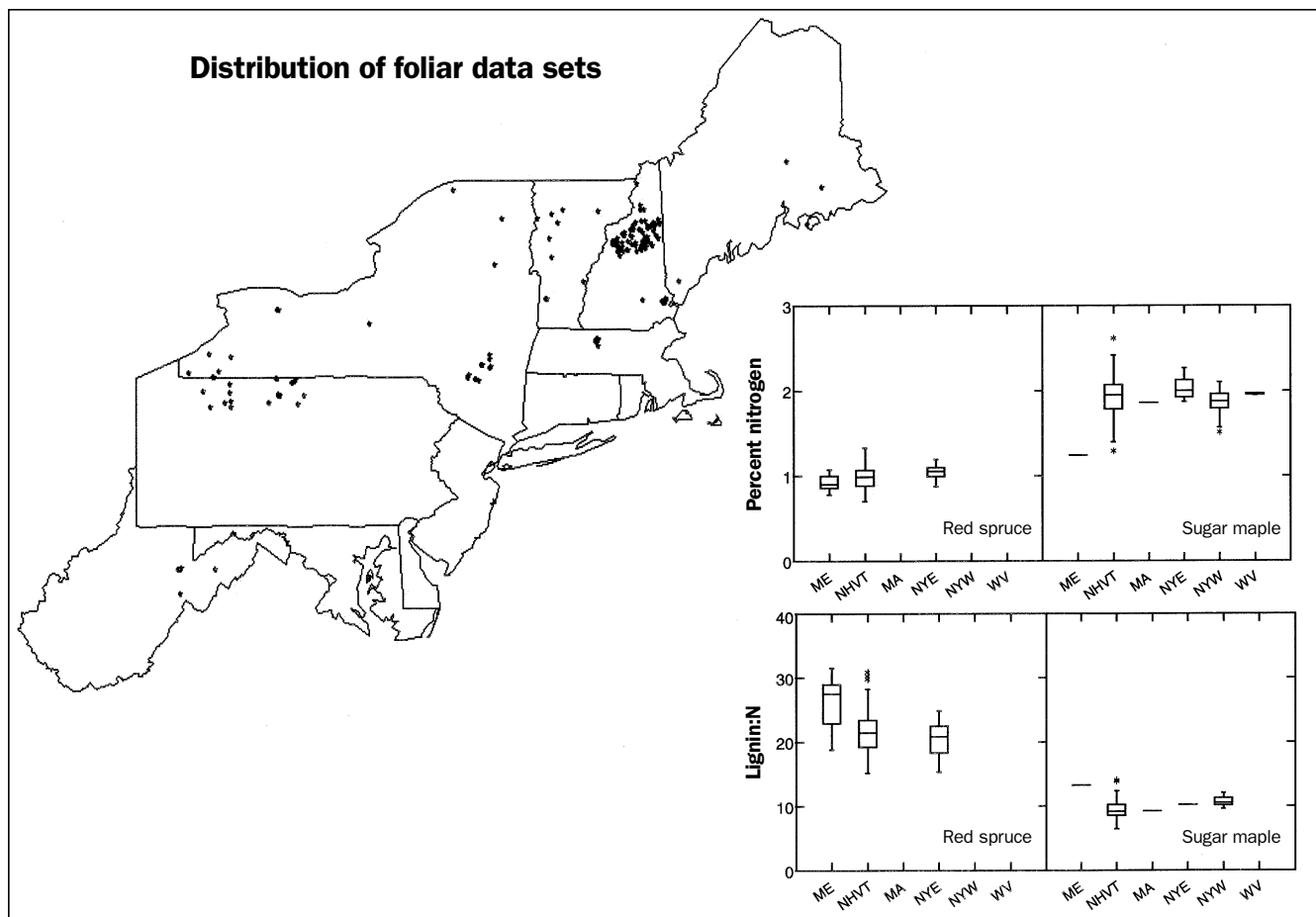
Mass-based foliar nitrogen concentration (grams [g] of foliar N per 100 g) of red spruce ranged from 0.7% to 1.3%, foliar lignin ranged from 15% to 28%, and the ratio of foliar lignin to N ranged from 15 to 31. For sugar maple, percent N values ranged from 1.2% to 2.6%, lignin values from 13% to 22%, and lignin-to-N ratios from 6.5 to 14. We used linear regression analysis to examine relationships among foliar concentrations, modeled N deposition, and variables related to patterns of N deposition in the Northeast, including latitude, longitude, elevation, and associated gradients of temperature and precipitation.

Differences in leaf-level chemistry in this regional data set were most strongly associated with elevation and with climatic variables that vary predictably with elevation (table 2, figure 3). Response to these gradients varied by species. In sugar maple, foliar N concentration increased with increasing elevation, as has been generally reported, especially in species

**Table 1. Location and number of plots of sugar maple and red spruce in the foliar chemistry data set for the northeastern United States.**

Location	Number of plots		Year	Reference
	Sugar maple	Red spruce		
Massachusetts	1	—	1992, 1999	ACCP 1994, Martin and Aber 1997
Maine	1	26	1988, 1992	McNulty et al. 1991, ACCP 1994
New Hampshire	95	132	1987–1998	McNulty et al. 1991, ACCP 1994, Goodale and Aber 2001, Ollinger et al. 2002, Smith et al. 2002
Vermont	6	10	1987–1988 1994	McNulty et al. 1991, 1996
Eastern New York	9	35	1996–1998	Lovett and Rueth 1999, Horsley et al. 2000
Western New York and Pennsylvania	45	—	1996	Horsley et al. 2000
West Virginia	2	—	1997	Mary Beth Adams, USDA Forest Service, Parsons, WV, personal communication, 2001
Total	159	203		





**Figure 2.** Distribution of foliar chemistry sampling plots across the northeastern United States. Box plots indicate the median, quartile, and range of measured foliar nitrogen (N) concentration and lignin:N ratios for red spruce and sugar maple in the growing season, along an east-to-west longitudinal gradient, in Maine (ME), New Hampshire and Vermont (NHVT), Massachusetts (MA), eastern New York (NYE), western New York and Pennsylvania (NYW), and West Virginia (WV).

whose leaves have a short life span (Yin 1994, Haxeltine and Prentice 1996). In contrast, in red spruce the ratio of lignin to N, rather than N concentration alone, was most highly correlated with climatic, geographic, and N deposition gradients across the region. As with sugar maple, the strongest relationships were with elevation and associated gradients of temperature and precipitation, although the relationship with N deposition was nearly as strong.

