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ANNUAL NITROUS OXIDE FLUXES FROM TEMPERATE FOREST SOILS IN THE NORTHEASTERN UNITED STATES

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Abstract. Nitrous oxide (N_2O) fluxes were measured from soils in a red pine plantation and a mixed hardwood stand in the northeastern United States. On an annual basis, the pine stand had an efflux of 0.010 ± 0.015 kg N/ha/yr and the hardwood stand had an efflux of 0.017 ± 0.017 kg N/ha/yr. Low net nitrification rates in both stands are suggested as the reason for the low rates of N_2O emissions. Slight seasonal trends were noted in both stands, with highest efflux rates in early summer and late fall. Both stands showed small but extended periods of uptake during summer and early fall. Diel patterns were not observed in either stand. This study suggests that on a global basis, a lower limit for N_2O emissions from temperate forests is 0.012 Tg N/yr, contributing less than 1% of the estimated total emissions from terrestrial sources.

Introduction

Nitrous oxide is a trace gas that has received considerable attention because of its importance in atmospheric chemistry and its influence in controlling the global heat budget. Nitrous oxide functions as a greenhouse gas by absorbing long-wave infrared radiation, which leads to energy retention within the atmosphere [Wang et al., 1976; Donner and Ramanathan, 1980; Cicerone, 1987]. Additionally, this gas contributes to stratospheric ozone depletion [Crutzen, 1974, 1976; Crutzen and Ehhalt, 1977; Cicerone, 1987], further influencing the global atmospheric thermal balance. Interest in N_2O has accelerated recently because the atmospheric concentration of this gas has been increasing at 0.2 - 0.3% per year [Weiss, 1981; Khalil and Rasmussen, 1983; Rasmussen and Khalil, 1986; Brasseur and Hitchman, 1988]. Small atmospheric increases can have long lasting effects because nitrous oxide has an atmospheric residence time of 100-175 years [Crutzen, 1976; McElroy et al., 1976; Khalil and Rasmussen, 1983].

A number of possible contributors to increased atmospheric N_2O have been investigated. Considerable attention has focused on effects of increased use of nitrogen fertilizers in agriculture [Crutzen, 1976; Crutzen and Ehhalt, 1977; McElroy et al., 1977; Liu et al., 1977; Seiler and Conrad, 1981; Firestone, 1982], and on contributions from biomass burning [Crutzen et al., 1979, 1985]. Fossil fuel combustion was considered to be a large source of atmospheric N_2O [Pierotti and Rasmussen, 1976; Crutzen and Ehhalt, 1977; Hao et al., 1987] but recent measurements indicate that it may contribute only a tenth of previous estimates [Muzio and Kramlich, 1988].

The magnitude of natural N_2O sources, including temperate forests, is still largely unknown. Globally, temperate forests occupy approximately 24% of total forested land. Numerous measurements of N_2O fluxes from temperate forests have been made, but there is a wide range of annual flux estimates, even for similar types of systems [Bowden, 1986]. Factors influencing these rates include vegetation types, soil and climatic conditions, differences in the sources of N_2O (e.g., nitrification, denitrification, other sources) and the diversity of techniques used to measure fluxes. Most annual estimates have also been derived from relatively few measurements. Some annual studies have been reported [Keller et al., 1983; Goodroad and Keeney, 1984a; Schmidt et al., 1988], but they are based on measurements taken only during daytime hours or during growing seasons. Strong diel or seasonal variations in fluxes may affect the accuracy of these estimates.

The purpose of this study was to determine the annual N_2O flux from a mixed hardwood stand and a pine plantation in the temperate forest region of the northeastern United States. Environmental factors and soil processes controlling flux rates, as well as seasonal and diel flux patterns, were examined.

Site Description

The study site is located at the Harvard Forest, Petersham, Massachusetts. Two adjacent 2.3-ha stands with well-documented histories were used: a 62-year-old pine plantation and a circa 80-year-old mixed hardwood stand. Both were established on Entic Haplorthods (spodosols) of the Gloucester series following abandonment from pastures in the early 1900s. The soil is of glacial origin, described as a stony, well-drained sandy loam. Soils in both stands are mors, although the hardwood stand has a deeper forest floor (6.5 ± 1.3 cm (S.E., n=9)) than the pine plantation (4.6 ± 0.3 cm (S.E., n=9)). Forest floors in both stands are acidic, with a mean pH value of 3.2 in the pine stand and 3.3 in the hardwood stand.

