Foliar Nitrogen Responses to Elevated Atmospheric Nitrogen Deposition in Nine Temperate Forest Canopy Species

BRENDEN E. MCNEIL,*,†
JANE M. READ,† AND
CHARLES T. DRISCOLL‡

Department of Geography, Maxwell School of Citizenship and Public Affairs, Syracuse University, Syracuse, New York 13244, and Department of Civil and Environmental Engineering, Syracuse University, Syracuse, New York 13244

Despite its ecological importance, broad-scale use of foliar nitrogen as an indicator of ecosystem response to atmospheric N deposition has heretofore been obscured by its poorly understood intrinsic variability through time, space, and across species. We used a regional survey of foliar N conducted within a single growing season to observe that eight of nine major canopy tree species had increased foliar N in response to a gradient of N deposition in the Adirondack Park, New York. These results (1) add important foliar N evidence to support N saturation theory, (2) strongly reinforce the conclusion that N deposition is affecting the N status of forest ecosystems in the northeastern U.S., and (3) extend N saturation theory by identifying that temperate forest canopy species differ in their foliar N response to N deposition. Interestingly, species-specific differences were strongly related to two functional traits that arise from within-leaf allocations of N resources—leaf mass per area (LMA) and shade tolerance. Thus, combining species-specific knowledge of these functional traits with existing foliar N-centered remote sensing and ecosystem modeling approaches may provide a much-needed avenue to make broad-scale assessments of how persistently elevated rates of N deposition will continue to affect temperate forest ecosystems.

Introduction

The fundamental control played by foliar nitrogen (N) on processes of photosynthetic production (*1*, *2*) and litter decomposition (*3*, *4*) make it a sensitive indicator of forest ecosystem response to atmospheric N deposition (*5*). Unfortunately, its broad-scale use as an ecosystem indicator has heretofore remained elusive because of its intrinsic variability through space, time, and across species (*6*, *7*). Furthermore, while each plant species invests in foliar N according to an adaptive tradeoff between leaf-based strategies of production versus persistence (*7*), little is known about how these investment strategies affect the response of foliar N to N deposition.

* Corresponding author: Present address: Department of Forest Ecology and Management, University of Wisconsin–Madison, Madison, Wisconsin, 53706. email: bmcneil@wisc.edu; phone: 608-263-9107; fax: 608-262-9922.
† Department of Geography.
‡ Department of Civil and Environmental Engineering.

Regardless of its leaf-based strategy for photosynthetic production, a plant species can be expected to physiologically acclimate, or genetically adapt, its investment in foliar N in response to the growing conditions in its environment (*8*). Since atmospheric N deposition can play a dominant role in determining the nutrient environment experienced by a tree, it is not surprising that elevated foliar N is one of the most consistent responses observed across spatial gradients of N deposition and within experimental N amendment studies (*5*, *9*–*11*). Given the wide range of production strategies employed by trees (*7*), it seems reasonable to anticipate that different species will vary in their foliar N response to N deposition. However, most studies to date have examined this response within a selectively chosen set of species (e.g., *9*, *10*, *12*). We are not aware of any work that has systematically examined the controls on species-specific foliar N responses to N deposition. This narrow focus is understandable given that previous research addressed the theoretical and now well-established biogeochemical linkages between N deposition and foliar N (*5*, *9*, *10*, *12*). Nevertheless, our ability to generalize this biogeochemical linkage to complex, mixed-species forest environments will depend upon establishing predictive relationships on the response of foliar N of a given species to changes in N deposition.

Here, we present results from a regional survey of foliar N conducted over a single growing season in temperate forest canopy trees of the Adirondack Park, New York. The elevated emissions of N from the upper Midwest with additions from local sources, and their north-eastward atmospheric transport and deposition onto the mountainous landscape of the Adirondack Park, cause a regional southwest to northeast N deposition gradient (ref *13*); Figure 1. Similar to a recent study by Aber and colleagues (*7*), we used a dense network of sample plots along this spatial deposition gradient (Figure 1) in an attempt to overwhelm variability attributable to other factors (e.g., geology, disturbance history, topographic position) and detect a foliar N response to N deposition. However, our approach differs from that of Aber and colleagues in that we control for interannual temporal variability by making all measurements within the peak of a single growing season, a period when foliar N is known to be relatively stable. This temporal constraint is significant, as long-term studies in the northeastern United States have shown foliar N to vary by as much as 25% between years (*12*), and Aber and colleagues identified that this variation likely confounded their recent attempt to uncover similar relationships between N deposition and foliar N (*6*). Moreover, our study is novel in that we measure foliar N in 10 species with widely varying leaf-based photosynthetic production strategies. These two features of our survey provide a unique opportunity to not only evaluate foliar N as a regional-scale indicator of ecosystem response to N deposition, but also begin to describe the species-specific nature of this response.

