JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 96, NO. D5, PAGES 9321-9328, MAY 20, 1991

EFFECTS OF NITROGEN ADDITIONS ON ANNUAL NITROUS OXIDE FLUXES FROM TEMPERATE FOREST SOILS IN THE NORTHEASTERN UNITED STATES

Richard D. Bowden[1], Jerry M. Melillo, and Paul A. Steudler

The Ecosystems Center, Marine Biological Laboratory, Woods Hole, Massachusetts

John D. Aber

Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, Durham

Abstract. Nitrous oxide (N_2O) fluxes between aerobic soils and the atmosphere were measured each month for 2 years (except during the period of snow cover) in control and N-fertilized plots in a red pine plantation and a mixed hardwood forest in the northeastern United States. Nitrogen was added as NH_4NO_3; application rates were 0 (control), 37 (low-N), and 120 (high-N) kg N/ha/yr in year 1. In year 2, application rates were 0 (control), 50 (low-N), and 150 (high-N) kg N/ha/yr. A total of 2520 individual flux measurements were made over the 2 years. Mean emission rates ($\mu g\ N/m^2/h$) from the control plots were low (pine: year 1 = 0.17, year 2 = 1.10; hardwood: year 1 = 0.27, year 2 = 0.29). Fertilization resulted in only small increases in N_2O effluxes even in the second year, with low rates in the low-N plots (pine: year 1 = 2.19, year 2 = 3.87; hardwood: year 1 = 1.16, year 2 = 0.77) and also in the high-N plots (pine: year 1 = 0.71, year 2 = 5.24; hardwood: year 1 = 1.12, year 2 = 0.90). The maximum N_2O loss was 0.350 kg N/ha/yr from the red pine high-N plot. Weak seasonal flux trends were noted in both stands, with highest efflux rates in spring and mid to late summer. No statistically significant relationships between fluxes and soil temperatures, soil moisture, or soil NO_3^- or NH_4^+ were detected. Low rates of net nitrification in both the control plots and the fertilized plots are thought to be responsible for the low N_2O emissions. Small increases in net nitrification were measured in the forest floor of the the pine stand, with rates in the control, low-N, and high-N plots of 1.5, 4.5, and 8.5 kg N/ha/yr, respectively. Net nitrification rates in the hardwood plots were 0.1, 0.9, and 0.6 kg N/ha/yr in the control, low-N, and high-N plots, respectively. Low NH_4^+ availability in the forest floor and low NH_4^+ concentrations in soil solution below the rooting zone (0.5 - 0.7 m) suggest that NH_4^+ availability for nitrification is limited by competition with plant uptake and microbial immobilization, and that this competition is not alleviated by 2 years (50 - 150 kg N/ha/yr) of N addition.

Introduction

Disturbances to forest ecosystems influence rates of soil processes contributing to N_2O production in forest soils, resulting in increased emissions of N_2O to the atmosphere [e.g., Bowden and Bormann, 1986; Luizao et al., 1989; Matson et al., 1987; Levine et al., 1988]. Disturbances can be physical, such as forest cutting or burning, or chemical, such as N additions. High nitrogen (N) deposition due to anthropogenically increased emissions is a chemical disturbance that has become important in some regions of North America and Europe [Bonis et al., 1980; Melillo et al., 1989] and is suggested to be increasing in other areas of the world [Galbally and Gillett, 1988; Mayewski et al., 1990]. Chronic high-N deposition may lead to nitrogen saturation; that is, the availability of ammonium and nitrate in excess of plant uptake and microbial nutritional demand [Aber et al., 1989; Schulze, 1989]. One possible effect of this disturbance on forest ecosystems is an increase in the rate of nitrogen cycling, and hence the potential for an increase in the emission of nitrous oxide (N_2O) from soil to the atmosphere.

Nitrous oxide is a radiatively important tropospheric trace gas that absorbs long-wave infrared radiation and thus influences the global heat budget [Wang et al., 1976; Donner and Ramanathan, 1980; Cicerone, 1987]. It also contributes to stratospheric ozone depletion [Crutzen, 1974, 1976; Crutzen and Ehhalt, 1977; Cicerone, 1987], further affecting the global atmospheric chemical and thermal balance. This gas has received considerable attention because its atmospheric concentration has been increasing at 0.2 - 0.3% per year [Weiss, 1981; Khalil and Rasmussen, 1983; Rasmussen and Khalil, 1986; Brasseur and Hitchman, 1988; Prinn et al., 1990], and increases in the atmosphere can have long-lasting effects. Nitrous oxide has a long (100-175 years) atmospheric residence time [Crutzen, 1976; McElroy et al., 1976; Khalil and Rasmussen, 1983; Cicerone, 1989], and it is 180 times more effective as a greenhouse gas than CO_2 [Lashof and Ahuja, 1990].

Nitrous oxide emissions from forest soils have been related positively to short-term nitrogen additions [Keller et al., 1988] and to nitrification [Matson and Vitousek, 1987], but the response of nitrification and N_2O emissions to chronic N additions is less well known. Recently, McNulty et al. [1990] found a strong relationship between atmospheric N deposition and nitrification in spruce forests, and Bowden et al. [1990] and Melillo et al. [1989] indicated that nitrification is a key factor controlling N_2O emissions from temperate aerobic forest soils. Brumme et al. [1989] also documented high rates of nitrification and N_2O emissions from forests in Germany that have been subjected to chronic high rates of atmospheric N deposition.

The objective of this 2-year study was to determine the effect of ammonium nitrate additions on annual N_2O fluxes from a mixed hardwood forest and a red pine plantation in the temperate forest region of the northeastern United States. The purpose of the N addition was to increase the rates of nitrogen cycling, particularly mineralization and nitrification, thus mimicking long-term chronic N deposition. Environmental factors and soil processes controlling flux rates, as well as seasonal and diel flux patterns, were examined.

Site Description

The study plots, a 2.3-ha 62-year-old red pine (Pinus resinosa Ait.) plantation and an adjacent 2.3-ha 80-year-old mixed hardwood stand, are located at the Harvard Forest in Petersham, Massachusetts (42°30'N, 72°10'W) and have a well-described history [Bowden et al., 1990]. Soil is an Entic Haplorthod (spodosol) of the Gloucester series, is of glacial origin, and is described as a stony, well-drained, sandy loam. It is acidic with a mean forest floor pH value of 3.2 in the pine stand and 3.3 in the hardwood stand. The hardwood stand has a deeper forest floor (6.5 cm) than the pine plantation (4.6 cm).

