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BIOGEOCHEMICAL RESPONSE OF FOREST ECOSYSTEMS TO SIMULATED CHRONIC NITROGEN DEPOSITION

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Abstract. Reported in this paper are foliar chemistry, tree growth (above- and belowground), soil chemistry, nitrogen cycling (net mineralization and nitrification) and soil N_2O flux responses to the first 6 yr of chronic nitrogen amendments at the Harvard Forest (Massachusetts, USA). A 70-yr-old red pine (*Pinus resinosa* Ait.) stand and a 50-yr-old mixed hardwood stand received control, low nitrogen (50 $kg \cdot ha^{-1} \cdot yr^{-1}$), high nitrogen (150 $kg \cdot ha^{-1} \cdot yr^{-1}$), and low nitrogen plus sulfur treatments, with additions occurring in six equal doses over the growing season as NH_4NO_3 and Na_2SO_4. Foliar N concentrations increased up to 25% in the hardwood stand and 67% in the pines, and there was no apparent decrease of N retranslocation due to fertilization. Wood production increased in the hardwood stand in response to fertilization but decreased in the pine stand. Fine-root nitrogen concentrations increased with N additions, and fine roots were a significant sink for added nitrogen. Nitrate leaching losses increased continuously over the 6-yr period in the treated pine stands but remained insignificant in the hardwoods. Annual net N mineralization increased substantially in response to treatments in both stands but declined in the pine high-N plot by the end of year six. Net nitrification increased from 17% of net mineralization in 1988 to 51% in 1993 for the pine high-N plot. Only a slight increase in net nitrification was measured in the hardwood stand, and only in 1993. Extractable NH_4 was consistently higher in treated plots than in controls in both stands, where extractable NO_3 was higher than controls only in the treated pine plots. Soil extracts yielded <1.5 kg/ha of NO_3-N for all plots in the hardwood stand throughout the experiment. Effluxes of N_2O were consistently greater in the pine high-N plot than in the pine control plot, but there were no observed large-scale increases in N_2O emissions immediately following fertilizer application. Calculated nitrogen budgets for the first 6 yr showed extremely high N retention (85–99%). Of the retained N, 50–83% appears to be in the long-term, recalcitrant soil pool. The relative importance of biotic and abiotic mechanisms of N incorporation into soils remains uncertain. Size, kinetics, and uptake capacity of this soil pool are critical and largely unknown factors determining ecosystem response to increased N loading and may be related to land-use history.

Key words: ammonium nitrate additions; foliar and litterfall N concentration; forests, red pine cf. mixed hardwood; Harvard Forest, Massachusetts, USA; N retention and saturation; net N mineralization rates; net nitrification in pine vs. hardwood stands; nitrate leaching losses; wood production, effects of fertilizer on.

INTRODUCTION

Atmospheric nitrogen deposition in the Northeastern United States is currently 10–20 times above historic background levels (Galloway et al. 1984, Shannon and Sisterson 1992, Ollinger et al. 1993). In spite of recent regulations on NO_x emissions (e.g., Clean Air Acts Amendments of 1990 [Air Pollution Prevention and Control. United States Code Title 42, Chapter 85]), current deposition levels are not expected to decrease in the near future (EPA 1995). This has led to concerns regarding the ability of terrestrial systems to absorb and accumulate N. It is hypothesized that excessive nitrogen inputs may saturate the nitrogen retention capacity of forest ecosystems (Ågren and Bosatta 1988, Aber et al. 1989, Aber 1992, Stoddard 1994). Potential effects of N saturation include increased nitrate leaching and simultaneous base cation losses (Hultberg et al. 1994, Hedin et al. 1995), soil acidification (van Breemen et al. 1984), altered fluxes of trace gases (Steudler et al. 1989, Bowden et al. 1991, Castro et al. 1995) and possible forest decline (Schulze 1989, McNulty et al. 1996).

Strengthening our ability to predict long-term effects of increasing N deposition on forested ecosystems is critical for making sound, science-based resource management decisions. The purpose of this study is to increase our understanding of ecosystem nitrogen dynamics in response to elevated N inputs. Long-term,

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TABLE 1. Initial soil characteristics of the chronic-nitrogen-addition plots at the Harvard Forest (Massachusetts, USA). Soil pH measurements were made in 0.01 mol/L $CaCl_2$ solution.

	Pine stand	Hardwood stand
Forest floor pH	3.2	3.3
Mineral soil pH	3.8	3.8
Forest floor depth (cm)	4.6	6.5

	C:N ratio†					
	Pine stand			Hardwood stand		
	Control	Low N	High N	Control	Low N	High N
Forest floor	22.17 (1.19)	25.75 (1.94)	23.68 (1.69)	25.86 (0.87)	22.86 (1.03)	23.65 (0.98)
Mineral soil	20.47 (0.59)	25.08 (1.17)	21.51 (0.98)	23.45 (0.99)	18.86 (1.27)	21.17 (0.85)

Note: Initial data were not recorded for N+S plots since they were initiated later than the other treatments.

† The C-to-N ratio is given as the mean, with 1 SE in parentheses.

low-level ("chronic") nitrogen additions were used to simulate increased N loading on a whole-ecosystem scale (Beier and Rasmussen 1994). Reported in this paper are the responses of foliar chemistry, tree growth (above- and belowground), soil chemistry, nitrogen cycling (net mineralization and nitrification) and N_2O flux to the first 6 yr of ammonium nitrate fertilizer additions.

MATERIALS AND METHODS

Study sites

The chronic-nitrogen-addition plots are located at the Harvard Forest in central Massachusetts (42°30′ N, 72°10′ W), a National Science Foundation Long-Term Ecological Research site since 1988. Historical land-use practices for the tract are well documented and show that most of the area (≈85%) was in cultivation or pasture in the mid-1800s (Foster 1992). The dominant soil types are stony to sandy loams formed from glacial till. Annual temperatures range from an average of 19°C in July to an average of −12°C in January. Average annual precipitation is 112 cm (Van Cleve and Martin 1991). Nitrogen deposition to the forest is ≈ 6 $kg \cdot ha^{-1} \cdot yr^{-1}$ wet and 2 $kg \cdot ha^{-1} \cdot yr^{-1}$ dry (Ollinger et al. 1993).

Two adjacent stands were chosen for the study: an even-aged red pine (*Pinus resinosa* Ait.) stand planted in 1926 and a 50-yr-old mixed-hardwood stand that had regenerated naturally after clearcutting. The hardwood stand is dominated by black and red oak (*Quercus velutina* Lam. and *Q. rubra* L., respectively) with significant amounts of black birch (*Betula lenta* L.), red maple (*Acer rubrum* L.), and American beech (*Fagus grandifolia* Ehrh.). Soils in both stands are Entic Haplorthods of the Canton series. Additional soil characteristics are listed in Table 1.

Treatments

Four treated plots were established within each stand: control, low N, low N plus sulfur (N+S), and high N. Each plot measured 30 × 30 m (0.09 ha) and was divided into 36 subplots (each 5 × 5 m). Fertilizer additions of NH_4NO_3 and Na_2SO_4 began in 1988 as six equal applications over the growing season. Fertilizer was weighed, mixed with 20 L of water (equivalent to 0.002 cm rainfall) and applied using a backpack sprayer. Two passes were made across each plot to ensure an even distribution of fertilizer.