Climatic gradients and N deposition covary in this region, and it is difficult to separate their individual effects. In this analysis, strong relationships with elevation reflect a preponderance of samples from the White Mountain region. These samples cover a wide elevational range (231 to 1295 m for red spruce samples, 31 to 885 m for sugar maple samples), spanning nearly the full range in elevation of samples analyzed from across the region. Wider geographic sampling of foliar chem-

istry across the Northeast and among a wider array of species is needed before the often covarying and confounding effects of climate and N deposition on the nutrient status of forest canopies can be fully described.

**Soils.** We compiled data from 15 data sets (table 3). Sites ranged geographically from the mid-Atlantic states to northern

**Table 2.** Foliar chemistry regression statistics.

Variable	Red spruce		Sugar maple	
	Percent N (n = 203)	Lignin:N (n = 202)	Percent N (n = 159)	Lignin:N (n = 123)
Latitude	NS	0.06	NS	0.06
Longitude	0.07	0.19	NS	0.08
Elevation	NS	0.38	0.22	NS
Nitrogen deposition	NS	0.33	NS	NS
Mean annual temperature	NS	0.36	0.08	0.07
Mean July precipitation	NS	0.37	0.32	NS

N, nitrogen; NS, not significant.

Note: Values expressed as  $R^2$ . All values shown are significant at  $p < 0.01$ .

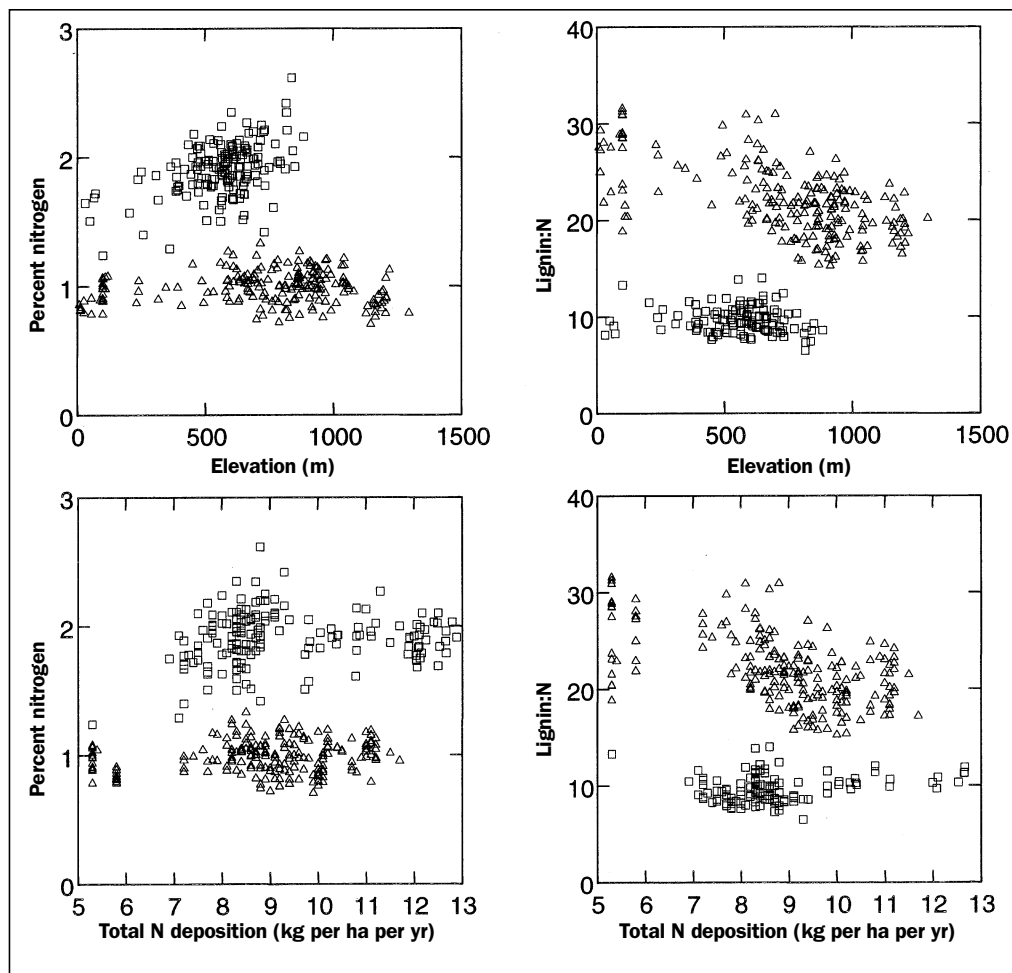
Table 3. Soil nitrogen mineralization and soil carbon-to-nitrogen (C:N) data sets.