The pine plantation is nearly pure red pine (97% of the total basal area (50.0 m^2/ha)) and the hardwood stand is dominated by black oak (Quercus velutina Lam.) which comprised 76% of the total basal area (21.8 m^2/ha). The remaining vegetation in the hardwood stand is mostly black birch (Betula lenta L.), paper birch (Betula

[1]Now at Department of Environmental Science, Allegheny College, Meadville, Pennsylvania.

Paper number 91JD00151.
0148-0227/91/91JD-00151$05.00

papyrifera Marsh.), and red maple (Acer rubrum L.). Understory vegetation is sparse in both stands.

Methods

Chronic N deposition was simulated by adding NH_4 and NO_3 to two 30 x 30 m plots in each stand. A low-N plot received 37 kg N/ha/yr in year 1 (1988-1989) and 50 kg N/ha/yr in year 2 (1989-1990) and a high-N plot received 120 kg N/ha/yr in year 1 and 150 kg N/ha/yr in year 2. One plot in each stand served as a control. Plots were separated by corridors at least 5 m wide. Ammonium nitrate was applied in liquid form with a backpack sprayer in six equal treatments throughout the year from May through October. The total water volume applied throughout the year was equivalent to 0.13 mm of rainfall, or less than 0.012% of the 30-year (1950-1980) mean annual precipitation of 108 cm (National Weather Service, personal communication, 1990).

Each plot was divided into 5 x 5 m subplots, and the outer subplots in each plot served as buffer strips and were not sampled. Three interior subplots from each plot were randomly selected for measurement of gaseous fluxes. Soil sampling was restricted to the remaining subplots and was not conducted on the subplots used for gas flux measurements.

Nitrous oxide fluxes were measured using an in situ chamber incubation technique [Steudler et al., 1989; Bowden et al., 1990]. Chambers, constructed of 1.8-cm-thick polyvinyl chloride plastic, consisted of a 28.7-cm-diameter x 4.0-cm-tall chamber top and a 5.2-cm-tall lower portion (anchor) attached to the forest floor with stainless steel stakes. Anchors were pushed no more than 1 cm into the forest floor to minimize disturbance, and the volume within each anchor was measured. Anchors were removed during the winter of 1988-1989, but were left in place throughout the year in 1989-1990. Incubations were conducted by placing chamber tops, colored white to minimize solar heating, on the anchors and holding them in place with two 4.5-kg weights. Total chamber volume, measured when anchors were implanted 1 cm, was approximately 5.4 L. Gas samples were withdrawn from the headspace via a stainless steel luer-lock needle that extended 2.5 cm into the headspace.

Fluxes were measured 6 times (every 4 hours) in a 24-hour period and were timed to provide sampling at approximate minimum and maximum daily soil temperatures. During each 30-min flux measurement, headspace air samples (20 cm^3) were collected at the beginning of the incubation and at 10-min intervals thereafter. Samples were withdrawn with 20-mL nylon syringes. Chamber tops were removed from anchors upon completion of each flux measurement.

Flux measurements began in May 1988 and were conducted for 2 years (year 1=1988-1989, year 2=1989-1990). Measurements were made at least once per month during nonwinter months, and twice per month during most spring and fall months; fluxes were measured at least 2 weeks after each fertilizer application. Fluxes were considered to be zero during winter (December 14, 1988, to April 2, 1989, December 15, 1989, to March 13, 1990) when soils were snow-covered and soil temperatures were at or below freezing. Incubations resumed in the spring when the upper 5 cm of soil began to thaw. Mean spring soil temperatures (0-2.5 cm depth) in the pine plantation were 4.6°C in year 1 and -1.5°C in year 2. In the hardwood stand, spring soil temperatures were 6.5°C and 0°C in years 1 and 2, respectively. Soil below 5 cm was still frozen in the pine stand, and the upper 5 cm of soil in the hardwood stand had thawed within the previous week. The net annual N_2O flux for each plot was calculated as the mean of the annual fluxes of the three chambers. Each sampling date was considered the midpoint of a sampling period, and the net annual flux was the sum of all sampling periods. Annual flux periods for years 1 and 2 were thus 256 and 278 days, respectively.

Laboratory tests showed that chambers and syringes were inert to N_2O, showed no "memory effect" after exposure to high concentrations, and stored samples satisfactorily [Bowden et al., 1990]. Gas samples were generally analyzed within 4 days of collection. Sample preservation could be extended up to 21 days by compressing the 20-cm^3 syringe volume to 15 cm^3 and holding the syringe plunger in place with a rubber band. This technique also prevented diffusion of atmospheric N_2O from the atmosphere into the syringes. Coefficients of variation of triplicate headspace samples taken from the same chamber at the same time ranged from 0.5% to 6%.

Nitrous oxide was measured using a Shimadzu gas chromatograph equipped with an electron capture detector [Bowden et al., 1990]. Carrier gas (95% Ar, 5% CH_4) flowed through the separatory column (Poropak Q, 80/100 mesh) at 20 cm^3/min. Injection and detector temperatures were 275°C and the column was 50°C. Standard curves prepared from certified N_2O standards (299.7, 500, and 999 ppb N_2O in N_2, Scott Specialty Gases, Plumsteadville, Pennsylvania) were linear over the range used.

Nitrous oxide concentrations were corrected to STP conditions and fluxes were calculated using the linear portion of the N_2O concentration increase within the chamber (usually all four samples over the 30-min incubation). Concentration changes were generally linear during the first 10 min of the incubation. Initial concentrations within the chambers at the beginning of each incubation averaged 305 $\pm$ 1 (SE, n=462) ppb during 1988-1989. The minimum detection limit for individual flux measurements was 0.57-1.50 μg N_2O-N/m^2/h, depending upon combined field and analytical precision on each sampling date.

Ambient air (at 1 m), chamber air, and soil temperatures (0-2.5 cm and 2.5-5.0 cm) were measured during each flux measurement. Air temperature was measured at one location in each stand, and chamber air and soil temperatures were measured at one chamber in each plot. Forest floor samples (n=3-9) were analyzed for moisture, NO_3^--N pools, and NH_4^+-N pools during each sampling [Bowden et al., 1990]. Soil NO_3^- and NH_4^+ pools were determined by extracting soil samples with 1 N KCl [Aber et al., 1983]. Net N mineralization and net nitrification rates for the forest floor (O horizon) and topmost 5 cm of the mineral soil (B horizon) were measured from May 1988 until May 1989 using the buried bag technique [Melillo, 1977; Aber et al., 1983]. In this technique, nine soil samples in each plot were incubated in situ in polyethylene plastic bags; net changes in NO_3^- and NH_4^+ indicate net nitrification and mineralization rates, respectively. Incubations lasted 4 to 6 weeks from May through October and 7 months during the overwinter period. Net mineralization and net nitrification were not measured in year 2. Soil solution NO_3^- and NH_4^+ concentrations below the rooting zone (0.5 - 0.7 m) were analyzed monthly during year 2 using porous cup tube lysimeters; three lysimeters were established in each plot in July 1988. Soil solutions were collected from lysimeters to which a suction of 3.5 kg/cm^2 had been applied for 24 hours. Nitrate and NH_4^+ were analyzed as described above [Aber et al., 1983].

