ECOSYSTEMS

A New Conceptual Model of Nitrogen Saturation Based on Experimental Nitrogen Addition to an Oak Forest

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ABSTRACT

The dominant conceptual model of nitrogen (N) saturation in forests predicts the temporal patterns of key N cycling indicators as an initially N-limited forest is progressively enriched in N. We present the results from a long-term N addition experiment in an oak forest in southeastern New York State, USA, which do not conform to the predictions of the conceptual model in several ways. In contrast to the predictions of the conceptual model, the foliar N concentrations in the N-treated stands of our study increased to about 20% above the levels in the control stands and then remained essentially constant, and nitrogen leaching from the treated stands increased almost immediately after the start of the experiment, prior to the onset of elevated nitrification. Concentrations of N in soil solution of the N-treated stands peaked at over 150-fold greater than the concentrations in the control stands. There were no significant changes in potential net N mineralization. Tree mortality increased in the treated stands, but the tree mortality did not appear to be the primary cause of the excess nitrate leaching. Based on these results and those of other recent studies, we present a new conceptual model of the N saturation process focused on the mass balance of N rather than the temporal dynamics of N cycling indicators. The

mass balance is characterized by inputs of N from atmospheric deposition and fertilization, internal sinks in the vegetation and soils, and outputs to leaching and gaseous losses. The key points of the conceptual model are (1) added N can flow simultaneously to all sinks and losses in the system, (2) the fate of the added N and the temporal patterns of flow of N depend on the strength of the sinks and the factors that control them, and (3) the movement of N to the various sinks determines how N saturation is manifested in the ecosystem. We distinguish *capacity* N saturation, in which the sinks in the vegetation and soil are zero or negative, from *kinetic* N saturation, in which the sinks are positive but lower than the N input rate. The sink strengths in the vegetation and soil have two components, one due to carbon (C) accumulation in the system and the other due to change in the stoichiometry (C:N ratio) of the pool. Further work quantifying the magnitudes and controlling factors for the N sinks will allow better prediction of the dynamics of N saturation in different types of forested ecosystems.

Key words: nitrogen; saturation; forest; leaching; oak; nitrification; nitrogen mineralization; foliar nitrogen; fertilization.

INTRODUCTION

Elevated levels of atmospheric nitrogen (N) deposition are a problem throughout the developed world, and are increasingly becoming a problem in

developing countries (Galloway and Cowling 2002). Atmospheric N compounds emanating from both fossil fuel combustion and agricultural activities are transported downwind and may be deposited on ecosystems far from the source of emission (Vitousek and others 1997). This excess N deposition may have a wide range of effects on ecosystems, including: fertilization effects, nutrient imbalances, accumulation of N in plants, soils and sediments, enhanced losses of gaseous nitrogen oxides, and acidification of soils and surface waters caused by leaching of nitrate (Fenn and others 1998; Driscoll and others 2003).

Nitrogen saturation is the term used to describe the changes that occur in a terrestrial ecosystem as it is continually exposed to elevated levels of N deposition (Agren and Bosatta 1988; Aber and others 1989). The dominant conceptual model of N saturation was proposed by Aber and colleagues (Aber and others 1989, 1998) as a hypothetical series of changes that would occur as an N-limited ecosystem accumulates deposited N (Figure 1). In this model, the ecosystem is viewed as progressing through a series of stages of N saturation, in which stage 0 is a system that is strongly N limited, stage 1 is characterized by changes indicating increasing N sufficiency, stage 2 represents the initial symptoms of N saturation such as elevated N leaching, and stage 3 represents N-induced decline in which the productivity of the forest decreases and nitrate leaching accelerates (Aber and others 1998). As this sequence progresses, enhanced N deposition makes N available to N-limited plants, which enriches plant tissues and litter, the litter N is transferred to soil organic matter, stimulating N mineralization and nitrification, and eventually resulting in elevated nitrate leaching from the ecosystem. T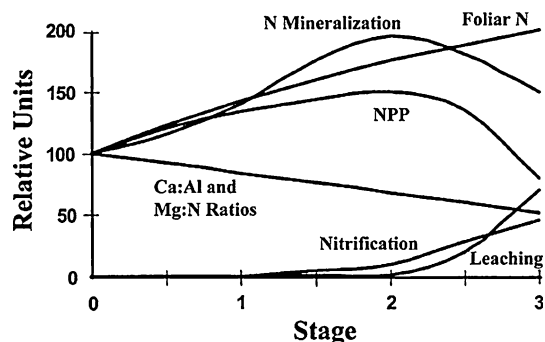he authors hypothesize a strong, continual increase in foliar N concentration through the progression of these stages, leading to an increase in net primary productivity (NPP) of the N-limited system, an initial increase and then a decrease in N mineralization, and decreasing ratios of calcium:aluminum and magnesium:nitrogen in the foliage. In stage 3, the system undergoes a decline in NPP and an increase in nitrate leaching. Though not explicitly diagrammed in Figure 1, Aber and others (1998) recognized that the major sink for added N in forest ecosystems is in the soil, and they described several potential mechanisms by which N could be incorporated in soil organic matter.

The fate of N added to an ecosystem is constrained by the N mass balance as expressed in the following equation

$$I - dN_v/dt - dN_s/dt = L + G \qquad (1)$$

where I = the N input rate to the system (deposition plus N amendments), dN_v/dt is the vegetation sink rate (the net rate of incorporation of N into vegetation), dN_s/dt is the soil sink rate (the net rate of incorporation of N into detritus and soil), L is the rate of leaching loss, and G is the rate of gaseous loss of N.

It is important to note that the mass balance in equation (1) is expressed in terms of rates, and that losses from the ecosystem can occur when the *rate* of N addition exceeds the *rate* at which N can be incorporated into the vegetation and soil sinks, even if the capacity of those sinks is not saturated. This suggests that we should distinguish *kinetic N saturation*, in which the rate of N input exceeds the vegetation and soil N net sink rates, from *capacity N saturation*, in which N has accumulated in the vegetation and soils to the point that further net N retention in these sinks does not occur. In kinetic N saturation, the vegetation and soil sinks are still active, though N losses will occur simultaneously with N retention. In capacity saturation, N retention in the vegetation and soils is negligible and system inputs should equal outputs. Both types of saturation are related to the accumulation of N in organic matter in the ecosystem. Capacity saturation occurs when the accumulation of N in the ecosystem is zero (or negative, in the case of a deteriorating system), and kinetic saturation occurs when the accumulation rate is positive but less than the N input rate. Capacity saturation is rarely observed in real forests unless disturbance reduces the vegetation sink rate or turns the vegetation into a net source of N within the system.

Ecosystem sinks for N are associated with a specific time frame, either implicitly or explicitly, and

Figure 1. Aber and others' (1998) hypothesized patterns of response of forest ecosystem properties to continuing N additions. Used with permission from Aber and others (1998).

the time frame determines which processes are important. For instance, incorporation of N into microbial biomass which then dies and is remineralized over several days, may represent a sink or source of N on an hourly or daily time scale, but does not contribute to N sinks evaluated over annual time scales. For the purposes of this paper, we define the time scales of interest for the vegetation and soil sinks in equation (1) to be on the order of years to decades or longer, because this scale represents a policy-relevant time frame for ecosystem sequestration of pollution-derived N.

The responses of forests to elevated rates of N deposition can be evaluated by several approaches, each of which has strengths and weaknesses. The most straightforward approach is long-term observation of forests subjected to N deposition (for example, Peterjohn and others 1996). However, this requires decades of measurements and may be confounded by simultaneous changes in other factors affecting the forests, such as climate or disturbance. A second approach is comparative or gradient studies, in which ecosystem properties are measured in ecosystems receiving different rates of N deposition (for example, McNulty and others 1991; Lovett and Rueth 1999; Boggs and others 2005; Thomas and others 2010). This eliminates the difficulties of long-term studies but introduces uncertainty caused by potential differences in the ecosystems caused by factors other than the rate of N deposition. In gradient studies, inferring the temporal sequence requires accepting the assumptions of the space-for-time substitution, that is, that processes that occur first in a temporal sequence also occur first along a gradient from low to high N deposition. A third approach is experimental studies, in which the rates of N deposition are enhanced by adding N to experimental plots (for example, Magill and others 1996, 2004; Wallace and others 2007). Careful pairing of experimental and control plots can minimize the influence of factors other than the level of N addition. However, because the experimental N addition rate is higher than ambient rates, these experiments run the risk of altering the processes controlling the fate of N in the ecosystem. In particular, experiments enhance kinetic saturation by increasing N input rates without necessarily increasing the N sink strengths in the ecosystem. These approaches can also be used in combination, for example, long-term experiments and experiments done across gradients.

The Aber and others' (1998) conceptual diagram (Figure 1) focuses on which ecosystem properties are expected to respond to elevated N deposition, and the temporal sequence in which they respond.

All three types of studies can be used to evaluate this conceptual model. Aber and others compared their conceptual model with results from several experimental studies in the eastern U.S. including studies in Massachusetts (Magill and others 2004), Vermont (McNulty and others 1996) and Maine (Magill and others 1996) as well as a gradient study in spruce-fir forests across northern New York and New England (McNulty and others 1991). In general the experimental responses agreed with the hypothesized trends, if one assumes that forest ecosystems start at different places along the N saturation continuum and may proceed at different rates, depending on the degree of initial N limitation induced mainly by past land use (Aber and others 1998).

