Ecosystems (2000) 3: 238–253
DOI: 10.1007/s100210000023

ECOSYSTEMS

Long-Term Nitrogen Additions and Nitrogen Saturation in Two Temperate Forests

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Abstract

This article reports responses of two different forest ecosystems to 9 years (1988–96) of chronic nitrogen (N) additions at the Harvard Forest, Petersham, Massachusetts. Ammonium nitrate (NH_4NO_3) was applied to a pine plantation and a native deciduous broad-leaved (hardwood) forest in six equal monthly doses (May–September) at four rates: control (no fertilizer addition), low N (5 g N m^{-2} y^{-1}), high N (15 g N m^{-2} y^{-1}), and low N + sulfur (5 g N m^{-2} y^{-1} plus 7.4 g S m^{-2} y^{-1}). Measurements were made of net N mineralization, net nitrification, N retention, wood production, foliar N content and litter production, soil C and N content, and concentrations of dissolved organic carbon (DOC) and nitrogen (DON) in soil water. In the pine stand, nitrate losses were measured after the first year of additions (1989) in the high N plot and increased again in 1995 and 1996. The hardwood stand showed no significant increases in nitrate leaching until 1995 (high N only), with further increases in 1996. Overall N retention efficiency (percentage of added N retained) over the 9-year period was 97–100% in the control and low N plots of both stands, 96% in the hardwood high N plot, and 85% in the pine high N plot. Storage in aboveground biomass, fine roots, and soil extractable pools accounted for only 16–32% of the added N retained in the amended plots, suggesting that the one major unmeasured pool, soil organic matter, contains the remaining 68–84%. Short-term redistribution of ^{15}N tracer at natural abundance levels showed similar division between plant and soil pools. Direct measurements of changes in total soil C and N pools were inconclusive due to high variation in both stands. Woody biomass production increased in the hardwood high N plot but was significantly reduced in the pine high N plot, relative to controls. A drought-induced increase in foliar litterfall in the pine stand in 1995 is one possible factor leading to a measured increase in N mineralization, nitrification, and nitrate loss in the pine high N plot in 1996.

Key words: ammonium nitrate; biomass production; foliar chemistry; net mineralization; net nitrification; nitrogen deposition; nitrogen saturation; soil solution chemistry.

Introduction

N deposition was first proposed as a possible threat to forest ecosystems nearly 15 years ago (Nihlgård 1985). Since that time, a number of coordinated studies in Europe under the NITREX and EXMAN programs (Wright and van Breemen 1995; Wright and Rasmussen 1998) and individual experiments in the US (for example, Christ and others 1995; Magill and others 1996, 1997; McNulty and others 1996; Peterjohn and others 1996; Fenn and others 1998; Norton and Fernandez 1999) have provided much of the data necessary for an increased under-

Received 2 April 1999; Accepted 29 October 1999.
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standing of the process of N saturation. In a recent article (Aber and others 1998), the results of European and North American studies were summarized into an inclusive theory where N saturation is defined as a series of nonlinear changes in key ecosystem processes in response to elevated nitrogen inputs. The primary indicators of N saturation include increases in nitrate leaching losses, net nitrification rates, and foliar N content, and an initial increase and subsequent decline in both net nitrogen mineralization and net primary productivity. Observation and measurement of such changes can be used to estimate the stage of N saturation, with each stage being characterized by a set of ecosystem response variables (see Aber and others 1989, 1998).

In contrast to the similarities observed among the N deposition experiments mentioned above is a significant amount of variation in the sensitivity of different forest types to N deposition. For example, it appears that conifer stands move through the stages of N saturation more rapidly than broadleaved deciduous forests (Stoddard 1994; Aber and others 1998). Where a forest lies within the continuum of N saturation is a function of several factors including community type, soil properties (especially C:N ratios; Gundersen and others 1998a; Gundersen and others 1998b), land-use history, and the rate and duration of N loading (Aber and others 1998). Still, little is known about the mechanisms that drive N retention and control the rate at which N saturation proceeds.

The Chronic Nitrogen Amendment experiment at the Harvard Forest in central Massachusetts (USA) is one of the longest running N addition experiments to date and has provided some of the essential data for understanding ecosystem responses to N additions (Aber and others 1993; Castro and others 1995; Currie and others 1996; Magill and others 1997; McDowell and others 1998; Magill and Aber 1998, Berntson and Aber forthcoming; Nadelhoffer and others 1999a; Yano and others 1998). The comparison of two forest types that have markedly different sensitivities to N deposition, coupled with an experimental gradient of N addition rates, make the Chronic N Amendment experiment an excellent platform for elucidating the mechanisms of N immobilization in response to N deposition.

This article summarizes 9 years (1988–96) of forest ecosystem N cycling, N and C storage, and forest productivity in the Chronic Nitrogen Amendment experiment. The long-term record of N fluxes and storage allowed us to examine several important questions including: (a) how does interannual climatic variability, including extreme events, such as drought, influence the ontogeny of N saturation; (b) can we discern whether rates of N deposition or cumulative N deposition have a greater impact on the progression of N saturation?; (c) do different forest types show a convergence or a divergence in responses to long-term N deposition?

METHODS

Study Site and Experimental Design

The Chronic Nitrogen Addition experiment is located at the Harvard Forest in central Massachusetts (42°30'N, 72°10'W) and is one of the core experiments of the Harvard Forest Long-Term Ecological Research (LTER) program. Two forest stands were studied: a 70+-year-old red pine (*Pinus resinosa* Ait.) plantation (planted in 1926) and a 50+-year-old mixed hardwood stand. The hardwood stand is dominated by black and red oak (*Quercus velutina* Lam.; *Q. rubra* L.) with varying amounts of black birch (*Betula lenta* L.), red maple (*Acer rubrum* L.), and American beech (*Fagus grandifolia* Ehrh.). The dominant soil types are stony to sandy loams, classified as Typic Dystrochrepts according to soil pit descriptions completed in December 1995. The pine stand soil is a Montauk variant and the hardwood soil is a Canton variant. Mean monthly temperatures range from 19°C in July to -12°C in January (Van Cleve and Martin 1991). Average annual precipitation is 112 cm. N deposition to the forest is approximately 0.8 g m^{-2} y^{-1} (0.6 g m^{-2} y^{-1} wet and 0.2 g m^{-2} y^{-1} dry; Ollinger and others 1993).

Four 30 x 30–m plots were established in each stand receiving the following treatments: control (no N added), low N (5 g N m^{-2} y^{-1}), high N (15 g N m^{-2} y^{-1}), and N+S (5 g N m^{-2} y^{-1} plus 7.4 g S m^{-2} y^{-1}). Fertilizer additions were divided into six equal monthly doses (May–September) beginning in 1988 (see Table 1) and were applied as a concentrated solution of NH_4NO_3 plus Na_2SO_4 for the N+S treatment. Each plot was divided into 36 5 x 5–m subplots, and only the center 16 subplots were used for sample collection.

Soil Water

Soil water samples were collected approximately monthly, May through November, from five porous cup lysimeters installed in each plot at 60 cm depth. A suction of 50 centibar was applied to each lysimeter for approximately 24 h, samples were collected, and a 20-mL subsample was frozen for subsequent analysis. Due to drought conditions in 1995, the soil

was too dry to collect soil water from the lysimeters between May 31 and October 17.