We report measurements of foliar N for all major overstory canopy species (Table 1) that occurred within a diverse set of 75 forest plots located along the N deposition gradient in the Adirondack Park (Figure 1). Since a supporting multiple linear regression analysis (*14*) revealed that the spatial patterns of foliar N along the deposition gradient could not be explained by any combination of other factors that may affect foliar N (e.g., temperature, disturbance history, moisture availability, geology), we use univariate linear regression to directly relate the spatial pattern of foliar N to modeled estimates of total inorganic N in wet deposition. Our discussion not only considers the significant ecological implications of the observed foliar N responses to N
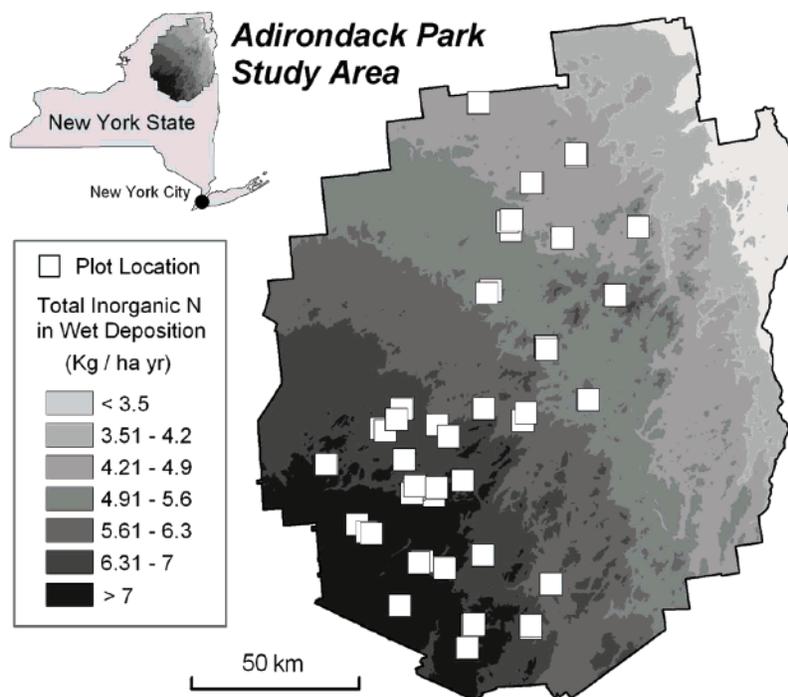
FIGURE 1. Plot locations along the Adirondack Park N deposition gradient. Many plots are hidden due to their proximity to other plots.

TABLE 1. Species Characteristics and Regression Parameters[a]

common name	scientific name	mean foliar N	LMA (gm-2)	shade tolerance	N	slope	intercept	species response metric
Paper birch	*Betula papyrifera*	2.53	74.40	1	7	0.34	0.56	13
Yellow birch	*Betula alleghaniensis*	2.96	66.33	2	44	0.38	0.62	13
Red maple	*Acer rubrum*	2.50	71.30	3	29	0.20	1.28	8
Sugar maple	*Acer saccharum*	2.60	62.58	4	34	0.22	1.24	8
American beech	*Fagus grandifolia*	2.96	61.08	4	52	na	na	na
White pine	*Pinus strobus*	1.52	173.67	2	7	0.13	0.78	9
Eastern hemlock	*Tsuga canadensis*	1.56	169.87	4	25	0.13	1.09	8
Balsam fir	*Abies balsamea*	1.47	203.90	4	24	0.10	0.90	7
Red spruce	*Picea rubens*	1.21	304.67	4	41	0.074	0.75	6

[a] LMA and shade tolerance for each species are from literature sources (see text). Regression parameters describe relationships in Figure 2. Species response metrics (see Methods) are scaled to integers by multiplying by 100.

deposition, but also examines the need for future research on the species-specific nature of foliar N response to N deposition. In order to begin to test a hypothesis stating that the species-specific nature of foliar N response to N deposition results from the diverging strategies used by tree species in allocating N resources to different leaf constituents, we relate variation in the species-specific rates of foliar N response to N deposition to two functional trait descriptors of a species' leaf-based resource allocation strategy—leaf mass per area (LMA) and shade tolerance.