However, not all studies have supported the hypotheses diagrammed in Figure 1. For example, in experimental studies in Europe at sites in which N was either experimentally added or removed, nitrogen leaching was often the first variable to respond to the treatment, rather than the last as suggested by Figure 1 (Emmett 2007). Recent studies across N deposition gradients in the eastern and midwestern U.S. have shown variable concordance with the hypotheses illustrated in Figure 1. Tree growth increases with N deposition for some tree species but not others (Thomas and others 2010). Foliar nitrogen sometimes increases along an N deposition gradient (Pardo and others 2006; McNeil and others 2007) and sometimes does not (Aber and others 2003). Likewise, net N mineralization and nitrification sometimes increase along a nitrogen deposition gradient (McNulty and others 1991; Aber and others 2003) and sometimes do not (Lovett and Rueth 1999). Across the Northeastern U.S., N leaching tends to be low in watersheds that receive less than about 8 kg N ha^{-1} y^{-1} of atmospheric N deposition (Aber and others 2003). For deposition rates above 8 kg N ha^{-1} y^{-1}, some sites show N leaching and some do not (Aber and others 2003). Whether these watersheds leach N or not is controlled in part by tree species composition, soil C:N ratio, and history of disturbance (Goodale and others 2000; Lovett and others 2002). It is often difficult to determine if the absence of trends in these gradient studies results from a true lack of response in the system, or an inability to discern the signal within the noise of spatial variation inherent in gradient studies. Overall, however, the literature suggests substantial variation in the degree to which forest ecosystems conform to the hypotheses expressed in Figure 1.

In this study, we tested the hypothetical series of changes proposed by Aber and others (1998) with

an N addition experiment in a mixed-oak forest in southeastern New York State, USA. The research site had similar vegetation to the Harvard Forest (Massachusetts) oak site used as one of the test sites for the conceptual model by Aber and others (1998) but was in a ridgetop rather than a valley location. Our goal was to determine if the temporal sequence of changes in key N cycling variables matched those illustrated in Figure 1, and if not, to propose a different conceptual model in better agreement with the observations.

METHODS

Research site

The research site is located at the Cary Institute of Ecosystem Studies (41.797°N, 73.734°W) in the Hudson Valley of southeastern New York State, USA. The forest is an upland mixed-oak woods dominated by red oak (*Quercus rubra* L.), chestnut oak (*Quercus prinus* L.) and several species of hickory (*Carya* sp.) regenerated from selective cutting associated with use as a woodlot early in the twentieth century (Glitzenstein and others 1990). The annual average temperature at the site is 9.6°C and precipitation averages 111 cm y^{-1}. The bedrock is mostly shale and slate and soils are predominantly thin and well-drained silt loams of the Nassau and Woodlawn series (Glitzenstein and others 1990) classified as lithic dystrudepts. Mean annual precipitation pH at the site ranged from 4.0 to 4.4 between 1984 and 2004 (Kelly and others 2002). Ambient inorganic N deposition (wet + dry) in the period of study (1996–2006) averaged approximately 9 kg N ha^{-1} y^{-1}. Wet deposition was collected continuously with a wet-only precipitation collector, and dry deposition was estimated from continuous filter-pack measurements of air concentrations of HNO_3 vapor and particulate NO_3^- and NH_4^+, applying a dry deposition model parameterized with meteorological data at the site (Kelly and others 2002).

We chose 6 pairs of plots, each 20 m in diameter. The two plots of each pair are within 40 m of each other. Pre-treatment measurements were made in July–August 1996, and the N addition began in November 1996. One plot of each pair was treated with granular NH_4NO_3 fertilizer at a rate equivalent of 100 kg N ha^{-1} y^{-1} from 1996 to 1999, and then the N addition rate was decreased to 50 kg N ha^{-1} y^{-1} from 2000 to 2006. This annual application was divided into 4 equal doses each year. Fertilizer was applied to the forest floor; although application to the canopy would have

been more realistic, it would be technically much more difficult, and prior data from these forests indicate that canopy uptake of ambient N deposition is negligible (Lovett and others 2000a).

Sample Collection and Processing

Foliar chemistry was monitored on full-sun leaves of the dominant oak trees in the plot collected with a shotgun annually in late July or early August. Potential net nitrogen mineralization and nitrification were measured annually or biennially in laboratory assays using a 14-day aerobic incubation following the methods detailed in Lovett and Rueth (1999). Soils for these assays were collected in July using 8 cores per plot (2.5-cm diameter, 10-cm deep) composited into 4 samples per plot, separated into 0–3 and 3–10 cm depth increments. The top 3 cm corresponds to the average depth of the forest floor at the sites; thus the 0–3 cm sample approximates the organic horizon and the 3–10 cm sample represents the top 7 cm of the mineral soil (typically a B horizon). We separated the cores by measured depth rather than by soil horizon to provide more consistent sampling from year to year, given that multiple individuals sampled the plots over the course of this long-term study. Soils were sieved through an 8-mm screen and incubated at constant temperature (20°C) and moisture (60% of field capacity). Soils were extracted at the beginning and end of the incubations using 2 M KCl (10 g moist soil/100 ml of KCl). Soil solution was collected using low-tension soil lysimeters installed in the lower B horizon (average depth ~40 cm), below the predominant rooting zone. A constant tension of 0.1 atm (10 kPa) was maintained on the lysimeters using a hanging column system as described by Johnson and Lindberg (1992). Our experience in these sites indicates that zero-tension lysimeters collect soil water unreliably, and that these constant, low-tension lysimeters provide a reasonable compromise between collection efficiency and obtaining a fraction of soil water that represents potential leachate. Samples were collected bi-weekly, stored at 4° C with chloroform as a preservative, and pooled into monthly samples for analysis. Leaching N fluxes were estimated from monthly concentrations and monthly water flow estimated from the Brook90 hydrologic model (Federer 1995), parameterized with data from the Cary Institute weather station approximately 800 m from the plots. Pools of C and N in the soil were measured using quantitative excavation of soil pits (20 × 20 cm, 4 pits per plot) in 2006.

Tree growth was measured by repeated diameter measurements of all trees larger than 5 cm diameter on every plot. Wood N concentration was measured on tree core samples taken in 2004 (Wallace and others 2007). Percent C and N were analyzed on segments of the cores representing the pre-treatment period (1989–1995) and the treatment period (1996–2004).

Chemical Analysis

Soil solution nitrate was measured using a Dionex ICS-2000 ion chromatograph, soil solution NH_4^+ and soil extract NH_4^+ and NO_3^- were measured with a Lachat QuickChem 8000 FIA Ion Analyzer, NH_4^+ using the phenate method, and NO_3^- using the cadmium reduction method. Soil solution total N was measured as NO_3^- on the Lachat Ion Analyzer after persulfate oxidation. Carbon and N concentrations in foliage, wood, and soil were measured on dried and ground samples using a CE Elantech Flash EA1112 Elemental Analyzer.

RESULTS

The patterns of response of this forest to long-term N addition (Figure 2) differ in some important ways from those hypothesized in the existing conceptual model of N saturation (Figure 1). To facilitate comparison with the hypotheses, we plot the temporal trends in key N cycling variables in Figure 2 as ratios of treatment plots/control plots. The N concentration in the foliage increased shortly after the experiment began as predicted by Figure 1, but the increase was only about 10–20% above control plot values and was essentially flat after the second year of treatment, with a decline in 2005, the last year for which we have measurements (Figures 2A, 3). This stands in contrast to Figure 1, in which foliar N is hypothesized to continue rising and nearly double in concentration. The hypotheses diagrammed in Figure 1 indicate that the N mineralization rates should increase initially and then decrease, whereas in our study the potential N mineralization showed no trend through the study (Figure 2B). The mean N mineralization rate in the treated plots was about 50% higher than in the control plots, but that difference existed before the treatments began (summer 1996), and the treated and control plots were not significantly different from one another in any year. [We note that these are net N mineralization rates, and in some cases gross N mineralization can be stimulated in N saturated sites without concomitant increases in net rates (Tietema 1998). In
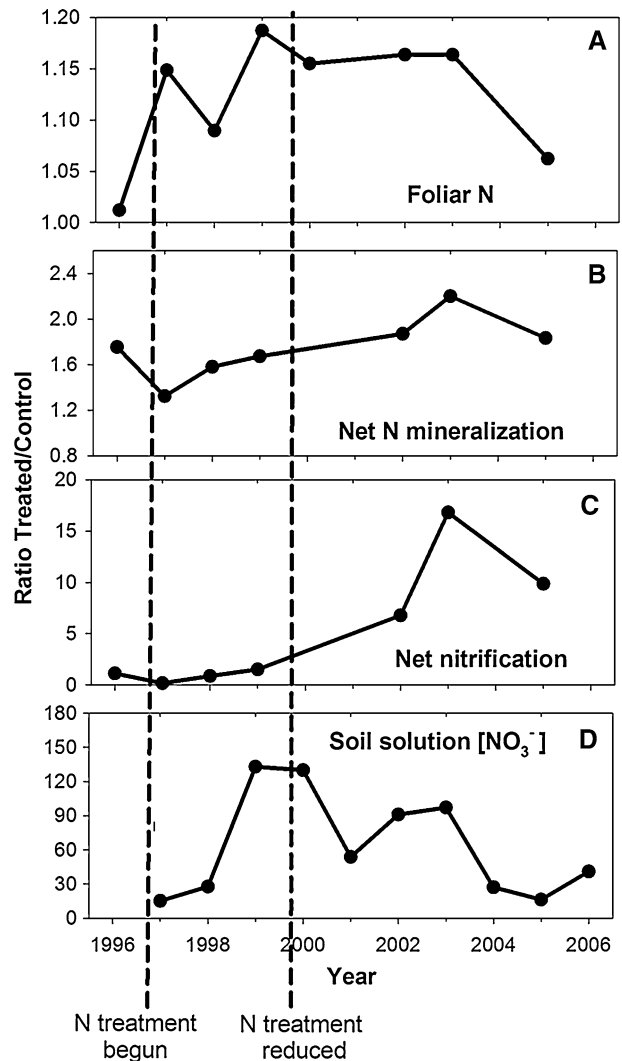

Figure 2. Temporal patterns of forest response to N addition in an oak forest in southeastern New York State. Values represent the ratio of the mean of treated plots to the mean of the untreated plots. **A** Foliar N concentration, **B** potential net N mineralization, **C** potential net nitrification, **D** soil solution NO_3^- concentration (B horizon).