Lysimeter samples were filtered and analyzed for NO_3-N and NH_4-N by using a Bran and Luebbe (Technicon) TrAAcs 800 Autoanalyzer. Ammonium was analyzed using the Berthelot Reaction chemistry (Technicon Method 780-86T); nitrate was determined using hydrazine sulfate reduction (Technicon Method 782-86T). Detection limits for both nitrate-N and ammonium-N are 0.2 mg L^{-1} by using these techniques. Dissolved inorganic nitrogen (DIN) was determined as the sum of NO_3-N and NH_4-N. Nitrate flux below the rooting zone was calculated using monthly water drainage flux estimates from the PnET model (Aber and others 1995b) parameterized with climatic data over the same period.

From 1993 to 1996, when sufficient volume of water was collected, lysimeter samples also were analyzed for dissolved organic carbon (DOC) and nitrogen (DON) concentrations. DOC was measured with a Shimadzu TOC-5000 Total Organic Carbon analyzer. Samples were combusted at 680°C with a Pt catalyst and CO_2 measured using a nondispersive infrared (NDIR) detector. Total dissolved nitrogen (TDN) was measured on the same machine modified with an Antek chemiluminecent detector (Merriam and others 1996). DON was calculated as TDN—DIN. The intermittent nature of sample availability does not allow the determination of seasonal trends or volume weighted average concentrations per lysimeter. Rather, data will be presented as annual mean concentrations for each treatment.

Trace Gas Emissions

Nitrous oxide fluxes were measured on 20 May 1996 on the control and high N treatment plots only. This date was chosen because intensive sampling over the first 3 years (Bowden and others 1990, 1991), and subsequent intermittent sampling (for example, Magill and others 1997) showed that changes in N_2O flux were only detectable in midspring during periods of moderate soil temperature and high soil moisture content. The static chamber method used in previous studies was used here (Bowden and others 1990, 1991). Sites were sampled twice at approximately 0600 and 01400 h; chamber tops were placed over rings, and four air samples collected over a 30-minute period. Air samples were analyzed using gas chromatography with flame ionization detection. Fluxes were calculated using the initial linear portion of the change in gas concentration in the chamber headspace (n = 3 chambers per plot). For detailed method, see Bowden and others (1990, 1991).

Net N Mineralization and Nitrification

Net nitrification and net N mineralization were measured in situ on all plots by using the buried bag technique (Nadelhoffer and others 1983; Pastor and others 1984, Aber and others 1993). Soils were incubated for a period of 4 to 6 weeks during the growing season, with an over-winter incubation from October to May. All incubations were initiated a minimum of 2 weeks after the most recent fertilizer addition. Net N mineralization and net nitrification were measured for a full year in 1988, 1990, 1993, and 1996. In 1991, growing season measurements were made and overwinter net mineralization and net nitrification were estimated as a percentage of growing season values by using data from 1988, 1990, and 1993.

In the laboratory, soil samples were homogenized through a 5.6-mm mesh sieve, and a 15-g sample was extracted in 150 mL of 1 N KCl for 48 hours. A subsample was oven-dried at 105°C for 48 hours to determine moisture content. Both initial and incubated cores were analyzed for extractable nitrate and ammonium; extracts were analyzed in the same manner as the lysimeter samples.

Net N mineralization was calculated as the difference between extractable nitrate-N plus ammonium-N in the incubated sample and extractable nitrate-N plus ammonium-N in the initial sample. Net nitrification was calculated as the difference between nitrate-N in the incubated sample and nitrate-N in the initial sample. Annual totals were calculated as the sum of 4- or 6-week buried bag incubations in each of the nine soil subplots for each treated plot (May through May). This provided nine replicate values of annual net N mineralization and net nitrification in each plot for statistical analyses. Data from 1988 were omitted due to a high number of missing sample points.

Soil Mass and Carbon and Nitrogen Content

An estimate of soil mass for the organic horizon and top 10 cm of the mineral soil was used to convert C and N concentration to total pools and for conversion of net mineralization rates from mg N g^{-1} soil to g N m^{-2}. A mean dry soil core weight value was calculated for 270–288 cores/horizon from each plot, collected between 1988 and 1991 as follows: [total sieved core weight (g)] * [wet weight to dry weight conversion] / [area of soil core collector (m^2)] . Using total sieved soil core weight corrects for inclusions and removes the need to use bulk density corrections. In 1996, we acquired subsamples from all soil samples used to measure initial extractable nitrate and ammonium for N mineralization. A total of 54 samples was collected per soil

horizon and plot. Samples were aggregated to 21 samples within each horizon and plot and analyzed for C and N concentration by using a Fisons CHN analyser (Milan, Italy).

Foliar and Woody Biomass

Aboveground litterfall was collected three times per year on or near June 1, September 1, and November 15 from each of nine plastic baskets per plot (0.2345 m^2 in size). Litter samples were sorted by species (red and black oak combined), dried for 48 hours at 70°C, weighed, and ground using a Wiley mill with a 1-mm mesh screen. Ground samples were dried overnight at 70°C and analyzed for nitrogen, lignin, and cellulose content by using near-infrared (NIR) spectroscopy (McLellan and others 1991; Bolster and others 1996). Only a partial collection was made in 1988 (November). June and September collections were estimated for each stand as a percentage of the November collection, by using 1990 control plot data. The 1988 data therefore were not used in statistical analyses.

All trees greater than 5 cm in diameter at 1.5 m aboveground level in 1988 were numbered with aluminum tags, and diameter breast height (DBH) was measured 2.5 cm above the tag in October 1988. Trees were measured in November 1990, November 1993, and January 1997. Aboveground woody biomass was calculated from tree diameter measurements by using allometric equations (Pastor and others 1983; Nadelhoffer and others 1985). Estimated annual aboveground net primary production over the entire measurement period (eight growing seasons) was calculated as the sum of woody biomass increment and aboveground litter inputs.

Foliar Chemistry

Fresh foliage was collected during the first week of August each year. Needle samples from 20 different red pines in each plot were pooled into five samples for analysis. Oak, red maple, and black birch were sampled in the hardwood stand. A total of four composite samples per species was collected in each plot. Each sample included leaves from the upper canopy, midlevel, and understory. Red oak and black oak were pooled together for all vegetation sampling. Fresh foliage samples were dried, then ground in the same manner as litterfall; fresh weights were not recorded. Foliar samples were analyzed for percentage nitrogen, lignin, and cellulose content by using near-infrared spectroscopy (McLellan and others 1991; Bolster and others 1996).

Statistical Analyses

The effects of the N deposition treatments were compared separately for the two forest types by using one of two approaches: nested, repeated-measures ANOVAs or simple ANOVAs. All measurements that were made at multiple times at the same location, without aggregating samples, were analyzed using a nested, repeated-measures analysis of variance (including soil water sampling and buried bag measurements). Nitrogen deposition was treated as a main (fixed) factor, replicate sampling locations (for example, lysimeter, treatment subplot) were nested in the nitrogen deposition treatment, and time was a subplot (fixed) factor. Post hoc comparisons were performed using least significant differences (LSDs) corrected for multiple comparisons by using the Bonferroni method (Day and Quinn 1989). Measurements for which individual samples had been aggregated and/or samples that were collected through time but not from the same location/individual (for example, green leaf nitrogen concentration) were analyzed using a two-way ANOVA with time and nitrogen deposition treatment as fixed factors. Soil C:N samples were collected seven times during the year, and nine replicate samples in each treatment were aggregated into three bulk samples. These measurements were analyzed using a nested, repeated-measures ANOVA because samples were always taken from the same subplots and aggregate samples were made using the same group of subplots. Normal probability plots were used to assess the homoscedasticity and normality of residuals (Velleman 1994).

Budget

Input/Output/Retention budgets were calculated for all plots for the entire 9-year period. Total inputs were equal to fertilizer additions plus estimated N deposition calculated from regional deposition data (Ollinger and others 1993). Total outputs were the sum of gaseous and leaching losses. Gaseous losses were measured directly as described above. Leaching losses were calculated as N concentration (mg N L^{-1}) measured in tension lysimeters (60 cm depth) times estimated monthly runoff calculated using the PnET model (Aber and others 1995b).