Methods

Research Design. As described in detail elsewhere (*14*), our plot selection within the approximately 2.5 million hectare Adirondack Park was designed to maximize spatial variation among major factors hypothesized to affect foliar N (i.e., temperature, species composition, disturbance history, geology, soil moisture, atmospheric N deposition). This approach has the inherent limitation that the effects of N deposition could be confounded by spatial variation in other factors affecting foliar N. However, in every instance but one, we found that the spatial variation explained by N deposition was independent (i.e., orthogonal in a multiple regression) of variation explained by other factors (*14*). In the one exception (which only occurred in the two *Acer* species),

lower temperature and higher N deposition appeared to interactively contribute to increased foliar N at high elevations. This interaction may have slightly inflated the magnitude of these species foliar N response along the gradient of N deposition. However, because of the regional nature of the deposition gradient in the Adirondack Park (Figure 1), approximately 85% of the spatial variation in N deposition is independent of elevation (*14*). Thus, the interaction of temperature and N deposition is highly unlikely to affect the qualitative interpretation of the relationships between foliar N and N deposition reported here.

Foliar N Data. We collected leaf samples from 73 forest plots (30 × 30 m) between July 1 and August 24, 2003, and an additional two plots within a high elevation spruce-fir forest on September 5, 2003. Plots were intentionally sampled to minimize any covariation among geographic location and sample date, and we found no significant effect of sample date on foliar N. We measured the latitude, longitude, and elevation of each plot centerpoint with a survey-grade GPS unit (±1 m accuracy). We collected fresh leaf material from two to five (typically three) individuals of each canopy species in the plot by using a shotgun to obtain small twigs from multiple canopy heights (*15*). Relatively rare canopy species were not included in our analysis because of their small sample size (*N* < 4 plots) and poor distribution along the N

deposition gradient. All leaf samples were dried at 60 °C for 48 h and ground to pass through a 1 mm mesh screen. We analyzed each sample for foliar N using the CHN combustion method, maintaining quality control checks within ±5% error. We averaged individual measurements to obtain one foliar N value for each species in a plot. The full foliar N dataset and plot coordinates are available online from the Foliar Chemistry Database (www.folchem.sr.unh.edu).

N Deposition. We predicted total inorganic N in wet deposition using a slightly improved version of a well-validated spatial regression model (*13*) and our GPS measurements of the latitude, longitude, and elevation of each plot. Details of our improvements to the model and model validation for our specific study period are provided in the supplementary methods. We note that the unknown patterns of dry deposition, cloudwater and fog deposition, and organic N deposition are likely to affect the actual spatial pattern of atmospherically derived N received by our sampled trees. Ongoing modeling work by Kathleen Weathers and colleagues within northeastern North America suggests that these additional atmospheric sources of N are likely to be small relative to wet deposition (15−30% of wet deposition) and are likely to have largely coincident spatial patterns (K. Weathers, personal communication). Thus, the roughly 4 to 8 kg N ha^{-1} yr^{-1} gradient of wet deposition used in our analysis is likely to represent a total N deposition gradient of 5 to 10 kg N ha^{-1} yr^{-1}, although total N deposition may be higher in evergreen forests, high elevations, and windward aspects that receive increased dry deposition, cloudwater, and fog inputs of N (*16*, *17*).

Statistical Analysis. We performed univariate regressions between the N deposition predicted at each plot and the plot-average foliar N value of each species. We identified a statistical outlier (Jackknife distance = 3.5, $N = 8$, $p < 0.05$) within the white pine regression and removed it from the analysis because the plot location was within a pine plantation that was fertilized or actively managed. Either form of forest management made this plot not part of the unmanaged forest requirement describing our statistical population.