A partial application was made in year 1 (1988). Total fertilizer applied was 38 kg $N \cdot ha^{-1} \cdot yr^{-1}$ to the low-nitrogen treatment and the nitrogen portion of the N+S treatment, 113 kg $N \cdot ha^{-1} \cdot yr^{-1}$ to the high-nitrogen treatment, and 74 kg $(SO_4\text{-}S) \cdot ha^{-1} \cdot yr^{-1}$ to the N+S treatment. Applications for all following years were at the rate of 50 kg $N \cdot ha^{-1} \cdot yr^{-1}$ to the low and N+S plots and 150 kg $N \cdot ha^{-1} \cdot yr^{-1}$ to the high-addition plots. Sulfur additions remained the same as year 1.

Sample collection and analysis

Foliage.—Litterfall was collected three times per year on or near 1 June, 1 September, and 15 November. Nine permanent litter baskets (0.023 ha in size) were located in each plot. Baskets were removed from the plots during fertilization to avoid potential contamination.

Green foliage was collected once per year during the first week of August. Only red pine was sampled in the pine stand; all trees were approximately the same height (20 m) and were dominant in the canopy. Needle samples from 20 different trees in each plot were pooled into five samples for analysis. Each species in the hardwood stand was sampled at three canopy heights: co-dominant, mid-canopy, and understory. A total of four composite samples from 12 trees per species were collected in each plot. Red oak and black oak were treated as a single species in all vegetation sampling.

Both litter and green leaf samples were analyzed for nitrogen, lignin, and cellulose content using near-infrared spectroscopy (McLellan et al. 1991, Bolster et al. 1996). Litter samples were sorted by species (red

and black oak combined), dried for 48 h at 70°C, weighed, and ground through a 1-mm mesh. Green leaf samples were dried and ground in the same manner without weighing. Samples were re-dried overnight at 70°C prior to analysis.

Total annual litterfall (in units of $kg \cdot ha^{-1} \cdot yr^{-1}$) for each plot was calculated as the average mass of leaves from the nine litter baskets. Only a partial-year collection was made in 1988 (November). June and September collections were estimated for each stand as a percentage of the November collection, using 1990 control-plot data. Retranslocation during foliar senescence was calculated as follows: [(green foliage percentage N) minus (litterfall percentage N)] divided by green foliage percentage N.

Tree increments and net primary production above ground.—All trees >5 cm in diameter at 1.5 m above ground level (diameter at breast height) in 1988 were numbered with aluminum tags. Diameter was measured 2.5 cm above the tag. Measurements were made in October 1988 and November 1990, 1992, and 1993. Woody biomass increment in all plots was determined by applying allometric equations (Whittaker et al. 1974, Pastor et al. 1984) to tree-diameter measurements. Estimated annual aboveground net primary production over the entire measurement period (five growing seasons) was calculated as the sum of woody biomass increment over this period and aboveground litter inputs for the years 1989–1993 divided by 5.

Fine roots.—Fine-root sampling was conducted in the control, low-N, and high-N plots of both stands in May 1988 and 1991. In 1988, prior to initiation of fertilizer additions, nine soil cores (7.5 cm diameter and extending to a mineral soil depth of 10 cm) were collected from randomly selected locations within each plot and frozen for future processing. In 1991 the basic sample unit was a soil monolith 10 cm wide × 20 cm long and extending to a mineral soil depth of 10 cm. Soil monoliths (five from the red pine plots and eight from the hardwood plots) were collected from random locations within a designated 5 × 5 m subplot within each plot. Monoliths were refrigerated (2°C) for no more than 4 d prior to fine-root sorting.

Both soil cores and monoliths were separated into organic and mineral horizons. In 1988 each sample was placed in a double layer of 1-mm mesh screen and soaked in distilled water to remove mineral particles. Residual tissues (live and dead) were rinsed and tree roots ≤2 mm in diameter were saved for analysis. In 1991 each monolith horizon was hand-sorted to remove only living fine-root tissues, here defined as primary ramifications without external suberization or bark development (generally corresponding to diameters of ≤0.5 mm and ≤1 mm for the hardwood and red pine roots, respectively). Following removal of any nontarget species, such as occassional hardwoods in the red pine stand and forbe roots in both stands, samples were placed in a 1-mm mesh sieve and rinsed with tap water for ≈15–20 sec. Sorted fine-root samples were oven-dried (105°C in 1988 and 70°C in 1991) to a constant mass and weighed. Roots were then ground and measured for total carbon and nitrogen using a Perkin-Elmer model 240B CHN analyzer.

Lysimeters.—Five porous-cup lysimeters were installed at 60 cm depth in each plot in August 1988. Sampling began in May 1989 and continued monthly throughout the growing season of each year. Prior to sample collection, tubes were drained of existing water using a syringe, and tension of 344.7×10^3 Pa (50 psi [pounds per square inch]) was applied to each lysimeter. After ≈24 h, samples were collected and volume was recorded. A 20-mL subsample was frozen for analysis of NO_3 and NH_4.

Lysimeter samples were filtered and analyzed for NO_3-N and NH_4-N using a Bran & Luebbe (formerly Technicon) TrAAcs 800 autoanalyzer (Bran & Luebbe, Buffalo Grove, Illinois, USA). 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 0.20 mg/L using these techniques.

Soils.—Net nitrification and net mineralization were measured in situ on all plots using the buried-bag technique (Nadelhoffer et al. 1983, Pastor et al. 1984). Soils were incubated for a period of 4–6 wk during the growing season, with an over-winter incubation from October to May. Both initial and incubated cores were analyzed for extractable nitrate and ammonium (see Aber et al. [1993] for specific sampling technique). Net N mineralization and nitrification were measured for a full year in 1988, 1990, 1991, and 1993. A 6-wk incubation was performed in midsummer of 1989 and 1992. All incubations were initiated a minimum of 2 wk after the most recent fertilizer addition. A partial year was measured for the N+S plots in 1988. In the laboratory, soil samples were homogenized and extracted in 1 mol/L KCl solution for 48 h. A subsample was oven-dried at 105°C for 48 h (for specific laboratory technique see Aber et al. 1993). 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 are calculated as the sum of all buried-bag incubations for a given year (May through May).

Percentage carbon and nitrogen were determined for both the organic horizon and mineral soil in all but the N+S plots, using a Perkin-Elmer 2300 CHN analyzer. Total N (as kilograms per hectare) and the C:N ratio were calculated.

Trace gases.—Exchanges of N_2O between the soil and the atmosphere were measured throughout the

study (Bowden et al. 1990, 1991). All measurements were made at least 2 wk following fertilizer applications, using a static-chamber technique. Three 28.7-cm-diameter rings were permanently anchored 1 cm into the soil surface in each plot. During flux measurements, a chamber top with headspace of 5.4 L was attached to the ring for 20–30 min incubations and four air samples were removed via syringe during the incubation. 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 headspace ($n = 3$ measurements). For more thorough explanation see Bowden et al. (1990, 1991).

To assess the possibility that trace-gas fluxes showed large, immediate responses to fertilizer applications, gas exchanges in the red pine control and high-N plots were compared for the period immediately before, and for 1 mo after, a midsummer N fertilization in 1994. Fluxes were measured daily for the 1st wk following fertilization, and then approximately twice weekly thereafter.

Budget calculations

Estimated N budgets for all eight plots were calculated as a total input–output budget with changes in storage for measured ecosystem pools. In addition, estimated average annual internal cycling rates were determined for all but the N+S plots.

In the first case, total 6-yr fertilizer inputs were added to estimated N inputs that were calculated from estimated wet and dry deposition data (Ollinger et al. 1993). Gaseous N_2O losses were estimated using previously reported values (Bowden et al. 1991) and 1994 estimated losses. Leaching losses were estimated as lysimeter inorganic-N concentrations multiplied by the volume of water leaching below the rooting zone, the latter calculated using the monthly carbon-and-water-balance model known as "PnET II" (Aber et al. 1995). Total losses (trace gas plus leaching) were subtracted from total inputs, yielding the total amount of N retained by the ecosystem.