Net sinks for added N within the plots were determined for directly measurable pools, as the difference between 1996 and 1988 standing pools . These included (a) soil extractable N, calculated as mean annual NO_3-N + NH_4-N; (b) foliar N calculated as (litterfall mass * green foliage %N), with a needle retention constant of 2.5 years for pine (Wessman and others 1988); (c) woody biomass calculated as [increment mass] * [N content of wood

Table 1. Total Ammonium Nitrate Added Since the Initiation of the Experiment

Year	Control	Low N	High N	N + S Nitrogen	N + S Sulfur
1988	0	3.8	11.3	3.8	7.4
1989–1996 (amount each year)	0	5	15	5	7.4
Total N or S Added	0	43.8	131.3	43.8	66.6

Fertilizer was added in six equal doses from May to September. Only a partial year's N treatment was added in 1988. Measurements are in g N m^{-2}.

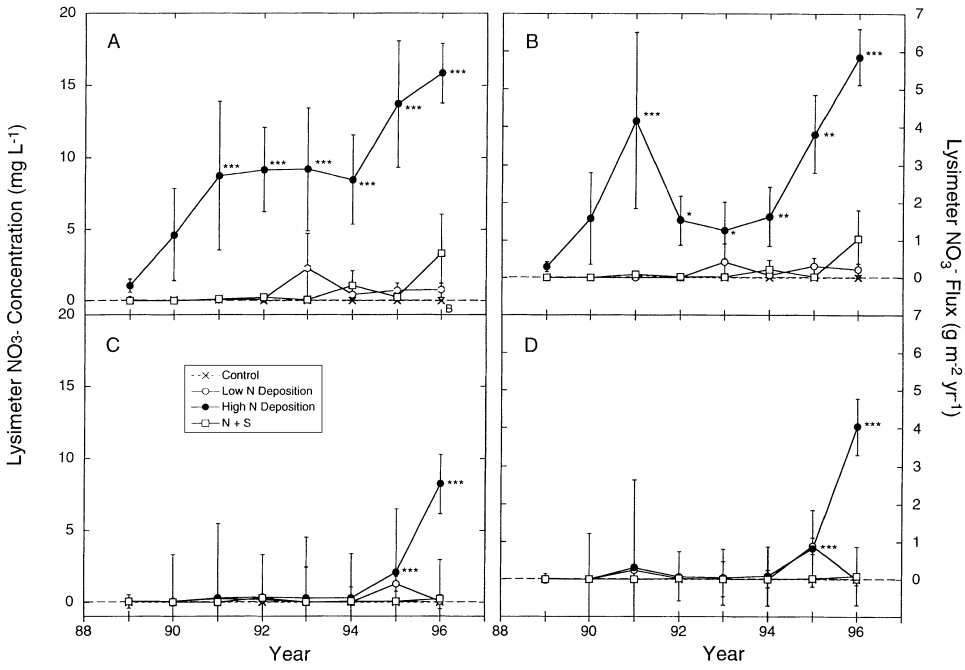

Figure 1. Mean annual nitrate concentrations (A and C, mg N L^{-1}) and nitrate losses (B and D, g N m^{-2} y^{-1}) from mineral horizon lysimeters (60 cm) for the pine plots (A and B) and hardwood plots (C and D). Values are the means of monthly collections made during the growing season from four or five lysimeters in each plot. Asterisks indicate whether any of the N deposition treatments were significantly different than control at any given time. $P < .1$; *, $P < 0.05$; **, $P < 0.01$; and ***, $P < 0.001$).

tissue] where mean wood N concentration was 0.063% for pine and 0.155% hardwoods (Nadelhoffer and others 1999a); and; (d) fine roots (not remeasured in 1996, 1993 data used; Magill and others 1997).

Results and Discussion

Nitrogen Inputs and Losses

Nitrogen Additions. Background nitrogen deposition at the Harvard Forest (0.8 g Nm^{-2} y^{-1}) is moderate for the northeastern US (Ollinger and others 1993; Lovett and Lindberg 1993) and substantially lower than in many experimental sites in Europe (Dise and Wright 1995; Gundersen and others 1998b). Ammonium nitrate additions to the chronic N plots have increased total N inputs by approximately 6- and 19-fold (Table 1).

Nitrate Leaching. Nitrate leaching is one of the most easily measured indicators of N saturation (Aber and others 1989, 1998). The pine and hardwood stands have shown very different patterns of leaching in response to N additions. In the pine high N plot, both nitrate concentrations and calculated fluxes of nitrate below the rooting zone (60-cm lysimeters) began to increase in 1990 and have been significantly higher than control values since 1991 (Figure 1A and B). Whereas concentrations responded in a roughly linear pattern, total calculated flux showed an initial increase and peak, due to high growing season water flux in 1991, followed by a second increase in 1995 and 1996. In the hardwood high N plot, neither nitrate concentration nor flux were increased significantly above control levels until 1995. Over 100 g N m^{-2} had been added by this time. Nitrate losses increased substantially above 1995 levels in 1996 (Figure 1C and D). No significant increases in nitrate loss have been measured in the low N plots in either stand.

While changes in nitrate losses over time were a central part of the original hypotheses on N saturation (Aber and others 1989), differential time delays before the initiation of nitrate losses were not. Because soil microbial activity is generally carbon limited rather than nitrogen limited (Flanagan and van Cleve 1983; Soderstrom and others 1983; Nohr-

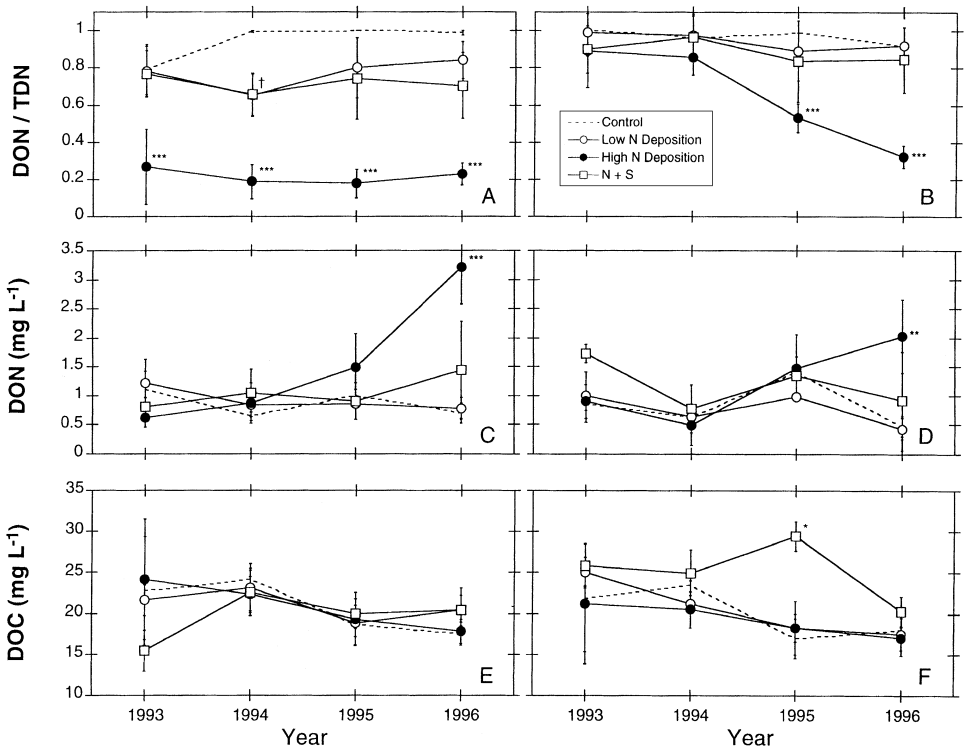

Figure 2. Mean annual fraction of total dissolved N (TDN), which is organic DON (A and B), DON concentrations (C and D), and DOC concentrations (E and F) from mineral horizon lysimeters (60 cm). Pine plots are the left-hand column (B, D, F). Values are the means of monthly collections made during the growing season from four or five lysimeters in each plot. Asterisks indicate significant difference from control plot. †, $P < 0.1$; *, $P < 0.05$; **, $P < 0.01$; ***, $P < 0.001$).