We quantified the variability in species foliar N response to N deposition by developing a species response metric. Since the magnitude of foliar N increase with N deposition is strongly affected by its intrinsic magnitude of foliar N investment (*14*), we calculated the species response metric for each species as the slope of its relationship between N deposition and foliar N divided by its mean foliar N (Table 1). We obtained LMA and shade tolerance information for each of our species from the literature (*15*, *18*), and used univariate and multiple linear regression to evaluate the degree to which these variables explain the variability in species response to N deposition. We coded shade tolerance class as continuous integer data ranging from intolerant as "1" to very tolerant as "4".

Results and Discussion

Foliar N as an Indicator of Ecosystem Response to N Deposition. Mean foliar N values ranged from 1.21 %N for red spruce to 2.96 %N for American beech and yellow birch (Table 1). As expected, we observed lower mean foliar N in species with high LMA, and vice versa (Table 1, (*7*, *19*)).

Foliar N values were significantly related (at $p < 0.1$) to N deposition for eight of nine species (Figure 2). Despite the differing evolutionary histories and leaf types of these species, these significant relationships for almost all major canopy species in this ecosystem indicate that once interannual variability and species effects are constrained, foliar N can serve in its much-anticipated capacity of a broad-scale indicator of ecological response to N deposition (*5*, *6*). Furthermore, given the large spatial variation within the sample, the strong linear form of the relationships implies that the effect of N deposition on foliar N occurs independently of the species' immediate growing environment (Figure 2). This finding suggests that the effect of additional spatial controls on foliar N (e.g., disturbance history, soil moisture availability) is likely to operate independently of N deposition, and that knowledge of these controls could help explain the spatial variability in foliar N unaccounted for by N deposition. Moreover, since N deposition appears to play an important role in structuring the spatial variability of foliar N (Figure 2), the effects of N deposition may be detectable within broad-scale, high-resolution maps of foliar N derived from hyperspectral image analysis (see, e.g., *20*).

In addition to supporting the use of foliar N as a broad-scale indicator of ecological response, the significant increases in foliar N observed in eight species across the Adirondack Park N deposition gradient (Figure 2) add substantial support to Aber and colleagues' recent conclusion that N deposition is altering the N status of northeastern U.S. forests (*6*). Moreover, foliar N data from our study complement recent surface water and soil surveys conducted within the same Adirondack Long-Term Monitoring (ALTM) watersheds where we collected over 75% of our foliar N measurements. Similar to Aber and colleagues' assessment of N status across the slightly broader gradient of N deposition in the northeast U.S. (*6*), the comparison of these foliar, soil, and surface water indices are useful for assessing the response of Adirondack ecosystems response to N deposition.

Surface water measurements indicate that N deposition exerts a moderate degree of control ($R^2 = 0.21$, $N = 52$, $p = 0.0006$) in causing increased surface water export of NO_3^- from ALTM lake-containing watersheds (*21*). Measurements of organic horizon soils indicate a weak pattern of declining C:N and increasing N with increasing N input along the deposition gradient ($R^2 = 0.05$, $p = 0.002$ for soil C:N and $R^2 = 0.13$, $p < 0.0001$ for soil N; data from ref *22* analyzed in ref *14*). In comparison, results from our study suggest that foliar N responses to N deposition are species-specific and range from extremely weak to very strong ($R^2 = 0.04$ to $R^2 = 0.85$, Figure 2). These surface water, foliar, and soil indicators all support the conclusion that N deposition is affecting the N status of Adirondack forests. Yet, the differing strength of the response observed in each indicator complicates our ability to directly assess the severity of the impact that N deposition is having on Adirondack forested ecosystems. Of the many factors affecting each indicator (see review in ref *6*), our assessment of these studies leads us to suggest that future research on three particular factors may be particularly valuable: (1) in-lake processes of denitrification and planktonic NO_3^- depletion (*21*), (2) intraannual variability of foliar and surface water N measurements (*23*, *24*), and (3) effects of species composition on soil N, soil C:N, and foliar N (*25*, *26*).