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The red pine stand is nearly pure red pine (Pinus resinosa Ait.), which comprises 97% of the total basal area (50.0 m^2/ha). The hardwood stand is dominated by black oak (Quercus velutina Lam.), comprising 76% of the total basal area (21.8 m^2/ha). Most of the remaining vegetation consists of black birch (Betula lenta L.), paper birch (Betula papyrifera Marsh.), and red maple (Acer rubrum L.). Total stem density is 1080 stems/ha in the pine stand and 1770 stems/ha in the hardwood stand. Understory vegetation is sparse in both stands, consisting of scattered seedlings and saplings of hardwoods and pines.

Methods

A 30x30 m plot was established in each stand, and each plot was divided into 5x5 m subplots. 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; no soil was removed from subplots used to measure gas fluxes.

Nitrous oxide fluxes were measured using two-piece 0.065 m^2 polyvinyl chloride (PVC) plastic chambers [Steudler et al., 1989] to cover the soil surface (Figure 1). Chambers had an inner diameter of 28.7 cm and a wall thickness of 1.8 cm. The lower half of the chamber (anchor) was 5.2 cm in height, was beveled at the base to provide a sealing surface of 2.7 cm, and was held to the forest floor with stainless steel rods. The O horizon was left intact; thus litter was not removed from the anchors. 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 left in place throughout the year except during winter when samples were not collected. During sampling, the chambers were assembled by placing a 4.1-cm-tall chamber top upon the anchor. The bottom of the chamber top was grooved and fitted with a Viton O-ring and an effective seal between the top and the anchor was created by placing two 4.5-kg weights on the chamber top. Total chamber volume, when anchors were implanted approximately 1 cm, was 5.4 L. The outside of the chambers was colored white to minimize solar heating. Gas samples were withdrawn from the headspace via a 16-gauge stainless steel luer-lock needle that extended 2.5 cm into the headspace.

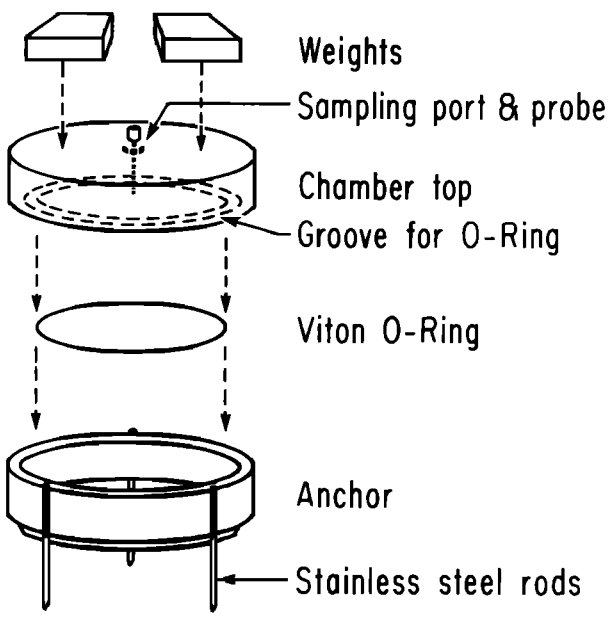

Fig. 1. Diagram of chamber used for measurement of N_2O fluxes.

Fluxes were measured six times (every four hours) in a 24-hour period and were timed to provide sampling at approximate minimum and maximum soil temperatures. Incubations lasted 30 min, with headspace samples collected at the beginning of each incubation and at 10-min intervals thereafter. Twenty mL air samples were withdrawn with 20-mL nylon syringes (SESI syringes, VWR Scientific Company) modified with Viton O-rings and equipped with one-way valves (Pharmaseal, Incorporated). Syringes were flushed with chamber air three times to mix the chamber atmosphere. Triplicate samples were withdrawn from at least one chamber during most samplings throughout the year to assess combined field handling and analytical variability. Chamber tops were removed from anchors upon completion of each incubation.