other cases, gross rates are unaffected by N additions (for example, Venterea and others 2004; Christenson and others 2009).] In Figure 1, nitrification rate is hypothesized to increase in the late stages of N saturation, followed by an increase in NO_3^- leaching. In our study, potential nitrification rate in the N-treated plots was significantly elevated over the control plots by year 6 (2002) (Figures 2C, 4). However, NO_3^- leaching, expressed as concentration of NO_3^- in soil solution, increased well before nitrification did. Nitrate leaching in treated plots was significantly higher than controls in the second year of treatment and
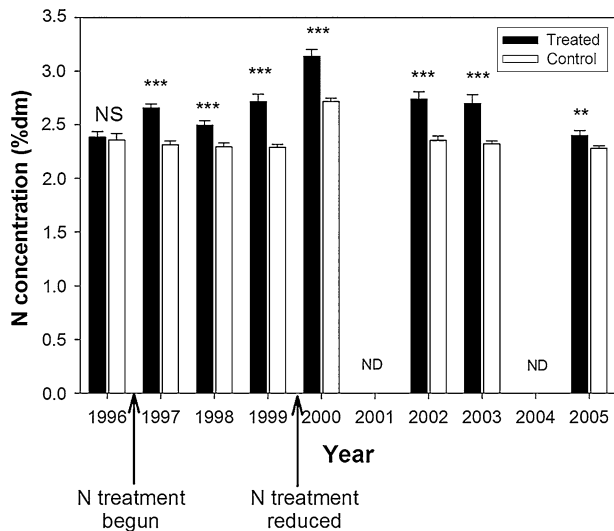
Figure 3. Nitrogen concentration (% dry mass) measured in sunlit oak foliage on the study plots. *NS* not significant, **$P < 0.01$, ***$P < 0.001$.

rose to almost 150-fold greater than the control by year 3 (1999) (Figure 2D). Estimated N (inorganic + organic) leaching fluxes reached almost 50 kg N ha^{-1} y^{-1} in 1999 (Figure 5), after which the N application rate was reduced from 100 to 50 kg N ha^{-1} y^{-1}. Leaching fluxes remained at high levels (10–40 kg N ha^{-1} y^{-1}), though they varied from year to year (Figure 5), primarily depending on water flow through the soil.

The control plots showed very little N leaching and retained 88% of the approximately 9 kg N ha^{-1} y^{-1} added to the site from ambient N deposition. By contrast, the treated plots retained only 65% of N inputs from deposition and the N amendments (Table 1). Ammonium and dissolved organic N (DON) were a significant fraction of the total N leaching in the control plots (25% and 38%, respectively) but were relatively much less important (2 and 6%, respectively) in the treated plots because of the elevated NO$_3^-$ concentrations (Table 1). Mass balance calculations of N inputs, hydrologic losses, and tree N accumulation indicate that about 55% of the added N was retained in the soil in the treated plots during the 8 years of the study. (This budget assumes that N gas losses (NH$_3$, NO, N$_2$O, N$_2$) are negligible in these acidic and well-aerated soils. We are in the process of testing this assumption.) In the untreated plots, however, the measured atmospheric deposition was insufficient to supply even the N accumulated in the wood over the study period, indicating that N was probably extracted from the soil N pool by the plants. Our best estimate is that this soil N "mining" activity totaled about 15 kg N ha^{-1} over the 8 years (Table 1).

Although these N budget calculations indicate incorporation of N into the soil in the treated plots and mining of N in the untreated plots, we observed no significant treatment effect on soil %N, C:N ratio or soil C or N pools (Table 2), probably because the soil pools are large and spatially variable, with expected changes in these pools
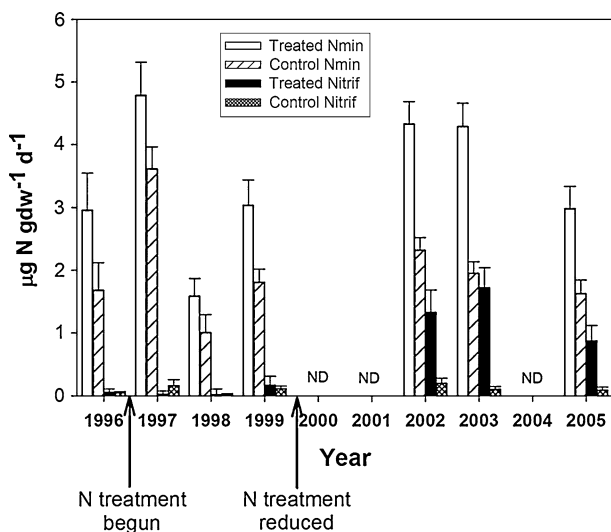

Figure 4. Mean rates of potential net N mineralization (Nmin) and nitrification measured in the study plots in the 0–3 cm depth samples. Error bars are standard errors. *ND* no data. For the 3–10 cm depth, the patterns were similar but the rates were lower (data not shown).
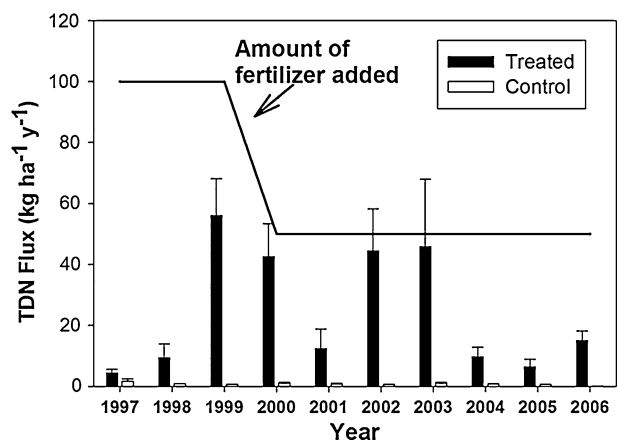

Figure 5. N leaching flux (inorganic + organic N) in treated and control plots based on measured soil solution concentrations and modeled water flows. *Solid line* shows rate of fertilizer addition to the treated plots. All plots also received an average of 8.6 kg N ha^{-1} y^{-1} from wet + dry atmospheric deposition.

Table 1. Plot N Budgets for the Period 1997–2004

		Treated	Untreated
Inputs	Atmospheric deposition (wet + dry)	69	69
	Fertilizer N added	550	0
Change in pools	N increment in wood (aboveground)	64	76
	Estimated N increment in soil[1]		
	0–3 cm	26	0
	3–10 cm	83	0
Outputs N Leaching	NH_4^+	4	2
	NO_3^-	200	3
	DON	12	3
	Total	216	8
Sum inputs		619	69
Sum outputs + pool increments		389	84
Difference (gaseous loss or change in subsoil pool)		230	−15
Retention % (= 100*(inputs − outputs)/inputs)		65%	88%

Values are cumulative means of treated and untreated plots, in kg N ha^{-1} for the entire 8-year period.
[1]Soil N pools were not measured in 1996. These calculations assume that the soil N increment was 0 in the untreated plots, and that the N pool in the untreated plots represents the pre-treatment (1996) pool in the treated plots. Thus the difference between the treated and untreated plots is the estimate of the N accumulation in the soil of the treated plots.

Table 2. Concentrations and Pools of C and N in Wood and Soils

	Treated (mean (SE))	Untreated (mean (SE))
Wood		
%N (1997–2004)	0.30 (0.03)[1]	0.22 (0.01)[1]
N pool 2004 (kg N ha^{-1})	409.8 (98.8)	543.6 (105.6)
C pool 2004 (kg C ha^{-1})	98,390 (28,609)	123,961 (17,180)
N increment (kg N ha^{-1} y^{-1})	8.0 (1.5)	9.5 (1.5)
C increment (kg C ha^{-1} y^{-1})	1359 (285)	1891 (251)
Soil (0–3 cm)		
%N	0.83 (0.09)	0.73 (0.03)
%C	15.8 (1.8)	14.3 (1.0)
C:N (mass)	18.9 (0.3)	19.6 (0.9)
N pool (kg N ha^{-1})	921 (48)	896 (67)
C pool (kg C ha^{-1})	17,356 (820)	17,319 (921)
Soil (3–10 cm)		
%N	0.37 (0.04)	0.37 (0.03)
%C	6.6 (0.6)	7.0 (0.4)
C:N (mass)	18.1 (0.4)	19.0 (0.6)
N pool (kg N ha^{-1})	1297 (81)	1214 (90)
C pool (kg C ha^{-1})	23,442 (1309)	22,827 (120)
Soil total (0–10 cm)		
N pool (kg N ha^{-1})	2219 (119)	2109 (152)
C pool (kg C ha^{-1})	40,798 (1876)	40,145 (1908)

Values are means of 6 plots, with standard error in parentheses. %N in wood is from wood accrued during the N treatment period. N and C increments in wood are based on sampling in 1996 and 2004. Wood pools and increments include both live and standing dead trees on the plot, and include only aboveground wood. Soil depth intervals are measured from the surface; 0–3 cm approximates the O horizon, and 3–10 cm approximates the top 7 cm of the mineral horizon.
[1]%N in wood was significantly higher in treated plots than in untreated plots (paired t-test, P = 0.015); for the other variables in the table there was no significant treatment effect.

smaller than the precision with which they can be measured. We did observe a significant increase in wood N concentration in the treated plots (Table 2). However, because of the tree mortality in the treated plots (see Wallace and others 2007 and discussion below), the increased N concentration did not result in a significantly elevated N pool in wood (live + standing dead) in the treated plots (Table 2). Similarly, C pools in wood did not differ significantly between treatments.

DISCUSSION

The data from this site do not entirely support the general conceptual model diagrammed in Figure 1, in which enhanced N deposition is taken up by N-limited plants and incorporated into their tissues, and then transferred to soil organic matter as plant litter, stimulating mineralization, nitrification, and then N leaching. We suggest that the amount of N that can flow through this plant–litter–soil pathway in mature forests is often quite limited because of both the restricted stoichiometric plasticity of most plants, and constraints on productivity imposed by stand age and the availability of light, water and other nutrients. The limited stoichiometric plasticity of these trees is illustrated by the foliar N concentration, which increased by a maximum of 20%, and then did not rise further, despite continued N additions. Other N addition studies in the U.S. and Europe have reported similar levels of foliar N enrichment (Gundersen and others 1998; Magill and others 2004), but some N-amended pine and spruce stands in the eastern U.S. have shown foliar N concentrations up to 100% greater than controls (Magill and others 2004, McNulty and others 2005).