stedt and others 1989), we hypothesized that plant sinks for added N would provide most of the resistance to N losses and therefore would be saturated fairly quickly. The lack of measurable nitrate losses in the hardwood high N plot through 1994 indicate that this first stage of N saturation can be delayed significantly by initial, predeposition conditions of the stand. The eventual increases in 1995 and 1996 suggest that "nitrate breakthrough" will eventually occur in even the most N limited stands.

Increased mobility of nitrate would be expected to increase losses of base cations as well, eventually affecting soil and foliar chemistry (van Breemen and others 1984; Reuss and Johnson 1986; Foster and others 1989). Increased movement of calcium, magnesium, and potassium between the forest floor and mineral soil in all treated plots has been measured (Currie and others 1996, 1999), as well as significant decreases in both the Mg:N and Ca:Al ratios in foliage (Aber and others 1995a; Magill and others 1997). The longer-term effects of chronic N additions on nutrient cation fluxes, foliar element ratios, and biochemical stress indicators are summarized in Minocha and others forthcoming.

DON and DOC Losses. Chronic N additions have significantly altered the relative importance of DON and DIN fractions of nitrogen in soil water below the rooting zone (Figure 2A and B). This is primarily a function of increased nitrate in leachate. DON concentrations in the tension lysimeters (60 cm depth) show relatively small differences between treatments (significant only in 1996, Figure 2C and D). DOC concentrations were variable and generally did not change with treatment (Figure 2E and F) resulting in a reduction in DOC:DON ratio of the soil solution in the high N treated plots in 1996. These results are similar to those reported for soil solution samples from zero tension lysimeters (ZTLs) located below the Oa horizon (Currie and others 1996; McDowell and others 1998), although DON increases at 60 cm occurred after increases were measured in Oa leachate. This difference in timing of DON loss below the forest floor and at 60 cm is most likely due to the ability of mineral soil to adsorb dissolved organic matter and therefore delay DON losses to lower horizon (Qualls and Haines 1991). The very high fraction of N loss as DON in control and low N plots emphasizes the need to measure DON fluxes when describing N balances in N-limited stands. However, the very small changes in DON concentrations reported here suggest that this is not an important component of ecosystems subject to high N deposition.

N_2O Losses. Gaseous fluxes of N as N_2O remain nearly undetectable (Table 2). Assuming that the highest rate measured (pine high N stand) continued for the entire 180-day frost-free period, the total resulting efflux still would be less than 0.1 g N m^{-2} y^{-1}. Gundersen (1998) found no significant differences in N_2O losses between an ambient (15–20 kg

Table 2. N_2O Flux from Control and High N Plots Only Collected May 20, 1996

Plot	Mean	SD	Minimum	Maximum
Hardwood control	−3.25	2.66	−6.33	−1.67
Hardwood high N	1.91	3.50	−2.09	4.43
Pine control	−3.50	2.12	−5.87	−1.80
Pine high N	7.98	7.89	−0.33	15.37

Flux measurements are given as μg m^{-2} h^{-1}. Treatments within a stand are significantly different (Pine stand, $P < 0.10$; Hardwood stand, $P < 0.05$). Negative values indicate a net sink for N_2O.

ha^{-1} y^{-1}) and an NH_4NO_3 fertilized (ambient plus 35 kg ha^{-1} y^{-1}) Norway spruce stand in Denmark. Denitrification rate was estimated at 0.04 kg N ha^{-1} y^{-1} by using these N_2O measurements. In contrast; Tietema and others (1991) measured N_2O losses of 2 g N m^{-2} y^{-1} in an N-saturated Dutch forest with a seasonal change in depth to the water table. Results from the Tietema study show that for gaseous losses to be substantial, soil conditions may need to be wetter than will likely ever be found at the chronic N plots.

Internal Sinks for Added N

Foliage. Foliar N concentration in untreated plots showed interannual variation (calculated as residuals of annual means relative to 9-year average, expressed as a percentage of 9-year average) that ranged from a high of 37% in red pine to a low of 11% in red maple (Figure 3A). When normalized for this variation (expressed as a fractional increase over controls) foliar N concentrations in treated plots were consistently higher than controls (Figure 3B–E). All species, except for red maple, showed an overall trend in leaf N concentrations of high N > low N = N + S > control ($P < 0.05$).

Red pine foliage showed the most dramatic change from controls, increasing by 80% and 30% in the high N and low N plots, respectively, by 1996. Hardwood foliar N levels increased as well, and differences between control and high N values have been significant for most species since 1990 although lower than red pine. Only red pine foliar N was consistently significantly different between the control and low N plots. Overall, increases in foliar biomass and N concentration accounted for 1.4–12.3 g N m^{-2} across all treatments and stands (Table 3).

Extractable NH_4 and NO_3. Extractable N pools increased by 0.2–3.5 g N m^{-2} across treatments over the 9-year period. Mean annual extractable N in both the organic and mineral horizons generally increased with increasing N additions.

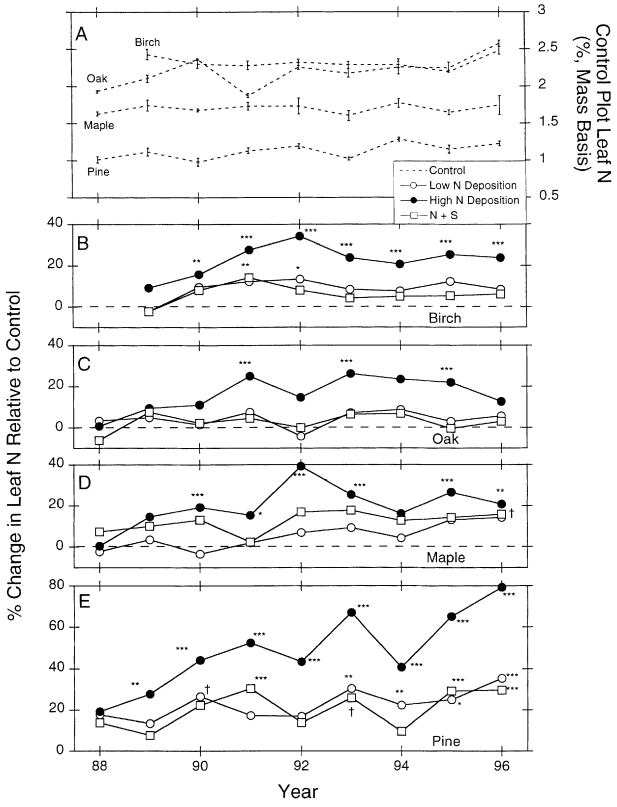

Figure 3. (A) Average control plot foliar nitrogen (mmol N g^{-1}) collected in August of each year. Pine foliage is from the pine stand, and hardwood species are from the hardwood stand. Black Birch data not available for 1988. Foliar nitrogen concentration in treated plots, shown as percent difference from control for each species: (B) black birch, (C) black oak, (D) red maple foliage from the hardwood stand, and (E) red pine from pine stand. Significant differences from control plot leaf N content are indicated by †, $P < 0.1$; *, $P < 0.05$; **, $P < 0.01$; ***, $P < 0.001$.