Net change in N storage was calculated for each ecosystem component by taking the difference between year 6 and year 1 values (extractable N, woody biomass, foilar N). Foliar N storage for a given year was calculated by species as percentage N in green foliage times litterfall biomass. Red pine values were multiplied by 2.5 to account for estimated needle-retention time.

Calculating changes in fine-root N standing stocks over the period was more complicated since different methods were used for biomass estimation in 1988 and 1991. The 1988 sampling included larger size classes (0–2 mm in 1988 vs. 0–1 mm in 1991) and contained no treatment effects. The 1991 sampling was less inclusive, but did compare treatment effects over the first 3 yr. In order to calculate changes in the fine-root nitrogen pool over the treatment period, we assumed that the initial survey (1988) provided a more accurate estimate of total fine-root mass whereas the 1991 study reflected the effects of treatments on relative biomass and N concentration. Total N content in 1988 was therefore estimated as measured biomass times measured N concentration, and averaged to get one initial value for each stand. In 1991 the control plot total N content was calculated as 1988 biomass times 1991 control N concentration. To add the apparent effects of treatment on changes in biomass, 1991 total fine-root N content in the treated plots was estimated as follows: (1988 biomass times the ratio of 1991 treatment biomass)/(1991 control biomass times 1991 treatment N concentration). This may overestimate actual differences if the 1–2 mm roots did not change in N concentration as much as the 0–1 mm roots, but is conservative in that the 1988 to 1991 changes are not predicted into the future.

Mean annual N cycling rates for the 6-yr treatment period were estimated in order to determine the fraction of added N which could have been incorporated into soil organic matter through increased plant uptake and subsequent litter production. A simple budget was constructed for each plot using mean annual measured N fluxes of net mineralization, litterfall, wood production, leaching, and deposition + fertilization, along with mean annual changes in pool sizes for foliage, roots, and the extractable soil pool. Fine-root litter input to the soil was estimated as the average root pool size multiplied by a turnover rate. The turnover rates, which were estimated using total N availability in each site and data from Aber et al. (1985), were 0.75, 1.0, and 1.5 for the pine control, low, and high plots, respectively, and 0.6, 0.8, and 1.2 for hardwood control, low, and high plots, respectively. N cycling rates were not calculated for the N+S treated plots.

Statistical calculations

For all time-series data, we used the least significant difference (LSD) calculation, given as:

$$s\sqrt{2/n} \times t_{h(n-1)}$$

where s in the within-sample estimate of σ_0 and h $(n - 1)$ is the number of degrees of freedom of this estimate (Snedecor and Cochran 1967, Miller and Miller 1988). All LSD values were calculated at $P = 0.05$. Analysis of variance was used to determine significant differences between treatments for soil N pools and trace-gas data at the $P = 0.05$ and $P = 0.005$ levels, respectively.

Results

Foliage

Foliar N concentration in the hardwood stand increased over the course of the experiment, in response to nitrogen treatments (Fig. 1a,b,c). By year 6, per-

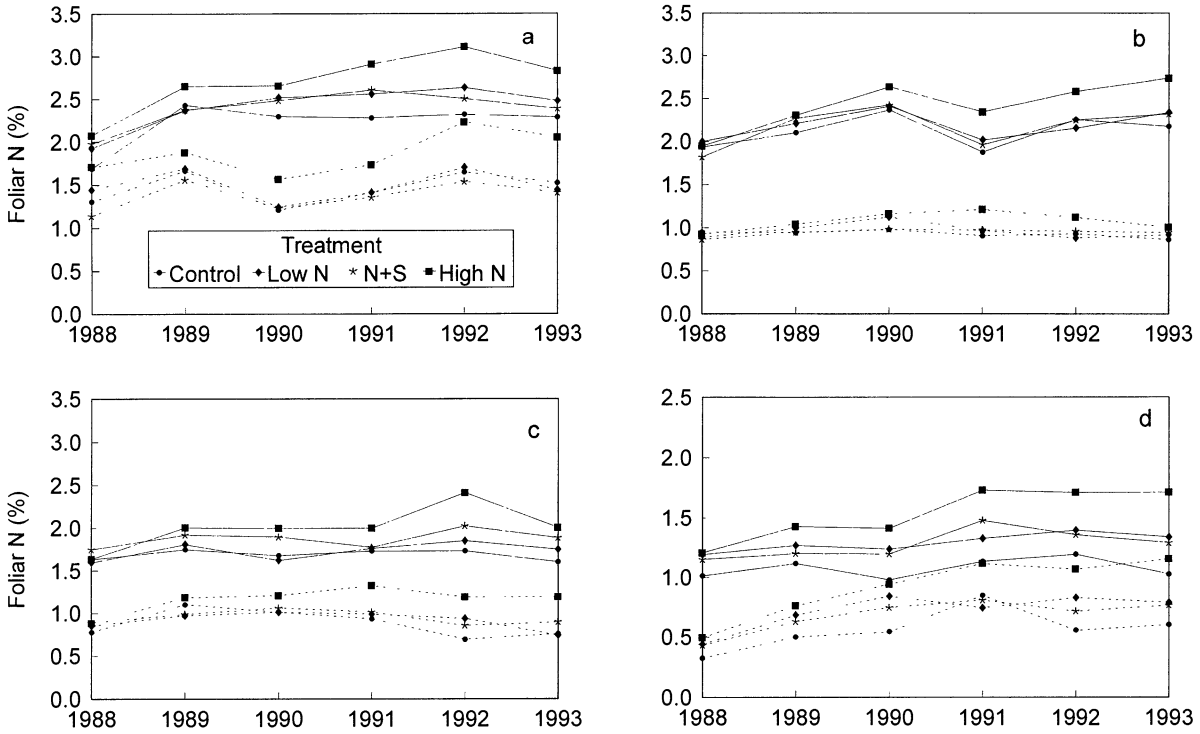

FIG. 1. Green foliar and litterfall nitrogen values for the dominant tree species on the chronic-N-addition plots in Harvard Forest (Massachusetts, USA): (a) black birch, (b) black oak, and (c) red maple from the hardwood stand and (d) red pine from the pine stand. In each graph, the upper four lines represent green-foliage N and the lower four lines represent litterfall N. Least significant difference (LSD) values are as follows: (a) black birch green foliage LSD = 0.093, litterfall LSD = 0.085; (b) black oak green foliage LSD = 0.099, litterfall LSD = 0.042; (c) red maple green foliage LSD = 0.093, litterfall LSD = 0.051; (d) red pine green foliage LSD = 0.066, litterfall LSD = 0.049.

centage N in black oak, red maple, and black birch foliage averaged 25% higher in the high-N plot than in the controls. Black birch percentage N in the control was anomalously low in the first year, resulting in a large difference between the high and control plots for that year only. In the pine stand (red pine only) differences between treatments were apparent in the 1st yr and increased substantially over the 6-yr period (Fig. 1d). By 1993, foliar N in the low-N and high-N plots were 30% and 67% higher, respectively, than in the control. Absolute percentage N concentrations in red pine foliage increased in the high-N plot for the first 4 yr and remained stable from 1991 through 1993. Differences between treatments occurred against inter-annual background variation of as much as 25% in the control plots for all species of green foliage and all hardwood litterfall. Annual litterfall N concentration in red pine fluctuated up to 50% between years. There was no apparent decrease in N retranslocation with increased N availability due to fertilization (Table 2).