The reduction in N retention efficiency in the N-treated plots indicates that the N retention mechanism in the soil was overwhelmed or inhibited by the N additions. Nitrogen retention in forest soils may occur abiotically or microbially (Johnson and others 2000; Dail and others 2001; Fitzhugh and others 2003). Microbial immobilization of added N may be low in soils with a C:N ratio below about 20, assuming that microbes are about 40–50% efficient and have C:N ratios of around 8–10 (Cleveland and Liptzin 2007). Moreover, microbial immobilization of NO_3^- can be suppressed by NH_4^+ additions (Betlach and others 1981) and this has been suggested as the mechanism by which N leaching has responded quickly to N treatments in other field experiments (Moldan and others 2006; Emmett 2007).

After 2001, several of the treated plots showed elevated tree mortality, primarily of red oaks, and most likely associated with soil acidification due to the NO_3^- leaching (Wallace and others 2007). This onset of ecosystem decline is consistent with stage 3 of the Aber and others (1998) N saturation model, and has been observed in some N-amended conifer stands in the eastern U.S. (Magill and others 2004; McNulty and others 2005) but had not previously been observed in hardwood stands. We do not believe that the decline in tree vigor precipitated the increase in NO_3^- leaching, for two reasons.

First, elevated NO_3^- leaching began in year 2 of the experiment, well before the tree mortality was observed (Figure 5). Second, the capacity for wood N storage by healthy trees (estimated from the untreated plots) could account for only about 12% of the N inputs to the treated plots (Table 1), thus reduction in tree uptake would have made only a minor difference in N leaching.

Other lines of evidence also suggest inconsistencies between the conceptual model illustrated in Figure 1 and experimental results. For instance, field studies in which a ^{15}N tracer is added to forest plots consistently show that the forest floor, rather than the vegetation, is the strongest initial sink for the added N (for example, Nadelhoffer and others 1999; Templer and others 2005). Experimental N addition and exclusion studies have shown that N leaching is often one of the first variables to respond to N manipulations, rather than the last (Emmett 2007), and cross-site comparisons have shown N leaching to be one of the most sensitive response variables across gradients of N deposition (for example, Aber and others 2003). Aber and others (1998) discussed the importance of soil N retention in controlling N leaching and evaluated several possible mechanisms by which N could be incorporated into SOM without first flowing through plant biomass, but these mechanisms are not included in the dynamic responses illustrated in Figure 1.

Rethinking the Conceptual Model

The conceptual model illustrated in Figure 1 focuses on the temporal dynamics of N saturation, particularly the sequential passage of N from plant to litter to soil. Although this conceptual view of the process may be sufficient in some cases, we do not believe it is general enough to explain the results of our study or other recent experimental studies. Our data show that the added N can reach all three measured fates (vegetation, soils, and leaching) simultaneously (we do not have data on gaseous losses). This makes it very difficult to predict temporal patterns of response of various parts of the system (as in Figure 1), because the patterns depend on the relative strength of the sinks, rather than the timing of transfer between one sink and the next. To move our predictive capacity forward, we need to consider the sink strengths within the context of the underlying mass balance of N in the forest ecosystem, as expressed in equation (1).

Our conceptual model of the N saturation process is diagrammed in Figure 6, which shows that N added to a forest ecosystem can flow simulta-
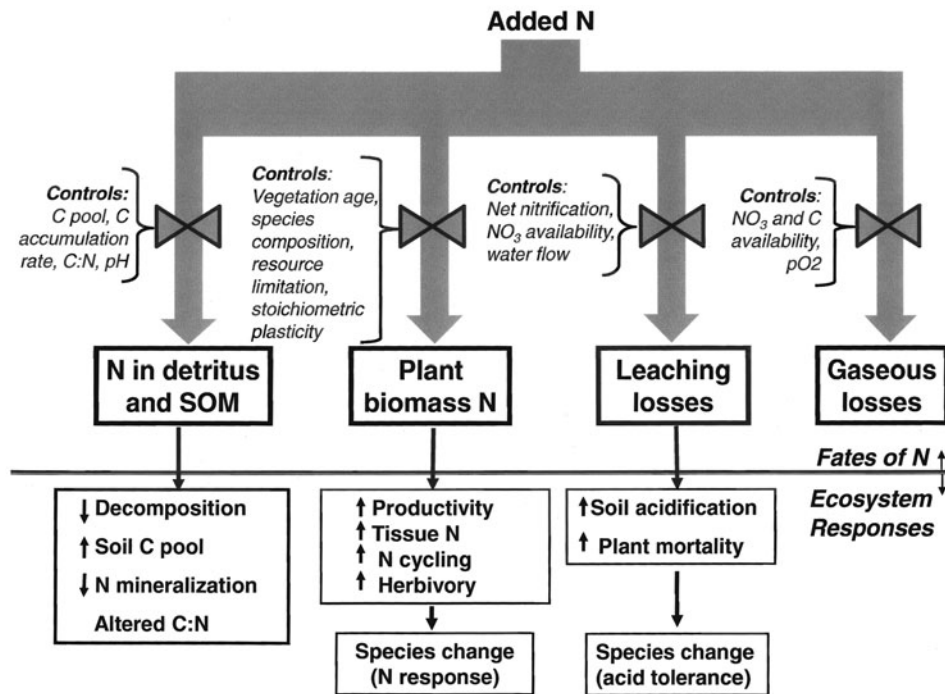
Figure 6. Conceptual model of N saturation in an ecosystem, illustrating N flow to various fates (internal sinks and losses from the ecosystem), examples of controlling factors, and ecosystem responses associated with N flow to those fates. Detritus includes the forest floor and woody debris, and SOM represents soil organic matter.

neously to four possible fates: incorporation into vegetation, incorporation into to the detritus and soil organic matter, leaching loss, or gaseous loss. The rate of movement to any of these fates is variable among ecosystems and controlled by properties of the ecosystem, some of which are well understood based on decades of research on the N cycle, and others of which are poorly understood.

In contrast to Figure 1, this diagrammatic model emphasizes (1) the multiple possible fates of N in the system, (2) the controls on movement of N to these fates, and (3) the variable ecosystem responses depending on which sinks or loss processes receive the added N. The temporal pattern of response of the system to N saturation (of which Figure 1 represents one possible example) will depend on the extent to which N flows to these various fates, each of which is associated with specific responses (Figure 6). For example, nitrate leaching can cause soil acidification, root damage, and tree mortality, and can lead to species change in favor of species tolerant of acid conditions (McNulty and others 2005; Hogberg and others 2006; Wallace and others 2007). Nitrogen accumulation in the forest floor and soil can reduce the activity of some microbial enzymes used in decomposition and N mineralization (Carreiro and others 2000; DeForest and others 2004; Sinsabaugh and others 2005), decrease decomposition rates of late-stage or lignin-rich litter or older soil organic matter (Berg 2000; Neff and others 2002; Knorr

and others 2005), cause an increase in soil carbon stocks (Hyvonen and others 2008; Pregitzer and others 2008; Nave and others 2009; Liu and Greaver 2010), and either increase or decrease the C:N ratio of the forest floor, depending on the balance between C accumulation and N accumulation. Nitrogen accumulation in biomass may lead to increased productivity, increased herbivory, increased N cycling from plant to forest floor, altered root:shoot ratios, and species change in favor of nitrophilic species (for example, Stevens and others 2004; Throop and Lerdau 2004; Hyvonen and others 2008; Thomas and others 2010). Species changes are likely to be most evident in ecosystems that have plants with short generation times (for example grasslands, Stevens and others 2004), whereas in forests such species changes may initially be most evident in herbaceous or understory species rather than the more long-lived trees (Gilliam 2006). At any particular site, whether and when specific effects are manifested depends on the movement of N into the pathway that causes those effects. This conceptual model, though very simple, explicitly incorporates the known sinks and losses for N, allowing better explanation of recent experimental evidence and providing a focus for future research. Expressing N saturation effects in this way allows us to focus on when and why systems may differ in their response to N additions, rather than expecting a uniform pattern of temporal response as suggested by Figure 1.

Estimating Sink Strengths

Although the sink strengths shown in Figure 6 are simple to diagram, they can be difficult to measure. Leaching losses are typically measured either by sampling soil solution chemistry and estimating water flow below the rooting zone with a hydrologic model, or by using stream water flux in gauged small watersheds (Lajtha 2000). Gaseous losses are typically measured in small chambers placed on the soil, but the rates are highly uncertain because of high spatial and temporal variability in the nitrification and denitrification processes that produce the gases, and the difficulty in measuring N_2 flux, which can be a major component of the total gaseous flux (Groffman and others 2006; Kulkarni and others 2008).

The vegetation and soil sinks represent the accumulation of N within the system, and each can be thought of as having two components: (1) a *carbon accumulation sink*, in which N is accumulated with C in a constant C:N ratio, and (2) a *stoichiometric sink*, in which N is accumulated without corresponding C accumulation by changing the C:N ratio of the organic matter. The two types of sinks can occur separately or together. In combination, the C accumulation sink and the stoichiometric sink form the total capacity for N accumulation within the vegetation or soil. The well-known hypothesis put forth by Vitousek and Reiners (1975) predicting that N export should be low in ecosystems with high net ecosystem production, is essentially a statement about the effectiveness of the C accumulation sink for N in ecosystems.