Fine Roots. Fine root biomass and N content were not remeasured in 1996. Measurements made in 1993 (Magill and others 1997) were used for budget calculations and accounted for 0.2–7.5 g N m^{-2} (Table 3). Differences in fine root N content between stands in 1993 were due entirely to differences in N concentrations in tissues, because no

Table 3. Nine-Year Nitrogen Budget, 1988–96

	Pine Stand				Hardwood Stand			
	Control	Low N	N + S	High N	Control	Low N	N + S	High N
N inputs (g/m^2)								
Atmospheric dep.	7.2	7.2	7.2	7.2	7.2	7.2	7.2	7.2
Fertilization	0	43.8	43.8	131.3	0	43.8	43.8	131.3
Total inputs	7.2	51.0	51.0	138.5	7.2	51.0	51.0	138.5
N losses (g/m^2)								
Gaseous (N_2O)	<0.1	<0.1	<0.1	0.5	<0.1	<0.1	<0.1	0.5
Leaching (NO_3-N)	0.04	1.05	1.42	20.13	0.01	1.15	0.15	5.40
Total losses	0.04	1.05	1.42	20.63	0.01	1.15	0.15	5.90
Total N retention	7.2	50.0	49.6	117.9	7.2	49.9	50.9	132.6
N retention (%)	99	98	97	85	100	98	100	96
Change in N storage (g/m^2)								
Soil extractable N	0.3	1.1	0.9	1.4	0.2	0.6	0.7	3.5
Woody biomass[a]	1.7	1.6	1.3	1.0	6.4	6.5	6.5	11.8
Foliage[b]	2.2	5.8	7.2	12.3	1.4	2.2	3.2	3.6
Fine roots[c]	0.2	4.9	4.9	4.1	0.8	5.9	5.9	7.5
Total measured changes	4.4	13.4	14.3	18.8	8.8	15.2	16.3	26.4
Nonextractable soil pool (by difference)[d]	2.8	36.6	35.3	99.1	−1.6	34.7	34.6	106.2
Soil retention (%)[e]	39	73	71	84	−22	70	68	80

Inputs are the total N received by each plot over 9 years.
[a]Woody pool = increment mass × wood %N. Percent N values are means from Nadelhoffer and others (1999a).
[b]Foliage pool = litterfall mass × greenleaf %N. Final pine plot values were multiplied by 2.5 to account for needle retention. The mean of 1994, 1995, and 1996 litterfall mass was used as the 9-year total because 1996 litterfall mass was very low due to excessive needle drop with drought in 1995. See Results for complete description.
[c]Fine roots were not measured in 1996. Data from 1993 is used for budget calculation. Estimates using a root N pool size increase of 50% (1993 values × 1.5) changed percent soil retention by less than 10%.
[d]Nonextractable soil pool = total N retained − total measured changes in N pools.
[e]% soil retention = nonextractable soil pool/total N retained × 100.

differences in biomass were detected. Whereas declining fine root biomass has been identified as a key response to N saturation and forest decline in European and North American studies (for example, Persson and others 1998), this response had not occurred in the chronic N plots by 1993. Resampling of fine root biomass is planned in the twelfth year of the study.

Woody Biomass. In the pine stand, cumulative wood production over 9 years was lowest in the high N plot and greatest in the control plot (Figure 4A). In contrast, hardwood stand wood increment was nearly 50% higher in the high N plot than in all the other plots, with the largest increase occurring between 1993 and 1996 (Figure 4B). The simultaneous increases in wood production and nitrate leaching in the hardwood stand over the last 3 years may indicate both increasing N cycling and reduced N limitations. Reduced wood production in the pine high N plot resulted in lower estimates for N sequestration than in the pine control and low N plots (Table 3). The opposite is true for the hardwood stand where the high N plot had the highest woody biomass production and sequestered 11.8 g N m^{-2} over 9 years.

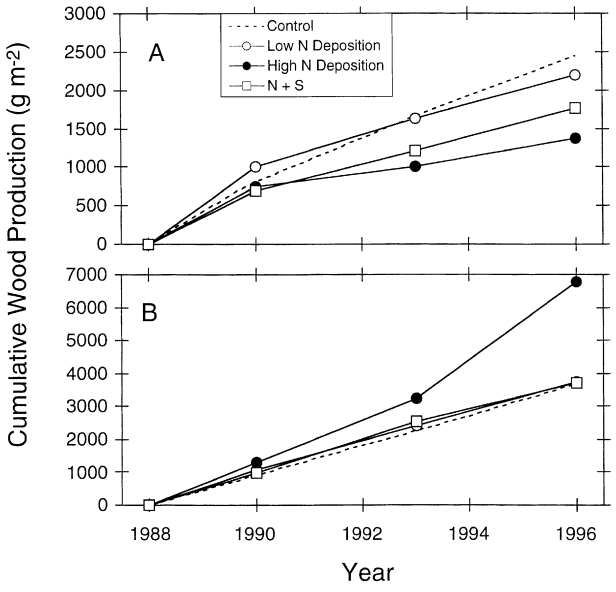

Figure 4. Cumulative wood production from 1988 to 1996 in g m^{-2} for the pine stand (A) and hardwood stand (B).

Table 4. Comparison of Two Methods for Estimating the Distribution of Added N in Control and Low N Plots

	Pine Control		Pine Low		Hardwood Control		Hardwood Low	
Sinks (%)	Budget	^{15}N	Budget	^{15}N	Budget	^{15}N	Budget	^{15}N
Foliage	30.5	0.5	12.7	9.6	19.4	0.5	5.3	5.9
Wood	23.6	0.1	2.9	1.4	88.9	0.5	12.7	4.4
Roots	2.8	1.0	9.6	8.7	11.1	2.1	11.6	13.7
Total plant	56.9	1.6	25.2	19.7	119.4	3.1	29.6	24.0
Soil (0–20 cm)	—	31.4	—	49	—	42.3	—	71.9
Soil (0–60 cm)[a]	43.1	(98.4)	74.8	(80.3)	−19.4	(96.9)	70.4	(76.0)
Grand total	100	33.1 (100)	100	68.7 (100)	100	45.4 (100)	100	95.9 (100)

Values are percent of total N retained in each pool. The Budget column uses data collected as described for Table 3. The ^{15}N data represent percent recovery by compartment after 2 years of natural abundance-level ^{15}N additions to control and low N plots (Nadelhoffer and others 1999a). The (estimated 0–60 cm) values assume that the unrecovered ^{15}N resides in the soil compartment in all cases but could not be fully detected due to low concentrations and sampling limited to 20 cm depth.

[a]Mineral soil pool recovery for 0–60 cm was estimated as follows: budget method = (0–10 cm pool × 6); ^{15}N method = (100 − measured total plant pool).

Declines in productivity or increases in mortality have been reported for other coniferous evergreen forests in the US receiving elevated N deposition (Aber and others 1995a; McNulty and others 1996), and increases in growth in response to removal of N and S from throughfall have been seen in European coniferous evergreen forests (for example, Beier and others 1998; Boxman and others 1998). At the Solling site, earlier studies linked forest decline to reduced Mg:N ratios in foliage (Schulze 1989), a response that also has been seen in the chronic N plots (Magill and others 1997; Minocha and others forthcoming). Together, these results suggest that the cumulative effects of N deposition at moderate-to-high levels may have negative impacts on biomass production in commercial evergreen forests.