An assessment of the role of nitrification versus denitrification as the source of N_2O was made by measuring fluxes during May and October 1988 on similar nearby red pine and mixed hardwood stands (see McClaugherty et al. [1982] for description) to which KNO_3 had been added for 3 years at the rate of 100 kg N/ha/yr. By adding a large source of NO_3^- only, increases in N_2O emissions due to added N could result primarily from denitrification, not from both denitrification and nitrification as might be expected in the plots to which both NH_4^+ and NO_3^- had been added.

Results

Mean annual flux rates from the control plots in the hardwood stand were similar over the 2 years, 0.27 μg N/m^2/h in year 1 (1988-1989) and 0.29 μg N/m^2/h in year 2 (1989-1990). The mean flux in the pine plantation, however, was 6 times greater in year 2 (1.10 μg N/m^2/h) than in year 1 (0.17 μg N/m^2/h). In both stands and in all treatments, we observed both net daily uptake and net daily effluxes (Figure 1). The range of fluxes (μg N/m^2/h) in the hardwood stand was similar among the three plots during the year (control: -3.71 to +5.14; low-N: -3.78 to +3.24; high-N: -3.30 to +5.20). The pine stand, however, had higher fluxes (μg N/m^2/h) in the fertilized plots, as well as lower rates of net daily uptake (control: -1.24 to +5.34; low-N: -1.28 to +22.70, high-N: -1.31 to +34.1).

There were no statistical differences (analysis of variance, $P < 0.05$) in annual N_2O emissions between fertilized and control plots;

however, there were discernible trends. In both forest stands, mean annual effluxes in fertilized plots were greater than effluxes in control plots. The hardwood stand showed only a small increase in N_2O emission due to N addition (Table 1), with the highest rates observed in the fertilized plots in the first year. There was no difference in rates between the two years. In the pine plantation, mean rates of net emissions were higher in the fertilized plots than in the control plot, with the largest fluxes evident in year 2, when rates were nearly 5 times greater than rates in the control plot, and over 7 times greater than rates in year 1 (Table 1). Total N_2O losses from all plots were small compared to N additions. For example, the total emission in the pine high-N plot in year 2 was only 0.35 kg N/ha/yr, compared to 150 kg N that was added per year.

Fluxes showed only weak seasonal trends (Figure 1), with the highest emission rates generally occurring in July or August, and again in the spring (March or April). Spring values were highest in the pine plantation, where the flux in the control plot in 1990 was 5 times the mean annual rate, and the fluxes in the fertilized plots were 6 times the mean annual rate. As reported previously [Bowden et al., 1990], predictable diel patterns were not evident. Periods of net uptake and net efflux were often observed on the same sampling date in the same chamber, but the time of maximal daily uptake or emission was not consistent.

Nitrous oxide fluxes showed no strong relationship to soil parameters (Table 2), including moisture, air or soil temperatures, and N concentrations. High rates of efflux observed in July and August did correspond to the period of highest soil (0-2.5 cm depth) temperatures (Figure 1), but soil temperatures were low in the spring when high flux rates were again observed. Mean daily soil temperatures showed no relationship to mean daily fluxes in any of the plots (Table 2). On any given sampling date, maximum uptake or efflux rates did not correspond to maximum or minimum daily temperatures. No relationships between N_2O fluxes and available (via KCl extraction) soil NO_3^- or NH_4^+ concentrations were observed either. Strong seasonal available soil N concentration patterns were not evident in any of the plots in either the pine plantation or the hardwood stand.

Flux rates from the plots fertilized only with KNO_3 were generally similar to fluxes at the NH_4NO_3-fertilized plots. In the pine plantation, net uptake was measured in the KNO_3 plots in May (Table 3), compared to net emissions in the NH_4NO_3 plots, whereas in October, efflux rates from the KNO_3 plots were higher than the NH_4NO_3 plots. In the hardwood stand, fluxes from the KNO_3 plot in May were within the range observed in the NH_4NO_3 plots, but were higher than the rates in the NH_4NO_3 plots in October.

Net N mineralization during the first year increased in the fertilized plots in both stands (Figure 2). Net nitrification during this year also increased only slightly in the forest floors of the fertilized hardwood plots but showed greater increases in the forest floor and mineral soil of the fertilized plots in the pine plantation. Net nitrification as a percentage of net mineralization in the forest floor was also greater in the fertilized pine plots than in the control plot, increasing from 3.9% in the control to 8.9% and 11.6% in the low-N and high-N plots, respectively.

Concentrations of NH_4^+ in soil solution collected by lysimeters from below the rooting zone were low at nearly all sampling times in all plots (Figure 3). Slightly higher concentrations were observed in the fertilized plots in July, particularly in the hardwood stand, but most measurements were below detection values (0.03 mg N/L). Nitrate concentrations were highest in the high-N pine plot, but were generally low in the other pine plots and in the hardwood stand. As observed for NH_4^+, NO_3^- concentrations were often below detection values (0.02 mg N/L).

Standing stocks of available nitrogen were low in both stands. Extractions of NH_4^+ and NO_3^- from the forest floor showed extremely low net NO_3^- availability, with only slightly higher concentrations observed in the fertilized pine plots compared to the hardwood plots (Table 4). Ammonium concentrations also remained essentially unchanged between the control and fertilized plots.

Discussion

Annual N_2O fluxes in the control plots remained low throughout the 2 years of study. A sixfold increase was measured in the pine control plot from year 1 (0.17 μg N/m^2/h) to year 2 (1.10 μg N/m^2/h), but the higher rate observed in year 2 is still a small rate of efflux compared to other forest ecosystems [Bowden, 1986]. For example, an estimate of the annual flux rate in a northern hardwood forest at the Hubbard Brook Experimental Forest in New Hampshire is 1.7 μg N/m^2/h [Goreau, 1982; Keller et al., 1983], based on measurements during the growing season only. At the Harvard Forest, annual N losses via N_2O emissions from the control plots were less than 0.1% of the annual net N mineralization.