TABLE 2. Percentage nitrogen retranslocation for each species and treatment. Data are means for the 6-yr period, with 1 SE in parentheses.

Stand	Species	Treatment			
		Control	Low N	N+S	High N
Hardwood	Black oak	56 (1.6)	56 (1.4)	56 (1.6)	55 (2.0)
	Red maple	48 (3.6)	47 (2.7)	49 (2.2)	42 (2.4)
	Black birch	34 (3.5)	38 (4.0)	42 (2.4)	31 (3.6)
Pine	Red pine	48 (5.9)	44 (4.2)	47 (3.5)	41 (4.2)

Note: N retranslocation was calculated as follows: [(green-foliage percentage N) − (litterfall percentage N)] ÷ (green foliage percentage N).

Biomass

Annual aboveground net primary production (NPP; litterfall plus wood) in the hardwood stand was greatest for the high-N plot, due primarily to increases in woody increment (Table 3). In the pine stand, total litterfall in the high-N plot increased, but tree growth (averaged over the 5-yr period) was lower than in all other plots. Previously reported values for the first 3 yr of the experiment (Aber et al. 1993) showed no difference in aboveground NPP between treatments in the pine stand.

Fine roots

Measured fine-root nitrogen content (Table 4) in control stands was comparable to values measured by McClaugherty et al. (1982) in a similar set of paired

TABLE 3. Total aboveground annual net primary production (in units of $Mg \cdot ha^{-1} \cdot yr^{-1}$) in the chronic-N-addition plots at Harvard Forest. Data are 5-yr averages for the years 1989 through 1993.

	Pine stand treatments				Hardwood stand treatments			
	Control	Low N	N+S	High N	Control	Low N	N+S	High N
Woody increment	3.3	3.3	2.4	2.0	4.5	4.8	5.1	6.5
Litterfall	3.2	3.7	3.8	4.1	2.9	2.9	3.0	3.3
Total	6.5	7.0	6.2	6.1	7.4	7.7	8.1	9.8

stands at the Harvard Forest (42 and 65 kg N/ha in live roots, in a red pine and mixed-hardwood stand, respectively). Nitrogen concentrations increased with N additions and the relative increases were higher in the pine stand than in the hardwood stand. In contrast, differences in estimated fine-root biomass were greater in the hardwoods than in the pines. Taken together, changes in both biomass and N concentration in this compartment result in a substantial estimated increase of total N in fine roots in response to chronic N additions.

Lysimeter N concentrations

Nitrate concentrations in the pine high-N plot began to increase in the latter part of 1989 and averaged ≈5 mg NO_3-N/L throughout 1990 (Fig. 2a). Concentrations increased every year to a maximum of 25 mg NO_3-N/L in 1993 (n = 4 or 5). The low-N plot showed no detectable NO_3 (limit of detection 0.2 mg/L) until 1993, when samples ranged from 2 to 6 mg NO_3-N/L. Small amounts of NH_4-N were detected in the pine high-N plot in 1993 (Fig. 2c). Nitrate and ammonium concentrations in the hardwood stand were near zero for all but one collection (30 July 1992) through 1993 (Fig. 2b and d).

Net mineralization and nitrification

Net N mineralization rates in control plots were relatively constant from year to year (Fig. 3a and b). In 1988, annual net mineralization increased significantly in the N-treated plots, for both the pine and hardwood stands. Net mineralization rate in the pine high-N plot was 30% lower in 1990 than in 1988 whereas the pine control plot was only 12% lower. By 1993, net mineralization in the pine high-N plot (96 $kg \cdot ha^{-1} \cdot yr^{-1}$) was lower than net mineralization in the pine low-N plot (110 $kg \cdot ha^{-1} \cdot yr^{-1}$). In the hardwood stand, annual N mineralization in the high-N plot increased 50% between 1990 and 1991, and by 1993 total mineralization had doubled its 1990 value.

Nitrification rates doubled in the pine high-N plot between 1988 and 1993, with most of this increase occurring in the mineral soil (Fig. 3c). As a result, nitrification increased from 17% of mineralization in 1988 to 51% of mineralization in 1993 for the pine high-N plot. Nitrification response was small in the hardwood stand, with only a slight increase occurring in the high-N plot in 1993 (Fig. 3d).

Extractable ammonium and nitrate

Mean annual extractable NH_4-N was greatest in the high-treatment plots of both stands. Pine high-N plot NH_4-N concentrations were 60% higher than the control in 1988 and 120% higher than the control in 1993. Similarly, differences between the high-N and control plots in the hardwood stand were 15% and 150% for 1988 and 1993, respectively (Fig. 4a and b). Extractable NO_3-N concentrations in the pine high-N plot increased five-fold from 1988 to 1993 whereas control plot concentrations decreased from 1988 to 1993 (Fig. 4c and d). Soil extracts yielded <1.5 kg/ha of NO_3-N

TABLE 4. Comparison of pre-treatment (1988) and year 4 (1991) fine-root characteristics in chronic-N-addition plots at Harvard Forest. Mineral soil was sampled to a depth of 10 cm; roots were not collected from N+S plots. Biomass differences between 1988 and 1991 are not directly comparable due to (1) inclusion of both live and dead roots in 1988; (2) inclusion of only select species in 1991; and (3) size-class differences between 1988 (0–2 mm) and 1991 (0–1 mm).

		Pine stand				Hardwood stand			
		Initial	1991			Initial	1991		
		1988	Control	Low N	High N	1988	Control	Low N	High N
Percentage N	Organic	1.6	1.7	2.1	2.5	1.2	1.3	1.4	1.4
	Mineral	1.2	1.2	1.7	2.2	0.9	1.1	1.1	1.3
Biomass (kg/ha)	Organic	1445	332	528	308	3324	427	765	713
	Mineral	1594	526	830	744	2609	289	387	509
Estimated total N content (kg/ha)†	Organic	23.1	23.8	47.8	33.5	39.9	43.2	84.6	79.4
	Mineral	19.1	19.8	43.5	49.4	23.5	27.9	36.7	58.8
	Total	42.2	43.6	91.3	82.9	63.4	71.1	121	138

† Values for 1988 are percentage N times biomass; 1991 values are estimated. See *Materials and methods: Budget calculations* for the 1991 calculations.

