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Decadal-scale fates of ^{15}N tracers added to oak and pine stands under ambient and elevated N inputs at the Harvard Forest (USA)

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Abstract

We used isotope mass balances to track the fates of ecosystem-scale ^{15}N tracer additions to control non-fertilized and nitrogen (N)-fertilized plots in oak-dominated hardwood and red pine-dominated pine over nine growing seasons. The plots used in our tracer study are the same as of those used in the Chronic Nitrogen Amendment study (see Magill et al., this volume) at the Harvard Forest in central Massachusetts, USA. Nitrogen tracers were added monthly during growing seasons (April–October) of fertilization years 4 and 5 (1991 and 1992) as either $^{15}NH_4$ or $^{15}NO_3$ to control and to N-fertilized plots (low N treatment, receiving additions of 5 g N m^{-2} per year as NH_4NO_3). Seven years after the end of ^{15}N additions (1999), tracer recoveries were highest in organic soil, ranging from 34 to 83% of additions across ^{15}N ion form, N loading rate and forest type. Mineral soil (0–20 cm depth) was the second largest sink, with tracer recoveries ranging from 15 to 34% of additions. Tree biomass (foliage, woody tissue, plus fine roots) accumulated 7–16% of tracer additions. Patterns of ^{15}N recoveries after 7 years differed from recovery patterns at the end of the 2-year labeling period. At the end of labeling in 1992, recoveries in tree biomass were higher under chronic N fertilization than under ambient atmospheric N loading (control plots), higher with $^{15}NO_3$ than $^{15}NH_4$ additions, and higher in hardwoods than in pines. Seven years after the end of labeling, recoveries in trees were still higher with $^{15}NO_3$ than with $^{15}NH_4$ labeling, and trees in control plots continued to accumulate ^{15}N tracers. In contrast, tracer recoveries in trees on the chronically fertilized plots were lower after 7 years than at the end of the labeling period. This was due mainly to transfers of ^{15}N tracer from leaf and root tissue to other ecosystem pools during the years since labeling. Wood produced during the 2-year labeling period and the 7-year post-labeling interval continued to accumulate ^{15}N tracers in control and chronically fertilized plots in both forest types. However, recoveries were low in woody tissues (wood formed from 1990 through 1999 plus bark), ranging from 1.1 to 6.1% of ^{15}N additions across forest types and treatments. Our comparisons of ^{15}N tracer recoveries at the end of a 2-year labeling period and at 7 years following the end of labeling suggest that soils are the dominant sinks for N deposition on temperate forests and that N does not contribute substantially to enhanced carbon uptake into tree biomass.

Keywords: N cycling; ^{15}N tracers; Temperate forests

1. Introduction

Forest responses to elevated inputs of reactive nitrogen (sensu Galloway, 1998) have been subject

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to considerable investigation during the past two decades (e.g. Nihlgård, 1985; van Breemen and van Dijk, 1988; Aber et al., 1989, 1998; Schulze, 1989; Wright et al., 1995; Gundersen et al., 1998). Labeling forests with ^{15}N, the heavier and less abundant of the two stable isotopes of nitrogen (^{14}N and ^{15}N), has provided information about the fates of N inputs to forests (Preston et al., 1990; Emmett and Quarmby, 1991; Nadelhoffer et al., 1992; Preston and Mead, 1994; Buchmann et al., 1995; van Dam and van Breemen, 1995; Jordan et al., 1997; Seely and Lajtha, 1997; Tietema et al., 1998; Schleppi et al., 1999a) and has led to insights regarding the influences of N deposition on forest and global-scale carbon budgets (Nadelhoffer et al., 1999b). Measurements of tracer recoveries from ^{15}N-labeled forest ecosystems also have been used to develop and test biogeochemical models of N dynamics (van Dam and van Breemen, 1995; Currie and Nadelhoffer, 1999; Currie et al., 1999).

We report here on recoveries of ^{15}N tracers applied to control and chronically fertilized plots sampled 7 years after the end of a 2-year labeling period (1991 and 1992 growing seasons). The large plots to which ^{15}N was added are a subset of the plots in a hardwood and a pine forest receiving additions of ammonium nitrate (NH_4NO_3) fertilizer from 1988 through the present as described by Magill et al. (this volume). We also compare the 7-year post-labeling results with tracer recoveries measured at the end of the 1991–1992 labeling period. Our objective was to determine (1) if the dominance of soils over trees as a sink for N inputs had shifted nearly a decade after labeled N was applied to plots, (2) if a greater proportion of N inputs resided in tree biomass under chronic fertilization, and (3) whether the greater sink strength of trees for nitrate than for ammonium additions was maintained during the decade following tracer additions. Finally, our ^{15}N recovery data were used to test model-based predictions of the effects of chronic N deposition on forest C and N cycling (Currie et al., this volume).

2. Methods

2.1. Study site

The hardwood and pine forests we investigated here are located in the Prospect Hill tract of the Harvard Forest (42°30′N, 72°10′W) in Petersham, Massachusetts, USA. The hardwood forest regenerated after cutting in the mid 1940s and is dominated by oaks (*Quercus velutina* Lam.; *Q. rubra* L.) with admixtures of red maple (*Acer rubrum* L.), black birch (*Betula lenta* L.), paper birch (*Betula paperifera* L.), and beech (*Fagus grandifolia* Ehrh.). The pine forest is a red pine (*Pinus resinosa* Ait.) plantation established in 1926. Soils are Inceptisols (Typic Dystrochrepts) on stony till with sandy-loam textures. More detailed descriptions of the site, plots, and treatments are reported elsewhere (Aber et al., 1993; Magill et al., 1997).

2.2. ^{15}N tracer and N fertilizer additions

As described in detail by Magill et al. (this volume), 30 m × 30 m plots in hardwood and pine forests have been fertilized with NH_4NO_3 in 6 monthly additions during each growing season from 1988 through the present time. In each forest there are four experimental treatments: control, low N, low N + S, high N. Control plots are subject to atmospheric N deposition, estimated at 0.8 g N m^{-2} per year. Low N and low N + S plots are subject to deposition plus 5.0 g N m^{-2} per year, while high N plots receive atmospheric deposition plus 15.0 g N m^{-2} per year, yielding total N loadings of 5.8 or 15.8 g N m^{-2} per-year on chronically fertilized plots.

In 1991 and 1992 (fertilization years 4 and 5), we added ^{15}N tracers to the control and low N plots, but to no other treatment plots. The isotopic composition of the N applied to fertilized plots was similar to that of the atmospheric N_2 standard (0.3663 at.% ^{15}N or $\delta^{15}N = 0$[1]). The mean $\delta^{15}N$ value of four fertilizer batches applied to plots from 1988 through 1992 was 0.0‰ (S.E.M. = 0.47) and values ranged from −1.3 to 0.8‰. Fertilizer NH_4-N and NO_3-N were not analyzed separately and we assume that the $\delta^{15}N$ values of each N ion were similar to the 0.0‰ value of the NH_4NO_3 fertilizer. In tracer addition years, the NH_4NO_3 fertilizer added to one 15 m × 30 m half of each low N plot was labeled with enough $^{15}NH_4Cl$ to

[1] We follow the convention of expressing ^{15}N contents of natural materials using $\delta^{15}N$ notation where units are expressed as per mil deviations from the atmospheric standard of 0.3663 at.% ^{15}N and are calculated: $\delta^{15}N = [(\text{at.\%}\ ^{15}N\ \text{sample}/0.3663) - 1] \times 1000$.

increase the ammonium $\delta^{15}N$ value from 0 to 965‰ (or from 0.3663 to 0.7173 at.% ^{15}N). Fertilizer applied to the other half of each low N plot was labeled with enough $K^{15}NO_3$ to increase the nitrate $\delta^{15}N$ to 761‰ (from 0.3663 to 0.6433 at.% ^{15}N). Control plots received the same amounts of ^{15}N excess as were applied to low N plots: 17.6 mg $^{15}N\ m^{-2}$ per year applied as dissolved $^{15}NH_4Cl$ (17.6 mg N m^{-2} as 99.1 at.% ^{15}N) and 13.9 mg $^{15}N\ m^{-2}$ per year applied as dissolved $K^{15}NO_3$ (14.1 mg N m^{-2} as 98.6 at.% ^{15}N) to separate halves (15 m × 30 m) of each non-fertilized 30 m × 30 m plot. Tracers were applied to the controls six times per growing season on the same days as ^{15}N-labeled fertilizers were applied to the low N plots, using backpack sprayers and the same amounts of water as on fertilized plots.

2.3. Sampling

Plant and soil samples were collected from $^{15}NH_4$- and $^{15}NO_3$-labeled halves of the control and low N plots in order to develop ^{15}N mass balances. We also collected samples from the low N + S and the high N plots in both forest types in order to determine the degree to which additions of non-labeled N fertilizer might have altered the natural patterns of ^{15}N abundances in these stands. We sampled from reference areas located at least 20 m from any labeled or fertilized plot for comparison to pre-labeling reference samples taken on labeled plots.

2.3.1. Plant tissues

Tree foliage, bark, and wood samples were collected from three dominant trees within the four interior subplots of the $^{15}NH_4$- and $^{15}NO_3$-labeled halves of the labeled plots in July 1999. In the hardwoods, we sampled foliage, bark and wood from oaks, and foliage only from the maples. In the pines, we only sampled red pines, the only species occupying the canopy. Needles from each tree were separated into two cohorts, current and prior year. Bolewood was sampled at two heights, 1.25 and >6 m, on each sampled tree to test the uniformity of ^{15}N distribution within trees. Bolewood cores were taken at both heights with a 1.9 cm diameter hole saw. Cores were separated into bark and three wood increments: wood grown in 1990 through 1992, 1993 through 1995, and 1996 through 1999. Branches and twigs were also sampled for comparison of ^{15}N contents with bolewood. Live fine roots (intact and fibrous) were separated from Oe and Oa horizon samples, but not the Oi horizons, which contained few or no roots. Fine roots were also separated from samples of 0–5, 5–10, and 10–20 cm mineral soil layers.