The effect of N additions on N_2O fluxes, even after 2 years of N application, was small in both forest stands. In each year, annual emission rates in the high-N plots were 3-5 times greater than fluxes in the control plots, but the higher emission rates in the fertilized plots were still low relative to other unfertilized temperate forest soils. In Germany, annual flux rates ranged from 3 to 11 μg N/m^2/h in several different hardwood forests [Schmidt et al., 1988], and in Wisconsin,

TABLE 1. Mean 1988-1989 (Year 1) and 1989-1990 (Year 2) Fluxes of N_2O from Control Plots and Plots Receiving NH_4NO_3 in a Red Pine Plantation and a Mixed Hardwood Stand at Harvard Forest

Stand	Plot	N_2O Flux (μg N/m^2/h)				N_2O Flux (kg N/ha/yr)			
		Year 1		Year 2		Year 1		Year 2	
Hardwood	control	0.27	(0.28)	0.29	(0.37)	0.02	(0.02)	0.02	(0.03)
Hardwood	low-N	1.16	(0.30)	0.77	(0.28)	0.07	(0.02)	0.05	(0.02)
Hardwood	high-N	1.12	(0.20)	0.90	(0.12)	0.07	(0.01)	0.06	(0.01)
Pine	control	0.17	(0.25)	1.10	(0.03)	0.01	(0.02)	0.07	(0.002)
Pine	low-N	2.19	(1.37)	3.87	(0.12)	0.13	(0.08)	0.26	(0.01)
Pine	high-N	0.71	(0.32)	5.24	(1.87)	0.04	(0.02)	0.35	(0.13)

Standard error in parentheses; n = three chambers measured during the year in each plot. Low-N plots received 37 kg N/ha/yr in year 1 and 50 kg N/ha/yr in year 2; high-N plots received 120 kg N/ha/yr in year 1 and 150 kg N/ha/yr in year 2. Years 1 and 2 means based on nonwinter flux periods of 256 and 278 days, respectively.

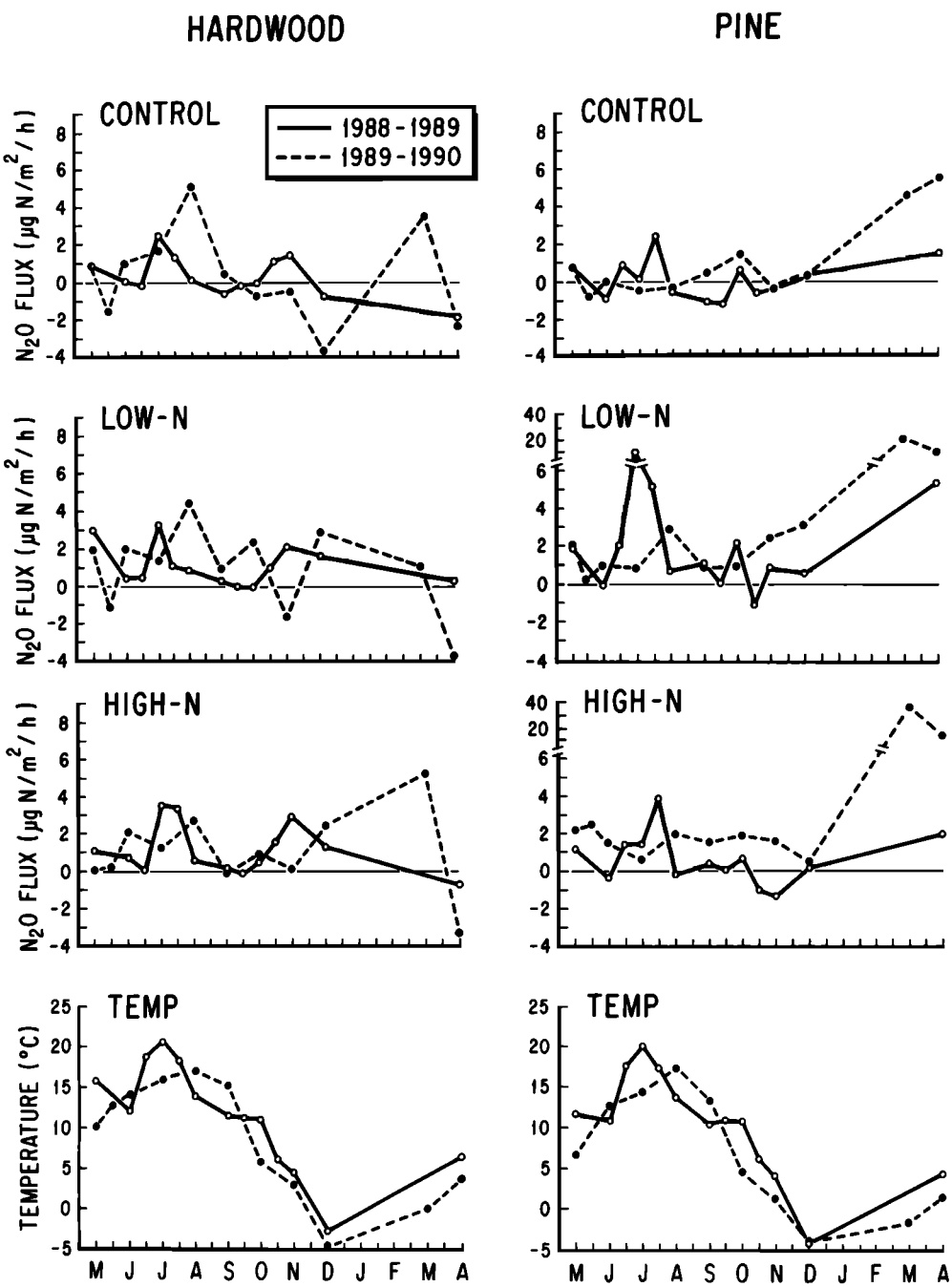


Fig. 1. Nitrous oxide fluxes and soil temperatures (0-2.5 cm depth) on dates of measurements in a mixed hardwood stand and a red pine plantation in control plots and plots receiving NH_4NO_3 at Harvard Forest. Low-N plots received 37 kg N/ha/yr in year 1 and 50 kg N/ha/yr in year 2; high-N plots received 120 kg N/ha/yr in year 1 and 150 kg N/ha/yr in year 2. Fluxes were measured at least 2 weeks after each N addition. Positive fluxes are emissions from the soil to the atmosphere; negative fluxes are uptake. Fluxes are means of three chambers per plot. Note scale changes.

emission rates were 5.4 μg $N/m_2/h$ in a pine plantation and 27.7 μg $N/m^2/h$ in a hardwood stand [Goodroad and Keeney, 1984]. Much higher flux rates have also been reported from fertilized plots in a tropical forest. In a short-term study [Keller et al., 1988] following a one-time N addition (200 kg N/ha) to soil in Brazil, N_2O emissions increased from 21 μg $N/m^2/h$ in control plots to 456 μg $N/m^2/h$ in NO_3^--amended plots and 113 μg $N/m^2/h$ in NH_4^+-amended plots. After 13 days, however, flux rates from the NO_3^--amended plots were reduced by 90%, and fluxes from the NH_4^+-amended plots were similar to rates in the control plots.