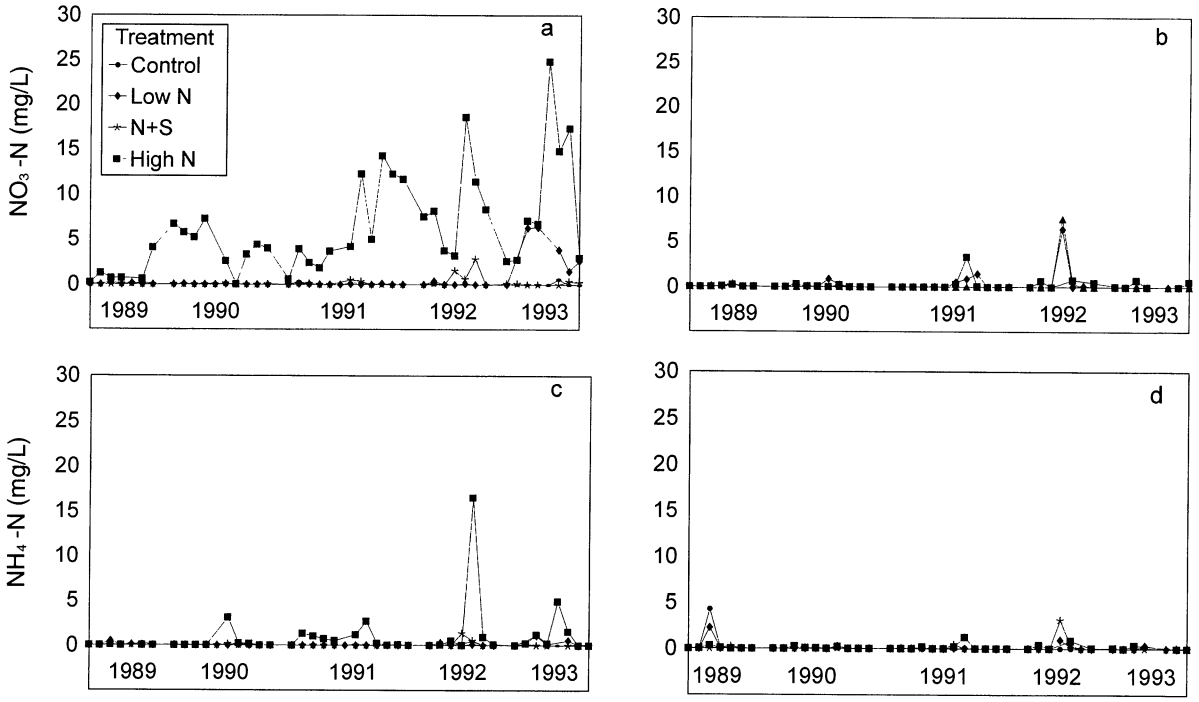

FIG. 2. Soil-solution nitrate concentrations in the (a) pine stand and (b) hardwood stand, and ammonium concentrations in the (c) pine stand and (d) hardwood stand from 60-cm tension lysimeters for the four different treatments. Lysimeters were sampled approximately every 4 wk from May through November of each year. Least significant difference (LSD) values are as follows: pine nitrate LSD = 4.13; hardwood nitrate LSD = 0.819; pine ammonium LSD = 0.735; and hardwood ammonium LSD = 0.361.

for all plots in the hardwood stand throughout the experiment.

Total N and the C:N ratio

Initial N content was not significantly different ($P = 0.05$) between treatments within the same stand for either soil horizon (Table 5). Total N was calculated as percentage N of individual samples times a mean soil-core dry mass determined for each plot ($n = 282$ soil cores). Due to the small number of initial samples ($n = 6$ and $n = 9$ for the organic and mineral soils, respectively), variability was high and significant differences between years within a plot were not discernible.

Trace gases

The average rate of N_2O emissions (Fig. 5) from the pine high-N plot (9.43 ± 0.54 $\mu g \cdot m^{-2} \cdot h^{-1}$; mean ± 1 SE) was significantly greater than the average rate from the pine control plot (4.17 ± 0.42 $\mu g \cdot m^{-2} \cdot h^{-1}$) and is twice the maximum summer flux rate (3.88 ± 2.19 $\mu g \cdot m^{-2} \cdot h^{-1}$) observed at these plots during the first 2 yr of the study (Bowden et al. 1991). An approximate total growing-season N_2O flux was calculated using the average summer rate (9.43 $\mu g \cdot m^{-2} \cdot h^{-1}$) × 24 h/d × 180 d during the growing season, and equaled ≈0.065 kg $N \cdot ha^{-1} \cdot yr^{-1}$. Although effluxes of N_2O were consistently greater in the pine high-N plot than in the pine control plot, there was no observed N_2O pulse immediately following fertilizer application.

Nitrogen budgets

Calculated 6-yr nitrogen input–output and pool storage budgets (Table 6) changed substantially from similar calculations for the first 3 yr of treatments (Aber et al. 1993). Nitrogen leaching losses increased dramatically in the pine high-N plot (22 kg N/ha for the first 3 yr, 105 kg N/ha for the second 3 yr). Foliar N storage increased substantially, particularly in the hardwood stand, resulting from increases in both foliar percentage N and total litterfall biomass. One of the largest measured N storage pools was fine roots, a pool that was ignored in calculating the 3-yr budget due to lack of data.

Mean annual N cycling rates (Fig. 6) suggest that a large portion of the added N that now resides in soil organic matter could have been incorporated through increased litter production. The cycle in the hardwood control plot is balanced within the error limits of the measurements and calculations performed, and all N was accounted for. In the hardwood low-N plot, calculated N uptake increased by 75% due to increases in fine-root turnover, a large annual increase in N content of soil organic matter was predicted, and the cycle was also nearly balanced. In the hardwood high-N plot, fine-root turnover increased further as did soil organic N, but there was still an excess of N inputs to the available soil pool beyond what was accounted for by estimated plant uptake, leaching losses, and changes in the ex-

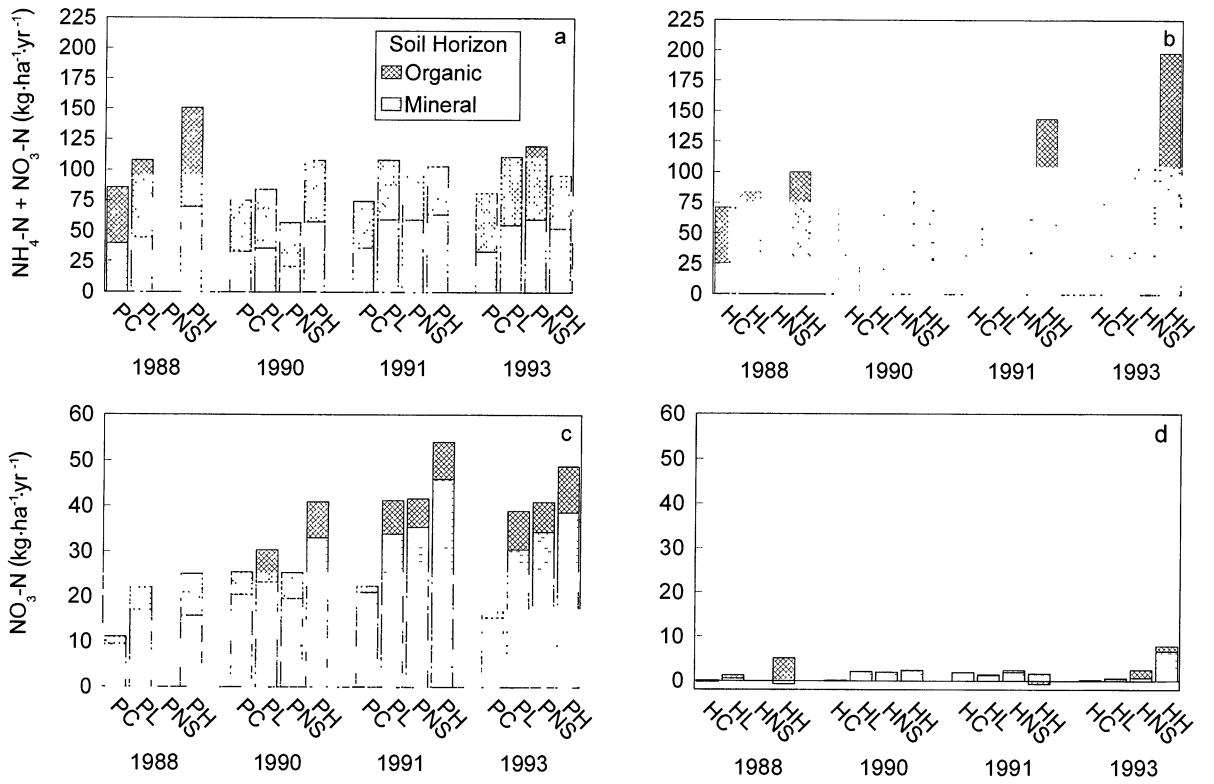

FIG. 3. Annual net mineralization (NH_4-N + NO_3-N) for the (a) pine stand and (b) hardwood stand, and annual net nitrification (NO_3-N) for the (c) pine stand and (d) hardwood stand. Values are the sums of buried-bag incubations for the 12-mo period from May of the indicated year to May of the following year for both the organic and mineral horizons. Letters below bars distinguish N treatments: P = pine, H = hardwood, C = control, L = low N, NS = N+S, H = high N. Complete data are not available for the N+S plots in 1988.

tractable pool. Similar patterns were seen in the pine stand, although an excess of available N was predicted for the control plot as well as the treated plots. Although ^{15}N tracer techniques suggest increased turnover rates in roots in treated stands (Hendricks 1994), estimates of fine-root turnover rate are the largest source of error in these budgets.