2.3.2. Soils

Organic and mineral soil samples were collected from random locations within interior 5 m × 5 m subplots of the labeled 15 m × 30 m half plots using a 5.1 cm (2 in.) diameter AMS™ split core sampler. Single organic + mineral soil cores were taken to at least 20 cm depth in mineral soils. Organic and mineral soils were separated immediately after sampling at the boundary of the Oa (lowest organic) and mineral soil. Organic layers were further separated into Oi (<1-year-old litter), Oe (partially decomposed tissues), and Oa (well-decomposed organic material). Mineral soil was separated into 0–5, 5–10, and 10–20 cm layers. Organic horizons and mineral soil layer masses were estimated using dry weights (70 °C) per unit area corrected for coarse fragments (>2 mm). Due to a systematic error in sampling of the pine low N soils in 1999, soils on this plot were resampled in the spring of 2001. In order to test for comparability of ^{15}N and elemental analyses between July 1999 and spring 2001, soils were resampled at the hardwood low N plot for comparison with July 1999 results. Comparisons of hardwood low N soil and root samples for ^{15}N contents failed to show differences in ^{15}N between these two sample times. Therefore, we used results of the spring 2001 sampling of the pine low N plot to estimate the 1999 ^{15}N contents of soils and roots in this plot.

2.4. Elemental and ^{15}N analysis

Plant and soil samples were dried to a constant mass at 70 °C, weighed, and then stored at 4 °C until processing. Samples were processed for elemental analysis by grinding to a fine powder, tested to pass through a no. 100 mesh sieve, drying again at 70 °C, and cooling in an evacuated desiccator immediately prior to analysis. C and N concentrations were measured by dry combustion on a Carlo-Erba™ NC 2100 at the Appalachian Laboratory in Frostburg, Maryland. We performed dry weight corrections (105 °C)

on each sample and expressed C and N concentrations on a dry weight, ash-included basis.

Splits of samples used for C and N analyses were analyzed at the Marine Biological Laboratory (Woods Hole, Massachusetts) for ^{15}N using either a dual-inlet Finnigan™ MAT Delta S isotope ratio mass spectrometer with a cryogenic trapping box preparation system (Fry et al., 1992) or a continuous-flow PDZ Europa™ Model 20-20 mass spectrometer.

2.5. Ecosystem C and N pools

We estimated dry masses and element contents of ecosystem pools on each plot using our own elemental analyses (above), combined with data on foliar, fine root, and woody biomass that was provided by Magill et al. (this volume) of the University of New Hampshire. Bark biomass was estimated using allometric regressions from TerMikaelian and Korzukhin (1997).

2.6. ^{15}N mass balances

We estimated ^{15}N tracer movements into forest components using ecosystem N pool size estimates based on field measures (above), changes in ^{15}N content of ecosystem pools following tracer additions, and ^{15}N mass recoveries. Percent recoveries within pools of total tracers applied to plots were calculated as follows:

$$^{15}N_{rec} = \frac{m_{pool}(\text{at.}\%\ ^{15}N_{pool} - \text{at.}\%\ ^{15}N_{ref})}{\text{at.}\%\ ^{15}N_{tracer} - \text{at.}\%\ ^{15}N_{ref}} \quad (1)$$

where $^{15}N_{rec}$ is the mass of ^{15}N tracer recovered in the labeled N pool (g N m^{-2}), m_{pool} the N mass of the labeled N pool (g N m^{-2}), at.% $^{15}N_{pool}$ the at.% ^{15}N in the labeled N pool, at.% $^{15}N_{ref}$ the at.% ^{15}N in the reference (pre- or non-labeled) N pool, and at.% $^{15}N_{tracer}$ the at.% ^{15}N of the applied tracer or labeled fertilizer.

Masses of tracers recovered in pools ($^{15}N_{rec}$) were expressed as percents of total ^{15}N tracer masses applied to a plots, designated as PR ^{15}N.

2.6.1. Correction

Our earlier reports of ^{15}N recoveries in control plots at the end of the 1992 labeling (Nadelhoffer et al., 1999a) were in error due to assumptions that ^{15}N additions to control plots were the same as ^{15}N additions to low N plots. Low N plots received additions of 35.7 mg ^{15}N m^{-2} to the $^{15}NH_4$-labeled half and 32.0 mg ^{15}N m^{-2} to the $^{15}NO_3$-labeled half during the two seasons of labeling. However, ^{15}N additions to the control plots were less than half of ^{15}N additions to low N plots because no background fertilizer ^{15}N was applied to controls. Total additions of ^{15}N to control plots for the 1991–1992 labeling period were 17.6 mg ^{15}N m^{-2} to the $^{15}NH_4$-labeled half and 13.9 mg ^{15}N m^{-2} to the $^{15}NO_3$-labeled half. Correcting for this error increases percent recovery estimates for individual pools on control plots (referred to as 'Ambient' plots in Nadelhoffer et al., 1999a) by a factor 2.04 for $^{15}NH_4$-labeled plot halves and 2.29 for $^{15}NO_3$-labeled plot halves. Correcting for this error improves our overall estimates of tracer recoveries, which never exceed 55% of tracer applications to control pots (Nadelhoffer et al., 1999a). However, it does not alter either the overall patterns of recovery or conclusions of the earlier study.

2.7. Statistical analyses

As in our prior study (Nadelhoffer et al., 1999a), we assume here that N pool sizes on the plots are well characterized due to intensive sampling. We also assumed, in this and our previous study, that the ^{15}N contents of ecosystem pools in non-labeled forests were well characterized based on the small variances in N isotope distributions observed before tracers were added (Tables 1 and 2). Our statistical analyses, therefore, assume fixed values for total N mass and pre-labeled ^{15}N content (m_{pool} and at.% $^{15}N_{ref}$ in Eq. (1)) of each pool within a plot and that variances in estimates of tracer recovery are due to variances in our estimates of ^{15}N contents of post-labeled pools (at.% $^{15}N_{pool}$ in Eq. (1)).

Neither the fertilization treatments nor plot labeling are replicated within either forest at our site. Our measurements of ^{15}N recovery are replicated within experimental plots and statistical extrapolations are made at the plot scale. We used two-way analysis of variance (SYSTAT® version 10.00) on log-transformed data to test for effects of N loading rate (ambient deposition versus chronic fertilization) and form of N tracer ($^{15}NH_4$ versus $^{15}NO_3$) on percent ^{15}N tracer recoveries in pools within forest types.

Table 1
^{15}N abundances in hardwood reference samples and plots subjected to N inputs of 0.8, 5.8, and 15.8 g m^{-2} per year, but with no ^{15}N tracer additions

Ecosystem pool	Treatment and N input rate									$\Delta\delta$ (treatment − ref)	
	Reference samples (0.8 g N m^{-2} per year)			Low N + S (5.8 g N m^{-2} per year)			High N (15.8 g N m^{-2} per year)			Low N + S (‰)	High N (‰)
	$\delta^{15}N$ (‰)	S.E.M.	*n*	$\delta^{15}N$ (‰)	S.E.M.	*n*	$\delta^{15}N$ (‰)	S.E.M.	*n*		
Foliage	−2.6	0.2	4	−2.9	0.6	3	−0.5	0.2	3	−0.3	2.1
Wood formed in											
1996–1999	−1.5	0.5	4	−1.6	0.4	3	0.5	0.1	3	−0.1	2.0
1993–1995	−1.8	0.6	4	−3.1	0.6	3	0.5	0.4	3	−1.2	2.4
1990–1992	−2.1	0.3	4	−0.5	0.6	3	−0.9	0.5	3	1.6	1.2
Bark	−2.5	0.3	3	−2.7	0.1	3	−1.4	0.3	3	−0.2	1.1
Fine roots in											
Oe	−2.5		1	n.d.			0.0	0.5	2		2.5
Oa	−0.6	0.3	4	n.d.			0.4	0.3	3		1.0
0–5 cm mineral	1.3	0.4	4	n.d.			1.6	0.2	3		0.3
5–10 cm mineral	2.1	0.7	4	n.d.			2.5	0.3	3		0.4
10–20 cm mineral	1.7	0.7	3	n.d.			1.2	0.4	3		−0.5
Soil											
Oi (fresh litter)	−3.6	0.4	3	n.d.			−1.1	0.1	3		2.5
Oe	−2.3	0.1	4	n.d.			−1.4	0.1	3		0.9
Oa	0.9	0.5	4	n.d.			−0.7	0.3	3		−1.6
0–5 cm mineral	4.6	0.5	4	n.d.			2.7	0.5	3		−1.9
5–10 cm mineral	5.8	0.5	4	n.d.			5.4	0.4	3		−0.4
10–20 cm mineral	7.1	0.3	4	n.d.			6.3	0.4	3		−0.8

Although the fertilizer treatments are not replicated within either forest, the small variances in ^{15}N contents in ecosystem pools both before and after ^{15}N additions indicate that significant differences in ^{15}N recoveries are due to treatments rather than to natural variations in ^{15}N abundances. Therefore, we use our estimates of tracer movements within plots to make inferences about the fates of atmospheric ammonium and nitrate inputs to these forests.

3. Results

3.1. ^{15}N abundances in non-labeled plots

Patterns of natural ^{15}N abundances in both the hardwood (Table 1) and the pine stand (Table 2) are typical of those reported in other temperate forests (Nadelhoffer and Fry, 1988; Gebauer and Dietrich, 1993; Garten and Van Miegroet, 1994; Nadelhoffer and Fry, 1994; Högberg et al., 1996) with plant tissue and soil $\delta^{15}N$ values bracketing the atmospheric standard of 0‰ (or 0.3663 at.% ^{15}N). Leaves and woody tissues were depleted in ^{15}N relative to atmospheric N, with $\delta^{15}N$ values ranging from −3.0 to −0.2‰. Fine root $\delta^{15}N$ values increased with sampling depth from −2.5 to +2.1‰ in hardwoods and from −1.3 to +1.2‰ in pines. Soil $\delta^{15}N$ values increased with age and depth from values similar to those of plants at the organic soil surface to maximum values of 7.1‰ in hardwoods and 5.4‰ in pines.