Soil moisture content and temperature often exert strong influences on N_2O fluxes [Payne, 1981a, b; Sahrawat and Keeney, 1986], but because flux rates were extremely low, no such relationships were observed at Harvard Forest. This suggests that other controls are important in determining rates of N_2O emissions by these soils. Soil NO_3^- and NH_4^+ concentrations also showed no relationship to N_2O fluxes (Table 2), but the instantaneous amount of N (soil NO_3^- or NH_4^+ concentrations) in soil pools may be less important than the amount cycling through the soil pools in determining the rate of N_2O production. It is therefore important to examine the processes

TABLE 2. Correlations (r^2) Between N_2O Fluxes and Soil Moisture, Temperature, and Forest Floor NH_4^+ and NO_3^- Concentrations in Control, Low-N, and High-N Plots Within a Red Pine Plantation (P) and a Mixed Hardwood Stand (H) at Harvard Forest

	Control		Low-N		High-N		All Plots Combined	
	P	H	P	H	P	H	P	H
N_2O flux versus								
Temperature	0.07	0.15	0.13	0.06	0.26	0.17	-	-
Moisture	0.00	0.04	0.00	0.00	0.11	0.05	-	-
NH_4^+-N	-	-	-	-	-	-	0.0003	0.08
NO_3^--N	-	-	-	-	-	-	0.015	0.0004

Within each plot n = 17; n = 51 when all plots within each stand were combined.

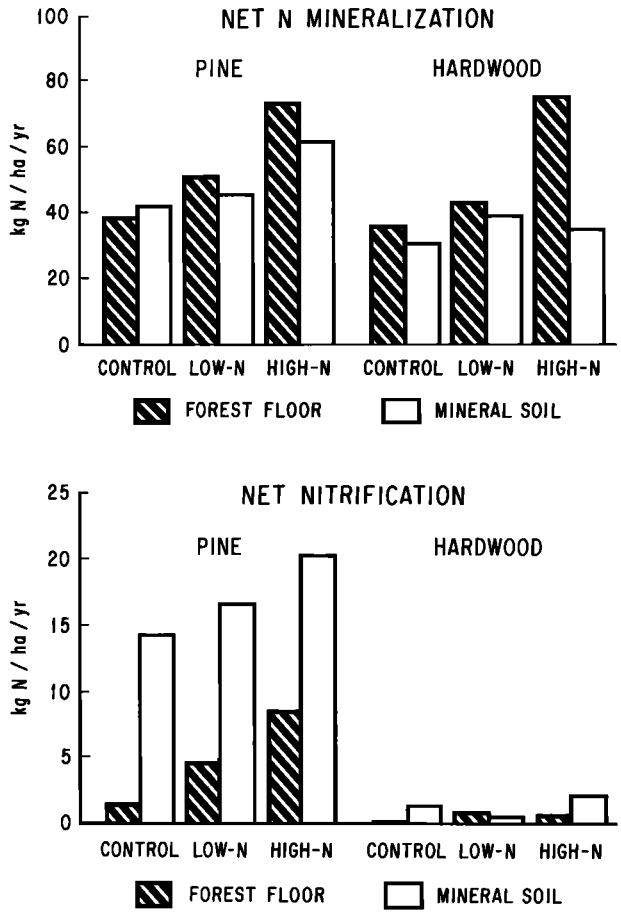
TABLE 3. Comparison of N_2O Fluxes at NH_4NO_3 and KNO_3^--Fertilized Red Pine Plantation and Mixed Hardwood Plots at Harvard Forest in 1988

Stand	Treatment	N Added kg N/yr	N_2O Flux $\mu g\ N/m^2/h$		Difference Between Treatment and Control $\mu g\ N/m^2/h$	
			May	October	May	October
Pine	NH_4NO_3	0	-0.10	-0.08	-	-
Pine	NH_4NO_3	50	+0.95	+0.39	+1.05	+0.47
Pine	NH_4NO_3	150	+0.39	-0.16	+0.49	-0.08
Pine	KNO_3	0	+0.12	+0.01	-	-
Pine	KNO_3	100	-0.40	+5.49	-0.52	+5.48
Hardwood	NH_4NO_3	0	+0.49	+0.53	-	-
Hardwood	NH_4NO_3	50	-0.53	+0.44	-1.02	-0.09
Hardwood	NH_4NO_3	150	+1.72	+1.00	+1.23	+0.47
Hardwood	KNO_3	0	+0.42	+1.24	-	-
Hardwood	KNO_3	100	+0.88	+2.64	+0.46	+1.40

Positive fluxes are emissions from the soil to the atmosphere; negative fluxes are uptake.

responsible for the production of N_2O from forest soils. In most soils, these are thought to be nitrification and denitrification [Sahrawat and Keeney, 1986; Tiedje, 1987; Robertson, 1989]. Chemodenitrification and nonrespiratory denitrification can produce N_2O; however, contributions from chemodenitrification are generally considered to be small [Bremner and Nelson, 1968; Nelson and Bremner, 1970] and the importance of nonrespiratory denitrifiers is not well understood [Bleakley and Tiedje, 1982; Robertson and Tiedje, 1987].

Denitrification does not appear to represent an important potential source of N_2O from the soils at Harvard Forest, even if sufficient NO_3^- is added through fertilization or through increased nitrification. The coarse texture and well-drained moisture status of the soil results in predominantly aerobic conditions, considered the most important factor limiting denitrification in many soils [Tiedje, 1987; Groffman et al., 1988; Robertson, 1989]. Even in the plots where NO_3^- had been the sole source of added nitrogen, and where



the duration of nitrogen addition was much larger (5 years) than the NH_4NO_3-amended plots (2 years), we still do not see consistently higher rates of N_2O emissions. This suggests strongly that denitrification is not an important N_2O source in these soils.