Laboratory tests showed that the chambers were inert to N_2O. Chambers filled with ambient air and with elevated N_2O concentrations (1250 ppb) showed no significant changes in concentration after a 30-min incubation, indicating no desorption or sorption of N_2O by the chamber walls. Additionally, chambers showed no "memory effect" due to exposure to high concentrations. Chambers were sealed and filled with 999-ppb N_2O for 30 min and then opened to ambient air for two hours. When resealed with ambient air, N_2O concentrations within chambers remained unchanged during a 30-min incubation.

Syringes were also inert to N_2O and stored samples satisfactorily. Syringes that were filled for two days with 999-ppb N_2O, opened and exposed to ambient air for two days, and then filled for two days with a 299.9-ppb standard, showed no contamination from the high N_2O concentrations. Storage tests showed that syringes filled with a subambient 299.9-ppb N_2O standard remained unchanged for 9 days. Syringes filled with an above-ambient 999-ppb N_2O standard showed a 3% concentration decrease after 4 days and a 5.6% decrease after 9 days. Gas samples were generally analyzed within 4 days of collection.

Nitrous oxide was measured using electron capture gas chromatography [Conrad and Seiler, 1980; Melillo et al., 1983; Bowden and Bormann, 1986]. Nitrous oxide was analyzed on a Shimadzu Mini-2 gas chromatograph (GC) equipped with a 3 m x 3.2 mm diameter stainless steel column filled with Poropak Q (80/100 mesh), and a ^{63}Ni electron capture detector. Carrier gas (95% Ar, 5% CH_4) was maintained at 20 mL/min. Injection and detector temperatures were 275°C and column temperature was 50°C.

Samples and standards were transferred into the GC via a stainless steel sampling valve (Valco Instruments Incorporated) equipped with a

1-mL sampling loop. A 20 mm long x 4 mm ID Si gel water trap (0.5 g, 80/100 mesh) attached to the sampling valve was used to reduce water vapor carried into the GC, thus maintaining clear separation of chromatographic peaks. There was no difference between samples and standards analyzed with or without the water trap. The trap was replaced after each set of analyses (approximately 600 samples), well before the moisture-reducing capacity of the Si gel had been reached. Inefficiency of the water trap could be detected in the size of the water peak. Certified N_2O standards (299.7, 500, and 999-ppb N_2O in N_2) were obtained from Scott Specialty Gases, Plumsteadville, Pennsylvania. Standard curves were linear over the range used, with r^2 values generally in excess of 0.992. During sample analyses, all standards were run approximately every 4 hours, or after approximately each 60 samples. Standard curves showed little variation throughout each day of analysis.

Nitrous oxide concentrations were corrected to STP conditions. Fluxes were calculated using the Statistical Package for the Social Sciences (SPSS) to determine regressions for the linear portion of the N_2O concentration increase within the chamber, which in most cases was for the entire incubation (four samples over the 30-min incubation). Samples collected at 2.5-min intervals during the first 10 min of several incubations throughout the year indicated that changes in N_2O concentrations during the first part of the incubation were not curvilinear. Initial concentrations within the chambers at the beginning of each incubation averaged 305 ± 1 (S.E., n=462) ppb over the entire year.

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. Precision of triplicate samples taken from the same chamber at the same time ranged from 0.5% to 6%, depending on the magnitude of the change in concentration.

Ambient air, chamber air, and soil temperatures (0-2.5 cm and 2.5-5.0 cm) were measured during each sampling. Ambient air temperature was measured at 1 m above the forest floor at one location in each stand, and soil and chamber headspace temperatures were measured at one chamber in each stand. Three to nine forest floor samples were analyzed for moisture, NO_3-N pools, and NH_4-N pools during each sampling. Samples were 5.5 cm in diameter and extended to the mineral soil. Soil was analyzed gravimetrically for moisture content, and by extraction with 2 N KCl for NO_3-N and NH_4-N [Melillo et al., 1983]. Analyses were conducted within 48 hours after return from the field. Net mineralization and net nitrification rates for the forest floor were measured using the buried bag technique [Melillo, 1977; Aber et al., 1983]. Buried bag incubations lasted 4-6 weeks from May through October and seven months during the overwinter period.