DISCUSSION

Chronic nitrogen additions at the Harvard Forest, specifically the high-N treatments, have resulted in considerable alterations of the nitrogen cycle in both the pine and hardwood stands. Substantial changes were observed in nearly all ecosystem pools and processes investigated. The four primary results discussed here are: (1) changes in nitrogen cycling rates; (2) differences in response between the two forest stands; (3) the large amounts of nitrogen still being retained by both stands; and (4) possible mechanisms for N retention.

Changes in N cycling

Many of the changes in nitrogen pools and processing that were hypothesized as indicators of N saturation (Aber et al. 1989) are beginning to occur. Foliar nitrogen levels continue to increase after 6 yr of treatments in all but the pine high-N plot, where a maximum percentage N appears to have been reached. Higher foliar N concentrations result in increased N-cycling rates as more nitrogen is returned to the forest floor in litterfall.

Soil response includes nitrification as an increasing portion of total net mineralization and subsequently larger nitrate pools in the pine plots. Increases in net N mineralization and extractable NH_4 pools in the hardwood high-N plot suggest that nitrification may become significant in the near future, assuming that nitrifiers are currently NH_4 limited. Nitrate may be assimilated by the vegetation, but only if the species present can afford the high energy costs involved in nitrate reduction in roots, or can utilize extra reductant from the light reactions of photosynthesis to reduce nitrate in foliage (Nadelhoffer et al. 1984). If nitrate is not taken up by plants or incorporated by the microbial population, it will leach from the soil accompanied by base cations such as calcium and magnesium (Smith 1990, Schlesinger 1991). Removal of these other nutrients, and their replacement with hydrogen, could lead to a reduction in soil nutrient status and reduced plant growth, despite the availability of additional nitrogen.

Effects of these changes may already be starting in the pine high-N plots, evidenced by both high levels of nitrate leaching and the reduction in woody biomass

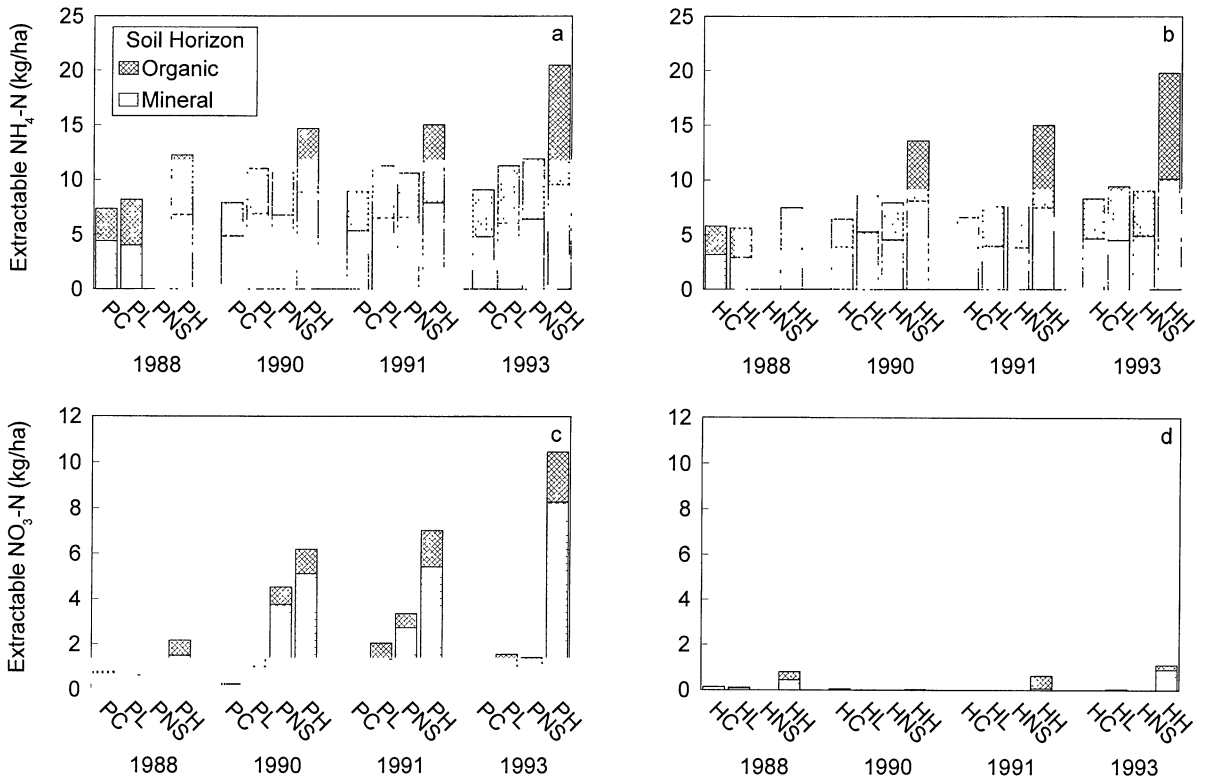

FIG. 4. Mean annual extractable ammonium for the (a) pine stand and (b) hardwood stand, and mean annual extractable nitrate for the (c) pine stand and (d) hardwood stand. Values are the averages of initial buried-bag samples for both the organic and mineral horizons. Complete data are not available for the N+S plots in 1988. Least significant difference (LSD) values are as follows: pine ammonium organic LSD = 1.38, mineral LSD = 1.53; hardwood ammonium organic LSD = 1.00, mineral LSD =1.13, pine nitrate organic LSD = 0.341, mineral LSD = 1.30; and hardwood nitrate organic LSD = 0.150, mineral LSD = 0.233. Bar letter codes are as in Fig. 3.

accumulation as compared with other plots in the pine stand. Wood growth in the pine high-N plot was only two thirds of the control plot over the 6-yr period, indicating that excessive nitrogen additions may actually be detrimental to plant growth. Similar results have been reported for spruce forests in central Europe (Schulze 1989) and in N-amended spruce–fir forests in Vermont (McNulty et al. 1996).

TABLE 5. Total soil nitrogen (in units of kg/ha) pools in the chronic-N-addition plots at the Harvard Forest. Values are means with 1 SE in parentheses. Mineral soil was sampled to a depth of 10 cm. Sample size varied: 1988 organic horizon, n = 6; 1988 mineral horizon, n = 9; 1992 all samples, n = 12.