Notwithstanding the need for future research in these areas, the combination of the foliar, soil, and surface water indicators suggests that at the present time, most Adirondack ecosystems are in the earlier "fertilization" stages of ecosystem N saturation (*5*). From the N saturation hypotheses (*5*, *11*), we would expect Adirondack ecosystems at more advanced stages of N saturation to have continued strong foliar N responses and exhibit stronger soil and surface water responses to N deposition. In light of the continued high rate of N emissions and the resultant N deposition onto Adirondack ecosystems, results from our study emphasize that continued foliar, soil, and surface water monitoring of the N status of Adirondack ecosystems will be essential for attempts to (1) predict the onset of the later stages of N saturation and (2) develop air pollution policy capable of mitigating the many deleterious environmental effects that are expected to occur with continued loading of N onto Adirondack ecosystems.
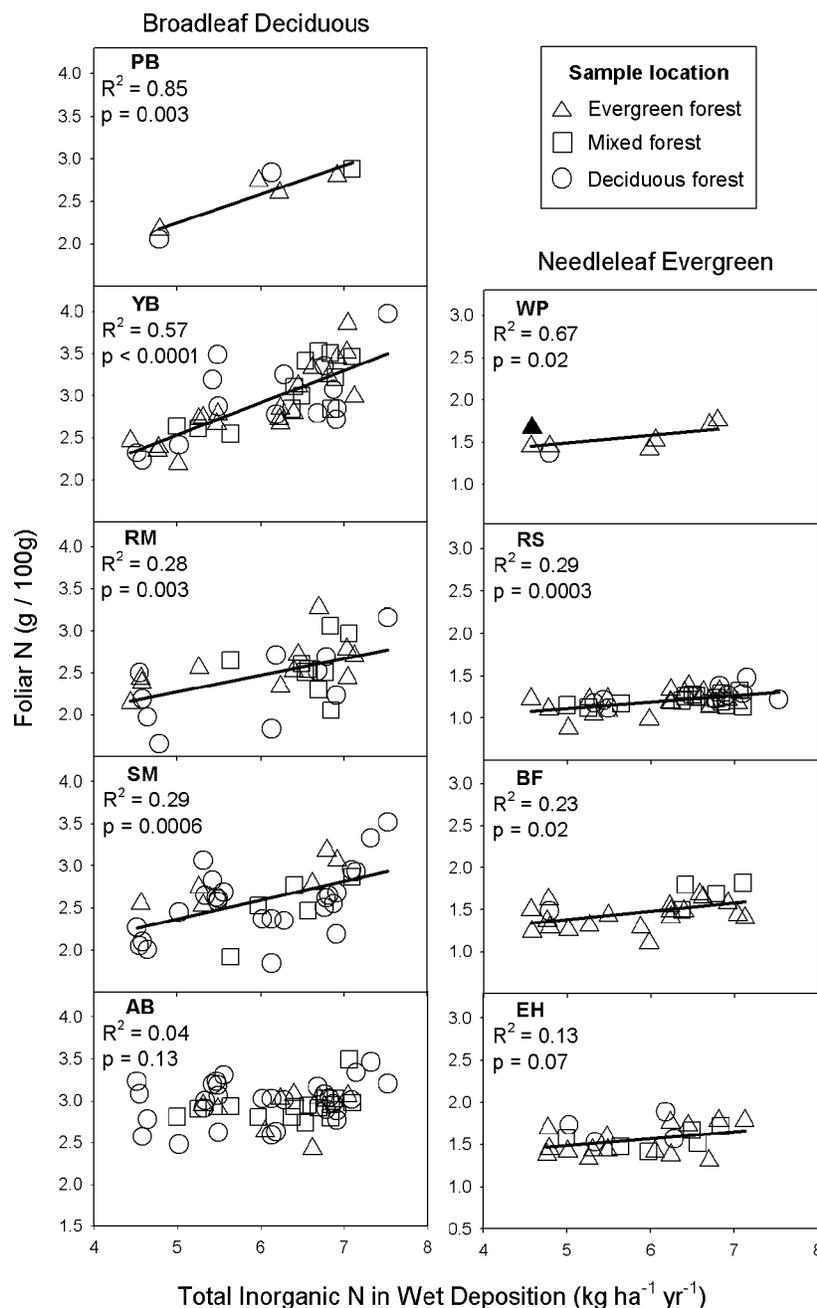
FIGURE 2. Species-specific relationships of foliar N and N deposition. Species are organized in columns by functional group and increase in shade tolerance down each column. Although the Y-axis is shifted downward in the needleleaf evergreen column, its magnitude is the same as in the broadleaf deciduous column. This enables visual comparison of slopes. The filled-in symbol within the WP panel denotes a statistical outlier that was removed from the analysis (see Methods). Scientific names, and regression parameters are in Table 1. Species are abbreviated by their common name given in Table 1.