N Sink in Vegetation

The N sink associated with C accumulation in forest vegetation can be quite variable depending on species composition and growth rate of the stand. The untreated oak forest in our study accumulated N in woody biomass at a rate of 9.5 kg N ha^{-1} y^{-1} (Table 1). A similar oak stand at Harvard Forest in central Massachusetts accumulated woody biomass N at a rate of 7.1 kg N ha^{-1} y^{-1} over a 9-year period, whereas a nearby pine stand accumulated only 1.9 kg N ha^{-1} y^{-1} (Magill and others 2000). Forests that are growing rapidly can be expected to have a substantial C accumulation sink for N. For example, a young pine plantation in North Carolina accumulated N at a rate of 5–23 kg N ha^{-1} y^{-1}, depending on the year, with a 4-year average of 15 kg N ha^{-1} y^{-1} (Finzi and others 2002). Reiners (1992) reported that in the first 10 years of regrowth of a northern hardwood forest at Hubbard Brook, NH, following experimental deforestation, the forest accumulated N in biomass at a rate of 9.2 kg N ha^{-1} y^{-1}. In contrast, in a nearby 90-year-old forest at Hubbard Brook, the C accumulation sink for N is essentially zero because there has been no measurable C accumulation in aboveground biomass for approximately the past 25 years (Fahey and others 2005).

If forest growth is limited by N, the sink for N can be expected to increase with N fertilization. For example, in the Harvard Forest oak stand amended with 150 kg N ha^{-1} y^{-1}, the N accumulation increased to 13 kg N ha^{-1} y^{-1} (compared to 7.1 in the control stand) due to both increased C accumulation and increased N concentration in wood (Magill and others 2000). However, the N accumulation in the N-amended pine stand decreased to 1.1 kg N ha^{-1} y^{-1} (compared to 1.9 in the control pine stand) due to mortality of the trees (Magill and others 2000), similar to the decline seen in the present study (Table 1). Thomas and others (2010) report that N deposition enhances aboveground C accumulation in Northeastern U.S. forests by an average of 61 kg C ha^{-1} y^{-1} for each 1 kg ha^{-1} y^{-1} increase in N deposition rate. Thus an increase in N deposition of 8 kg N ha^{-1} y^{-1} (for example, from pre-industrial times to present in this area) would increase C sequestration by 488 kg C ha^{-1} y^{-1}, and at a C:N ratio of 250 (typical for woody biomass) this would correspond to an enhanced N sink of about 2 kg N ha^{-1} y^{-1}.

Vegetation can also produce a stoichiometric sink for N if plant tissue N concentrations change, for example, if they increase with N fertilization. However, the limited ability of plant tissues to use or absorb excess N may make this sink small and short-lived. The stoichiometric component of the sink strength (N accumulation rate) would decline to near zero when the concentration in tissues stabilizes, as occurred in the foliage of our study by year 3 of the experiment. Increases in wood and root N concentration can also provide a stoichiometric sink.

N Sink in Detritus and Soil

The forest floor and mineral soil can also have both C accumulation and stoichiometric sinks. A forest stand that is recovering from a disturbance such as harvesting or fire could have a significant N sink associated with the accumulation of C in the forest floor. For instance, in a century of forest regrowth after agricultural abandonment in Rhode Island, USA, Hooker and Compton (2003) reported a relatively rapid accumulation of N in the forest floor (11.6 kg N ha^{-1} y^{-1}). However, this forest floor

accumulation was roughly balanced by the decrease in N in the mineral soil (-12.1 kg N ha^{-1} y^{-1}) suggesting that the trees ''mine'' the mineral soil N and deposit it on the surface as plant detritus, producing a redistribution of N within the ecosystem rather than a net sink (Hooker and Compton 2003). Berg and Dise (2004) found N accumulation rates in humus ranging from 1.5 to 2.5 kg N ha^{-1} y^{-1} for coniferous forests in Sweden that ranged from 120 to 2900 years since the last disturbance. After the forest floor C pool comes to steady state, the principal N sink would be the stoichiometric sink associated with changes in forest floor C:N ratios.

In mineral horizons the slow rate of C accumulation as soil organic matter should produce a long-term sink for N. For example, if post-agricultural soils accumulate mineral soil organic matter at a rate of 100–300 kg C ha^{-1} y^{-1} (for example, Gaudinski and others 2000; Post and Kwon 2000) with a C:N ratio of 15–20, they would accumulate 5–20 kg N ha^{-1} y^{-1}. On an even longer time scale, Emmett and Reynolds (1996) estimated that soil under a spruce forest in Wales accumulated N at an average rate of 2.1 kg N ha^{-1} y^{-1} since the last glaciation, calculated by dividing the current soil N pool by the estimated 11,500 years that have elapsed since deglaciation. The C accumulation rate is a key factor governing the N accumulation rate in soil, but the ability of soils to retain added N has also been suggested to be related to the C pool size (Evans and others 2006) and the C:N ratio (Lovett and others 2002; MacDonald and others 2002) (Figure 6), both of which probably influence the ability of the soil to retain N through a stoichiometric sink. Changes in mineral soil C:N ratio can produce stoichiometric N sinks that may be slow in rate but high in capacity because of the large mass of soil C.

Measurement of the soil sinks is particularly problematic because soil pools of C and N are large and have high spatial variability, making a small difference resulting from N accumulation very difficult to detect by direct sampling (Huntington and others 1988). Other ways of estimating the soil N sink include N budget calculations in which the sink for N is estimated indirectly as the residual in a mass budget of N, and isotopic tracer methods, in which trace amounts of the stable isotope ^{15}N are added to plots and the amount of N incorporated into the soil is measured over time (for example, Nadelhoffer and others 2004; Templer and others 2005). In our Cary Institute site, despite the fact that our budget suggests that a substantial portion of the added N is retained in the soil, we observed no significant difference in soil C or N pools (to 10 cm depth) nor in C:N ratio between control and N-treated plots (Table 2), indicating that any C accumulation or stoichiometric sinks in the soil are not detectable through the noise of spatial variation. This illustrates that soil measurements may not be precise enough to observe what appears to be a major term in the budget. A similar situation was observed at the Harvard Forest N addition experiment (for example, Magill and others 2000), although other studies have been able to detect increases in C and N pools in the soil over the course of long-term N addition experiments (for example, Moldan and others 2006; Hyvonen and others 2008; Pregitzer and others 2008). We discuss the use of ^{15}N tracer measurements in the section ''Comparison of the Conceptual Model with Observations from Isotopic Tracers'' below.

Estimating Sinks in This Study

In our study, we can calculate some of the important sink strengths but not others. The N sink strength in aboveground vegetation was 9.5 kg N ha^{-1} y^{-1} in the untreated plots, and we interpret this as entirely due to a C accumulation sink because the wood N concentration did not change significantly during the study. In the treated plots the wood N increment was 8.0 kg N ha^{-1} y^{-1}. The wood C increment was 1359 kg C ha^{-1} y^{-1} and the C:N ratio of the wood prior to the treatment was 260, so we estimate that, of the 8.0 kg N ha^{-1} y^{-1} total sink, the C accumulation sink provided 5.2 kg N ha^{-1} y^{-1} or 65%, and the stoichiometric sink (caused by increasing wood N concentrations) contributed the remainder (2.8 kg N ha^{-1} y^{-1}). This calculation does not include any N storage in belowground woody tissues. Belowground woody biomass often averages roughly 20% of aboveground (Cairns and others 1997; Jenkins and others 2004); if belowground woody biomass accumulates in similar proportion to aboveground, woody roots might represent an additional sink of roughly 1.6 and 1.9 kg N ha^{-1} y^{-1} in the treated and untreated plots, respectively.

Calculation of the soil sinks is difficult because we do not have pre-treatment estimates of C or N pools in the soil, a common problem (for example, Pregitzer and others 2008; Hyvonen and others 2008). The sinks for the 0–10 cm layer of soil are estimated in Table 1 by assuming that the N increment in soil in the untreated plots during the 8-year experimental period was 0, and that the difference in mean N pools between the treated and untreated plots represents N accumulation in soil due to the N treatment. Although the mean soil N

pool in treated plots was about 5% higher than in the untreated plots, this difference was not statistically significant, therefore the calculation is highly uncertain. Nonetheless, this calculation suggests an N sink of about 14 kg N ha^{-1} y^{-1} in the top 10 cm of the soil.

The imbalance in the N budget (the "difference" term in Table 1) indicates that there was an additional sink of about 29 kg N ha^{-1} y^{-1} in the treated plots that must be the result of some combination of gaseous loss and N incorporation into the subsoil. We suspect that this imbalance is largely a result of N accumulation in the soil below 10 cm depth because the plots are well drained and therefore are probably not conducive to denitrification. Nitrogen accumulation in the subsoil could occur through turnover of roots or soil microbes, or by adsorption of dissolved organic N compounds. At the observed C:N ratio of about 18, it would require soil C storage of about 500 kg C ha^{-1} y^{-1} to produce this N sink if it were purely a C accumulation sink. This amount of soil C storage is large but not inconceivably so. Alternatively a strictly stoichiometric sink would decrease the soil C:N ratio to around 16, and some combination of C accumulation and stoichiometric sinks is likely. An N sink in the coarse woody debris, which we did not measure, is also a possibility.

Our N-treated plots are clearly undergoing *kinetic* N saturation, because the N sinks in the vegetation and soil are still active, but the total sink strength is exceeded by the input rate, so that N is also leaching from the system. Thus N is flowing simultaneously to the soil, vegetation and leaching pathways in Figure 6, and perhaps to the gaseous loss pathway as well, although we do not have measurements of gaseous loss to evaluate this possibility. By the second year of the study, we observed multiple ecosystem effects of N saturation, including N leaching (which led to soil acidification, Wallace and others 2007), increases in foliar N which led to fertilization responses (Wallace and others 2007), and retention of N in the soil, which we calculate from the budget but were unable to detect with field sampling because of the high spatial variability of soil N pools.