Flux measurements began on May 16, 1988 and were conducted twice per month during spring and early fall and once per month during the remainder of the year until December 5, 1989 when soils began to freeze. Sampling was suspended during winter (January - March) and resumed on April 17, 1989 when soils began to thaw.

Results

Mean flux rates for the pine plantation ranged from an uptake of 1.24 ± 0.85 (S.E., n=3) μg $N/m^2/h$ to an efflux of 2.36 ± 1.03 (S.E., n=3) μg $N/m^2/h$ (Figure 2). In the hardwood stand, mean fluxes ranged from an uptake of 1.92 ± 1.47 (S.E., n=3) μg $N/m^2/h$ to an efflux of 2.51 ± 1.64 (S.E., n=3) μg $N/m^2/h$ (Figure 2).

The net annual N_2O flux for each stand 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, weighted for sampling frequency, was the sum of all sampling periods. Fluxes were considered to have ended in mid-December (December 14, 1988) when soils were snow-covered and soil temperatures were at or near freezing, and to have resumed during the spring thaw (April 3, 1989) when the upper 5 cm of soil began to thaw. At that time, mean soil temperature at the 0-2.5 cm depth was 2.9°C in both stands (Figure 2). 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. Fluxes from December 15, 1988 to April 3, 1989 were considered to be zero, thus providing an annual flux period of 256 days. On an annual basis, the pine stand had a net N_2O-N emission of 0.010 ± 0.015 kg N/ha/yr, or a mean flux rate of 0.17 ± 0.25 μg $N/m^2/h$. The hardwood stand had a net emission of 0.017 ± 0.017 kg N/ha/yr, or a mean flux of 0.27 ± 0.28 μg $N/m^2/h$.

Fluxes were low throughout the year, and strong temporal patterns of N_2O fluxes were not observed. Fluxes showed slight seasonal trends in both stands (Figure 2) with emissions greatest in early July, uptake occurring during late summer and early fall months, and pulses of emissions evident again in the fall. Spring fluxes were different in the two stands. Emissions were observed in the pine stand whereas uptake occurred in the hardwood stand. Predictable diel patterns were not evident and the time of maximal daily uptake or emission was not consistent. On nearly all sampling dates, periods of both N_2O uptake and efflux were observed.

Environmental factors and soil processes considered to be important in regulating N_2O fluxes were examined. The factors and processes included were air and soil temperature, soil moisture, soil NO_3-N and NH_4-N pools, net nitrogen mineralization, and net nitrification. Environmental factors showed little relationship to the observed fluxes. The times of maximal N_2O uptake or efflux did not correspond to maximal or minimal air or soil temperatures. Maximum temperatures usually occurred in early afternoon with minima generally preceding sunrise, but as shown in the selected daily measurements (Figure 3) maximal flux rates and temperatures were not always synchronous. Annually, N_2O fluxes did not show strong relationships to air or soil temperatures or to soil moisture (Figure 2). Mean daily air and soil temperatures were greatest during July, and that was the time when the highest N_2O emission