Analyses of ^{15}N contents of samples collected from the low N + S plots, which received the same rate of N addition as the low N plots, but without ^{15}N tracers, reveal that 13 years of fertilization (fertilizer $\delta^{15}N = \sim 0‰$) with 5 g NH_4NO_3-N m^{-2} per year did not systematically alter natural patterns of ^{15}N abundance. Differences in $\delta^{15}N$ values within plant tissue types between low N + S and reference samples ($\Delta\delta$) ranged from −0.3 to +1.6‰ in hardwood leaves and

Table 2
^{15}N abundances pine reference samples and in pine plots subjected to N inputs of 0.8, 5.8, and 15.8 g m^{-2} per year, but with no ^{15}N tracer additions

Ecosystem pool	Treatment and N input rate									Δδ (treatment − ref)	
	Reference samples (0.8 g N m^{-2} per year)			Low N + S (5.8 g N m^{-2} per year)			High N (15.8 g N m^{-2} per year)			Low N + S (‰)	High N (‰)
	$\delta^{15}N$ (‰)	S.E.M.	*n*	$\delta^{15}N$ (‰)	S.E.M.	*n*	$\delta^{15}N$ (‰)	S.E.M.	*n*		
Foliage	−1.8	0.1	6	−1.4	0.1	3	1.8	0.6	3	0.5	3.6
Wood formed in											
1996–1999	−1.3	0.4	3	−0.7	0.5	3	2.2	0.6	3	0.6	3.5
1993–1995	−0.6	0.5	3	−0.7	0.4	3	1.5	0.1	3	−0.1	2.1
1990–1992	−0.2	0.2	3	−0.8	0.6	3	1.8	0.7	3	−0.6	2.0
Bark	−3.0	0.2	3	n.d.			−0.3	0.5	3		2.8
Fine roots in											
Oe	−1.3		1	−0.9		1	n.d.			0.4	
Oa	−0.1	0.1	3	0.1		1	n.d.			0.2	
0–5 cm mineral	0.6	0.3	3	1.0		1	4.2	0.2	3	0.4	3.6
5–10 cm mineral	1.0	0.6	4	1.2		1	2.6	0.5	3	0.2	1.6
10–20 cm mineral	1.2	0.6	3	1.0		1	2.0	0.6	3	−0.2	0.8
Soil											
Oi (fresh litter)	−3.1	0.2	3	−2.0	0.5	6	0.0	0.1	3	1.1	3.0
Oe	−1.2	0.4	4	−1.5	0.4	6	−1.0	0.1	3	−0.3	0.2
Oa	0.1	0.2	4	0.4	1.4	7	0.1	0.5	3	0.3	0.1
0–5 cm mineral	2.8	0.2	4	3.9	0.6	7	3.5	0.3	3	1.1	0.7
5–10 cm mineral	4.0	0.3	4	4.6	0.3	6	5.1	0.1	3	0.6	1.2
10–20 cm mineral	5.4	0.3	4	5.7	0.5	6	6.6	0.1	3	0.4	1.2

wood (Table 1) and from −0.6 to +1.1‰ in pine plant tissues and soils (Table 2).

The $\delta^{15}N$ values of foliage, wood and fine roots from organic horizons in the hardwood high N plot were 1.0–2.5‰ greater than values of the same pools collected from reference areas (Table 1). High N plot forest floor N was enriched in ^{15}N relative to reference samples only in the Oi (2.5‰) and possibly in the Oe horizon (0.9‰). The hardwood high N Oa horizon and mineral soil layers were slightly depleted in ^{15}N, perhaps due to illuviation of nitrified N from higher in the profiles. Foliage, wood and fine roots collected from the pine high N plot had $\delta^{15}N$ values ranging from 0.8 to 3.6‰ greater than values of reference samples. Soils in the pine high N plots did not differ markedly from reference samples in ^{15}N abundance.

3.2. Ecosystem N pools

Ecosystem N contents (excluding soils >20 cm deep and wood >10 years old) were 4.15 Mg ha^{-1} in the hardwood control and low N plots (Table 3). Total N contents in the pine stand were 4.60 Mg ha^{-1} in the control plot and 4.40 Mg ha^{-1} in the low N plot (Table 4). Mineral soil (0–20 cm depth) contained the most N, accounting for 70 (±5)% of total N across control and low N plots in both stands. Tree biomass (foliage, fine roots, bark, and wood produced from 1990 through 1999) accounted for 6–7% of the N mass in hardwood control and low N plots and 5–8% of N in pine control and low N plots. Wood formed from the onset of labeling through sampling (1990–1999) plus bark accounted for 2.9 (±0.3)% of total N in the hardwood and 1.5 (±0.2)% of total N in the pine plots. Wood formed prior to 1990 is not included in our ecosystem budget as it is likely to be only a minor sink for ^{15}N additions. Course and fine woody debris are also omitted from our budgets here. Total amounts of C and N in course plus fine woody debris are roughly equivalent to amounts of these elements in 1990–1999 bole wood plus bark pools and are reported elsewhere (Currie and Nadelhoffer, 2002).

Table 3
Masses and N contents of major ecosystem pools in ^{15}N-labeled plots in the hardwood stand of the Harvard Forest Chronic N Amendment Study

Ecosystem pool	Hardwood control				Hardwood low N			
	Mass (kg ha^{-1})	N (%)	C:N	N (kg ha^{-1})	Mass (kg ha^{-1})	N (%)	C:N	N (kg ha^{-1})
Foliage								
Oak	2629	2.35		61.7	2506	2.64		66.2
Red maple	175	1.90		3.3	288	1.93		5.6
Total foliage	2804			65.0	2794			71.7
Bole wood years								
1996–1999	10100	0.17	274	17.7	14700	0.22	217	32.5
1993–1995	10500	0.13	379	13.3	12300	0.13	375	15.8
1990–1992	13400	0.13	360	17.9	13500	0.13	372	17.4
Bark	15400	0.38	126	58.7	16400	0.40	120	65.3
1990–1999 wood + bark	49400			107.6	56900			131.0
Fine roots in								
Oe	217	1.62	30.4	3.5	956	1.74	30.2	16.6
Oa	2960	1.20	39.4	35.4	1801	1.26	37.2	22.7
0–5 cm mineral	1816	0.91	48.4	16.6	1454	0.94	48.0	13.7
5–10 cm mineral	1050	0.74	57.9	7.8	1517	0.83	52.6	12.5
10–20 cm mineral	1427	0.62	71.5	8.8	1500	0.64	60.3	9.6
Total fine roots	7469			72.2	7229			75.2
Tree biomass (=10 years old)	59673			244.8	66923			277.9
Organic horizon								
Oi (fresh litter)	5309	1.34	27.1	71	5932	1.69	21.7	100
Oe	13402	1.89	20.0	253	22100	2.12	18.9	468
Oa	54516	1.28	25.3	696	66835	1.26	21.6	839
Total organic horizon	73227			1021	94867			1407
0–5 cm mineral	325000	0.34	20.6	1117	320000	0.31	24.9	989
5–10 cm mineral	405000	0.22	20.2	876	395000	0.19	23.1	762
10–20 cm mineral	830000	0.14	19.8	1133	970000	0.10	20.2	988
Total 0–20 cm mineral soil	1560000			3126	1685000			2738
Total organic + 0–20 cm soil	1633227			4147	1779867			4146

3.3. ^{15}N tracer recoveries

Seven years after the end of ^{15}N additions to control and low N plots, tracers were clearly detectable in the hardwood (Table 5) and pine (Table 6) forests. Mean δ^{15}N values of tree tissue and organic soil samples from labeled plots were 8–40 ‰ greater than means of non- or pre-labeled samples. Differences in mean δ^{15}N values between mineral soil samples in labeled versus non- or pre-labeled plots were smaller, ranging from 0.9 to 4.0‰ in hardwood mineral soil layers and from 2.3 to 7.0‰ in pine mineral soil layers.

We used plot-specific information on ^{15}N contents of reference samples (Tables 1 and 2), N pool sizes (Tables 3 and 4), and ^{15}N contents of tree tissue, organic horizon and mineral soil samples collected in 1999 (Tables 5 and 6) to calculate the movements of ^{15}N tracers (applied as either $^{15}NH_4$ or $^{15}NO_3$) into ecosystem components of the hardwood (Table 7) and pine (Table 8) plots. Values are expressed as percent recoveries of total ^{15}N tracers added to plot halves under each combination of forest stand, N addition rate, and form of ^{15}N addition. We also included percent recoveries of tracers in fine woody debris (<5 cm diameter) as reported in Currie et al. (2002).