In comparison, for the first 9 years of the Harvard Forest N addition experiment (Magill and others 2000), there was only an average of 1.3 kg N ha^{-1} y^{-1} of N leaching in the hardwood low-N plot (the plot that was most similar to ours in vegetation and N addition rate), compared to 27 kg N ha^{-1} y^{-1} in our N-treated plots. The woody biomass increment was 7.2 kg N ha^{-1} y^{-1} compared to 8.0 in our plots, and the soil sink, calculated as the residual in the N budget, averaged 39 kg N ha^{-1} y^{-1} compared to 29 in our plots. This comparison suggests that the soil N sink at Harvard Forest was strong enough to prevent kinetic saturation at that site, whereas at our site, with its weaker soil sink, the inputs exceeded the total sink strength of the system, resulting in kinetic saturation and high levels of NO$_3^-$ leaching.

Comparison of the Conceptual Model with Observations from Isotopic Tracers

Several different stable isotopic approaches, including natural (^{18}O–NO$_3^-$) and introduced (^{15}N) tracers, have been used to make inferences about N saturation by providing information on the sources and fate of newly added N over various timescales. (In N saturation studies, natural abundance ^{15}N measurements have been most often used as qualitative indices of N saturation and ecosystem openness (for example, Pardo and others 2006) rather than for determination of quantitative sources and fates of N.) For example, atmospheric processes generate NO$_3^-$ that is far more enriched in ^{18}O than is the NO$_3^-$ produced by microbial nitrification, so that measurements of the natural abundance of ^{18}O–NO$_3^-$ in soil- and streamwater allows partitioning of the relative contributions of these two NO$_3^-$ sources (Kendall and others 2007). Most measurements of δ^{18}O–NO$_3^-$ in forested catchments point to microbial nitrification as the primary source of stream NO$_3^-$, even in catchments with moderately high NO$_3^-$ deposition and export (for example, Spoelstra and others 2001; Williard and others 2001; Burns and Kendall 2002; Pardo and others 2004; Piatek and others 2005; Sebestyen and others 2008; Goodale and others 2009). At first glance, the lack of an atmospheric signature in NO$_3^-$ from streamwater of high N deposition watersheds might appear contradictory to our conceptual model in which kinetic saturation of the soil N sink may lead to increased leaching of atmospherically deposited N. However, it is important to consider the time scales involved. Every "sink" has an implicit or explicit time scale, and we define the sinks in Figure 6 as those capable of sequestering N from years to decades or longer. Yet any microbial cycling of atmospheric N, no matter how rapid, that occurs in the ecosystem prior to its export as NO$_3^-$ will yield stream δ^{18}O–NO$_3^-$ values reflecting a microbial rather than an atmospheric source. Hence, the prevalence of the microbial signal of exported ^{18}O–NO$_3^-$ probably indicates rapid short-term microbial cycling (on times scales of hours to months) without accumulation in microbial biomass

or soil organic matter over time scales of a year or longer. Thus, the fast N cycling that sets the ^{18}O signature of NO_3^- does not necessarily reflect the activity of longer-term sinks.

It is also worth noting that many streamwater ^{18}O–NO_3^- studies detect increased contributions from atmospheric NO_3^- during snowmelt (Spoelstra and others [2001]; Pardo and others [2004]; Sebestyen and others [2008]; Goodale and others [2009]) or other high-flow events (Williard and others [2001]; Burns and Kendall [2002]) and in forest stands that are in decline (Durka and others [1994]). These responses are consistent with our conceptual model, in that periods of high flow are times when N is being delivered to vegetation and soil faster than it can be retained, thus increasing the leaching loss. Forests in decline would have a diminished or nonexistent N sink in biomass, also leading to increased leaching of atmospheric N.

Temporal scaling issues can also make tracer ^{15}N studies challenging to interpret. Using a ^{15}N tracer to measure the sink strength of soils or plants requires that the recovery of the tracer be measured on the temporal scales over which the sink is evaluated. Clearly, evaluation of sinks operating on annual to decadal time scales requires measurement of the recovery of the tracer on those same time scales (for example, Nadelhoffer and others [2004]), and shorter-term studies may be misleading.

Spatial scaling issues can also become important in interpreting ^{15}N tracer studies, especially if pools are not well mixed. For instance, one approach to the vexing problem of quantifying the soil N sink strength is to measure the rate of accumulation in the SOM of ^{15}N from an applied tracer. But the ''gross'' N retention measured by the tracer is not equal to the ''net'' N retention of N by the entire soil profile if the N immobilization at the soil surface indicated by ^{15}N retention is balanced in part by N mineralized and lost from deeper, unlabeled soil N pools. Only when the tracer has had time to mix thoroughly with the entire SOM pool will the tracer measurement provide an accurate picture of N retention in the soil sink. This same challenge applies to plant uptake, which may occur from both the soil surface where most ^{15}N tracers are applied, and from deeper mineralized N. Although ^{15}N tracers are powerful tools for discerning the fate of newly added N over a particular timescale, they provide less information on net ecosystem N balance.

Despite the probable over-estimation of soil sink strengths, several ^{15}N tracer studies in forests with accompanying fertilization experiments support the idea that as the N addition rate increases, the proportion of the added ^{15}N label that is retained in plant and soil pools declines (Tietema and others [1998]; Templer and others [2005]). This is consistent with kinetic saturation of those sinks leading to increased losses by leaching and/or gaseous efflux.

Critical Loads of N

Critical loads are defined as the amount of deposition of a substance that can be tolerated by an ecosystem without incurring harm, and are used in Europe and Canada as an assessment tool for determining emissions targets for S and N pollution (Burns and others [2008]). One of the most easily recognized and widely accepted indicators of harm from N deposition is excess N leaching into groundwater or surface water, and critical loads are often set to minimize the amount of N leaching (Bobbink and others [2003]; Fenn and others [2008]). In equation ([1]), the critical load (CL) for NO_3^- leaching would be the input rate (I) for which the leaching rate (L) is at some acceptable level (L_a), or:

$$CL = dN_v/dt + dN_s/dt + G + L_a \qquad (2)$$

This mass balance model is commonly used to calculate the critical load for N (for example, Reynolds and others [1998]). As a static mass balance may not be adequate if the system is changing over time, it is often preferable to implement the mass balance within a dynamic model to predict the temporal changes in N saturation and critical loads (for example, Langusch and Matzner [2002]; Davis and others [2008]; Fenn and others [2008]). Nonetheless, Figure [6] illustrates that progress in predicting critical loads for forests can be made by focusing studies on calculating these sink strengths and determining what factors cause them to vary among sites and over time. Most useful would be the development of quantitative relationships between these sink strengths and readily measured ecosystem properties that can be used to predict them, such as the variables listed as controlling factors in Figure [6]. In addition, the conceptual model in Figure [6] emphasizes the point that not all effects of N deposition are caused by N leaching. Even if deposition rates are low enough to allow all deposited N to be accommodated by the sink strengths in vegetation, soils and gaseous losses, there may nonetheless be ecological effects resulting from the accumulation of N in vegetation and soils. As mentioned above, accumulation of N in plant tissue can change plant growth rates, cause nutrient imbalances, and increase herbivory, whereas accumulation of N in soil can alter microbial processes, reduce late-stage

decomposition rates, and change plant species composition (Figure 6). As a result, multiple indicators of N saturation should be used together, including indicators of vegetation and soil N status (for example, Fenn and others 2008).

Experimental N Additions and Ambient N Deposition

Experimental N addition studies, such as the one reported here, provide realistic field settings with good control of potentially confounding factors, but the elevated input rates may cause changes in the partitioning of added N within the system. For this reason, experimental studies should be used with caution in predicting the effects of N saturation and determining critical loads in systems receiving ambient deposition. In experiments with artificially elevated N inputs, kinetic N saturation will be exacerbated relative to ecosystems receiving lower, ambient rates of N deposition. However, even in ecosystems receiving ambient N deposition, kinetic N saturation is much more common than capacity saturation, because most forests have some ability to retain N in soil or vegetation sinks. (Capacity saturation occurs only if the sinks for N in the system are zero or negative). Thus the N addition experiments represent a more extreme case of the type of N saturation that occurs under ambient conditions. Capacity and kinetic factors can interact; for instance, N retention rates in vegetation and soils may slow as these pools approach their capacity, thus altering the balance between inputs and retention rates, which determines kinetic saturation. Further, the effects of kinetic saturation may influence C accumulation in the ecosystem, thus affecting capacity saturation. For example, nitrate-induced cation leaching may reduce forest growth and thus the capacity of the vegetation to retain N. Thus the complex interplay between kinetic and capacity factors can be influenced by the artifacts of an N addition experiment.

From Conceptual to Predictive Models

Our conceptual model is broadly applicable to ambient deposition and N addition studies because it is based on a fundamental mass balance of N. The three key points of the conceptual model— (1) that added N flows to four principal sinks and loss processes in the ecosystem, and may flow to all of these fates simultaneously, (2) that the fate of added N and the temporal patterns of N flow depend on the strength of the sinks and loss processes and the factors that control them, and (3) the movement of N to the various sinks determines how N saturation is manifested in the ecosystem —are applicable to any terrestrial ecosystem receiving N deposition.

Moving from general conceptual understanding to predictive capacity is challenging, however, and is limited by incomplete understanding of the factors that regulate the movement of N to various fates, particularly the soil/detritus sink and gaseous loss. Our knowledge of the competition among sinks within an ecosystem is also rudimentary. For instance, we know that systems with rapid accretion of C in vegetation or high C:N ratios in the soil are likely to have strong vegetation and soil N sinks, respectively, which reduce leaching losses (Vitousek and Reiners 1975; Dise and Wright 1995). One is tempted to assume, as many ecosystem models do, that losses by leaching and gaseous efflux occur only if the N input rates exceed the retention rates of soil and vegetation. Yet studies across gradients of ambient N deposition indicate that NO_3^- leaching begins to increase at deposition rates at least as low as those that produce measurable changes in soil and vegetation (for example, Aber and others 2003), and organic N leaching can be substantial in ecosystems with little NO_3^- leaching (Lovett and others 2000b; Perakis and Hedin 2002). This suggests that some losses may be unavoidable, and that N inputs can flow to all fates, including leaching and gaseous loss, even at relatively low levels of N deposition.