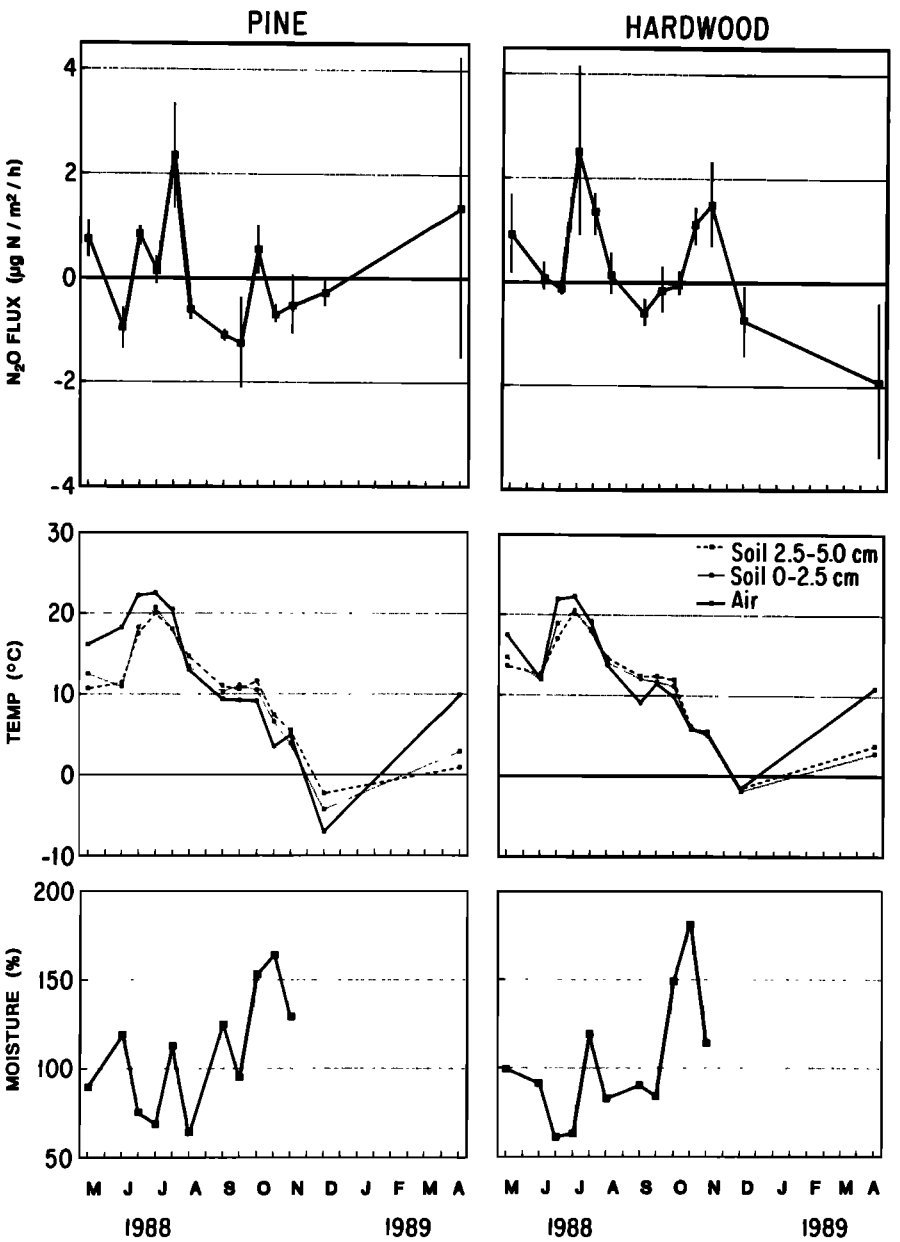

Fig. 2. Seasonal N_2O fluxes, air and soil temperatures, and soil moisture for pine plantation and hardwood stand at Harvard Forest. Soil moisture values are means (n=3-9). Flux rates for each date are means with S.E. bars of the three chambers in each stand. The rate for each chamber was calculated as the mean of the six diel measurements that were taken every four hours. Positive flux values indicate emissions from the soil. Negative values represent uptake.

rates were observed in each stand. However, when all measurements throughout the year were examined, there was no relationship between temperature and N_2O fluxes (pine: r^2=0.004, n=71; hardwood: r^2=0.006, n=72). Similarly, there was no relationship between soil moisture and N_2O flux (pine: r^2=0.02, n=33; hardwood: r^2=0.003, n=33). Soil moisture was highest in the spring and fall, but flux values did not correspond closely to this trend.

Soil N pools showed little relationship to annual variations in N_2O fluxes. Marked seasonal patterns in N pools were not evident in either stand (Figure 4) and soil N pools showed no correlations to N_2O fluxes. Soil NO_3-N concentrations in the hardwood stand were below detection limits (0.30 mg N/kg dry soil) at most times throughout the year. Dynamic soil processes also showed no correlation to N_2O fluxes. Net nitrification rates throughout the year were very low in each stand (Table 1), with slight activity in the spring in the pine plantation, and rates near zero throughout the year in the hardwood stand. Net mineralization showed neither a strong seasonal pattern, nor a close relationship to N_2O fluxes.