3.3.1. Foliage

By late summer 1999, mean ^{15}N tracer recoveries (averages of $^{15}NH_4$ and $^{15}NO_3$ additions) in hardwood foliage were 3.8% in the control and 3.1% in the low N

Table 4
Masses and N contents of major ecosystem pools in ^{15}N-labeled plots in pine stand of the Harvard Forest Chronic N Amendment Study

Ecosystem pool	Pine control				Pine low N			
	Mass (kg ha^{-1})	N (%)	C:N	N (kg ha^{-1})	Mass (kg ha^{-1})	N (%)	C:N	N (kg ha^{-1})
Foliage								
Oak	2623	0.84		22.0	3905	1.39		54.2
Red maple	3935	1.12		44.1	5858	1.71		100.2
Total foliage	6558			66.1	9763			154.3
Bole wood years								
1996–1999	10300	0.16	300	8.0	7600	0.17	275	10.5
1993–1995	7700	0.14	343	10.8	5700	0.14	350	7.83
1990–1992	8500	0.13	369	11.1	6300	0.08	601	5.03
Bark	20100	0.19	253	38.2	15200	0.19	253	28.9
1990–1999 wood + bark	46700			76.7	37000			56.7
Fine roots in								
Oe	574	1.70	27.7	9.7	982	2.08	21.7	20.5
Oa	949	1.62	28.7	15.3	1498	2.03	23.0	30.5
0–5 cm mineral	1387	1.20	37.1	16.7	1354	1.81	24.3	24.6
5–10 cm mineral	1247	0.88	47.3	10.9	1431	1.73	25.6	24.8
10–20 cm mineral	1984	0.77	52.4	15.3	1915	1.63	25.1	31.2
Total fine roots	6141			68.0	7181			131.5
Tree biomass (=10 years old)	59398			210.8	53943			342.6
Organic horizon								
Oi (fresh litter)	15475	1.93	31.6	299	16714	1.58	26.3	264
Oe	14557	1.86	22.8	271	25345	1.53	22.5	387
Oa	38676	1.37	20.6	531	65175	0.98	21.3	641
Total organic horizon	68708			1100	107234			1293
0–5 cm mineral	321880	0.35	19.1	1118	351041	0.29	29.2	1016
5–10 cm mineral	404379	0.26	19.6	1038	422471	0.21	28.8	894
10–20 cm mineral	793839	0.17	17.9	1343	892784	0.13	21.4	1200
Total 0–20 cm mineral soil	1520097			3498	1666296			3111
Total organic + 0–20 cm soil	1588805			4598	1773530			4404

plot (Table 7). Compared to the end of labeling in 1992, when recoveries in hardwood control and low N plots were 1.4 and 6.3% of additions (Nadelhoffer et al., 1999a), recoveries had increased by a factor of ~2 in control foliage and had decreased by a factor of ~2 in low N foliage across the 7-year sampling interval. In contrast to hardwoods, pine stand foliage retained more ^{15}N tracer on the low N than on the control plot (Table 8). Mean recoveries in pine control and pine low N by 1999 were 2.9 and 6.1% of additions, respectively. Recoveries in these same pools in 1992 were 1.3 and 11.5% of additions. As with the hardwoods, recoveries in foliage had increased on the control and decreased on the low N plot by factors of ~2 by 7 years after the end of labeling.

Recoveries in foliage differed with form ($^{15}NH_4$ versus $^{15}NO_3$) of tracer addition in 1999 (Tables 7 and 8), as was reported for 1992 at the end of labeling (Nadelhoffer et al., 1999a). Foliage accounted for 60% more tracer after $^{15}NO_3$ than after $^{15}NH_4$ additions to the hardwood control, and 23% more tracer after $^{15}NO_3$ additions to the hardwood low N plot (Table 7). Tracer recoveries in pine plots were 31 and 57% greater after $^{15}NO_3$ than after $^{15}NH_4$ additions to control and low N plots, respectively (Table 8). For all pine treatments, tracer recoveries were greater in older needles than in current-year needles. The opposite occurred at the end of tracer additions when current-year needles had more tracer than older needles.

Table 5
^{15}N abundances in ecosystem pools in the hardwood stand before and 7 years after the end of ^{15}N tracer additions

Ecosystem pool	Pre- or non-tracer samples			Hardwood control						Hardwood low N					
	$\delta^{15}N$ (‰)	S.E.M	n	$^{15}NH_4$			$^{15}NO_3$			$^{15}NH_4$			$^{15}NO_3$		
				$\delta^{15}N$ (‰)	S.E.M	n	$\delta^{15}N$ (‰)	S.E.M	n	$\delta^{15}N$ (‰)	S.E.M	n	$\delta^{15}N$ (‰)	S.E.M	n
Foliage															
Oak	−2.6	0.2	4	19.4	1.1	3	25.4	2.9	3	16.4	1.1	3	15.6	1.7	3
Red maple	−3.9	0.5	4	13.6	1.2	2	11.3	1.1	2	15.3	1.5	3	18.3	3.4	3
Bole wood years															
1996–1999	−1.5	0.5	4	19.1	0.6	6	22.1	1.7	6	17.2	0.3	6	17.9	0.6	6
1993–1995	−1.8	0.6	4	14.3	1.7	6	17.1	2.9	6	17.8	1.5	6	22.3	0.7	6
1990–1992	−2.1	0.3	4	7.4	2.5	6	6.7	1.1	6	10.0	1.2	6	12.9	1.0	6
Bark	−2.5	0.3	3	6.7	0.9	6	9.9	0.9	6	10.8	1.6	6	11.1	1.1	6
Fine roots in															
Oe	−2.5	–	1	31.6	6.2	4	35.9	9.1	3	20.5	2.7	4	21.6	–	1
Oa	−0.6	0.3	4	33.1	5.3	5	39.7	4.5	5	22.8	1.8	4	16.5	2.4	3
0–5 cm mineral	1.3	0.4	4	10.9	1.5	5	13.9	2.0	5	14.2	2.4	4	17.3	3.3	4
5–10 cm mineral	2.1	0.7	4	8.5	1.2	5	9.3	1.1	4	12.0	1.7	5	17.3	2.4	4
10–20 cm mineral	1.7	0.7	3	7.8	2.3	3	9.4	1.3	5	12.2	3.2	5	14.0	2.3	4
Soil															
Oi (fresh litter)	−3.6	0.4	3	15.5	1.4	5	12.9	4.2	5	11.5	1.2	5	13.0	0.9	5
Oe	−2.3	0.1	4	36.5	6.5	5	32.7	9.6	4	23.4	3.3	4	19.7	2.2	5
Oa	0.9	0.5	4	27.6	6.8	5	31.5	3.6	4	16.1	2.2	5	12.0	3.0	5
0–5 cm mineral	4.6	0.5	4	8.7	1.5	5	8.3	0.6	5	8.4	0.6	5	7.9	0.5	5
5–10 cm mineral	5.8	0.5	4	7.3	0.9	5	7.5	0.4	5	7.4	0.7	5	8.3	0.3	5
10–20 cm mineral	7.1	0.3	4	8.0	0.5	5	7.8	0.5	5	9.2	1.1	5	8.5	0.5	5

Pre-labeling values are means (and standard errors) of samples collected from treated and fertilized plots prior to additions.

3.3.2. Wood and bark

As for foliage, tracer recoveries in wood varied with N loading rate and tracer form. Recoveries in wood produced from 1990 through 1999 were greater on low N plots than on control plots and were greater after $^{15}NO_3$ than after $^{15}NH_4$ additions in both hardwood (Table 7) and pine stands (Table 8). Comparisons of recoveries among year classes of wood formation suggest that wood is becoming a greater sink for the tracers through the decade following tracer additions; percent recoveries increased with more recently formed wood within all stand treatment combinations except the pine control $^{15}NH_4$ plot. Recovery patterns for bark were similar to those in wood. In all cases, recoveries in bark were greater than in any of the wood age classes sampled and were always a large proportion of total percent recovery.

In the hardwoods, wood was approximately equal in importance as a sink for tracers as other plant pools across treatments (Table 7), whereas in the pines, wood was the smallest sink (Table 8). Percent ^{15}N recoveries in 1999 ranged from 3.0 to 6.1 in the hardwood and from 1.1 to 2.6 in the pine stand. Ranges in 1992 (for wood formed from 1988 to 1992 plus bark) were 0.9–5.7 in hardwoods and 0.2–2.0 in pines (Nadelhoffer et al., 1999a).

3.3.3. Fine roots

Tracer recoveries in hardwood fine roots by 1999 ranged from 2.7 to 5.0% of the 1991–1992 additions across all combinations of N loading and ^{15}N forms (Table 7). The Percent ^{15}N recoveries in roots were lower in 1999 than in 1992 when recoveries ranged from 3.2 (control, $^{15}NH_4$ added) to 18.3% (low N, $^{15}NO_3$ added) of tracer additions. Recoveries in pine roots by 1999 ranged from 3.2 to 5.6% of ^{15}N additions (Table 8). In contrast to hardwoods, the 1999 pine recoveries were in the mid-range of 1992

Table 6
^{15}N abundances in ecosystem pools of a pine stand before and 7 years after the end of ^{15}N tracer additions

Ecosystem pool	Pre- or non-tracer samples			Pine control						Pine low N					
	$\delta^{15}N$ (‰)	S.E.M.	n	$^{15}NH_4$			$^{15}NO_3$			$^{15}NH_4$			$^{15}NO_3$		
				$\delta^{15}N$ (‰)	S.E.M.	n	$\delta^{15}N$ (‰)	S.E.M.	n	$\delta^{15}N$ (‰)	S.E.M.	n	$\delta^{15}N$ (‰)	S.E.M.	n
Foliage															
Current year				16.3	1.2	3	17.7	0.9	3	13.9	0.2	3	17.7	1.4	3
>1-year-old				16.4	0.7	3	16.6	1.0	3	13.1	0.8	3	16.5	0.9	3
All age classes	−1.8	0.1	6	16.4		6	17.2		6	13.5		6	17.1		6
Bole wood years															
1996–1999	−1.3	0.4	3	10.0	1.9	6	17.4	1.2	6	12.7	1.0	6	17.0	1.2	6
1993–1995	−0.6	0.5	3	10.6	0.6	6	16.8	1.7	6	13.8	0.3	6	15.8	0.6	6
1990–1992	−0.2	0.2	3	4.9	1.2	6	8.6	0.3	6	8.2	0.7	6	9.4	0.8	6
Bark	−3.0	0.2	3	0.8	0.9	6	5.6	2.7	6	7.4	3.1	4	13.1	3.5	4
Fine roots in															
Oe	−1.3	–	1	39.2	–	1	28.0	3.3	3	23.0	2.5	5	18.6	1.2	5
Oa	−0.1	0.1	3	25.8	2.5	4	23.8	3.7	4	21.4	2.0	5	15.4	1.6	5
0–5 cm mineral	0.6	0.3	3	17.2	6.2	3	20.9	2.1	4	16.7	1.9	5	13.5	2.0	5
5–10 cm mineral	1.0	0.6	4	21.7	7.1	2	14.3	2.2	5	13.9	1.6	5	18.3	1.6	5
10–20 cm mineral	1.2	0.6	3	17.3	1.5	4	14.3	3.1	5	13.3	1.4	5	16.8	1.6	5
Soil															
Oi (fresh litter)	−3.1	0.2	3	16.3	4.8	5	17.8	2.9	5	12.9	1.1	5	13.2	0.9	5
Oe	−1.2	0.4	4	38.8	5.0	5	37.4	7.1	5	19.7	1.7	5	12.4	1.8	5
Oa	0.1	0.2	4	15.3	3.0	5	11.4	2.8	5	7.7	1.9	5	5.1	0.9	5
0–5 cm mineral	2.8	0.2	4	9.9	1.7	5	7.3	0.4	5	6.7	0.8	5	6.1	0.6	5
5–10 cm mineral	4.0	0.3	4	9.0	2.3	5	7.0	0.3	5	7.4	0.8	5	7.3	0.3	5
10–20 cm mineral	5.4	0.3	4	7.7	1.1	5	7.8	1.5	5	8.3	0.6	5	8.5	0.4	5

Pre-labeling values are means (and standard errors) of samples collected from treated and fertilized plots prior to additions.

recoveries, when values ranged from 1.7 (control, $^{15}NH_4$ added) to 10.3% (low N, $^{15}NO_3$ added) of tracer additions.