The key to predicting N saturation is understanding the N sinks and loss processes in the ecosystem and the factors that regulate them. Both comparative and experimental studies should focus on measuring rates and regulation of these sinks and loss processes. With that knowledge, it should be possible to predict the types of N saturation impacts that are likely in particular ecosystems and the rates of N deposition that would cause those impacts.

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REFERENCES

Aber JD, Nadelhoffer KJ, Steudler P, Melillo JM. 1989. Nitrogen saturation in northern forest ecosystems. Bioscience 39:378–86.

Aber J, McDowell W, Nadelhoffer K, Magill A, Berntson G, Kamakea M, McNulty S, Currie W, Rustad L, Fernandez I. 1998. Nitrogen saturation in temperate forest ecosystems: hypotheses revisited. Bioscience 48:921–34.

Aber JD, Goodale CL, Ollinger SV, Smith ML, Magill AH, Martin ME, Hallett RA, Stoddard JL. 2003. Is nitrogen deposition altering the nitrogen status of northeastern forests? Bioscience 53:375–89.

Agren GI, Bosatta E. 1988. Nitrogen saturation of terrestrial ecosystems. Environ Pollut 54:185–97.

Berg B. 2000. Litter decomposition and organic matter turnover in northern forest soils. For Ecol Manage 133:13–22.

Berg B, Dise N. 2004. Calculating the long-term stable nitrogen sink in northern European forests. Acta Oecol 26:15–21.

Betlach MR, Tiedje JM, Firestone RB. 1981. Assimilatory nitrate uptake in Pseudomonas fluorescens studied using N-13. Arch Microbiol 129:135–40.

Bobbink R, Ashmore MR, Fluckiger W, Van den Wyngaert IJJ. 2003. Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update. In: Achermann B, Bobbink R, Eds. Empirical critical loads for nitrogen, expert workshop. Zurich: Swiss Agency for the Environment, Forests and Landsape (SAEFL). p 43–170.

Boggs JL, McNulty SG, Gavazzi MJ, Myers JM. 2005. Tree growth, foliar chemistry, and nitrogen cycling across a nitrogen deposition gradient in southern Appalachian deciduous forests. Can J For Res 35:1901–13.

Burns DA, Kendall C. 2002. Analysis of delta N-15 and delta O-18 to differentiate NO_3^- sources in runoff at two watersheds in the Catskill Mountains of New York. Water Resour Res 38. doi:10.1029/2001wr000292.

Burns DA, Blett T, Haeuber R, Pardo LH. 2008. Critical loads as a policy tool for protecting ecosystems from the effects of air pollutants. Front Ecol Environ 6:156–9.

Cairns MA, Brown S, Helmer EH, Baumgardner GA. 1997. Root biomass allocation in the world's upland forests. Oecologia 111:1–11.

Carreiro MM, Sinsabaugh RL, Repert DA, Parkhurst DF. 2000. Microbial enzyme shifts explain litter decay responses to simulated nitrogen deposition. Ecology 81:2359–65.

Christenson LM, Lovett GM, Weathers KC, Arthur MA. 2009. The influence of tree species, nitrogen fertilization, and soil C to N ratio on gross soil nitrogen transformations. Soil Sci Soc Am J 73:638–46.

Cleveland CC, Liptzin D. 2007. C:N:P stoichiometry in soil: is there a ''Redfield ratio'' for the microbial biomass? Biogeochemistry 85:235–52.

Dail DB, Davidson EA, Chorover J. 2001. Rapid abiotic transformation of nitrate in an acid forest soil. Biogeochemistry 54:131–46.

Davis SC, Hessl AE, Thomas RB. 2008. A modified nitrogen budget for temperate deciduous forests in an advanced stage of nitrogen saturation. Global Biogeochem Cycles 22:12.

DeForest JL, Zak DR, Pregitzer KS, Burton AJ. 2004. Atmospheric nitrate deposition, microbial community composition, and enzyme activity in northern hardwood forests. Soil Sci Soc Am J 68:132–8.

Dise NB, Wright RF. 1995. Nitrogen leaching from European forests in relation to nitrogen deposition. For Ecol Manage 71:153–61.

Driscoll CT, Whitall D, Aber J, Boyer E, Castro M, Cronan C, Goodale CL, Groffman P, Hopkinson C, Lambert K, Lawrence G, Ollinger S. 2003. Nitrogen pollution in the northeastern United States: sources, effects, and management options. Bioscience 53:357–74.

Durka W, Schulze ED, Gebauer G, Voerkelius S. 1994. Effects of forest decline on uptake and leaching of deposited nitrate determined from 15N and 18O measurements. Nature 372:765–7.

Emmett BA. 2007. Nitrogen saturation of terrestrial ecosystems: some recent findings and their implications for our conceptual framework. Water Air Soil Pollut Focus 7:99–109.

Emmett BA, Reynolds B. 1996. Nitrogen critical loads for spruce plantations in Wales: Is there too much nitrogen? Forestry 69:205–14.

Evans CD, Reynolds B, Jenkins A, Helliwell RC, Curtis CJ, Goodale CL, Ferrier RC, Emmett BA, Pilkington MG, Caporn SJM, Carroll JA, Norris D, Davies J, Coull MC. 2006. Evidence that soil carbon pool determines susceptibility of semi-natural ecosystems to elevated nitrogen leaching. Ecosystems 9:453–62.

Fahey TJ, Siccama TG, Driscoll CT, Likens GE, Campbell J, Johnson CE, Battles JJ, Aber JD, Cole JJ, Fisk MC, Groffman PM, Hamburg SP, Holmes RT, Schwarz PA, Yanai RD. 2005. The biogeochemistry of carbon at Hubbard Brook. Biogeochemistry 75:109–76.

Federer CA. 1995. Brook 90: a simulation model for evaporation, soil water, and streamflow. Version 3.25. Durham, NH: USDA Forest Service.

Fenn ME, Poth MA, Aber JD, Baron JS, Bormann BT, Johnson DW, Lemly AD, McNulty SG, Ryan DF, Stottlemeyer R. 1998. Nitrogen excess in North American ecosystems: predisposing factors, ecosystem responses and management strategies. Ecol Appl 8:706–33.

Fenn ME, Jovan S, Yuan F, Geiser L, Meixner T, Gimeno BS. 2008. Empirical and simulated critical loads for nitrogen deposition in California mixed conifer forests. Environ Pollut 155:492–511.

Finzi AC, DeLucia EH, Hamilton JG, Richter DD, Schlesinger WH. 2002. The nitrogen budget of a pine forest under free air CO_2 enrichment. Oecologia 132:567–78.

Fitzhugh RD, Lovett GM, Venterea RT. 2003. Biotic and abiotic immobilization of ammonium, nitrite, and nitrate in soils developed under different tree species in the Catskill Mountains, New York, USA. Global Change Biol 9:1591–601.

Galloway JN, Cowling EB. 2002. Reactive nitrogen and the world: 200 years of change. Ambio 31:64–71.

Gaudinski JB, Trumbore SE, Davidson EA, Zheng SH. 2000. Soil carbon cycling in a temperate forest: radiocarbon-based estimates of residence times, sequestration rates and partitioning of fluxes. Biogeochemistry 51:33–69.

Gilliam FS. 2006. Response of the herbaceous layer of forest ecosystems to excess nitrogen deposition. J Ecol 94:1176–91.

Glitzenstein JS, Canham CD, McDonnell MJ, Streng DR. 1990. Effects of environment and land-use history on upland forests

of the Cary Arboretum, Hudson Valley, NY. Bull Torrey Bot Club 117:106–22.

Goodale CL, Aber JD, McDowell WH. 2000. The long-term effects of disturbance on organic and inorganic nitrogen export in the White Mountains, New Hampshire. Ecosystems 3:433–50.

Goodale CL, Thomas SA, Fredriksen G, Elliott EM, Flinn KM, Butler TJ, Walter MT. 2009. Unusual seasonal patterns and inferred processes of nitrogen retention in forested headwaters of the Upper Susquehanna River. Biogeochemistry 93:197–218.

Groffman PM, Altabet MA, Bohlke JK, Butterbach-Bahl K, David MB, Firestone MK, Giblin AE, Kana TM, Nielsen LP, Voytek MA. 2006. Methods for measuring denitrification: diverse approaches to a difficult problem. Ecol Appl 16:2091–122.

Gundersen P, Emmett BA, Kjonaas OJ, Koopmans CJ, Tietema A. 1998. Impact of nitrogen deposition on nitrogen cycling in forests: a synthesis of NITREX data. For Ecol Manage 101: 37–55.

Hogberg P, Fan HB, Quist M, Binkley D, Tamm CO. 2006. Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest. Global Change Biol 12:489–99.

Hooker TD, Compton JE. 2003. Forest ecosystem carbon and nitrogen accumulation during the first century after agricultural abandonment. Ecol Appl 13:299–313.

Huntington TG, Ryan DF, Hamburg SP. 1988. Estimating soil-nitrogen and carbon pools in a northern hardwood forest ecosystem. Soil Sci Soc Am J 52:1162–7.

Hyvonen R, Persson T, Andersson S, Olsson B, Agren GI, Linder S. 2008. Impact of long-term nitrogen addition on carbon stocks in trees and soils in northern Europe. Biogeochemistry 89:121–37.

Jenkins JC, Chojnacky DC, Heath LS, Birdsey RA. 2004. Comprehensive database of diameter-based biomass regressions for North American tree species. Newtown Square, PA: USDA Forest Service, Northeastern Research Station.

Johnson DW, Lindberg SE. 1992. Atmospheric deposition and nutrient cycling in forest ecosystems. New York: Springer-Verlag.

Johnson DW, Cheng W, Burke IC. 2000. Biotic and abiotic nitrogen retention in a variety of forested soils. Soil Sci Soc Am J 64:1503–14.