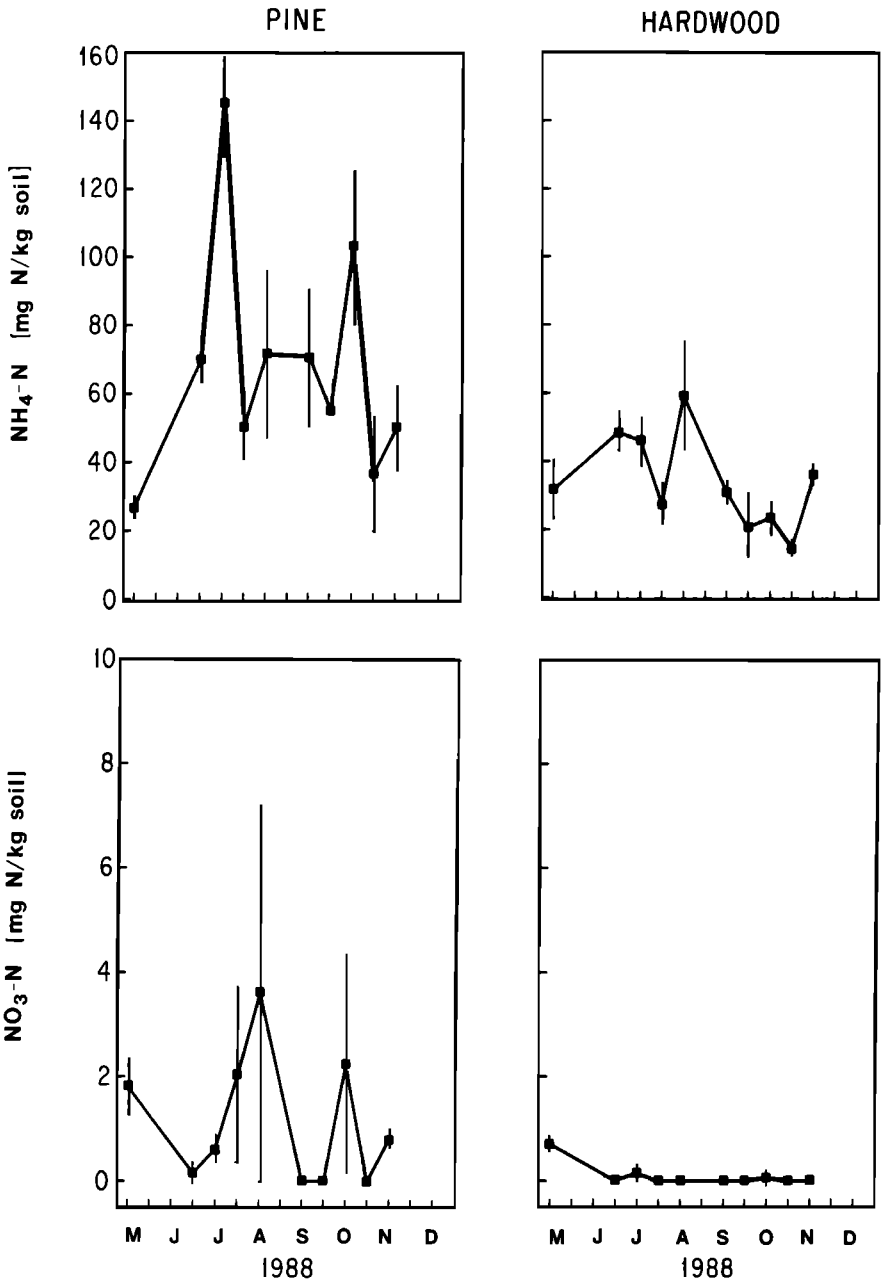

Fig. 3. Seasonal (1988) concentrations of soil NO_3-N and NH_4-N in pine plantation and hardwood stand at Harvard Forest. Soil is expressed on a dry weight basis.