Although tracer recoveries in hardwood fine roots were much greater with N fertilization at the end of labeling in 1992 (recoveries were 4.6 and 13.7% of ^{15}N additions to hardwood control and low N plots, respectively), recoveries no longer increased with N loading by 1999 (Table 7); mean recoveries (average of $^{15}NH_4$ and $^{15}NO_3$ additions) in 1999 were 4.2 and 3.1% of tracer additions to hardwood control and low N plots, respectively. For pines in 1999 (Table 8), recoveries were still greater in low N (5.1% of additions) than in control plots (3.4% of additions), but the difference was greater in 1992 when pine low N recoveries were 8.8% of additions to the low N plot and 2.3% of additions to the control plot (Nadelhoffer et al., 1999a). Percent recoveries in 1999 fine roots were higher after $^{15}NO_3$ additions in both forest stands under control and low N treatments, but differences were not as large as those in 1992 when recoveries were >2 times greater after $^{15}NO_3$ than $^{15}NH_4$ labeling.

Tracer recoveries varied by horizon or depth in soil from which roots were sampled. For example, recoveries in Oa horizon fine roots were disproportionately high in hardwood plots, particularly in control plots with only ambient inputs (Table 7). Tracer recoveries were more evenly distributed across roots from organic horizons and mineral soil layers in pines, particularly in the control plot (Table 8).

3.3.4. Organic soil

Organic soil (including fine woody debris and O horizons) was the dominant sink for ^{15}N tracers by 1999 in both forest types under all treatment

Table 7
Tracer ($^{15}NH_4$ and $^{15}NO_3$) recoveries in control and low N treatment (5 g N added m^{-2} per year) plots in the hardwood stand of the Harvard Forest Chronic N Amendment Study

Ecosystem pool	Hardwood control			Hardwood low N			Significant effects (two-way ANOVA, P values)[a]		
	$^{15}NH_4$	$^{15}NO_3$	Mean	$^{15}NH_4$	$^{15}NO_3$	Mean	N load	N form	Interaction
Foliage									
Oak	2.83	4.56	3.70	2.58	3.13	2.85	0.086	0.818	0.122
Other	0.11	0.12	0.12	0.20	0.30	0.25	**	0.103	0.980
Total foliage	2.94	4.69	3.81	2.78	3.43	3.11			
Wood									
1996–1999	0.83	1.29	1.06	1.38	1.81	1.59	***	***	0.134
1993–1995	0.49	0.75	0.62	0.69	1.06	0.88	*	**	0.941
1990–1992	0.40	0.46	0.43	0.48	0.74	0.61	0.273	0.183	0.992
Bark	1.23	2.12	1.67	1.92	2.48	2.20	***	0.199	0.745
Total 1990–1999 wood + bark	2.95	4.61	3.78	4.47	6.09	5.28			
Fine roots in									
Oe	0.25	0.36	0.30	0.80	1.05	0.92	*	0.200	0.371
Oa	2.49	3.78	3.14	1.10	1.02	1.06	***	0.230	0.088
0–5 cm soil	0.33	0.55	0.44	0.37	0.58	0.48	0.628	0.051	0.942
5–10 cm soil	0.10	0.15	0.13	0.26	0.50	0.38	***	**	0.416
10–20 cm soil	0.11	0.18	0.15	0.21	0.31	0.26	0.078	0.081	0.861
Total fine roots	3.29	5.02	4.15	2.73	3.47	3.10			
Total tree biomass (≤10 years old)	9.2	14.3	11.8	10.0	13.0	11.5			
Organic horizon									
Fine woody debris (diameter <5 cm)[b]	2.2	0.7	1.5	0.3	0.2	0.2			
Oi	2.8	3.1	3.0	3.1	4.4	3.8	0.144	***	0.917
Oe	20.5	23.5	22.0	25.0	27.1	26.0	*	0.858	0.858
Oa	38.7	56.2	47.4	26.4	24.4	25.4	*	0.455	0.158
Total organic horizon	64.2	83.5	73.9	54.9	56.1	55.4			
Mineral soil									
0–5 cm	9.6	10.9	10.2	7.8	8.7	8.2	0.654	0.326	0.622
5–10 cm	2.8	4.0	3.4	2.6	5.0	3.8	0.683	0.240	0.407
10–20 cm	2.1	2.1	2.1	4.4	3.7	4.0	0.436	0.402	0.587
Total 0–20 cm mineral soil	14.6	16.9	15.7	14.7	17.4	16.1			
Total organic + mineral soil	78.8	100.4	89.6	69.6	73.5	71.5			
Total	88.0	114.7	100.4	79.6	86.5	83.0			

Values are percent recoveries in 1999 of total ^{15}N additions to plots during 1991 and 1992.

[a] Values are percent recoveries of ^{15}N tracers at the end of 1992 that were applied in 1991 and 1992 as $^{15}NH_4$ or $^{15}NO_3$ to forest floors in separate halves of plots.

[b] From Currie et al. (2002).

* Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.05$.

** Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.01$.

*** Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.001$.

combinations (Tables 7 and 8). Total recoveries in the O horizon ranged from 34 (pine, low N, $^{15}NO_3$ additions) to 84% of additions (hardwood, control, $^{15}NO_3$ additions). The O horizon was also a strong sink for ^{15}N tracers at the end of labeling in 1992, but in all forest and treatment combinations, it had become a greater sink for tracers by 1999. In contrast to plant pools, where sink strength for tracers either remained constant or increased with N loading, the relative importance of the O horizon as a sink for N inputs

Table 8
Tracer ($^{15}NH_4$ and $^{15}NO_3$) recoveries in control and low N treatment (5 g N added m^{-2} per year) plots in the pine stand of the Harvard Forest Chronic N Amendment Study

Ecosystem pool	Pine control			Pine low N			Significant effects (two-way ANOVA, P values)[a]		
	$^{15}NH_4$	$^{15}NO_3$	Mean	$^{15}NH_4$	$^{15}NO_3$	Mean	N load	N form	Interaction
Foliage									
Current year	0.84	1.14	0.99	1.71	2.72	2.22	***	*	0.325
>1 year old	1.68	2.15	1.91	3.02	4.70	3.86	***	**	0.440
Total foliage	2.51	3.29	2.90	4.73	7.43	6.08			
Wood:									
1996–1999	0.30	0.88	0.59	0.43	0.69	0.56	0.071	0.380	0.953
1993–1995	0.30	0.56	0.43	0.27	0.38	0.33	0.264	*	0.391
1990–1992	0.18	0.34	0.26	0.16	0.22	0.19	0.092	0.082	0.641
Bark	0.29	0.86	0.57	0.61	1.21	0.91	0.490	*	0.327
Total 1990–1999 wood + bark	1.07	2.63	1.85	1.46	2.50	1.98			
Fine roots in									
Oe	0.82	0.75	0.79	1.03	1.07	1.05	0.079	0.986	0.705
Oa	0.83	0.97	0.90	1.36	1.24	1.30	**	0.830	0.290
0–5 cm soil	0.58	0.90	0.74	0.82	0.83	0.83	0.344	0.170	0.157
5–10 cm soil	0.47	0.38	0.43	0.67	1.13	0.90	***	0.290	0.037
10–20 cm soil	0.51	0.53	0.52	0.79	1.29	1.04	***	0.165	0.144
Total fine roots	3.21	3.53	3.37	4.67	5.57	5.12			
Total tree biomass (≤10 years old)	6.80	9.46	8.13	10.87	15.50	13.18			
Organic horizon									
Fine woody debris (diameter <5 cm)[b]	0.7	0.2	0.4	0.1	<0.1	0.1			
Oi	12.1	16.5	14.3	8.7	11.3	10.0	0.101	0.052	0.662
Oe	22.6	27.6	25.1	16.8	13.9	15.3	*	0.794	0.293
Oa	16.8	15.9	16.4	10.2	8.6	9.4	*	0.672	0.990
Total organic horizon	52.2	60.2	56.2	35.8	33.8	34.9			
Mineral soil									
0–5 cm	16.4	13.2	14.8	8.3	8.8	8.5	*	0.964	0.662
5–10 cm	10.8	8.2	9.5	6.4	8.0	7.2	0.498	0.451	0.558
10–20 cm	6.5	8.8	7.6	7.3	10.0	8.6	0.123	0.520	0.750
Total 0–20 cm mineral soil	33.7	30.2	31.9	21.9	26.7	24.3			
Total organic + mineral soil	85.9	90.4	87.7	57.7	60.5	59.2			
Total	92.7	99.9	96.2	68.6	76.0	72.4			

Values are percent recoveries in 1999 of total ^{15}N additions to plots during 1991 and 1992.

[a] Values are percent recoveries of ^{15}N tracers at the end of 1992 that were applied in 1991 and 1992 as $^{15}NH_4$ or $^{15}NO_3$ to forest floors in separate halves of plots.

[b] From Currie et al. (2002).

* Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.05$.

** Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.01$.

*** Significant main effects and interactions (2-way ANOVA) for individual ecosystem compartments: $P < 0.001$.

decreased with N loading. Percent recovery of tracers in the hardwood O horizon (averaged across $^{15}NH_4$ and $^{15}NO_3$) was 74 in the control and 55 in the low N plot (Table 7). Recoveries in pine control and low N O horizons were 56 and 35% of additions, respectively.

With respect to form of addition, O horizons in control plots of both forests retained more ^{15}N by 1999 after $^{15}NO_3$ than after $^{15}NH_4$ additions, but O horizons in low N plots retained less ^{15}N when $^{15}NO_3$ was added. This pattern was similar to that observed in

1992 and may reflect, in part, a saturation of O horizon capacities to retain NO_3-N additions under elevated N inputs.

3.3.5. Mineral soil

The greatest uncertainties in our estimates of tracer recoveries are in the mineral soil pools. This is because mineral soils are large N pools, typically containing >2/3 of total ecosystem N in our plots, and because ^{15}N tracers entering these large pools are diluted such that changes in ^{15}N abundances (as indicated by $\delta^{15}N$ values in Tables 5 and 6) are small. Despite the higher degree of uncertainty of our estimates of tracer movements into mineral soil, it appears that mineral soil was a greater sink for ^{15}N additions to pine than to hardwood plots. In hardwoods, recoveries across all four treatment combinations ranged from 15 to 17% of ^{15}N additions (Table 7), whereas in pines recoveries ranged from 22 to 34% of additions (Table 8). This difference in mineral soil recovery between forest types is consistent with the observation that hardwood O horizons were more effective sinks for ^{15}N tracers than were pine O horizons.

4. Discussion

4.1. ^{15}N natural abundances

Because ranges of $\delta^{15}N$ values for ecosystem pools within stands were narrow with small standard errors of means (Tables 1 and 2), and because fertilization without tracers (low N + S treatment) did not systematically alter $\delta^{15}N$ values, we assume (in this paper and in simulations by Currie et al., as reported in this volume) that ecosystem N pools were well defined with respect to isotopic compositions and that ^{15}N contents of individual pools were minimally influenced by low N fertilization per se. Rather, any detectable shifts in $\delta^{15}N$ values in low N treatment plots (receiving total inputs of 5.8 g N m^{-2} per year, with ^{15}N tracers) resulted from tracer retention and not from isotopic perturbations resulting from elevated N inputs or outputs.

In contrast to the minimal shifts in ^{15}N distributions after 13 years of fertilization with non-labeled N at 5 g N m^{-2} per year (low N + S, without tracers), the high N treatment (15.8 g N m^{-2} per year) appears to have altered ^{15}N contents of plant tissues and litter layers (Oi horizon) after 13 years in both stand types. However, Oe, Oa and mineral soils did not appear to have become enriched in ^{15}N under the high N treatment. The overall enrichment of vegetation pools in ^{15}N relative to soil pools is consistent with observations of foliar ^{15}N enrichment in forests subject to chronically elevated N deposition (Gebauer and Schulze, 1991; Emmett et al., 1998). Such ^{15}N enrichment of vegetation and rapidly cycling N pools could be indicative of "leaky" N cycling wherein presumably ^{15}N-depleted nitrate losses accompany forest disturbances (Garten and Van Miegroet, 1994; Garten et al., 1998; Pardo et al., 2002).

Comparisons of plant tissue and soil horizon $\delta^{15}N$ values (Tables 1 and 2) suggest that plant available N is isotopically lighter than bulk N within soil horizons. The $\delta^{15}N$ values of fine roots in control and low N plots increase along with soil $\delta^{15}N$ values, but the fine roots are typically 2–5‰ lighter than soils from which they were sampled. This could be due to fractionations that discriminate against ^{15}N during mineralization and nitrification reactions (see Nadelhoffer and Fry, 1988), to differences in ^{15}N contents of fast-cycling (plant available) versus slow-cycling (resistant) N pools, or to both factors. Fine root ^{15}N did not increase consistently with depth in high N plots, probably due to perturbations of N cycling patterns and increased downward transport of N in soil profiles under the highest N loadings.

4.2. ^{15}N tracer recoveries

Tracers recovered at any time after labeling are the net result of uptake into and losses from individual pools. As such, percent ^{15}N recoveries within a pool are a function of initial uptake, residence time, and subsequent inputs from and losses to other ecosystem pools. For example, because foliage turns over in less than 2 years (1 summer in hardwoods), virtually all of the ^{15}N recovered in foliage in 1999 was likely remobilized to wood or shed as litter within 2 years of labeling. Tracers we detected in foliage 7 years after labeling were recycled from other pools, probably soil, roots, and woody tissues. Even woody tissues, which accumulated ^{15}N tracers (albeit slowly), probably exported some ^{15}N to other pools (foliage, bark, older wood). Organic soil, the strongest initial sink for

labeled inputs, maintained high levels of ^{15}N tracer. Organic soil horizons certainly received tracer inputs from litterfall and root death, and lost tracer through mineralization, plant uptake, and export to underlying mineral soil. Model results (Currie et al., this volume) suggest that organic soils exported labeled N to mineral soil, where it is recycled at decadal scales to vegetation.

Estimates of ^{15}N percent recoveries reported here are likely better than those reported earlier (Nadelhoffer et al., 1999a) for several reasons. First, the 7-year post-label sampling yielded more detailed and reliable N pool estimates (Tables 7 and 8) than the 1992 N pool estimates reported earlier (Tables 3 and 4 in Nadelhoffer et al. (1999a)); soil sampling in 1999 used larger numbers of soil cores to estimate soil C and N pools by O horizons and mineral layers, whereas the 1992 sampling used fewer cores and fewer soil sub-horizons. Second, unlike the 1992 sampling in which soil cores were taken from 5 m × 5 m subplots located along plot edges, the 1999 cores were all taken >5 m from any plot edge. This minimized mixing of litter and roots from untreated areas outside plot boundaries and allowed for better matching with tree samples, all of which were taken >5 m from plot edges in both 1992 and 1999. Third, for control plots (but not low N plots), we wrongly assumed the masses of ^{15}N additions were approximately 2× those actually applied (see Section 2). Therefore, low percent recoveries in control plots at the end of the 1991–1992 labeling period were largely due to a calculation error (rectified here).

We reported earlier (Nadelhoffer et al., 1999a) that organic and mineral soils were stronger sinks for N additions than were trees at the end of labeling in 1992. At that time ^{15}N recoveries in tree biomass ranged from 13.1 to 31.5% of additions to low N plots. Recoveries in trees were much lower on control than on low N plots, even after correcting for the true masses of ^{15}N added to controls. Corrected ^{15}N recoveries in tree biomass (leaves, wood, fine roots) at the end of labeling in 1992 were 4.8% of $^{15}NH_4$ and 9.4% of $^{15}NO_3$ additions to the hardwood control plots (versus previous recovery estimates of 2.3 and 3.9%) and were 2.6% of $^{15}NH_4$ and 5.0% of $^{15}NO_3$ additions to the pine control (versus previously 1.2 and 2.1%).

At 7 years after labeling, as immediately after labeling, tracer recoveries in tree biomass were small relative to recoveries in soils (Fig. 1). Recoveries in tree biomass across treatments in 1999 ranged from 9.2 to 13.0% of ^{15}N additions to the hardwoods (Table 7) and from 6.8 to 15.5% of ^{15}N additions to the pines (Table 8). However, the considerably higher recoveries in trees under chronic fertilization that occurred at the end of labeling were no longer evident by 1999. In 1992, recoveries in trees were several-fold greater on the low N than on the control plots in both hardwoods and pines. By 1999, recoveries in low N trees were either similar to (hardwoods), or slightly greater than (pines) recoveries in control trees. In all cases, ^{15}N tracers continued to accumulate in woody tissues from 1992 through 1999 and either remained constant or increased slightly in leaves and roots on control plots. Under chronic fertilization (low N), however, ^{15}N recoveries in leaves and roots were lower in 1999 than in 1992. It is likely that the decreases in ^{15}N in leaves and roots under fertilization result from continued uptake of non-labeled N additions during the post-labeling period (see Currie et al., this volume).

Organic and mineral soils together were dominant sinks for $^{15}NH_4$ and $^{15}NO_3$ tracers at the end of labeling in 1992 in control (ambient N deposition) and low N plots in both forest types. The initial retention of labeled nitrate in soils, particularly in low N (fertilized) plots where energetic costs of NO_3^- assimilation are high, could have resulted from reduction driven by organic or inorganic proton sources (Davidson et al., 2003). Soils remained the dominant sink for N inputs seven years after tracers were applied, with ^{15}N recoveries in organic plus 0–20 cm mineral soils exceeding 70% of additions in all cases (Fig. 1, Tables 7 and 8). Uncertainties in percent recovery estimates are greater for soil than for plant pools due to the larger soil N pool sizes (Tables 3 and 4) and resulting weaker shifts in $\delta^{15}N$ values (smaller $\Delta\delta$) in soils than in smaller plant N pools. This is particularly true for mineral soils where differences in $\delta^{15}N$ values of samples between labeled and non-labeled plots were always <10‰ (Tables 5 and 6). Recovery estimates for organic soils are considerably more reliable than for mineral soils, as shifts in $\delta^{15}N$ values for Oi, Oe, and Oa horizons were nearly as great as were shifts for plant tissues. Therefore, most of the error in our ecosystem-level estimates of ^{15}N tracer recoveries is likely associated with uncertainties in mineral soils.