Kelly VR, Lovett GM, Weathers KC, Likens GE. 2002. Trends in atmospheric concentration and deposition compared to regional and local pollutant emissions at a rural site in southeastern New York, USA. Atmos Environ 36:1569–75.

Kendall C, Elliott EM, Wankel SD. 2007. Tracing anthropogenic inputs of nitrogen to ecosystems. In: Michener RM, Lajtha K, Eds. Stable isotopes in ecology and environmental science. Oxford: Blackwell. p 375–449.

Knorr M, Frey SD, Curtis PS. 2005. Nitrogen additions and litter decomposition: a meta-analysis. Ecology 86:3252–7.

Kulkarni MV, Groffman PM, Yavitt JB. 2008. Solving the global nitrogen problem: it's a gas!. Front Ecol Environ 6:199–206.

Lajtha K. 2000. Ecosystem nutrient balance and dynamics. In: Sala OE, Jackson RB, Mooney HA, Howarth RW, Eds. Methods in ecosystem science. New York: Springer. p 249–64.

Langusch JJ, Matzner E. 2002. Long-term modelling of nitrogen turnover and critical loads in a forested catchment using the INCA model. Hydrol Earth Syst Sci 6:395–402.

Liu LL, Greaver TL. 2010. A global perspective on belowground carbon dynamics under nitrogen enrichment. Ecol Lett 13:819–28.

Lovett GM, Rueth H. 1999. Soil nitrogen transformations in beech and maple stands along a nitrogen deposition gradient. Ecol Appl 9:1330–44.

Lovett GM, Traynor MM, Pouyat RV, Carreiro MM, Zhu WX, Baxter JW. 2000a. Atmospheric deposition to oak forests along an urban-rural gradient. Environ Sci Technol 34:4294–300.

Lovett GM, Weathers KC, Sobczak WV. 2000b. Nitrogen saturation and retention in forested watersheds of the Catskill Mountains, New York. Ecol Appl 10:73–84.

Lovett GM, Weathers KC, Arthur MA. 2002. Control of N loss from forested watersheds by soil carbon:nitrogen ratio and tree species composition. Ecosystems 5:712–18.

MacDonald JA, Dise NB, Matzner E, Armbruster M, Gundersen P, Forsius M. 2002. Nitrogen input together with ecosystem nitrogen enrichment predict nitrate leaching from European forests. Global Change Biol 8:1028–33.

Magill AH, Downs MR, Nadelhoffer KJ, Hallett RA, Aber JD. 1996. Forest ecosystem response to four years of chronic nitrate and sulfate additions to Bear Brooks Watershed, Maine, USA. For Ecol Manage 84:29–37.

Magill AH, Aber JD, Berntson GM, McDowell WH, Nadelhoffer KJ, Melillo JM, Steudler P. 2000. Long-term nitrogen additions and nitrogen saturation in two temperate forests. Ecosystems 3:238–53.

Magill AH, Aber JD, Currie WS, Nadelhoffer KJ, Martin ME, McDowell WH, Melillo JM, Steudler P. 2004. Ecosystem response to 15 years of chronic nitrogen additions at the Harvard Forest LTER, Massachusetts, USA. For Ecol Manage 196:7–28.

McNeil BE, Read JM, Driscoll CT. 2007. Foliar nitrogen responses to elevated atmospheric nitrogen deposition in nine temperate forest canopy species. Environ Sci Technol 41:5191–7.

McNulty SG, Aber JD, Boone RD. 1991. Spatial changes in forest floor and foliar chemistry of spruce-fir forests across New England. Biogeochemistry 14:13–29.

McNulty SG, Aber JD, Newman SD. 1996. Nitrogen saturation in a high-elevation spruce-fir stand. For Ecol Manage 84:109–21.

McNulty SG, Boggs J, Aber JD, Rustad L, Magill A. 2005. Red spruce ecosystem level changes following 14 years of chronic N fertilization. For Ecol Manage 219:279–91.

Moldan F, Kjonaas OJ, Stuanes AO, Wright RF. 2006. Increased nitrogen in runoff and soil following 13 years of experimentally increased nitrogen deposition to a coniferous-forested catchment at Gardsjon, Sweden. Environ Pollut 144:610–20.

Nadelhoffer K, Emmett B, Gundersen P, Kjonaas O, Koopmans C, Schleppi P, Tietema A, Wright R. 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 398:145–8.

Nadelhoffer KJ, Colman BP, Currie WS, Magill A, Aber JD. 2004. Decadal-scale fates of N-15 tracers added to oak and pine stands under ambient and elevated N inputs at the Harvard Forest (USA). For Ecol Manage 196:89–107.

Nave LE, Vance ED, Swanston CW, Curtis PS. 2009. Impacts of elevated N inputs on north temperate forest soil C storage, C/N, and net N-mineralization. Geoderma 153:231–40.

Neff JC, Townsend AR, Gleixner G, Lehman SJ, Turnbull J, Bowman WD. 2002. Variable effects of nitrogen additions on the stability and turnover of soil carbon. Nature 419:915–17.

Pardo LH, Kendall C, Pett-Ridge J, Chang CCY. 2004. Evaluating the source of streamwater nitrate using delta N-15 and delta O-18 in nitrate in two watersheds in New Hampshire, USA. Hydrol Process 18:2699–712.

Pardo LH, Templer PH, Goodale CL, Duke S, Groffman PM, Adams MB, Boeckx P, Boggs J, Campbell J, Colman B, Compton J, Emmett B, Gundersen P, Kjonaas J, Lovett G, Mack M, Magill A, Mbila M, Mitchell MJ, McGee G, McNulty S, Nadelhoffer K, Ollinger S, Ross D, Rueth H, Rustad L, Schaberg P, Schiff S, Schleppi P, Spoelstra J, Wessel W. 2006. Regional assessment of N saturation using foliar and root delta N-15. Biogeochemistry 80:143–71.

Perakis SS, Hedin LO. 2002. Nitrogen loss from unpolluted South American forests mainly via dissolved organic compounds. Nature 415:416–19.

Peterjohn WT, Adams MB, Gilliam FS. 1996. Symptoms of nitrogen saturation in two central Appalachian hardwood forest ecosystems. Biogeochemistry 35:507–22.

Piatek KB, Mitchell MJ, Silva SR, Kendall C. 2005. Sources of nitrate in snowmelt discharge: evidence from water chemistry and stable isotopes of nitrate. Water Air Soil Pollut 165:13–35.

Post WM, Kwon KC. 2000. Soil carbon sequestration and land-use change: processes and potential. Global Change Biol 6:317–27.

Pregitzer KS, Burton AJ, Zak DR, Talhelm AF. 2008. Simulated chronic nitrogen deposition increases carbon storage in Northern Temperate forests. Global Change Biol 14:142–53.

Reiners WA. 1992. 20 years of ecosystem reorganization following experimental deforestation and regrowth suppression. Ecol Monogr 62:503–23.

Reynolds B, Wilson EJ, Emmett BA. 1998. Evaluating critical loads of nutrient nitrogen and acidity for terrestrial systems using ecosystem-scale experiments (NITREX). For Ecol Manage 101:81–94.

Sebestyen SD, Boyer EW, Shanley JB, Kendall C, Doctor DH, Aiken GR, Ohte N. 2008. Sources, transformations, and hydrological processes that control stream nitrate and dissolved organic matter concentrations during snowmelt in an upland forest. Water Resour Res 44. doi:10.1029/2008wr006983.

Sinsabaugh RL, Gallo ME, Lauber C, Waldrop MP, Zak DR. 2005. Extracellular enzyme activities and soil organic matter dynamics for northern hardwood forests receiving simulated nitrogen deposition. Biogeochemistry 75:201–15.

Spoelstra J, Schiff SL, Elgood RJ, Semkin RG, Jeffries DS. 2001. Tracing the sources of exported nitrate in the turkey lakes watershed using N-15/N-14 and O-18/O-16 isotopic ratios. Ecosystems 4:536–44.

Stevens CJ, Dise NB, Mountford JO, Gowing DJ. 2004. Impact of nitrogen deposition on the species richness of grasslands. Science 303:1876–9.

Templer PH, Lovett GM, Weathers KC, Findlay SE, Dawson TE. 2005. Influence of tree species on forest nitrogen retention in the Catskill Mountains, New York, USA. Ecosystems 8:1–16.

Thomas RQ, Canham CD, Weathers KC, Goodale CL. 2010. Increased tree carbon storage in response to nitrogen deposition in the US. Nat Geosci 3:13–17.

Throop HL, Lerdau MT. 2004. Effects of nitrogen deposition on insect herbivory: implications for community and ecosystem processes. Ecosystems 7:109–33.

Tietema A. 1998. Microbial carbon and nitrogen dynamics in coniferous forest floor material collected along a European nitrogen deposition gradient. For Ecol Manage 101:29–36.

Tietema A, Emmett BA, Gundersen P, Kjonaas OJ, Koopmans CJ. 1998. The fate of N-15-labelled nitrogen deposition in coniferous ecosystems. For Ecol Manage 101:19–27.

Venterea RT, Groffman PM, Verchot LV, Magill AH, Aber JD. 2004. Gross nitrogen process rates in temperate forest soils exhibiting symptoms of nitrogen saturation. For Ecol Manage 196:129–42.

Vitousek PM, Reiners WA. 1975. Ecosystem succession and nutrient retention: a hypothesis. Bioscience 25:376–81.

Vitousek PM, Aber JD, Howarth RW, Likens GE, Matson PA, Schindler DW, Schlesinger WH, Tilman DG. 1997. Human alteration of the global nitrogen cycle: sources and consequences. Ecol Appl 7:737–50.

Wallace ZP, Lovett GM, Hart JE, Machona B. 2007. Effects of nitrogen saturation on tree growth and death in a mixed-oak forest. For Ecol Manage 243:210–18.

Williard KWJ, DeWalle DR, Edwards PJ, Sharpe WE. 2001. O-18 isotopic separation of stream nitrate sources in mid-Appalachian forested watersheds. J Hydrol 252:174–88.