Sum of Measured Sinks and N Retention Efficiency

Measured N retention in the four sinks described above (foliage, extractable soil, roots, wood), accounted for nearly all ambient N deposition in both the pine and hardwood control plots. Discrepancies of 1.6 and 2.7 g N m^{-2} may be due either to cumulative measurement errors or to minor changes in total storage in soil organic matter. This near-balance and the similarity in values for most processes between the low N and N+S plots provide some evidence for internal consistency in methods and calculations.

Overall DIN retention efficiency (percentage of added DIN retained) for the first 9 years was 97–100% in the control, low, and N+S plots (Table 3). Elevated nitrate losses in the hardwood high N plot in 1996 reduced the 9-year retention efficiency in that plot to 96%, whereas a longer period of nitrate loss in the pine high N plot yielded an overall DIN retention efficiency of 85%. Increases in directly measurable ecosystem N storage pools accounted for 16–32% of added DIN retained in the amended plots (Table 3). This suggests that the one major unmeasured pool, soil organic matter, contained the remaining 68–84% (last line in Table 3).

Nine-year budget estimates were compared with sink strengths estimated as percent recovery of natural abundance levels of ^{15}N additions (Nadelhoffer and others 1999a; Table 4). ^{15}N was added to the control and low N plots in both stands in 1991–92. The isotope was added as part of the monthly fertilizer additions in the low N plots and in a highly enriched form as subfertilizer additions in the control plots. Measurements of ^{15}N distribution were made in 1992.

Very different results are obtained by the two methods for the control plots. The budgeting technique suggests that most to all of the incremental additions of N in both control plots can be accounted for by increases in plant N pools. The ^{15}N method shows almost none of the added isotope is in plants. This discrepancy could reflect differences in the N environment immediately after N additions. In the control plots, ^{15}N was added as very low concentrations of highly enriched N, such that similar amounts of isotope were added to both control and low N plots, whereas total N applications to the control plots remained at subfertilizer levels. In the control plots, total N concentration remained low, and immobilization of this N directly into the litter/soil system appears to have been nearly complete. In the low N plots, the ^{15}N was added with a pulse of fertilizer N that may have temporarily overwhelmed the soil immobilization processes, allowing greater movement to plant roots. This comparison is consistent with a review of the N addition literature by Johnson (1992) who found that low level N inputs were immobilized by the microbial population and

that larger pulses of N were necessary before plants could compete effectively against soil immobilization processes.

Net increases in total plant N pools in the control plots must represent a longer-term accumulation of N by plants against the strong soil N sinks. The short-term pulse of ^{15}N into these systems may be rapidly immobilized and only slowly released in a form that can be accessed by plants. In the low N plots, the pulse of fertilizer addition apparently increased N mobility to roots immediately and resulted in a larger portion residing in the plant pool.

Given the potential for error in each method due to the number of factors affecting field level data collections, the agreement between the two estimates in the low N plots is reasonable. This is especially true if the unrecovered ^{15}N is assumed to reside in the lower, unsampled soil horizons, which accounts for a much larger fraction of added N in the pine low N plot than the hardwood low N plot. These results appear to be consistent and support the theory that most of the added N in these plots resides in the litter/soil system.

Results from three other field ^{15}N addition studies were compared with the chronic N results, and whereas total ^{15}N recovery varied significantly between studies, the fraction retained within the soil pool was similar. Total tracer recovery at four N deposition sites in Europe (Tietema and others 1998) ranged from 65 to 105%. In addition, they found that organic soils retained between 11 and 54% of added ^{15}N whereas mineral soil retention of ^{15}N ranged from 1 to 27% of the total N added. In a second study, watershed level additions of $^{15}NH_4$ at Bear Brook in eastern Maine, USA were measured in three different forest types (Nadelhoffer and others 1999b). Total recovery was low (34–40%) as was 0–5 cm soil retention (18–32%). A third study found that recovery of ^{15}N (both $^{15}NO_3$ and $^{15}NH_4$ labels) added to *Pinus radiata* stands in New Zealand as part of fertilizer treatments ranged from 72 to 100%, depending on presence or absence of pasture (Clinton and Mead 1994). Organic soil retention was 18–32%, and mineral soil retention (0-20 cm) of ^{15}N ranged from 22 to 31%. Overall, the variation in recovery rates illustrate the difficulties in working with ecosystem or plot level ^{15}N additions and measurements. However, they also demonstrate that soils usually exhibit a significant capacity for N retention.

Possible Mechanisms of N Retention. Near complete retention of long-term N additions is one of the most striking results of the chronic N experiment. Determining the mechanisms by which this has occurred remains the major challenge of this project. In a recent article (Aber and others 1998), we discussed three possible mechanisms for incorporation into soil organic matter: (a) N immobilization by free-living microbes; (b) abiotic reactions with soil organic matter; and (c) mycorrhizal assimilation and exudation of organically bound N. Simultaneous consideration of carbon and nitrogen cycling constraints on the mechanism of N retention suggested a greater importance for abiotic and mycorrhizal pathways than previously had been thought. Still, these remain hypotheses to be tested. A number of studies have measured abiotic or chemical NH_4 immobilization rates into soil organic matter sterilized soil samples (Nommik 1970; Axelsson and Berg 1988; Schimel and Firestone 1989; Sen and Chalk 1995). An initial set of measurements of nitrate cycling by ^{15}N pool dilution techniques (Berntson and Aber forthcoming) suggested that "fast" immobilization processes (generally associated with abiotic reactions) account for all gross nitrate immobilization in the pine high N, pine control, and hardwood high N plots. Only the hardwood control plot showed detectable "slow" gross immobilization, generally associated with microbial processes. Given that the hardwood control plot is the furthest from saturation, these results indicate that nitrate retention mechanisms may change with increasing N availability and stage of N saturation (Berntson and Aber forthcoming).

Direct Measurement of Soil C and N Pools. Attempts to directly measure changes in soil storage pools (as much as 100 g N m^{-2} in total N content) proved inconclusive (Table 5). Our measurements extended only to a depth of 10 cm into the mineral soil, such that estimation of changes in N content between 10 and 60 cm (the depth of the lysimeters) was not possible. However, due to high spatial variability, differences between plots were not statistically significant even for the organic horizons and the top 10 cm of mineral soil.

Increased N content could occur either as a decrease in the soil C:N ratio with no change in total organic matter content or as an increase in total soil organic matter alone or both. The latter would be consistent with measured reductions in decomposition rates accompanied by continued increases in N content of older litter cohorts as was seen in the N amended stands (Magill and Aber 1998). However, we cannot detect changes in either organic matter content or C:N ratio after 9 years. Previous studies have shown that a change of 100–200 kg N ha^{-1} would be required for detection with standard soil sampling techniques (Huntington and others 1988; Johnson 1995). Budget calculations (Table 3) indi-

Table 5. Total Soil Nitrogen, Total Soil Carbon, and C:N Ratio for 1996

	Pine Stand				Hardwood Stand			
	Control	Low N	N + S	High N	Control	Low N	N + S	High N
Total soil N								
Organic	108	111	112	105	112	125	128	153
Mineral (10 cm)	214	187	210	187	152	161	159	162
Total N storage	321	298	323	292	264	286	287	314
Total soil C								
Organic	2547	3007	3224	2999	2877	3131	3280	3816
Mineral (10 cm)	4309	4056	4752	4008	3464	3715	3544	3626
Total C storage	6856	7063	7976	7007	6341	6846	6823	7442
C:N ratio								
Organic	23.69	27.13	28.70	28.52	25.58	25.09	25.56	24.97
Mineral	20.16	21.72	22.61	21.40	22.79	23.11	22.32	22.45

Totals are in units of g N or C m^{-2}.

cate that increased total N content in the nonextractable soil pools in the high N plots (99-106 g N m^{-2}) are just beginning to approach detectable levels.