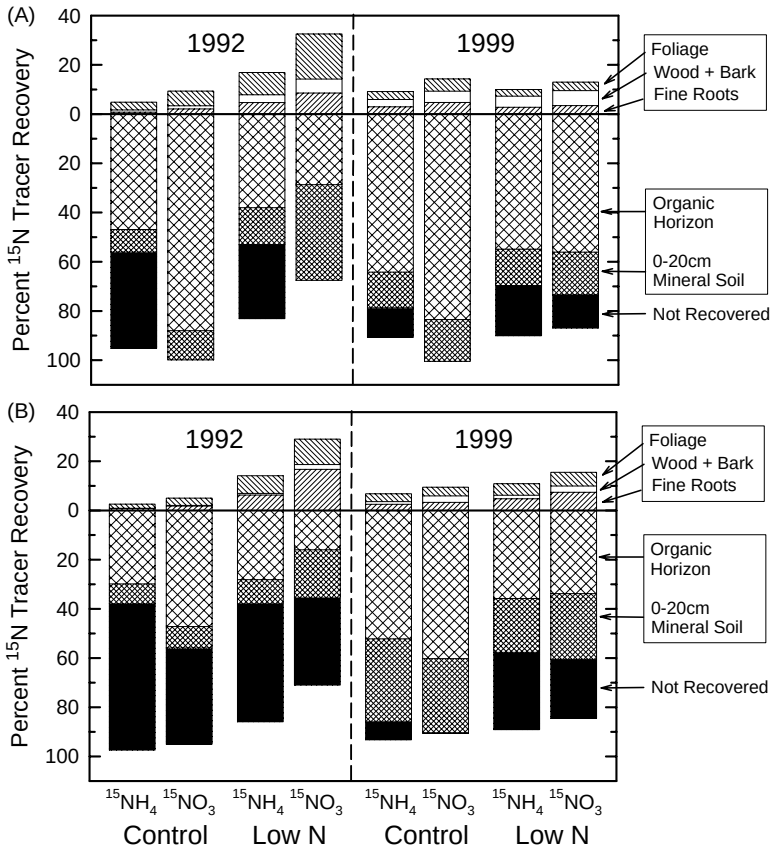

Fig. 1. Percent recoveries of ^{15}N tracers after 2 years of tracer additions (1992) and 7 years after the end of tracer additions (1999) in labeled plots subject to ambient N deposition (control) and 5 g N m^{-2} per year additions (low N). (A) Hardwoods and (B) pines.

Findings that organic soil was the largest measured sink for ^{15}N tracers by 7 years after labeling under all combinations of N loading and tracer form, and that the strength of this sink did not increase with N loading (Fig. 1), suggest that organic horizons in these forests exported large proportions of the fertilizer N inputs. As ^{15}N recoveries in trees either did not increase, or increased only slightly, with chronic fertilization, it is likely that most N exports were to mineral soils (see Currie et al., this volume). These exports were either not detected in the mineral soils due to small signal:pool size ratios, or were transported below the 20 cm soil sampling depth, or both.

Comparisons of ^{15}N contents of fine roots and of soils from which they were sampled suggest that the proportion of total soil N residing in fast-cycling pools is greater in organic than in mineral soils (Tables 5 and 6). Fine root δ^{15}N values were typically similar to values of O subhorizons from which they were sampled, whereas δ^{15}N values of fine roots in mineral soils were consistently greater than values of mineral soil layers from which they were sampled. The larger differences in fine root versus soil δ^{15}N values in mineral soils suggest that N uptake by roots in mineral soil layers is from an N pool that is smaller and more highly labeled than the bulk N pool.

Our end of labeling measures in 1992 showed that trees accumulated more ^{15}N after $^{15}NO_3$ than after $^{15}NH_4$ additions. This trend was still evident in 1999, but was much weaker than in 1992. In contrast to 1992, when most plant pools in both forests contained more ^{15}N after $^{15}NO_3$ than after $^{15}NH_4$ additions (Tables 4 and 5 in Nadelhoffer et al. (1999a)), the only significantly greater recoveries on $^{15}NO_3$-labeled plots were in foliage and roots on pine plots and in

woody tissues in both hardwood and pine plots (Tables 7 and 8). It is important to recognize that greater uptake of $^{15}NO_3$ than of $^{15}NH_4$ additions does not lead to the conclusion that trees are taking up more NO_3-N than NH_4 from soils. Where net nitrification rates are low, or where nitrate concentrations in soil are otherwise low, uptake of $^{15}NO_3$ tracers by tree roots can be high, even when nitrate uptake rates are low (Buchmann et al., 1995).

4.3. Other studies

The dominance of soils over trees as sinks for N inputs in the hardwood and pine forest plots at the end of the labeling period are consistent with results of earlier tracer studies done at forest plot (Preston et al., 1990; Nadelhoffer et al., 1995; Buchmann et al., 1996; Koopmans et al., 1996; Tietema et al., 1998; Perakis and Hedin, 2001; Zak et al., 2004) and catchment scales (Nadelhoffer et al., 1992; Schleppi et al., 1999b; Lamontagne et al., 2000). Recoveries of ^{15}N tracers in trees in these studies ranged from <5 to a maximum of 42% of tracer applications, but most values were less than 25% of ^{15}N additions, particularly when N inputs were <5 g N m^{-2} per year.

Studies comparing movements of NH_4 versus NO_3 inputs to forests using ^{15}N tracers are few in number and have yielded differing results. Buchmann et al. (1996) concluded that Norway spruce (*Picea abies*) in a 12-year-old forest in Germany competed better for NH_4 than for NO_3 additions. Perakis and Hedin (2001) found no differences in NH_4 versus NO_3 uptake by vegetation in a mixed-species forest in Chile. Preston and Mead (1994) reported greater uptake of pulse applications (on snow) of $^{15}NH_4$ than $^{15}NO_3$ by lodgepole pine (*Pinus contorta*) trees in western Canada, but total retention (in plants plus soil) of $^{15}NH_4$ was about two times greater than was $^{15}NO_3$ retention. Trees accounted $10 \pm 2\%$ of total forest ^{15}N retention at 1 year after labeling with either $^{15}NH_4$ or $^{15}NO_3$.

The only other study of which we are aware that has followed the fates of ^{15}N tracers to forest plots at decadal scales is Preston and Mead (1994) who fertilized single-tree plots with 10 g N m^{-2} with either ^{15}N-urea, $^{15}NH_4NO_3$, or $NH_4{}^{15}NO_3$. Eight years after applying labeled fertilizers, approximately two-thirds of applied ^{15}N was recovered on plots, but recoveries in trees did not exceed 9% of total tracer additions and were similar to recoveries in trees at 1 year after labeling. They concluded that most fertilizer N taken up by trees after a single high dose of application occurs within 1 year. This is largely consistent with our 7-year resampling, wherein tracer recoveries in trees either increased slightly (controls) or decreased (low N, 5 g N m^{-2} per year fertilization) due to N turnover in leaves and roots and subsequent uptake of non-labeled fertilizer.

5. Conclusions

Our measurements of ^{15}N tracer recoveries through 7 years following two seasons of ^{15}N additions to forest plots receiving background (0.8 g N m^{-2} per-year) and or chronically elevated NH_4NO_3 inputs (5.8 g N m^{-2} per year) provide information as to how rates and chemical forms of N additions influence the partitioning of N inputs among forest ecosystem components. Our decade-long tracer study at the Harvard Forest chronic N study site suggests:

- Soil is a stronger sink than is tree biomass for ammonium and for nitrate inputs; trees take up from <5% to a maximum of one-third of annual N inputs.
- The percentage of N inputs taken up by trees increases with the rate of N input. This is likely due to decreased competitive effectiveness of microbial and soil processes for N at high levels of N input.
- Nitrate-N is more likely to enter tree biomass pools than is ammonium-N when inputs are as ammonium nitrate, even when ammonium is the dominant form of N taken up by tree roots.
- Trees continue to accumulate N inputs from a given season for at least a decade. Cumulative assimilation of a single year's N inputs into wood over a decade is measurable; however, it is a small fraction of total N input.
- Surface organic soils are stronger sinks for N inputs than are underlying mineral soils at input rates of up to approximately 6 g N m^{-2} per year.

These conclusions apply directly to the hardwood and pine forests under investigation at the Harvard Forest. Some conclusions are less generalizable than others. For example, few studies of the effectiveness of

forest trees in competing for NH_4 versus NO_3 inputs have been conducted elsewhere. The limited number of studies comparing fates of $^{15}NH_4$ and $^{15}NO_3$ tracers have led to different conclusions as to how well trees compete for these two N forms.

Our Harvard Forest ^{15}N study, together with ecosystem-scale ^{15}N tracer studies in other forests, suggests that some patterns we observed are common to temperate forests. All studies of plot and catchment-scale ^{15}N additions to forests, particularly those adding N across growing seasons rather than as pulse additions, suggest that soils are stronger N sinks than trees in forests. Soils typically assimilate from 3 to >10 times more N deposition or fertilizer than do trees. The present study suggests that this is not a transient (1- to 3-year) response; rather, once N inputs enter forest soil pools they can be expected to have mean residence times greater than a decade. Modeling analyses of the time series presented here, however, suggests that some portion of the N retained in soil has a more rapid turnover time and thereby continues to enter plant biomass (and accumulate in wood) during the decade after input (Currie et al., this volume; Currie and Nadelhoffer, 1999). Given the dominance of soils as sinks for N deposition, further research on biotic and abiotic processes controlling N assimilation in soils is clearly needed.

The results we report here, as well as results of shorter term studies at Harvard Forest (Nadelhoffer et al., 1999a) and elsewhere (e.g. Nadelhoffer et al., 1992, 1995; Tietema et al., 1998; Schleppi et al., 1999) have important implications for assessing the effects of N deposition on temperate forest carbon balances. These studies suggest that small fractions of N inputs, generally much less than 10% of deposition, are likely to be assimilated into woody tissues (C:N > 200) in temperate forests. The generally low recoveries of ^{15}N tracers in woody tissues, and high recoveries in soils and other pools with low C:N ratios, suggest that N inputs across growing seasons or years are not likely to stimulate appreciable C accumulation in tree biomass. This is consistent with results of Magill et al. (this volume), who reported either a small (Harvard Forest hardwoods) or no increase (pines) in woody biomass after 15 years of fertilization, and with model predictions (Currie et al., this volume) of average annual accumulation of 25 g C m^{-2}, primarily in woody tissues, through 30 years of N deposition at 5 g m^{-2} per $year^{-1}$. Although the modeling analysis projected the greatest differential C storage as occurring in wood, the prediction is much smaller than previous stoichiometric projections.