Discussion

The mean annual rates of N_2O emissions, 0.17 μg N/m^2/h from the pine stand and 0.27 μg N/m^2/h from the hardwood stand, are the lowest reported rates of N_2O efflux from temperate forest soils. The rate in the hardwood stand is only one-sixth the rate (1.7 μg N/m^2/h) reported by Keller et al. [1983] for a northern hardwood forest at the Hubbard Brook Experimental Forest in New Hampshire, and much less than rates reported from other hardwood forests. Schmidt et al. [1988] reported annual effluxes of 3-11 μg N/m^2/h for deciduous forests in West Germany, and Goodroad and Keeney [1984a] reported efflux rates from a deciduous forest and a pine plantation in Wisconsin of 5.4 μg N/m^2/h and 27.7 μg N/m^2/h, respectively. Still higher rates of potential N_2O emissions have been reported from temperate forest soils where soil core incubation techniques have been employed [Melillo et al., 1983; Robertson and Tiedje, 1984]. However, because of the limitations of soil incubation techniques, the amount of the potential N_2O actually released to the atmosphere is unknown [Melillo et al., 1983].

The low rates of N_2O efflux from the stands at Harvard Forest are probably due to low rates of net nitrification in these soils. Nitrous oxide can be produced during the processes of

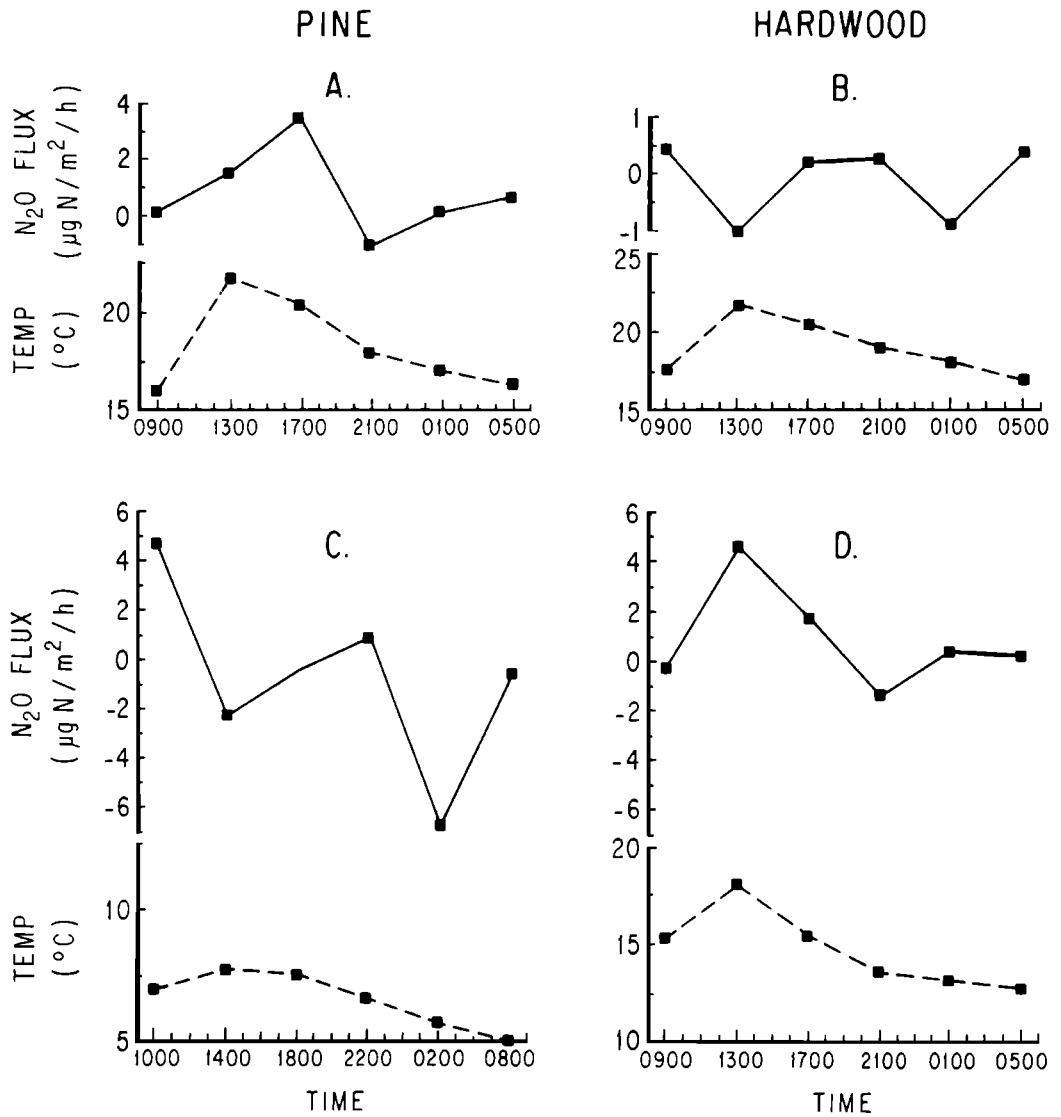

Fig. 4. Daily N_2O flux rates and temperatures for selected dates in the pine plantation and hardwood stand at Harvard Forest. (a) and (b) June 20, 1988; (c) October 24, 1988; (d) May 16, 1988. Note differences in scales.

nitrification [Bremner and Blackmer, 1978, 1979; Freney et al., 1978, 1979; Goodroad and Keeney, 1984a,b, 1985; Robertson and Tiedje, 1987] and denitrification [Wijler and Delwiche, 1954; Crutzen, 1981; Payne, 1981a,b]. In addition, other studies [Bleakley and Tiedje, 1982; Robertson and Tiedje, 1987] have shown that it can also be produced by other NO_3-N-utilizing organisms, possibly including fungi, yeasts, and non-denitrifying bacteria. The mineralization data showed that although NH_4-N is being produced by the Harvard Forest soils, net NO_3-N production is extremely low (Table 1). Thus, potential N_2O losses