	Pine stand			Hardwood stand		
Total N, kg/ha	Control	Low N	High N	Control	Low N	High N
1988						
O horizon	735 (61)	1071 (73)	1004 (124)	757 (62)	588 (66)	934 (141)
M horizon	1814 (180)	1857 (135)	1810 (157)	2426 (848)	1666 (159)	1356 (145)
Total	2549	2928	2814	3183	2254	2290
1992						
O horizon	1094 (75)	927 (75)	1008 (100)	1142 (87)	1100 (84)	1276 (136)
M horizon	2506 (158)	2172 (110)	2309 (161)	1855 (159)	2643 (248)	2845 (398)
Total	3600	3099	3317	2997	3743	4121
Change from 1992 to 1988	1051	171	503	−186	1489	1831

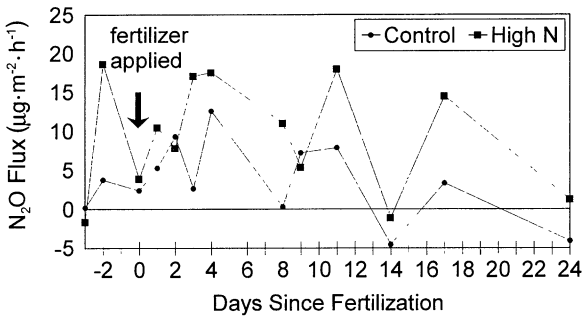

FIG. 5. Nitrous oxide fluxes prior to and following fertilizer applications in mid-July 1994 for the control and high-N plots in the Harvard Forest pine stand.

N_2O flux data further indicate the impact of changes in N cycling due to chronic N additions as flux rates increased by a factor of two over the course of the experiment. Whereas these rates are less than rates observed in other temperate forests (e.g., Goodroad and Keeney 1984, Schmidt et al. 1988) and are still an order of magnitude lower than rates observed in a German forest subjected to high levels of atmospheric N deposition (Brumme and Beese 1992), they do indicate that N deposition can stimulate N-cycling processes (particularly nitrification) that are important in controlling rates of N_2O emissions.

Influence of initial stand characteristics

The marked difference in response time between the pine and hardwood stands suggests that there were differences in initial ecosystem nitrogen status. Measured net N mineralization rates support this suggestion. The average annual net N mineralization rate in the hardwood control plot (74 $kg \cdot ha^{-1} \cdot yr^{-1}$) was low compared to 24 temperate hardwood stands (53–135 $kg \cdot ha^{-1} \cdot yr^{-1}$) in Wisconsin and New England (Aber et al. 1991). Mean net N mineralization rate for the pine control plot (79 $kg \cdot ha^{-1} \cdot yr^{-1}$) was among the highest rates for pine stands (28–81 $kg \cdot ha^{-1} \cdot yr^{-1}$).

Prior land-use history may be critical in determining current nitrogen status of soils and forests. Forest stand inventory records from the Harvard Forest archives indicate that the red pine plantation was plowed and pastured up to the time of planting in 1926. Higher mineralization rates in the pine stand may reflect soil/site enrichment prior to the plantation, although it is unclear the extent to which cropping, and therefore biomass and nutrient removal, occurred on the site. The hardwood stand was never plowed, as evidenced by high soil stone content and the lack of adjacent stone walls. However, it was clearcut as part of a clean-up effort several years after the 1938 hurricane and was then allowed to regenerate without further improvements. Its location next to a plowed field may have resulted in frequent coppicing for fuel and timber during the agricultural period. Thus, the combination of different inherent site fertilities, along with different land-use histories, may have influenced the differences in N-cycling rates between the two stands.

Potential nitrification rates in currently forested

TABLE 6. Inputs, outputs, retention, and estimated distribution of nitrogen after 6 yr of ammonium nitrate additions at the Harvard Forest. For more information on calculations, see *Materials and methods: Budget calculations.*

	Pine stand				Hardwood stand			
	Control	Low N	N+S	High N	Control	Low N	N+S	High N
N inputs (kg/ha)								
Atmospheric deposition	48	48	48	48	48	48	48	48
Fertilization	0	276	276	826	0	276	276	826
Total	48	324	324	874	48	324	324	874
N losses (kg/ha)								
Gaseous (N_2O)	<1	<1	<1	<1	<1	<1	<1	<1
Leaching (NO_3 + NH_4)	2	6	4	127	1	6	4	5
Total	2	6	4	127	1	6	4	5
N retention								
Total (kg/ha)	46	318	320	747	46	318	320	869
As % of N inputs	96	98	99	85	98	98	99	99
Change in N storage (kg/ha)								
Soil extractable N	1	4	4	17	2	4	5	13
Woody biomass	30	29	22	18	40	44	46	58
Foliage	13	25	18	50	21	21	59	45
Fine roots	2	49	49	41	8	59	59	75
Total measured N retention	46	107	93	126	71	128	169	191
Non-extractable soil pool retention†	0	211	227	621	−24	190	151	678
Soil retention (%)	0	66	71	83	(100)	60	47	78

† Non-extractable soil retention = unmeasured N retention, and is calculated as (total N retained) − (total measured N retention).

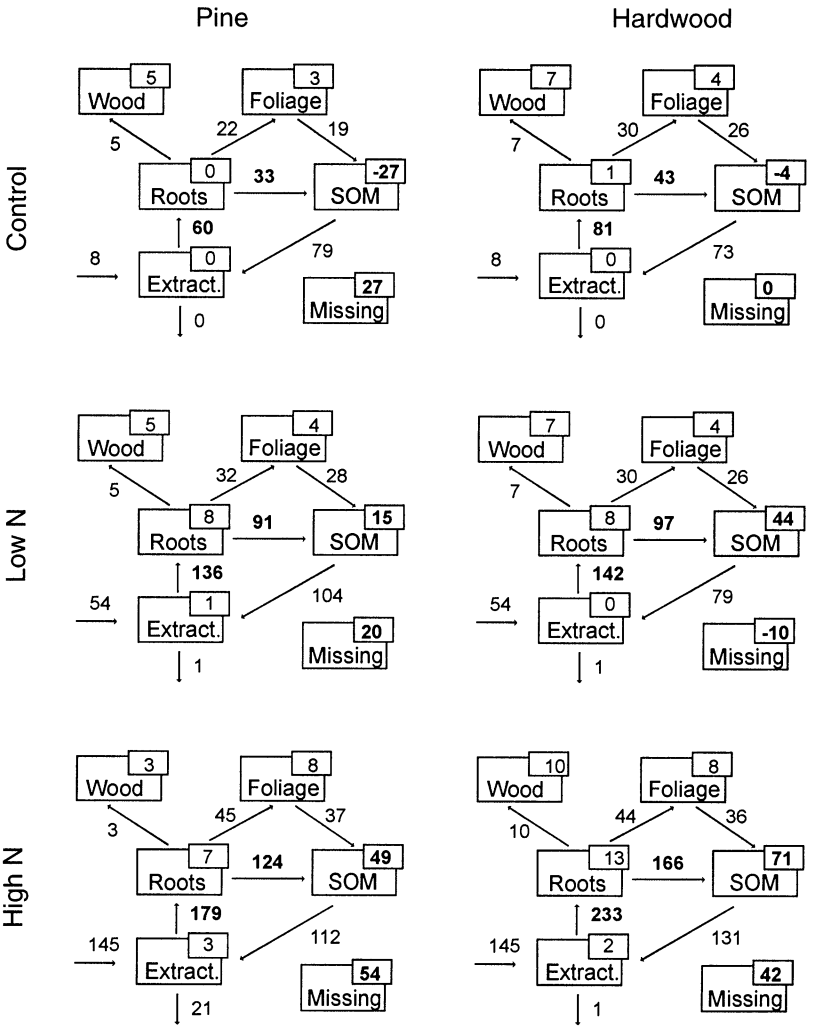

FIG. 6. Estimated average annual N cycling rates for the control, low-N, and high-N plots at the Harvard Forest. Bold numbers are calculated; all others are measured. Values within boxes are net annual changes in storage; flux values are adjacent to arrows; all data are kg/ha. "Missing" N is calculated as (net N mineralization + deposition + fertilization) minus (plant uptake + leaching loss + increase in extractable N pool). "SOM" is the soil organic matter pool, and "Extract." is the extractable soil nitrogen pool.

stands with differing prior land use were measured at the Harvard Forest. Formerly plowed or pastured sites, on which trees established ≈100 yr ago, had higher potential mineralization rates than those that had been in permanent woodlots (J. Compton, *personal communication*). These findings support the observed differences between the pine stand (previously pastured) and the hardwood stand (woodlot), and indicate that past disturbance may affect site fertility for decades.