Internal Nitrogen Cycling

N Mineralization and Nitrification. Significant interannual variability in net mineralization rates was observed in both the pines and hardwoods (Figure 5). In the pine stand, measured values across all treatments were statistically equivalent in 1990, 1991, and 1993 but were significantly greater in 1996 (Figure 5A, post hoc comparison, $P < 0.05$). There were no significant differences between treatments in the pine stand in any year. In the hardwood stand (Figure 5B), measured rates in 1990 and 1991 were equivalent, but significantly lower than 1993 and 1996 (post hoc comparison, $P < 0.05$). Significant overall increases in net mineralization rates were observed in both the hardwood high N and hardwood low N treatments relative to the control (no significant change in the N+S plot). These effects varied through time. In 1990 and 1991, there were no significant effects of N addition. By 1993, the high N plot showed elevated net mineralization rates, and by 1996 both high and low N plots were significantly higher than the control stand.

Annual net nitrification for the pine stand, expressed as a fraction of net N mineralization, was consistently greater in treated plots than in the control plot (Figure 6A) and was highest in 1996, where all three N amended plots showed significantly greater relative nitrification rates than the pine control plot. In the hardwood stand, only the high N plot showed elevated, relative nitrification rates (Figure 6B). This increase in nitrification occurred only in 1996, coincident with the first year of significant nitrate leaching in lysimeters at 60 cm (Figure 1A and B).

At the Fernow Experimental Forest in West Virgina, net nitrification was found to be between 90 and 100% of net mineralization in both an $(NH_4)_2SO_4$ fertilized and an untreated stand (Gilliam and others 1996). In this case, the investigators concluded that the entire watershed was N saturated, which is why a significant response was not seen with N treatment. Gundersen (1998) measured an 85% increase in net mineralization rate in the organic layer of an NH_4NO_3 fertilized Norway spruce stand, as compared with rates from an ambient plot, but measured insignificant net nitrification rates or net nitrate immobilization (0% of net mineralization). Soil net mineralization rate and other soil parameters are sensitive to both prior land-use history and current vegetative cover (Compton and others 1998). Disturbances such as logging or agriculture can have both short- and long-term impacts on soil N status (for example, Covington 1981; Hamburg 1984) and may partially account for the wide range of rates measured across different N deposition experiments.

Litterfall. Aboveground litter production in the pine stand was consistently higher in the high N and N+S plots than in the control or low N plots (Figure 7A). In 1995, a year of severe mid-summer drought, anomalously high litterfall (30–50% increase in all plots relative to the mean of previous years) was measured in the pine stand, with rates in all three amended plots being significantly greater than in the control plot. A large and significant ($P < 0.05$) decrease in litterfall occurred the following year (50% decrease from pre-1995 mean litterfall). In

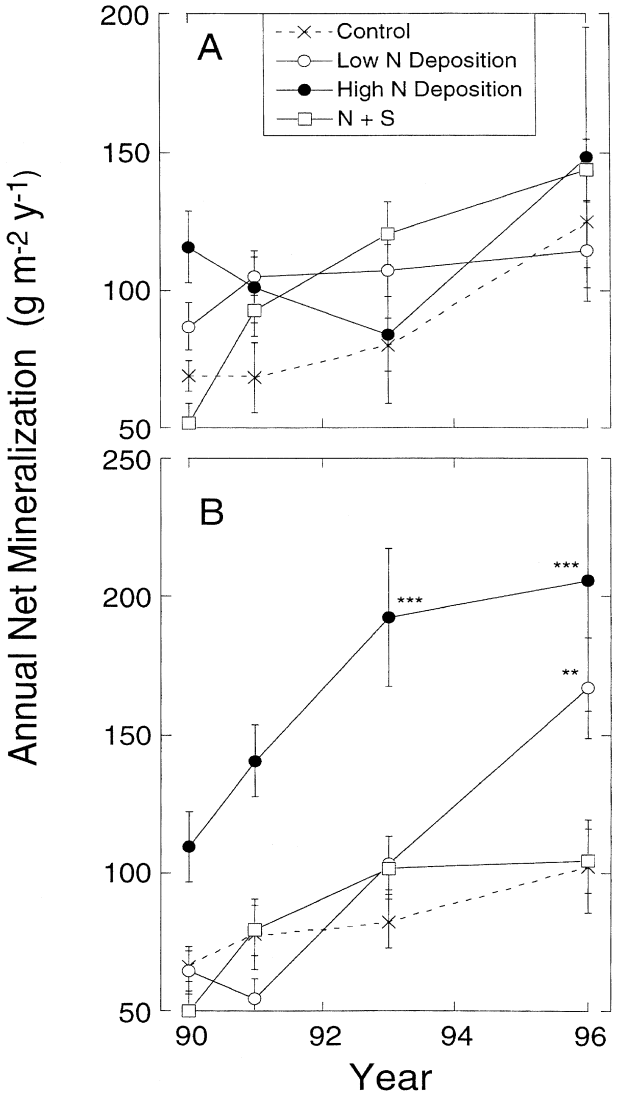

Figure 5. Total net annual N mineralization (NH_4^+ and NO_3^-) measured using in situ buried bags in the pine stand (A) and the hardwood stand (B). Asterisks indicate significant difference from control plot. **, $P < 0.01$; ***, $P < 0.001$).

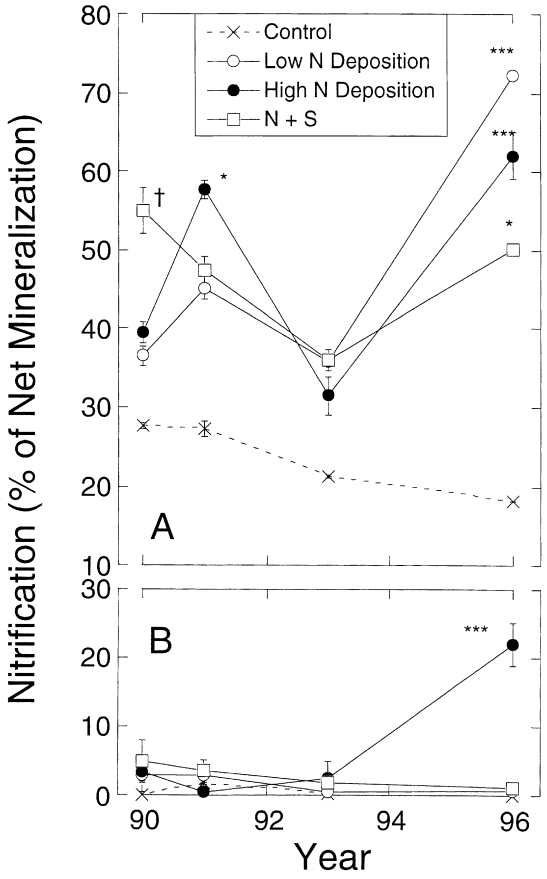

Figure 6. Net nitrification as a percent of total annual net N mineralization (Figure 5) in the pine stand (A) and the hardwood stand (B). Asterisks indicate significant difference from control plot. †, $P < 0.1$; *, $P < 0.05$; ***, $P < 0.001$).

the hardwood stand there are no significant interplot differences in litterfall in any year (Figure 7B). Unlike the pine stand, litter production did not vary significantly in 1995 relative to 1994 or 1996.