Nitrification is probably the most important N_2O-generating process in coarse-textured, aerobic soils [Keller et al., 1986] and appears to be the key process controlling N_2O emissions from the soil at Harvard Forest. Increases in N_2O effluxes were greatest in the fertilized pine plots (Table 1) where increases in net nitrification had also increased (Figure 2). Overall, however, net nitrification rates were extremely low in the forest floor of the control plots, and the rates were very low even in the high-N plots (pine: 8.5 kg N/ha/yr; hardwood: 0.8 kg N/ha/yr). In comparison, Melillo [1977] reported a rate of 13 kg N/ha/yr for the forest floor in northern hardwood soils at Hubbard Brook Experimental Forest and Aber et al. [1985] reported up to 82 kg N/ha/yr in pine stands and 135 kg N/ha/yr in hardwood stands in Wisconsin. Thus because of the low rate of net nitrification in the soil at Harvard Forest, N_2O losses are low.

Competition of nitrifying organisms with microbial N immobilization (i.e., net microbial N uptake) and plant uptake may be responsible for the low rates of net nitrification in the Harvard Forest soils because nitrifiers are poor competitors for NH_4^+ [Jones and Richards, 1977; Schmidt, 1982]. The potential for high rates of nitrification has been demonstrated in northern temperate forest soils where plant uptake has been reduced (e.g., forest cutting [Likens et al., 1969], laboratory soil incubations [Melillo, 1977], root-free forest

Fig. 2. Annual net N mineralization and net nitrification in forest floor and mineral soil (upper 10 cm) horizons in a mixed hardwood stand and a red pine plantation in control and fertilized plots at Harvard Forest from May 1988 to April 1989 (year 1). Low-N plots received 37 kg N/ha/yr in year 1 and high-N plots received 120 kg N/ha/yr in year 1.

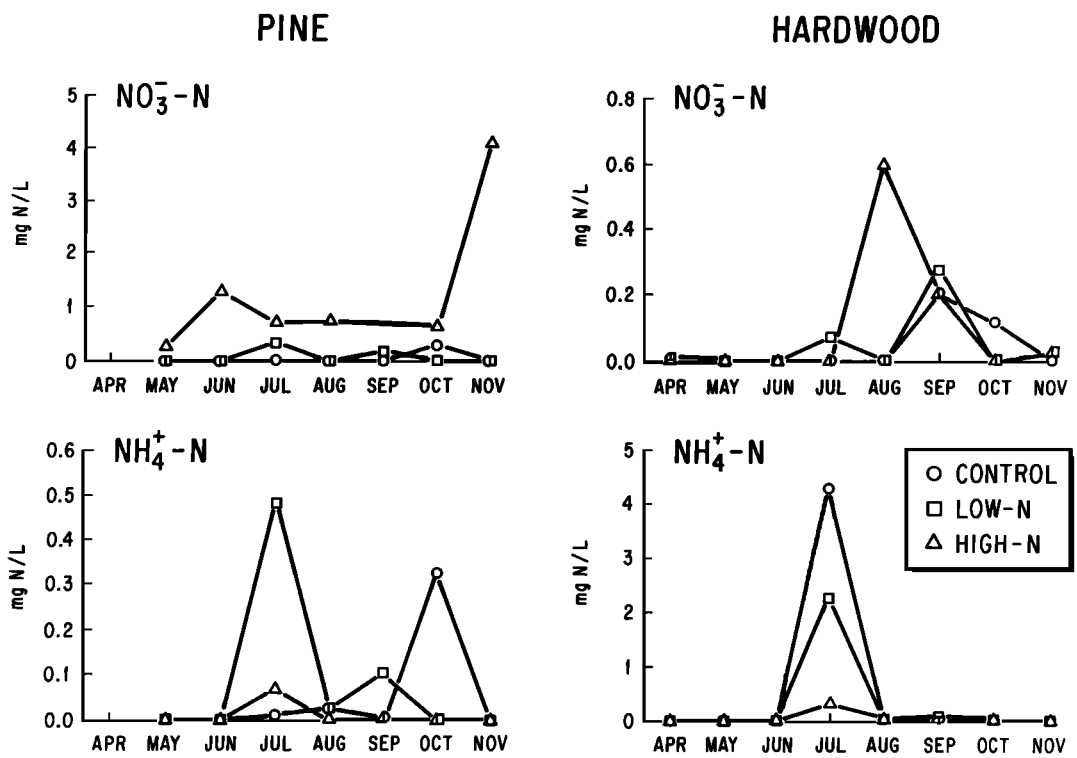


Fig. 3. Concentrations of NO_3^- and NH_4^+ in soil solution collected from lysimeters placed below the rooting zone (0.5 - 0.7 m) in a mixed hardwood stand and a red pine plantation in control and fertilized plots at Harvard Forest in 1989. Concentrations represent means of 4-5 lysimeters. Means are not weighted for volume.

TABLE 4. Mean Available (Determined by KCl Extractions) NH_4^+ and NO_3^- (mg N/kg Soil) in Forest Floor in Control and Fertilized Plots in a Red Pine Plantation and Mixed Hardwood Stand at Harvard Forest

		1988		1989		1988 - 1989		
Stand	Plot	Mean	SE	Mean	SE	Mean	SE	kg N/ha
				NH_4^+				
Hardwood	control	33.7	4.4	39.0	7.1	35.9	3.8	2.2
Hardwood	low-N	30.7	3.0	37.5	7.6	33.5	3.6	1.7
Hardwood	high-N	44.8	6.1	49.7	9.7	46.8	5.2	2.9
Pine	control	68.0	10.9	50.9	10.4	60.9	7.8	2.3
Pine	low-N	60.7	7.7	55.0	7.3	58.3	5.3	2.8
Pine	high-N	105.6	15.9	67.8	7.3	90.0	13.6	4.5
				NO_3^-				
Hardwood	control	0.1	0.0	0.004	0.003	0.1	0.04	0.006
Hardwood	low-N	0.2	0.1	0.05	0.05	0.1	0.07	0.005
Hardwood	high-N	2.0	1.2	0.8	0.7	1.5	0.07	0.09
Pine	control	1.1	0.4	0.4	0.2	0.8	0.3	0.03
Pine	low-N	3.2	1.2	1.3	0.7	2.4	0.8	0.1
Pine	high-N	18.1	8.0	6.4	1.9	13.2	4.9	0.7

Low-N = 37 kg N/ha/yr in year 1 and 50 kg N/ha/yr in year 2, high-N = 120 kg N/ha/yr in year 1 and 150 kg N/ha/yr in year 2 (n = 10 for 1988, n = 7 for 1989, n = 17 for 1988-1989).

plots [Vitousek et al., 1982]). Rapid increases in net nitrification have also been observed following cutting of tropical forests [Matson et al., 1987; Montagnini and Buschbacher, 1988], and Matson and Vitousek [1987] observed a strong relationship between nitrification potentials and N_2O emissions across a range of tropical soils.