Stand age and vitality is another factor to consider in determining the nutrient requirements of a forest (Pritchett 1979). An aggrading forest will more readily take up additional N inputs compared to a steady-state forest with slower growth and therefore lower nutrient requirements (Vitousek and Reiners 1975, Bormann and Likens 1979). In the hardwood stand (50 yr old), woody biomass production was 50% higher in the high-N plot than in the control plot, and nitrate was not detected in soil solution from any treatment in that stand. In contrast, tree growth in the pine stand (70 yr old) was lower in the high-N plot than in the control plot, and both nitrification and nitrate leaching increased substantially over the 6-yr period. This difference in response suggests that the pine stand was less N-limited initially and therefore closer to nitrogen saturation. Mitchell et al. (1992), in a comparison of two hardwood forests, found substantially higher NO_3 leaching in the mature forest (150–300 yr old) than the younger forest (100 yr old) even though the latter had shown signs of impact from acid deposition.

Ecosystem N retention

Budget calculations show that 85–99% of the added nitrogen was retained for both forest types (Table 6).

Retention in the pine high-N plot was 95% after the first 3 yr of applications but only 85% for the total 6-yr period, indicating that the retention capacity of the ecosystem has decreased since 1990. Increases in fine-root, foliar, woody biomass, and extractable soil nitrogen pools could account for only 17–22% of the retained N in both the pine and hardwood high-N plots. The remaining N is assumed to be in the recalcitrant, non-extractable soil pool.

Quantitatively, measured changes in total N content from 1988 to 1992 are large enough to account for the retained N. However, these data are too variable to be statistically significant. The inherent heterogeneity of soils, combined with uncertainty due to small sample size (n = 6 to 12) and the limited sensitivity of the CHN analysis, makes it impossible to confidently draw conclusions about changes in the soil pool. An additional study using a ^{15}N tracer was initiated on the chronic N plots in 1991 (K. Nadelhoffer, *personal communication*). Results from that study should aid in determining changes in N storage within the various soil pools.

Absence of an observable increase in N_2O emissions in the first 2 wk following fertilizer additions suggests that we did not miss important N_2O losses by measuring trace-gas fluxes monthly. Due to the well-drained nature of the chronic N soils, anaerobic conditions that typically favor denitrification (Robertson and Tiedje 1987, Firestone and Davidson 1989, Davidson et al. 1990) should not prevail at this site. Additionally, soil pH values are low enough (<4.0) to inhibit N_2 production during denitrification (Firestone and Davidson 1989). Ammonia volatilization losses are also expected to be minimal due to the low pH of the fertilizer (5.9) as well as the soils themselves (Bowden 1986, Killham 1994). Production of nitric oxide can occur during both nitrification and denitrification (Galbally 1989, Davidson et al. 1993), but net losses via this avenue are not well quantified for temperate forests. Although these other potential pathways of gaseous N loss were not measured, it is unlikely that any omitted fluxes would substantially influence the N budgets.

Mechanisms of N retention

Four mechanisms for incorporation of added N into soil organic matter (SOM) can be postulated: (1) litter production and decomposition; (2) increased incorporation into dissolved organic matter (DOM); (3) biotic immobilization into SOM and; (4) abiotic incorporation into SOM.

The calculated N-cycling rates in Fig. 6 suggest that increased plant uptake and litter production, particularly through fine roots, can account for a substantial fraction of N incorporation. Total plant N uptake is estimated to have nearly tripled between the control and high-N treatments in both stands. This calculation is based on both measured increases in the N concentration of roots (Table 4) and on estimated increases in fine-root turnover. Hendricks (1994) measured the dilution of ^{15}N in the live fine-root pool following the cessation of ^{15}N additions in the pine control and low-N plots and estimated that fine-root turnover had increased substantially with treatment.

Currie (1995, and Currie et al. 1996) measured 40–60% higher dissolved organic nitrogen (DON) concentrations in forest-floor leachate from the high-N plots, as compared with the control plots, for both stands. This occurred without any clear change in dissolved organic carbon (DOC) flux, but through a narrowing of the DOC:DON ratios. DON losses represented only 4–6 kg $N \cdot ha^{-1} \cdot yr^{-1}$, and are therefore not a significant N sink in the treated plots as compared with the 15- to 50-fold increase in NO_3-N and NH_4-N leaching losses.

Micks (1994) looked at the effect of N additions on microbial immobilization through measurements of soil respiration following N additions. CO_2 flux was measured (soda lime technique) over 24-h periods for 24 d in July and August of 1992, on the control and high-N plots as well as on a new set of high-N plots. The expected increase in CO_2 flux resulting from N incorporation into microbial biomass was estimated using measured rates of mineral-N disappearance and published ranges for microbial the C:N ratios and carbon-use efficiency (Schimel 1988). Differences in respiration rates between treatments were rarely significant, and were much lower than predicted by N disappearance rates. This suggests that if microbial uptake was important, it must have been accompanied by changes in the C:N ratios in the microbial population rather than by increased biomass production.

Abiotic incorporation of N into organic materials as a process of N retention has been known for decades (e.g., Burge and Broadbent 1961, Nommik and Vahtras 1982), but has received little attention in forest N-cycling studies (Johnson 1992). Short-term studies of ^{15}N immobilization into soils reveal that a significant portion is incorporated into non-extractable or recalcitrant pools (e.g., Vitousek and Matson 1985). Foster et al. (1985) measured forest-floor material in a jack pine stand following urea amendments and found chemically fixed N exceeded biotic incorporation. In the laboratory, Axelsson and Berg (1988) showed that abiotic incorporation of ^{15}N into leaf litter of different ages was directly related to N concentration in the amendment and inversely related to N concentration in the litter. Strickland et al. (1992) demonstrated rapid incorporation of ^{15}N into a non-microbial, physically shielded soil pool (slow turnover pool) but stated that the mechanisms by which that is accomplished need further study. Most ^{15}N pool-dilution studies show a near-immediate disappearance of labelled N, which has been attributed to abiotic processes (e.g., Schimel and Firestone 1989). Their estimated 20% abiotic incorporation value, if applied to the calculated N cycles (Fig. 6), would account for 50–60% of the "missing"

N in the pine low-N and high-N plots and in the the hardwood high-N plot.

Conclusions

It is evident from the results of this experiment that ecosystem response to 6 yr of chronic N additions is highly non-linear over time, and that very different conclusions would have been drawn from a study limited to 1 or 2 yr. Although many of the processes that drive ecosystem nitrogen dynamics are well understood, the central question regarding mechanisms of N incorporation and retention by the soil pool still remains unanswered. Alterations of the N cycle, as observed in these stands, could have serious implications for both forest and human health if such changes were to occur on a regional scale. The ability of forest soils to retain added N is the most critical remaining unknown in the understanding of nitrogen saturation. The effect of historical patterns of land use, extending decades to centuries into the past, may be an important determinant of soil N-retention capacity. Interactions between inorganic N and native organic matter in forest soils, and the relationship of these interactions to previous site history, is perhaps the area in which future research will yield the greatest rewards.

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