Site	Forest type	Number of plots	Samples collected <sup>1</sup>		Mineral soil depth (cm)	Incubation location	N cycling methods		Reference
			Forest floor	Number of plots			Duration (days)	C:N <sup>2</sup>	
Fernow, WV	HW	16	All		10	Field	56	O + M	Christ et al. 2002
Fernow, WV	HW	21	NA		5	Field	365	M	Gilliam et al. 1996
Dry Creek, NY	HW	1	All		15	Field	210	O + M	Doug Burns, US Geological Survey, Troy, NY, personal communication, 2001
Huntington, NY	HW and conifer	4	Oe + Oa		15	Field	365	O + M	Ohrui et al. 1999, Bischoff et al. 2001
NE transect	Conifer	12	Oa		10	NA		O + M	David and Lawrence 1996
NE transect	HW	31	Oe + Oa		10	Lab	28	O + M	Lovett and Rueth 1999
NE transect	Conifer	11	Oe + Oa		NA	Lab	28	O	McNulty et al. 1991
Harvard Forest, MA	HW and conifer	12	NA		15	Field	42	M	Compton and Boone 2000
Montague Plain, MA	HW and conifer	16	NA		15	Field	42	M	Compton et al. 1998
White Mountains, NH	HW and conifer	32	Oe + Oa		10	Field and lab	28–365	O + M	Ollinger et al. 2002
White Mountains, NH	HW	36	Oe + Oa		10	Lab	28	O + M	Goodale and Aber 2001
Maine (statewide)	Conifer	20	Oe + Oa		5	Lab	28	O + M	Ivan Fernandez, University of Maine, Orono, ME, personal communication, 2001
Maine (statewide)	HW and conifer	28	Oe + Oa		NA	Lab	28	O	Fernandez et al. 2000
Bear Brook, ME	HW and conifer	4	Oe + Oa		5	Lab	28	O + M	Parker et al. 2001; Lindsey Rustad, USDA Forest Service, Durham, NH, and Ivan Fernandez, University of Maine, Orono, ME, personal communications, 2001
Acadia National Park, ME	HW and conifer	2	Oe + Oa		5	Lab	28	O + M	Parker et al. 2001; Lindsey Rustad, USDA Forest Service, Durham, NH, and Ivan Fernandez, University of Maine, Orono, ME, personal communications, 2001

HW, hardwood; NA, not applicable; NE transect, East–West northeastern US transect (Maine to New York); Oe, well-decomposed organic material; Oa, semidecomposed, fragmented organic matter.

1. “Samples collected” indicates forest floor subhorizons and mineral soil sampling depths.

2. C:N indicates the soil horizon at which C:N ratios were calculated (O, organic layer; M, mineral layer).



**Figure 3.** Distribution of foliar nitrogen concentration and ratio of lignin to nitrogen for red spruce and sugar maple in relation to elevation and estimated total nitrogen deposition. Triangles, red spruce; squares, sugar maple.

Maine and included a total of 251 plots (figure 4). Sites varied in vegetation type (conifer versus hardwood), elevation, and soils. To be included in this analysis, each site had to pro-

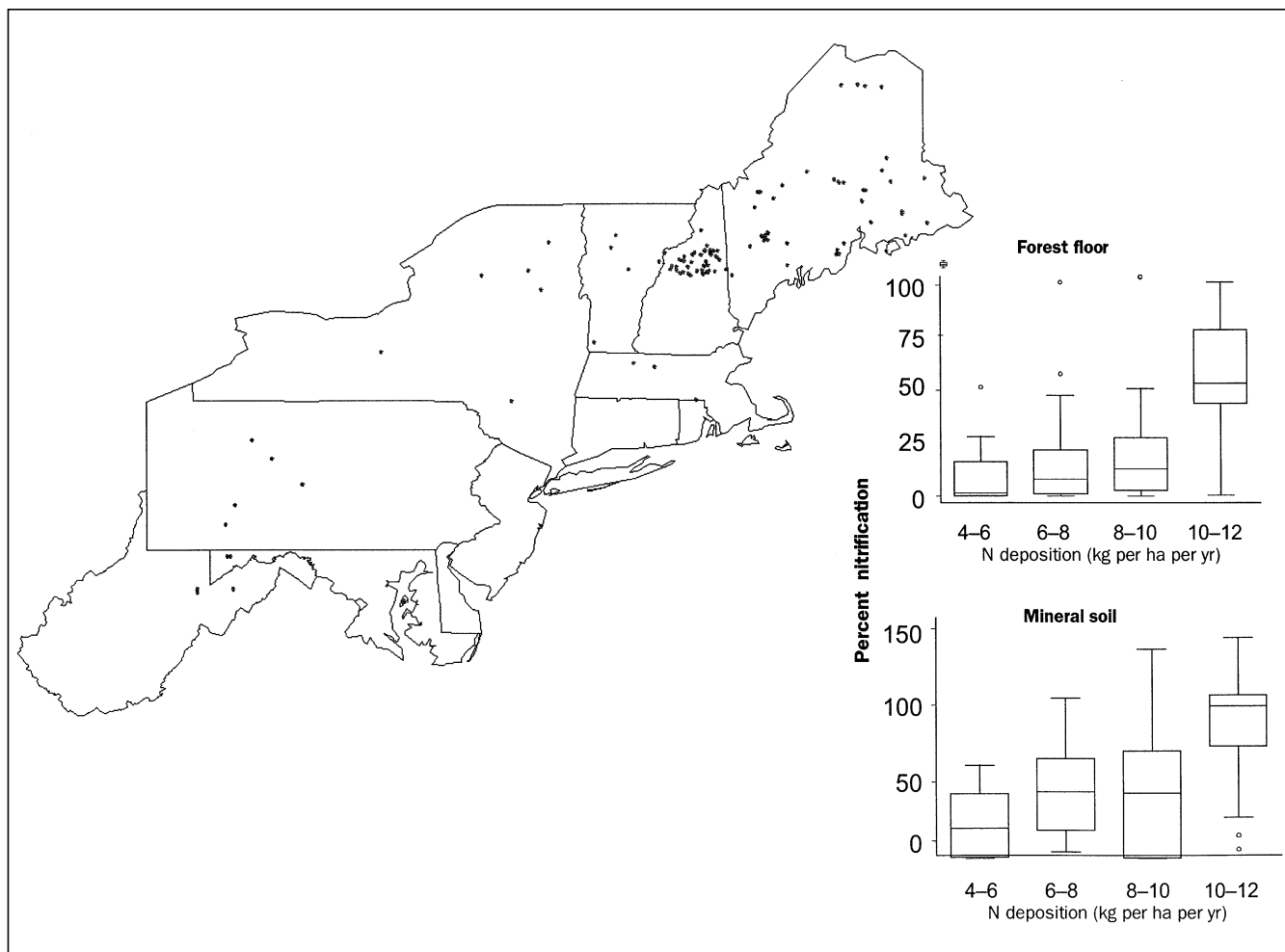
vide data for two or more of the variables shown in table 3. Methods for soil analysis were surprisingly consistent, but differences in methods of sample collection prevented some data from being used in all comparisons. Most studies sampled both the forest floor and the upper mineral soil (the top 5 to 15 centimeters), but some collected only one of these layers or combined both layers into a single analysis.