Species-Specific Nature of Foliar N Response. A most striking result to emerge from our analysis is that the nine temperate forest canopy species in our study have markedly different rates of foliar N response to N deposition (Figure 2). This species-specific nature of foliar N response to N deposition has obvious implications toward the selection of study species for future foliar N-based assessments of the ecosystem impacts of N deposition. For example, if our assessment was limited to the study species of American beech, our results would be in apparent conflict with the aforementioned soil and surface water indices suggesting that N deposition is altering the N status of our study ecosystem. This apparent conflict was also observed in a survey of European forests where beech and oak were found to have no foliar N response, whereas soil C:N, soil solution

NO_3^-, and foliar N in spruce, pine, and fir species all had significant responses to a natural gradient of N deposition.

These studies pose the question: Why do species differ in their foliar N response to N deposition? We began to address this question by calculating species response metrics for eight of our nine study species (Table 1). We did not calculate a species response metric for American beech since its slight, and statistically insignificant $(p > 0.1)$ increase in foliar N was inseparable from a relationship with mean annual temperature along the elevation component of the deposition gradient (14).

Species response metrics were highest in paper and yellow birch, and lowest in red spruce (Table 1). We found that 86% of the variability in species response metrics was accounted for by species characteristics of LMA and shade tolerance
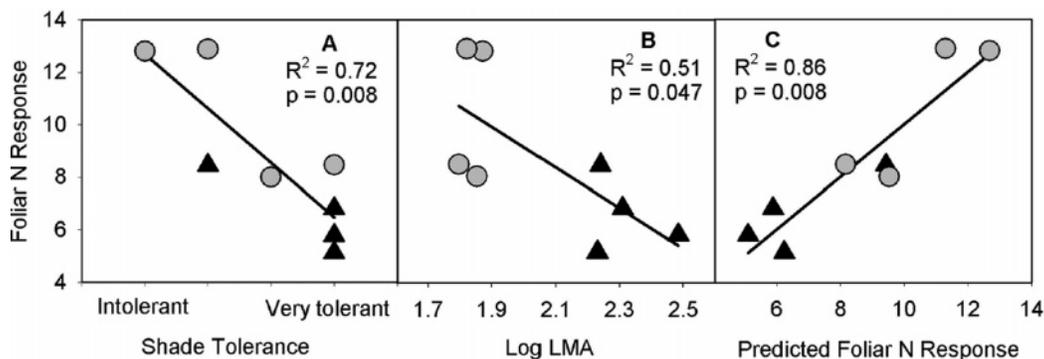
FIGURE 3. Prediction of species foliar N response to N deposition by LMA and shade tolerance. Broadleaf deciduous species are gray circles and needleleaf evergreen species are black triangles. Foliar N response is a unitless index representing the normalized slope of regressions shown in Figure 2 (see text). The regression equation in panel A has a slope of -2.07 and a y-intercept of 14.75, and panel B a slope of -7.72 and a y-intercept of 24.55. The predicted equation describing the x-axis of panel C: Foliar N response $= -22.66 - 4.46(LMA) - 1.61(shade\ tolerance)$.

(Figure 3). The significant effect of LMA was strongly associated with differences among broadleaf deciduous and needleleaf evergreen functional groups (Figure 3b). However, the effect of LMA also appeared to operate within a functional group, as the three very shade tolerant needleleaf evergreen species showed a trend of decreasing species response metric with increasing LMA (Table 1, Figure 3b). This trend is in agreement with other recent findings indicating that LMA is a more direct characterization of species differences in leaf-level physiology than functional type classifications (*7*, *27*). We note that the small sample size ($N = 8$ species) and use of literature derived values for LMA and shade tolerance precludes a more rigorous test of the role played by these characteristics in regulating foliar N response to N deposition. Nevertheless, it is conceptually striking that these two species characteristics result from fundamental tradeoffs that define a species leaf-based photosynthetic strategy.