An 18-year study of litterfall in a mixed hardwood forest in Northeast Iowa measured variation in litterfall mass of approximately 10% (CV) (Knutson 1997). Fluctuations in growing season precipitation were found to significantly effect litter decomposition rates but were not related to litterfall amounts. These stands did not include any coniferous species, however, which may be more sensitive to water stress. Gundersen (1998) measured litterfall and litter N content in an ambient (15–20 kg ha^{-1} y^{-1}) and an NH_4NO_3 fertilized (ambient plus 35 kg ha^{-1} y^{-1}) Norway spruce stand in Denmark. Measurements were made from 1991 to 1995, including three successive drought years (1992–94). Litterfall increased approximately 50% in the first year of drought (1992) under both treatments, dropping back to pretreatment levels in 1993 and increasing again in 1994. Significant increases in needle litter N content with N treatment also were measured.

The dramatic increase in pine litterfall, and mixed response of other studies cited above, led us to question the influence of drought on these forests. In 1995, summer rainfall was much lower than average from May through October, and a number of ecosystem processes were strongly affected (for example, Goulden and others 1996). In addition to litterfall, 1996 net mineralization rates increased as well as the fraction of mineralized N nitrified. These measured changes in litter inputs and mineralization coincide with the highest DIN flux from the pine high N plot. Although litterfall was not affected

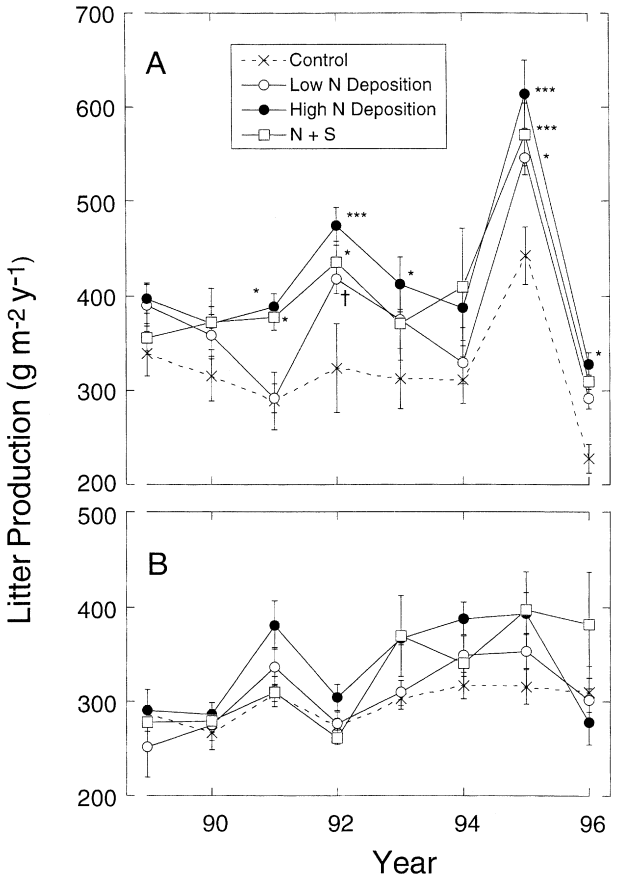

Figure 7. Total annual litterfall rates in the pine stand (A) and the hardwood stand (B). Asterisks indicate significant difference from control plot. †, $P < 0.1$; *, $P < 0.05$; ***, $P < 0.001$).

in the hardwood stand, nitrate losses were first detected in the hardwood high N plot in the fall of 1995 and increased in 1996 relative to previous measurements in 1993.

One of the oldest extant hypotheses for the induction of N saturation, nitrate leaching and forest decline, is the drought-induced "nitrate pulse" theory (Ulrich 1983). In this theory, extreme soil drought events induce nitrification that creates a pulse of nitrate loss and soil acidification, leading to forest decline. A number of drought experiments at the whole ecosystem level were conducted as part of the EXMAN network of sites in Europe. Lamersdorf and others (1998) summarized the results of several 2–4-year drought experiments by using artificial roofs to control precipitation inputs to forests and tested Ulrich's theory of nitrate pulse/acidification upon rewetting of the soil after a prolonged period of drought. They did not observe marked nitrification pulses at any of the sites and only single sampling locations were found to have increasing aluminum concentrations and decreasing pH values. The lack of response was attributed primarily to differences in soil properties, plant uptake, ambient N availability, and other properties that resulted in unique "buffering abilities" at each site.

A major difference between the European studies and the chronic N study is the simultaneous occurrence of high N additions and drought at the Harvard Forest. However, because response to drought was not experimentally tested, it is unknown whether drought-induced limitations on plant and soil function contributed to the loss of N retention capacity in the pine and hardwood high N plots in 1995 and 1996. Did initiation of nitrate losses in the hardwood stand or the increase in loss in the pine stand occur because of the extreme event, or was co-occurrence of the drought event and increases in nitrate leaching purely coincidental?

Many of the measured changes in the chronic N plots point to a response of the vegetation to water stress, for example, enhanced litterfall. Even though the theory of Ulrich cannot be proven, water stress can have an serious impact on vegetation function and health (Fitter and Hay 1987), potentially weakening trees and making them less resistant to the adverse effects of N deposition and N saturation (Aber and others 1992). Given that the final stage of N saturation is reached upon death or decline of the vegetation (Aber 1992), all factors that contribute to vegetative health have the potential to speed up the saturation process and move a forest along the N saturation continuum at a more rapid pace.

Conclusions

Long-term ecological experiments allow for interpretation of the response of an ecosystem to environmental effects under a more realistic and dynamic time scale than is possible under a 2–3-year study. Some of the important findings from this experiment are summarized as follows:

1. The simultaneous increase in nitrification, nitrate leaching and wood production in the hardwood high N plot suggests that that there is a strong link between the initiation of nitrification (as microbial immobilization capacity is saturated) and increased N availability for plant uptake, leading ultimately to the reduction of N limitations on plant growth and productivity. Because this response was not seen until year 8, short-term observations would have concluded that N additions were having no effect on the hardwood plots. The sharp contrast in response of the pine and hardwood stands is an indication that the mosaic of community types across the

landscape must be considered when determining regional scale responses to N deposition.

2. A continued decline in wood production in the pine high N plot, accompanied by increasing foliar N concentrations, suggests the existence of nutrient imbalances and the potential for reduction in photosynthetic rates. In a previous article (Magill and others 1997), foliar N content in the pine high N appeared to have reached a maximum after year 4 and did not increase through year 6. Continued measurement of this N sink, as presented in this article, revealed that the foliar capacity was greater than would have been concluded after 6 years of N additions.
3. Low N plots have shown minimal responses to N additions to date. If cumulative N dose is the determining factor in pushing these plots to saturation, it should take roughly three times as long for the low N plots to show the same response as the high N plots. A comparison of measured nitrate losses from the pine stand revealed that low N plot concentrations in year 9 were similar to nitrate concentrations in the high N plot in year 3, supporting the theory that cumulative dose is important. Because a leaching response was not seen until year 8 in the hardwood high N plot, (a cumulative dose of 120 g m^{-2}), the low N plot may not be affected until year 24 of additions. If the intensity of additions is also important, low N response would occur over even longer periods of time.

ACKNOWLEDGMENTS

This research was funded by the National Science Foundation Long-Term Ecological Research Program and the USDA National Research Initiative Competitive Grants Program. Many people have contributed to the success and longevity of this project, including Gloria Quigley, Steve Newman, Richard Bowden, Bill Currie, Joe Hendricks, Matt Kizlinski, and all the hourly employees of the Forest Ecosystem Group at the Complex System Research Center, UNH. Additional thanks goes to Chris Catricala and Kathy Newkirk for collection of trace gas data and Jeff Merriam for DOC and DON analysis.

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