Analytical methods for C and N concentration are relatively straightforward and were consistent across sites. As a result, the C:N ratio was available at nearly all sites for at least one of the two horizons. Among N cycling measurements, data were largely from approximately 1-month laboratory incubations. For net N mineralization, there is no acceptable way to normalize data obtained from different incubation methods. For net nitrification, however, different methods can be compared by expressing net nitrification as a percentage of net

**Table 4.** Results of multiple linear regression analysis of soil carbon-to-nitrogen ratio and percent nitrification versus nitrogen deposition, mean annual temperature, and elevation.

	<i>n</i>	Mean annual temperature	N deposition	Elevation	<i>R</i> <sup>2</sup>
<b>Hardwood</b>					
C:N—organic	103		X		0.19
C:N—mineral	103	X	X		0.06
Percent nitrification—organic	105	X	X		0.17
Percent nitrification—mineral	97	X	X	X	0.50
<b>Conifer</b>					
C:N—organic	48	X			0.27
C:N—mineral	37				NS
C:N—total	27				NS
Percent nitrification—organic	54			X	0.1
Percent nitrification—mineral	41		X	X	0.42

NS, not significant; X, significant contribution to final equation.



**Figure 4.** Distribution of soil samples and box plots showing percent nitrification across the nitrogen deposition gradient. Box plots show median values and quartile ranges. Vertical lines show the 90% range of means; circles are outliers.

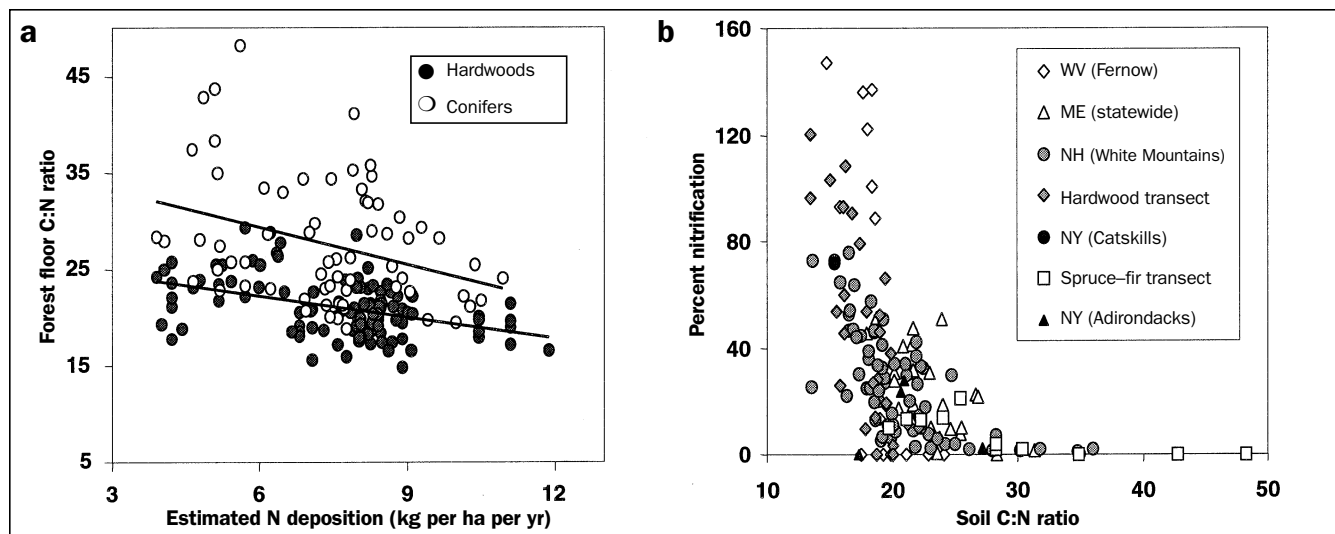
Across all sites, soil C:N ratios varied from 15 to 48 in the forest floor and from 10 to 39 in the mineral soil; they were generally higher in coniferous than in deciduous stands. In the forest floor, C:N ratios were significantly and inversely correlated with estimated N deposition (figure 5a, table 4). This trend differed between deciduous and coniferous stands but was significant for both forest types ( $P < 0.001$ ). Trends between N deposition and C:N ratios in mineral soils were weaker or nonsignificant (table 4). Both soil horizons showed strong inverse correlations between C:N ratios and nitrification, with net nitrification increasing sharply below a threshold C:N ratio of between 20 and 25 (figure 5b). This is consistent with a number of earlier studies from the northeastern United States, several of which are included in the present data set (Lovett and Rueth 1999, Goodale and Aber 2001, Ollinger et al. 2002). Similar trends have been reported across a range of European sites, with a C:N threshold of approximately 24 (Tietema and Beier 1995, Dise et al. 1998a, Emmett et al. 1998, Gundersen et al. 1998).