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References

Aber, J.D., Magill, A., Boone, R., 1993. Plant and soil responses to chronic nitrogen additions at the Harvard Forest, Massachusetts. Ecol. Appl. 3, 156–166.

Aber, J.D., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L., Fernandez, I., 1998. Nitrogen saturation in temperate forest ecosystems: hypotheses revisited. Bioscience 48, 921–934.

Aber, J.D., Nadelhoffer, K.J., Steudler, P., Melillo, J.M., 1989. Nitrogen saturation in northern forest ecosystems. Bioscience 39, 378–386.

Buchmann, N., Gebauer, G., Schulze, E.D., 1996. Partitioning of N-15-labeled ammonium and nitrate among soil, litter, below- and above-ground biomass of trees and understory in a 15-year-old Picea abies plantation. Biogeochemistry 33, 1–23.

Buchmann, N., Schulze, E.-D., Gebauer, G., 1995. 15N-ammonium and 15N-nitrate uptake of a 15-year old Picea abies plantation. Oecologia 102, 361–370.

Currie, W.S., Nadelhoffer, K.J., 1999. Dynamic redistribution of isotopically labeled cohorts of nitrogen inputs in two temperate forests. Ecosystems 2, 4–18.

Currie, W.S., Nadelhoffer, K.J., 2002. The imprint of land-use history: patterns of carbon and nitrogen in downed woody debris at the Harvard Forest. Ecosystems 5, 446–460.

Currie, W.S., Nadelhoffer, K.J., Aber, J.D., 1999. Soil detrital processes controlling the movement of N-15 tracers to forest vegetation. Ecol. Appl. 9, 87–102.

Currie, W.S., Nadelhoffer, K.J., Colman, B., 2002. Long-term movement of N-15 tracers into fine woody debris under chronically elevated N inputs. Plant Soil 238, 313–323.

Davidson, E.A., Chorover, J., Dail, D.B., 2003. A mechanism of abiotic immobilization of nitrate in forest ecosystems: the ferrous wheel hypothesis. Global Change Biol. 9, 228–236.

Emmett, B.A., Kjønaas, O.J., Gundersen, P., Koopmans, C., Tietema, A., Sleep, D., 1998. Natural abundance of ^{15}N in forests across a nitrogen deposition gradient. For. Ecol. Manage. 101, 9–18.

Emmett, B.A., Quarmby, C., 1991. The effect of harvesting intensity on the fate of applied 15N-ammonium to the organic horizons of a coniferous forest in N. Wales. Biogeochemistry 15, 47–63.

Fry, B., Brand, W., Mersch, F.J., Tholke, K., Garritt, R., 1992. Automated analysis system for coupled ë13C and ë15N measurements. Anal. Chem. 64, 288–291.

Galloway, J.N., 1998. The global nitrogen cycle: changes and consequences. Environ. Pollut. 102, 15–24.

Garten Jr., C.T., Van Miegroet, H., 1994. Relationships between soil nitrogen dynamics and natural 15N abundance in plant foliage from Great Smoky Mountains National Park. Can. J. For. Res. 24, 1636–1645.

Garten, C.T., Schwab, A.B., Shirshac, T.L., 1998. Foliar retention of N-15 tracers implications for net canopy exchange in low- and high-elevation forest ecosystems. For. Ecol. Manage. 103, 211–216.

Gebauer, G., Dietrich, P., 1993. Nitrogen isotope ratios in different compartments of a mixed stand of spruce, larch and beech trees and of understory vegetation including fungi. Isotopenpraxis Environ. Health Stud. 29, 35–44.

Gebauer, G., Schulze, E.-D., 1991. Carbon and nitrogen isotope ratios in different compartments of a healthy and a declining {IPicea abies} forest in the Fichtelbebirge, NE Bavaria. Oecologia 87, 198–207.

Gundersen, P., Emmett, B.A., Kjoenaas, O.J., Koopmans, C.J., Tietema, A., 1998. Impact of nitrogen deposition on nitrogen cycling in forests: a synthesis of NITREX data. For. Ecol. Manage. 101, 37–55.

Högberg, P., Högbom, L., Schinkel, H., Högberg, M., Johannisson, C., Wallmark, H., 1996. 15N abundance of surface soils, roots and mycorrhizas in profiles of European forest soils. Oecologia 108, 207–214.

Jordan, M.J., Nadelhoffer, K.J., Fry, B., 1997. Nitrogen cycling in forest and grass ecosystems irrigated with 15N-enriched wastewater. Ecol. Appl. 7, 864–881.

Koopmans, C.J., Tietema, A., Boxman, A.W., 1996. The fate of N-15 enriched throughfall in two coniferous forest stands at different nitrogen deposition levels. Biogeochemistry 34, 19–44.

Lamontagne, S., Schiff, S.L., Elgood, R.J., 2000. Recovery of superior 15N-labelled nitrate applied to a small upland boreal forest catchment. Can. J. For. Res. 30, 1165–1177.

Magill, A.H., Aber, J.D., Hendricks, J.J., Bowden, R.D., Melillo, J.M., Steudler, P.A., 1997. Biogeochemical response of forest ecosystems to simulated chronic nitrogen deposition. Ecol. Appl. 7, 402–415.

Nadelhoffer, K.J., Aber, J.D., Downs, M.R., Fry, B., Melillo, J.M., 1992. Biological sinks for nitrogen additions to a forested catchment. EPA/600/A-92/292.

Nadelhoffer, K.J., Downs, M.R., Fry, B., 1999a. Sinks for N-15-enriched additions to an oak forest and a red pine plantation. Ecol. Appl. 9, 72–86.

Nadelhoffer, K.J., Downs, M.R., Fry, B., Aber, J.D., Magill, A.H., Melillo, J.M., 1995. The fate of N-15-labeled nitrate additions to a northern Hardwood Forest in eastern Maine, USA. Oecologia 103, 292–301.

Nadelhoffer, K.J., Emmett, B.A., Gundersen, P., Kjønaas, O.J., Koopmans, C.J., Schleppi, P., Tietema, A., Wright, R.F., 1999b. Nitrogen deposition makes a minor contribution to carbon sequestration in temperature forests. Nature 398, 145–148.

Nadelhoffer, K.J., Fry, B., 1988. Controls on natural nitrogen-15 and carbon-13 abundances in forest soil organic matter. Soil Sci. Soc. Am. J. 52, 1633–1640.

Nadelhoffer, K.J., Fry, B., 1994. Nitrogen isotope studies in forest ecosystems. In: Lajtha, K., Michener, R. (Eds.), Stable Isotopes in Ecology. Blackwell Scientific Publications, Oxford, UK, pp. 22–44.

Nihlgård, B., 1985. The ammonium hypothesis: an additional explanation to the forest decline in Europe. Ambio 14, 2–8.

Pardo, L.H., Hemond, H.F., Montoya, J.P., Fahey, T.J., Siccama, T.G., 2002. Response of the natural abundance of N-15 in forest soils and foliage to high nitrate loss following clear-cutting. Can. J. For. Res. Revue Canadienne De Recherche Forestiere 32, 1126–1136.

Perakis, S.S., Hedin, L.O., 2001. Fluxes and fates of nitrogen in soil of an unpolluted old-growth temperate forest, southern Chile. Ecology 82, 2245–2260.

Preston, C.M., Marshall, V.G., McCullough, K., Mead, D.J., 1990. Fate of 15N-labelled fertilizer applied on snow at two forest sites in British Columbia. Can. J. For. Res. 20, 1583–1592.

Preston, C.M., Mead, D.J., 1994. Growth response and recovery of 15N-fertilizer one and eight growing seasons after application to lodgepole pine in British Columbia. For. Ecol. Manage. 65, 219–229.

Schleppi, P., Bucher-Wallin, L., Siegwolf, R., Saurer, M., Muller, N., Bucher, J.B., 1999a. Simulation of increased nitrogen deposition to a montane forest ecosystem: partitioning of the added N-15. Water Air Soil Pollut. 116, 129–134.

Schleppi, P., Bucher-Wallin, L., Siegwolf, R., Saurer, M., Muller, N., Bucher, J.B., 1999b. Simulation of increased nitrogen deposition to a montane forest ecosystem: partitioning of the added N-15. Water Air Soil Pollut. 116, 129–134.

Schulze, E.-D., 1989. Air pollution and forest decline in a spruce (*Picea abies*) forest. Science 244, 776–783.

Seely, B., Lajtha, K., 1997. Application of a 15N tracer to simulate and track the fate of atmospherically deposited N in the coastal forests of the Waquoit Bay Watershed, Cape Cod, Massachusetts. Oecologia 112, 393–402.

TerMikaelian, M.T., Korzukhin, M.D., 1997. Biomass equations for sixty-five North American tree species. For. Ecol. Manage. 97, 1–24.

Tietema, A., Emmett, B.A., Gundersen, P., Kjoenaas, O.J., Koopmans, C.J., 1998. The fate of ^{15}N-labelled nitrogen deposition in coniferous forest ecosystems. For. Ecol. Manage. 101, 19–27.

van Breemen, N., van Dijk, H.F.G., 1988. Ecosystem effects of atmospheric deposition of nitrogen in the Netherlands. Environ. Pollut. 54, 249–274.

van Dam, D., van Breemen, N., 1995. NICCCE a model for cycling of nitrogen and carbon isotopes in coniferous forest ecosystems. Ecol. Model. 79, 255–275.

Wright, R.F., Roelofs, J.G.M., Bredemeier, M., Blanck, K., Boxman, A.W., Emmett, B.A., Gundersen, P., Hultberg, H., Kjønaas, O.J., Moldan, F., Tietema, A., van Breemen, N., van Dijk, H.F.G., 1995. NITREX: responses of coniferous forest ecosystems to experimentally changed deposition of nitrogen. For. Ecol. Manage. 71, 163–169.

Zak, D.R., Pregitzer, K.S., Holmes, W.E., Burton, A.J., Zogg, G.P., 2004. Anthropogenic N deposition and the fate of $^{15}NO_3^-$ in a northern hardwood ecosystem, Biogeochemistry.