Low concentrations of NH_4^+ in the soil and in the lysimeters in the control and fertilized plots of both stands at Harvard Forest indicate that most NH_4^+ added in fertilization or produced through mineralization remains within the rooting zone, and is probably sequestered by plant uptake and microbial immobilization. Nitrification in this forest, therefore, may be limited by low NH_4^+ availability.

Evidence from other studies suggests that chronic N additions will eventually result in increased nitrification and elevated N_2O emissions. High rates of nitrification (50 kg N/ha/yr, R. Brumme, personal communication, 1989) and N_2O emissions (5.7 kg N/ha/yr) have been measured in the Solling region of Germany where high levels of N deposition have been occurring for several decades [Brumme, 1990]. Increases in net nitrification have been documented similarly at high elevation sites in the eastern United States exposed to elevated levels of N deposition, where net nitrification as a percentage of net mineralization is directly proportional to the amount of N deposition [McNulty et al., 1990]. Small but similar increases in the percentage of nitrified NH_4^+ were observed in the pine plantation at Harvard Forest during this study (3.9% in the control, 8.9% in the low N plot, 11.6% in the high-N plot), but nitrification appears limited by competition with microbial nitrogen immobilization and plant uptake. Increases in nitrification and N_2O emissions after 2 years at Harvard Forest were small on both a relative and an absolute scale, but do indicate that chronic N additions have the potential to alter the rates of nitrogen cycling processes and to change the rates of N_2O emissions from soils to the atmosphere.

Acknowledgments. This research was supported by the Ecosystems Studies Program of the National Science Foundation and is a contribution to the Harvard Forest Long Term Ecological Research program. We thank Anne (Molly) Boutwell, Tim Gregg, Michelle R. Miliefsky, Alison Magill, Andrea Ricca and Dan Sperduto for their field and laboratory assistance, and we thank two anonymous reviewers for their helpful suggestions. Dave Kicklighter is recognized for valuable contributions toward the development of data acquisition and analysis capabilities. We also appreciate the cooperation of the Harvard Forest staff.

References

Aber, J. D., J. M. Melillo, C. A. McClaugherty and K. N. Eshleman, Potential sinks for mineralized nitrogen following disturbance in forest ecosystems, Ecol. Bull., 35, 179-192, 1983.

Aber, J. D., J. M. Melillo, K. J. Nadelhoffer, C. A. McClaugherty, and J. Pastor, Fine root turnover in forest ecosystems in relation to quantity and form of nitrogen availability: A comparison of two methods, Oecologia, 66, 317-321, 1985.

Aber, J. D., K. J. Nadelhoffer, P. A. Steudler, and J. M. Melillo, Nitrogen saturation in northern forest ecosystems, BioScience, 39(6), 378-386, 1989.

Bleakley, B. H., and J. M. Tiedje, Nitrous oxide production by organisms other than nitrifiers or denitrifiers, Appl. Environ. Microbiol., 44, 1342-1348, 1982.

Bonis, K., E. Meszaros, and M. Putsay, On the atmospheric budget of nitrogen compounds over Europe, Idojaras, 84, 57-68, 1980.

Bowden, R. D., P. A. Steudler, J. M. Melillo, and J. D. Aber, Annual nitrous oxide fluxes from temperate forest soils in the northeastern United States, J. Geophys. Res., 95, 13997-14005, 1990.

Bowden, W. B., Gaseous nitrogen emissions from undisturbed terrestrial ecosystems: An assessment of their impacts on local and global nitrogen budgets, Biogeochemistry, 2, 244-279, 1986.

Bowden, W. B., and F. H. Bormann, Transport and loss of nitrous oxide in soil water after forest clearcutting, Science, 233, 867-869, 1986.

Brasseur, G., and M. H. Hitchman, Stratospheric response to trace gas perturbations: Changes in ozone and temperature distributions, Science, 240, 634-637, 1988.

Bremner, J. M., and D. W. Nelson, Chemical decomposition of nitrite in soils, Trans. Int. Congr. Soil Sci. 9th, 2, 495-503, 1968.

Brumme, R., Gasförmige N-Austräge aus Waldböden (PA-2), Ber. Forschungszentrum Waldokysteme, Reihe B., 7, 68-77, 1990.

Cicerone, R. J., Changes in stratospheric ozone, Science, 237, 35-42, 1987.

Cicerone, R. J., Analysis of sources and sinks of atmospheric nitrous oxide (N_2O), J. Geophys. Res., 94, 18265-18271, 1989.

Crutzen, P. J., Estimates of possible variation in total ozone due to natural causes and human activity, Ambio, 3, 201-210, 1974.

Crutzen, P. J., Upper limits on atmospheric ozone reductions following increased application of fixed nitrogen to the soil, Geophys. Res. Lett., 3, 169-172, 1976.

Crutzen, P. J., and D. H. Ehhalt, Effects of nitrogen fertilizers and combustion on the stratospheric ozone layer, Ambio, 6, 112-117, 1977.

Donner, L., and V. Ramanathan, Methane and nitrous oxide: Their effects on terrestrial climate, J. Atmos. Sci., 37, 119-124, 1980

Galbally, I. E., and R. W. Gillett, Processes regulating nitrogen compounds in the tropical atmosphere, in Acidification in Tropical Countries, edited by H. Rodhe and R. Herrera, pp. 73-116. John Wiley, New York, 1988.

Goodroad, L. L., and D. R. Keeney, Nitrous oxide emission from forest, marsh, and prairie ecosystems, J. Environ. Qual., 13, 448-452, 1984.

Goreau, R. J., The biogeochemistry of nitrous oxide, Ph.D. thesis, Harvard Univ., Cambridge, Mass., 1982.

Groffman, P. M., J. M. Tiedje, G. P. Robertson, and S. Christensen, Denitrification at different temporal and geographical scales, proximal and distal controls, in Advances in Nitrogen Cycling in Agricultural Ecosystems, edited by J. R. Wilson, pp. 174-192, Commonwealth Agricultural Bureau, Perth, Australia, 1988.

Jones, J. M., and B. N. Richards, Effects of reforestation on turnover of N-15 labelled ammonium plus nitrate in relation to change in soil microflora, Soil Biol. Biochem., 9, 383-392, 1977.

Keller, M., T. J. Goreau, S. C. Wofsy, W. A. Kaplan, and M. B. McElroy, Production of nitrous oxide and consumption of methane by forest soils, Geophys. Res. Lett., 10, 1156-1159, 1983.

Keller, M., W. A. Kaplan, and S. C. Wofsy, Emissions of N_2O, CH_4, and CO_2 from tropical forest soils, J. Geophys. Res., 91, 11791-11802, 1986.