These results suggest that patterns of N deposition across the region have had a measurable effect on the chemistry of

soil organic matter and that this change should produce corresponding changes in net nitrification (as seen in figure 5). However, results from the foliar analysis suggest the possibility of an additional climatic effect that could also contribute to regional patterns of N cycling in soils. For some species, foliar N concentrations increase with elevation, either because N deposition increases with elevation or because the leaves respond to decreasing temperature and growing season length with increasing N concentrations (Yin 1992, Haxeltine and Prentice 1996). This raises the question of whether climatic effects on foliar chemistry might also influence soil C:N ratios through the effect of litter chemistry.

To examine this possibility, we compared soil N variables with plot elevations and estimated MAT. In the forest floor, both elevation and MAT were significant correlates of the C:N ratio in coniferous stands, but the trends were only mildly significant or not significant for deciduous stands (table 4). In the mineral soil, N deposition on its own was not significant, but elevation and MAT were significant within both deciduous and coniferous forests. Trends between N deposition and nitrification showed similar results. In the forest





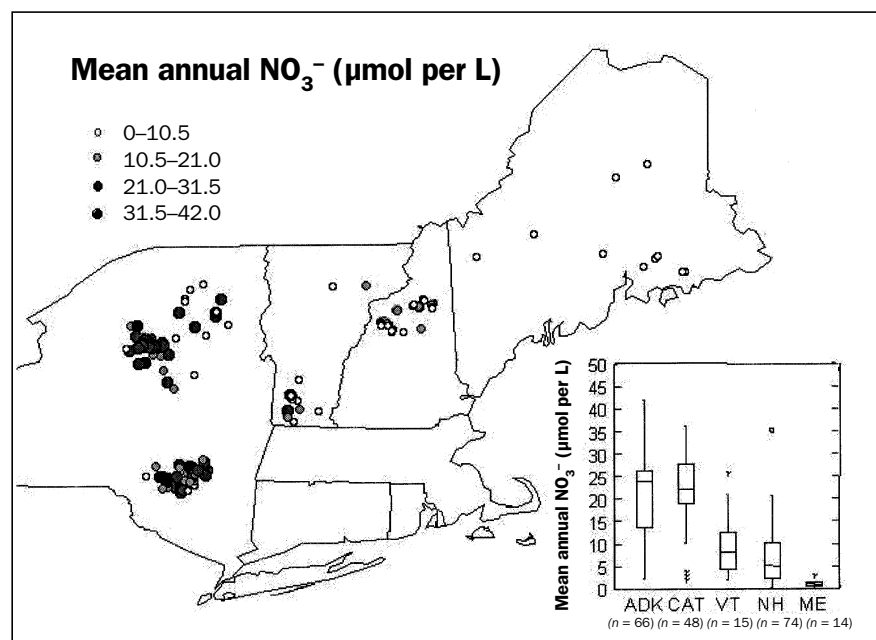
**Figure 5.** (a) Measured ratios of carbon to nitrogen (C:N) in the forest floor in relation to estimated nitrogen deposition, showing different trends for hardwood and conifer stands (hardwood stands,  $R^2 = 0.19$ ,  $P < 0.001$ ; conifer stands,  $R^2 = 0.27$ ,  $P < 0.001$ ). Trends were weaker or nonsignificant in mineral soils (see table 4). (b) Percent nitrification in combined organic and mineral soils in relation to soil C:N ratio. Study areas and sampling methods are described in table 3. Trends were significant ( $P < 0.001$ ) in organic, mineral, and combined soil layers, but data for combined soils are shown here because several studies did not report nitrification rates for individual horizons.

floor, percent nitrification increased significantly with increasing N deposition; the trend was stronger in deciduous than in coniferous stands. Trends between percent nitrification in the forest floor and elevation or MAT were either not significant or only weakly significant. In the mineral

soil, N deposition was the strongest correlate of nitrification in deciduous stands. Although the same was true for coniferous stands in a purely statistical sense, mineral soil nitrification showed an inverse relationship with N deposition, which ran counter to our expectations. Collectively, these

results suggest some additional influence of climate on soil variables but do not negate the significant effect of N deposition. As with the foliar data described above, the large degree of variability seen in all trends indicates that other factors play an important role in determining both soil C:N and nitrification fractions (table 4).

**Surface waters.** We assembled measurements of lake and stream chemistry from 354 upland forested catchments distributed across eastern New York, New England, and West Virginia (table 5). All were sampled during the mid- to late 1990s. Surface water  $\text{NO}_3^-$  concentrations generally peak during snowmelt and are lowest during the growing season, when biotic uptake is greatest. Changes in this seasonal pattern of stream  $\text{NO}_3^-$  concentration are key features for identifying stages of N saturation (Stoddard 1994), so we focused on  $\text{NO}_3^-$  status during the expected seasonal minimum (summer, considered here as June through September), maximum