There are several physiological mechanisms that likely cause LMA and shade tolerance to affect species response to N deposition. First, the self-shading structure, CO_2 diffusionary mechanics, or the requirement to invest N in structural compounds necessitate that species with high LMA must invest proportionately greater amounts of N to achieve a similar carbon gain (*28*–*30*). Since this cost makes it comparatively expensive for high LMA species to capitalize on N from deposition, high LMA species are likely to have a less sensitive (i.e., smaller magnitude) foliar N response to atmospheric N deposition. Second, shade tolerant trees invest proportionately more leaf N in light harvesting compounds than in the RuBP carboxylase (i.e., Rubisco) enzyme used to maximize carbon gain from foliar N input (*31*, *32*). This adaptive strategy causes shade tolerant species to have less plastic foliar N, and decreased potential for carbon gain in high resource environments (*31*). Because of this decreased potential for an investment of foliar N resulting in carbon gain, shade tolerant species are also likely to exhibit a less sensitive foliar N response to N deposition.

Thus, from this conceptual viewpoint of plant ecophysiology, and empirical view of the strong relationship observed in Figure 3, we find very promising support for the hypothesis that variability in species response to N deposition originates from fundamental differences in how species allocate N to different leaf constituents. Further testing this hypothesis suggests many future lines of research, and opens many novel research questions. For instance, we could expect an intermediate shade tolerant species that is investing comparatively higher amounts of N in Rubisco to receive a greater enhancement of photosynthetic production from N deposition relative to a more shade tolerant competitor. If so, these species differences in the leaf-level allocation of N may

provide a mechanism for N deposition to directly influence the community dynamics of temperate forest ecosystems. Whole-tree assessment of N allocation may also be important. Of particular interest here is the apparent *lack* of foliar N response in American beech (Figure 2). Since this widely abundant species does not appear to be increasing its foliar N in response to N deposition (Figure 2), we suggest that it is important to ask: how else might it be allocating the additional N made available by N deposition? To the extent that responses such as expanding leaf display or growing additional roots could not only be enabled by elevated N availability, but also fit within the growth strategy of this persistence-oriented, shade-tolerant species, the *lack* of foliar N response in American beech could suggest an alternative mechanism whereby N deposition could affect forest dynamics. Questions such as these merit attention with respect to predicting the future ecosystem effects of N deposition, as shifts in species composition can have important effects on the ability of temperate forest ecosystems to continue retaining N from atmospheric deposition (*26*, *33*).

Implications for Predicting Ecosystem Response to N Deposition. Much of our current knowledge regarding the response of forest ecosystems to N deposition is contained within the hypotheses of ecosystem N saturation (*5*, *11*). These hypotheses predict that ecosystem retention of N is ultimately driven by biotic demand, and that this demand may be best assessed through foliar analysis (*5*). Thus, the relationships between N deposition and foliar N presented here (Figure 2) add direct and regional-scale support to this central tenet of the N saturation hypotheses. Results from this study also extend the hypotheses of N saturation by indicating that use of foliar analysis to assess the effects of N deposition should be contextualized according to species differences in foliar N response. Finally, our analysis suggests that the expected degree of foliar N response may be characterized by two easily measurable factors, LMA and shade tolerance (Figure 3).

A species' LMA and shade tolerance characteristics arise from sub-leaf scale allocations of N. Yet, their constraint upon foliar N response to N deposition provides a promising approach to make regional-scale and long-term predictions of ecosystem response (e.g., N retention, carbon sequestration) to N deposition. The use of LMA and shade tolerance to predict species-level foliar N responses to N deposition may find valuable application toward regional analyses when combined with remotely obtained spatial data layers of whole-canopy foliar N (*20*, *34*, *35*) and species composition (*36*, *37*). For long-term projections of the effect of N deposition on ecosystem response and vegetation dynamics, the relationships presented in Figure 3 provide a quantitative basis

for including species responses to N deposition within lumped parameter ecosystem models (*38*) or individual-based gap-dynamic ecosystem models (*39, 40*). The successful application of these foliar N-centered remote sensing and ecosystem modeling approaches would enable forecasts of temperate forests' continued ability to retain excess atmospheric N and carbon (*41−44*) and thereby mitigate the myriad environmental problems caused by N pollution (*45*) and climate change. Given the sustained high rates of N emissions, and resultant N deposition to temperate forests in the U.S. (*46*) and worldwide (*47, 48*), such regional-scale and long-term forecasts are likely to become increasingly critical for the development of sound air pollution control policies.

Acknowledgments

Supporting Information Available

Further description and validation of the atmospheric N deposition model. This material is available free of charge via the Internet at http://pubs.acs.org.

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