Keller, M., W. A. Kaplan, S. C. Wofsy, and J. M. Da Costa, Emissions of N_2O from tropical forest soils: Response to fertilization with NH_4^+, and PO_4^{3-}, J. Geophys. Res., 93, 1600-1604, 1988.

Khalil, M. A. K., and R. A. Rasmussen, Increase and seasonal cycles of nitrous oxide in the Earth's atmosphere, Tellus, 35B, 161-169, 1983.

Lashof, D. A., and D. R. Ahuja, Relative contributions of greenhouse gas emissions to global warming, Nature, 344, 59-531, 1990.

Levine, J. S., W. R. Cofer, D. I. Sebacher, E. L. Winstead, S. Sebacher, and P. J. Boston, The effects of fire on biogenic soil emissions of nitric oxide and nitrous oxide, Global Biogeochem. Cycles, 3, 445-449, 1988.

Likens, G. E., F. H. Bormann, and N. M. Johnson, Nitrification: Importance to nutrient losses from a cutover forested ecosystem, Science, 163, 1205-1206, 1969.

Luizao, F., P. Matson, G. Livingston, R. Luizao, and P. Vitousek, Nitrous oxide flux following tropical land clearing, Global Biogeochem. Cycles, 3, 281-285, 1989.

Matson, P. A., and P. M. Vitousek, Cross-system comparisons of soil nitrogen transformations and nitrous oxide flux in tropical forest ecosystems, Global Biogeochem. Cycles, 1, 163-170, 1987.

Matson, P. A., P. M. Vitousek, J. J. Ewel, M. J. Mazzarino, and G. P. Robertson, Nitrogen transformations following tropical forest felling and burning on a volcanic soil, Ecology, 68, 491-502, 1987.

Mayewski, P. A., W. B. Lyons, M. J. Spencer, M. S. Twickler, C. F. Buck, and S. Whitlow, An ice-core record of atmospheric response to anthropogenic sulphate and nitrate, Nature, 346, 554-556, 1990.

McClaugherty, C., J. Aber, and J. Melillo, The role of fine roots in the organic matter and nitrogen budgets of two forested ecosystems, Ecology, 63, 1481-1490, 1982.

McElroy, M. B., J. W. Elkins, S. C. Wofsy and Y. L. Yung, Sources and sinks for atmospheric N_2O, Rev. Geophys., 14, 143-150, 1976.

McNulty, S. G., J. D. Aber, T. M. McLellan, and S. M. Katt, Nitrogen cycling in high elevation forests of the northeastern US in relation to nitrogen deposition, Ambio, 19, 38-40, 1990.

Melillo, J. M., Mineralization of nitrogen in northern forest ecosystems, Ph.D. thesis, Yale Univ., New Haven, Conn, 1977.

Melillo, J. M., P. A. Steudler, J. D. Aber, and R. D. Bowden, Atmospheric deposition and nutrient cycling, in Exchange of Trace Gases Between Terrestrial Ecosystems and the Atmosphere, edited by M. O. Andreae and D. S. Schimel, pp. 263-280, John Wiley, New York, 1989.

Montagnini, F., and R. Buschbacher, Nitrification rates in two undisturbed tropical rain forest and three slash-and-burn sites of the Venezuelan Amazon, Biotropica, 21, 9-14, 1988.

Nelson, D. W., and J. M. Bremner, Gaseous products of nitrite decomposition in soils, Soil Biol. Biochem., 2, 203-215, 1970.

Payne, W. J., The status of nitric oxide and nitrous oxide as intermediates in denitrification, in Denitrification, Nitrification, and Atmospheric Nitrous Oxide, edited by C. C. Delwiche, pp. 85-103, John Wiley, New York, 1981a.

Payne, W. J., Denitrification, John Wiley, New York, 214 pp, 1981b.

Prinn, R., D. Cunnold, R. Rasmussen, P. Simmonds, F. Alyea, A. Crawford, P. Fraser and R. Rosen, Atmospheric emissions and trends of nitrous oxide deduced from 10 years of ALE-GAGG data, J. Geophys. Res., 95, 18369-18385, 1990.

Rasmussen, R. A., and M. A. K. Khalil, Atmospheric trace gases: Trends and distributions over the last decade, Science, 232, 1623-1624, 1986.

Robertson, G. P., Nitrification and denitrification in humid tropical ecosystems: Potential controls on nitrogen retention, in Mineral Nutrients in Tropical Forest and Savanna Ecosystems, edited by J. Proctor, pp. 55-69, Blackwell Scientific, Boston, Mass., 1989.

Robertson, G. P., and J. M. Tiedje, Nitrous oxide sources in aerobic soils: nitrification, denitrification, and other biological processes, Soil Biol. Biochem., 19, 187-193, 1987.

Sahrawat, K. L., and D. R. Keeney, Nitrous oxide emission from soils, Adv. Soil Sci., 4, 103-148, 1986.

Schmidt, E. L., Nitrification in soil, in Nitrogen in Agricultural Soils, edited by F. J. Stevenson, pp. 253-288, Agron. Monogr. Vol. 22, American Society of Agronomy, Madison, Wis., 1982.

Schmidt, J., W. Seiler, and R. Conrad, Emission of nitrous oxide from temperate forest soils into the atmosphere, J. Atmos. Chem., 6, 95-115, 1988.

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

Steudler, P. A., R. D. Bowden, J. M. Melillo and J. D. Aber, Influence of nitrogen fertilization on methane uptake in temperate forest soils, Nature, 341, 314-316, 1989.

Tiedje, J., Ecology of denitrification and dissimilatory nitrate reduction to ammonium, in Environmental Microbiology of Anaerobes, edited by A. J. B. Zehnder, pp. 1-54, John Wiley, New York, 1987.

Vitousek, P. M., J. R. Gosz, C. C. Grier, J. M. Melillo, and W. A. Reiners, A comparative analysis of potential nitrification and nitrate mobility in forest ecosystems, Ecol. Monogr., 53, 152-177, 1982.

Wang, W. C., Y. L. Yung, A. A. Lacis, T. Mo., and J. E. Hansen, Greenhouse effects due to man-made perturbations of trace gases, Science, 194, 685-690, 1976.

Weiss, R. F., The temporal and spatial distribution of tropospheric nitrous oxide, J. Geophys. Res., 86, 7185-7195, 1981.

J. D. Aber, Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, Durham, NH 03824.

R. D. Bowden, Department of Environmental Science, Allegheny College, Meadville, PA 16335.

J. M. Melillo and P. A. Steudler, The Ecosystems Center, Marine Biological Laboratory, Woods Hole, MA 02543.

(Received August 27, 1990;
revised January 10, 1991;
accepted January 11, 1991.)