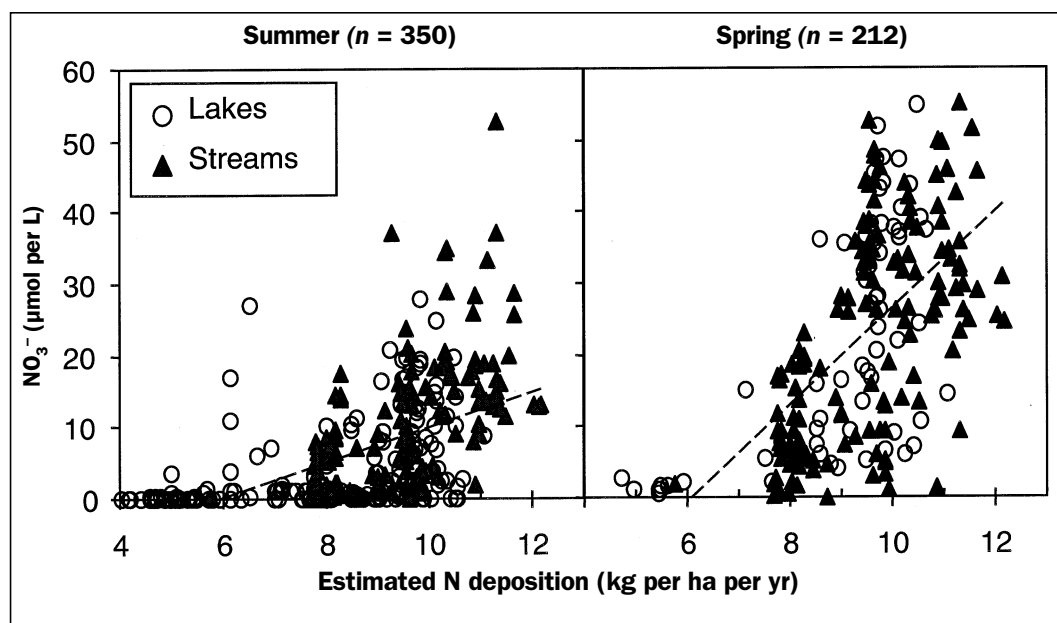


**Figure 6.** Mean annual nitrate ( $\text{NO}_3^-$ ) concentration in 220 lakes and streams across the northeastern United States. Inset indicates the median, quartile, and 90% range of mean annual  $\text{NO}_3^-$  in the Adirondacks (ADK), the Catskills (CAT), Vermont (VT), New Hampshire (NH), and Maine (ME).

Table 5. Surface water data sets.

Region	Data set	Surface water	Number of sites for which nitrate concentration was measured			Years included	Frequency of sampling	Reference
			Spring	Summer	Annual			
Maine	East Bear Brook	Stream	1	1	1	1993–1999	52/yr	Kahl et al. 1993a, 1999, Norton et al. 1999
Maine	LTM network	Lake	10	13	0	1993–1999	4/yr	Kahl et al. 1993b, Stoddard et al. 1998a
Maine	High-elevation lakes monitoring	Lake	0	39	0	1997–1999	1–4/yr	Kahl et al. 1991
White Mountains	Hubbard Brook	Stream	4	4	4	1995–1997	52/yr	Campbell et al. 2000
White Mountains	The Bowl	Stream	4	4	4	1994–1997	12/yr	Martin et al. 2000
White Mountains	MAP-BGC	Stream	18	19	0	1999–2000	4/yr	
White Mountains	Land-use history survey	Stream	25	25	25	1996–1997	12/yr	Goodale et al. 2000
White Mountains	Mount Moosilauke	Stream	21	21	0	1996–1997	4/yr	Goodale et al. 2003
White Mountains	Cone Pond inlet	Stream	1	1	1	1993–1999	52/yr	Campbell et al. 2000
Vermont	Sleepers River	Stream	1	1	1	1993–1999	52/yr	Campbell et al. 2000
Vermont	Lye Brook	Stream	3	3	3	1994–1995	26/yr	Campbell et al. 2000
Vermont	LTM network	Lake	11	11	0	1993–1999	4/yr	Stoddard and Kellogg 1993, Stoddard et al. 1998a
Adirondack	Arbutus inlet	Stream	1	1	1	1995–1998	> 52/yr	McHale et al. 2000, Mitchell et al. 2001
Adirondack	Adirondack Effects Assessment Program	Stream	20	20	0	1998–2000	4–5/yr	Greg B. Lawrence, US Geological Survey, Troy, NY, personal communication, 2001
Adirondack	LTM network	Lake	45	45	0	1993–1999	12/yr	Driscoll et al. 1995, Stoddard et al. 1998b
Catskills	Biscuit Brook	Stream	1	1	1	1993–1997	> 52/yr	Murdoch et al. 1998
Catskills	LTM network	Stream	8	8	0	1993–1999	> 12/yr	Murdoch and Stoddard 1992, 1993
Catskills	Catskill survey	Stream	39	39	39	1994–1997	4/yr	Lovett et al. 2000, 2002b
West Virginia	Fernow Experimental Forest	Stream	3	3	3	1995–1999	52/yr	Peterjohn et al. 1996, Adams et al. 1997
Northeast	TIME	Lake	0	95	0	1991–1995	1/yr	Kahl et al. 1991, Stoddard et al. 1998a, 1998b
Total			216	354	220			83

LTM, long-term monitoring of lakes and streams; MAP-BGC, mapping and analysis of productivity and biogeochemical cycles; TIME, temporally integrated monitoring of ecosystems project.



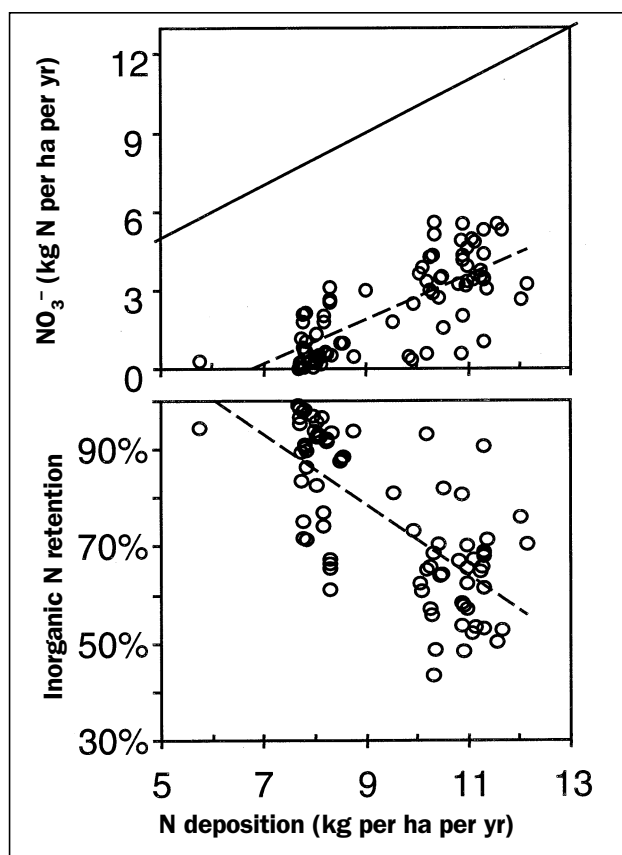
**Figure 7.** Surface water nitrate ( $\text{NO}_3^-$ ) concentrations increase with estimates of nitrogen (N) deposition at the base of each watershed in both summer and spring. Nitrogen deposition to the whole watershed may be 2 to 6 kilograms per hectare per year greater than at the base. Summer nitrate =  $2.5 \text{ N deposition} - 14.4$ ;  $R^2 = 0.30$ ,  $P < 0.001$ . Spring nitrate =  $6.7 \text{ N deposition} - 40.7$ ;  $R^2 = 0.38$ ,  $P < 0.001$ .

(spring, considered here as February through mid-May), and mean annual  $\text{NO}_3^-$  concentration. Of the 354 sites, 220 were sampled at least quarterly and were included in annual summaries. The other 134 are included here only to illustrate patterns of  $\text{NO}_3^-$  loss during the growing season. Export of  $\text{NO}_3^-$  and ammonium ( $\text{NH}_4^+$ ) has been estimated for 83 of the streams in kg N per ha per yr, although this total includes 28 sites with modeled streamflow and 43 sites with flow estimated from nearby gauged watersheds, along with 12 sites with precisely measured flow and frequently measured stream chemistry (table 5). Dissolved organic N export has been estimated for 80 of these streams, or all except the three streams at the Fernow Experimental Forest. Dissolved organic N has been shown to constitute a significant portion of N loss in many northeastern streams (Campbell et al. 2000, Lovett et al. 2002a).

Mean annual  $\text{NO}_3^-$  concentrations decreased across the northeastern United States, from 20 to 25 micromoles per liter ( $\mu\text{mol per L}$ ) in the Adirondack and Catskill Mountains in New York to 10  $\mu\text{mol per L}$  in the Green Mountains of Vermont, 5  $\mu\text{mol per L}$  in the White Mountains of New Hampshire, and barely 1  $\mu\text{mol per L}$  in scattered Maine lakes (figure 6). This trend of decreasing surface water  $\text{NO}_3^-$  concentrations from southwest to northeast closely parallels the spatial pattern of N deposition across the region (figure 1). Direct comparisons indicate that stream and lake  $\text{NO}_3^-$  increased with N deposition during both the growing season and the dormant season, although the increase was steeper during the dormant season (figure 7).

Although these relationships display substantial scatter, three important trends emerge. First,  $\text{NO}_3^-$  concentrations rarely exceeded 1  $\mu\text{mol per L}$  in watersheds receiving less than about 7 kg N per ha per yr at their base. Because N deposition generally increases with elevation, N deposition estimated for the base of the watersheds most likely underestimates N received on the whole catchment by 2 to 6 kg N per ha per yr, so that the threshold of 7 kg N per ha per yr for the base of a watershed is comparable to a threshold of about 9 to 13 kg N per ha per yr for the whole watershed. Second, the only lakes and streams with relatively high  $\text{NO}_3^-$  concentrations were those receiving relatively high N inputs, although responses to increased N deposition varied greatly. Watershed response to excess N deposition can vary greatly, depending on factors such as species composition (Lovett et al. 2000, 2002a), land use history (Vitousek and Reiners 1975, Goodale et al. 2000), bedrock mineralogy (Williard et al. 1997), and flowpaths (Burns et al. 1998, Creed and Band 1998), but  $\text{NO}_3^-$  losses are low unless N inputs are elevated to several times above preindustrial conditions. Three high-elevation ponds on Mount Katahdin, Maine, are the only exceptions to this trend. The relatively large fractions of rock cover in these watersheds may have made them particularly sensitive to even low amounts of N deposition, as suggested for rocky catchments in the Rocky and Sierra Mountains (e.g., Williams et al. 1995, Baron and Campbell 1997). Third, as N deposition increased, the variance in  $\text{NO}_3^-$  concentration increased along with the mean. Some streams have low  $\text{NO}_3^-$  concentrations even under relatively high N deposition conditions, and therefore differences among watersheds in  $\text{NO}_3^-$  leaching and its effects (e.g., stream acidification) are more pronounced in areas of higher N deposition.

Unlike the foliage and soil data, the surface water data appear to be free from confounding covariation between N deposition, elevation, and climate factors. Although elevation covaried modestly with N deposition ( $R^2 = 0.18$ ,  $P < 0.001$ ), relationships between N deposition and  $\text{NO}_3^-$  concentrations were far stronger than between elevation and spring



**Figure 8.** (a) Nitrogen (N) export in stream water increases as N deposition at the base of the watersheds increases above 7 kilograms per hectare per year. Nitrogen export =  $0.85 \text{ N deposition} - 5.8$ ;  $R^2 = 0.56$ ,  $P < 0.001$ . (b) Watershed N retention decreases as N deposition at the base of the watersheds increases (N retention =  $-0.07 \text{ N deposition} + 1.44$ ;  $R^2 = 0.50$ ).

( $R^2 = 0.01$ ,  $P = 0.13$ ) or summer ( $R^2 = 0.02$ ,  $P = 0.01$ )  $\text{NO}_3^-$  concentration. For the 94 watersheds with measurements of mean watershed elevation, the correlations between elevation and  $\text{NO}_3^-$  loss did not improve when the mean rather than the minimum watershed elevation was used.

Over the 83 streams with estimates of  $\text{NO}_3^-$  export,  $\text{NO}_3^-$  losses increased steeply with N deposition above 6.8 kg per ha per yr at the base of the watershed (figure 8) or about 10 kg per ha per yr for the whole watershed. Within each region (e.g., the Catskills or the White Mountains),  $\text{NO}_3^-$  export varied greatly and did not appear to vary strongly with deposition; however, when all of the data sets were combined, clear patterns emerged across the region (figure 8). Nitrate export increased from about 0.3 kg N per ha per yr at the East Bear Brook watershed in south-central Maine to over 5.0 kg N per ha per yr at Watershed 4 in the Fernow Experimental Forest, West Virginia, and at several streams in the Catskill Mountains. Estimated losses of dissolved organic N averaged 0.8 kg N per ha per yr, ranged from 0.1 to 2.2 kg N per ha per yr, and did not vary with N deposition ( $R^2 < 0.01$ ,  $P = 0.88$ ).

Variation in  $\text{NO}_3^-$  export explained 95% of the variability in total N loss. As  $\text{NO}_3^-$  losses increased, watershed inorganic N retention ( $[\text{inputs} - \text{losses}]/\text{inputs}$ ) decreased from over 90% retention in watersheds receiving little N deposition to 50% to 60% retention in watersheds receiving 10 to 12 kg N per ha per yr at the base of the watershed.

The relationship between N deposition and  $\text{NO}_3^-$  export observed here for the northeastern United States is remarkably similar to the relationships observed by Dise and Wright (1995) and Dise and colleagues (1998b) for European forests, despite differences in forest types and management. Dise and colleagues (1998b) found that inorganic N losses were close to zero for watersheds where N deposition was less than 10 kg per ha per yr, highly variable for watersheds receiving 10 to 30 kg N per ha per yr, and consistently elevated for watersheds receiving more than 30 kg N per ha per yr. Although the northeastern United States does not receive N deposition at the high end of this range, the pattern at the lower end is nearly identical; even the threshold of approximately 10 kg N per ha per yr is similar to observations from the Northeast, after the 7 kg N per ha per yr at the base of the watershed is scaled up to the whole watershed.

### Synthesis and conclusions

Our original question was, "Is the N status of northeastern forests being altered by N deposition?" Using correlational techniques and large sample size, our analysis suggests that the answer to this question is yes, although the degree of response varied greatly across the three different categories of indicators we examined. The surface water data suggest a strong relationship between  $\text{NO}_3^-$  concentration and flux across the N deposition gradient. The soil data show strong relationships between N deposition, soil C:N ratio, and nitrification in several cases, but the strength and significance of these trends differed among forest types and soil horizons. Finally, in the foliar data set, significant relationships with N deposition did not emerge beyond the covarying effects of climate and elevation.

One explanation for these differences is that the confounding factors described earlier (climate variation, disturbance, species composition, and hydrologic pathways) affect foliar, soil, and stream chemistry at different spatial and temporal scales. We suggest that the relative sensitivity of the three indicators to these confounding variables affects the strength of the relationship exhibited with N deposition. Specifically, foliar samples integrate over a very small area (one tree crown) and are subject to strong and unexplained interannual variation. We did not attempt to control for year of sampling in our foliar analyses, and we cannot be certain whether this would have been helpful, as we do not know if the factors controlling interannual variability are local or regional. In addition, although we included only the most widely distributed species in our analysis, the foliar data set was the least evenly distributed over the study region. Although soil characteristics do not change rapidly over time, they are also subject to large variation over very short spatial scales



because of the fine scale of disturbance forces and species distribution in the soil. The close correlation between C:N and nitrification suggests that N status is reflected in nitrification, while the weaker relationship between C:N and N deposition may reflect the influence of disturbance and species effects. Surface water samples integrate over much larger areas (watersheds), and although decadal changes in stream  $\text{NO}_3^-$  appear to occur, all samples used in this analysis were taken during the 1990s, when stream  $\text{NO}_3^-$  concentrations were relatively stable and generally low compared with those of earlier periods.

Of the three indicators discussed here, surface waters yield the most comprehensive view of the state of N saturation across the Northeast. After we controlled for time, surface waters integrated more effectively over space than did foliage or soils. Significantly larger sample sets for soils and foliage would be required to cover the same effective area as stream sampling; if spread more systematically across the region, these sample sets could also provide valuable insights. Synoptic, broad-scale, repeatable sampling, like that becoming possible for foliage through imaging spectrometry (Martin and Aber 1997, Smith et al. 2002), may be the only method by which foliar chemistry could be a valuable broad-scale spatial predictor of forest N status.

Using surface waters as an indicator of the degree of N saturation, it seems clear that N deposition is altering the N status of, and  $\text{NO}_3^-$  leaching from, forests in the northeastern United States.

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