via the process of nitrification are low, and the amount of NO_3-N potentially available for loss via denitrifying bacteria or other organisms is also low. Although NO_3-N does enter the system from bulk precipitation inputs, plant uptake and microbial immobilization apparently sequester sufficient nitrogen to keep soil NO_3-N levels low.

Net nitrification has been shown to be strongly correlated with N_2O production and loss in other studies. Melillo et al. [1983] found a strong relationship between N_2O production and available NO_3-N in northern hardwood forest soils in New Hampshire. Matson and Vitousek [1987] found a strong relationship between N_2O effluxes and net nitrification potential across a range of tropical forest sites in Costa Rica, Brazil, and Hawaii. At our site, annual rates of net nitrification in the forest floor are extremely low (0.11-1.5 kg N/ha/yr), as are the annual N_2O fluxes. In comparison, nitrification in the forest floor of the northern hardwood forest at the Hubbard Brook Experimental Forest in New Hampshire [Melillo, 1977] is one to two orders of magnitude greater (13 kg N/ha/yr), as are the rates of N_2O emission [Keller et al., 1983]. Extremely high rates of efflux (7 kg N/ha/yr) have also been measured in a beech stand in the Solling region of West Germany

TABLE 1. Net Mineralization and Nitrification Rates for Forest Floors

Incubation Period	Number of Days	Mineralization (mg N/m^2/day)	Nitrification (mg N/m^2/day)
		Pine Plantation	
May 16 - June 20	35	21.1 (3.7)	1.4 (0.3)
June 20 - July 26	36	14.4 (4.2)	1.1 (0.3)
July 26 - Aug. 30	35	28.0 (4.6)	0.9 (0.3)
Aug. 30 - Oct. 17	48	21.3 (2.1)	0.6 (0)
Oct. 17 - May 16	211	2.7	B.D.* (-)
Yearly mean		10.5	0.4
		Hardwood Stand	
May 16 - June 20	35	11.7 (2.3)	B.D. (-)
June 20 - July 26	36	23.3 (3.9)	B.D. (-)
July 26 - Aug. 30	35	25.1 (3.7)	B.D. (-)
Aug. 30 - Oct. 17	48	15.0 (1.7)	0.2 (0)
Oct. 17 - Apr. 17	180	2.7	B.D. (-)
Yearly mean		10.0	0.03

Rates are means with standard errors (n=3) in parentheses.
* Below detection: NO_3, 0.91 mg N/m^2/incubation period; NH_4, 1.36 mg N/m^2/incubation period.

where net nitrification rates are about 50 kg N/ha/yr [Brumme et al., 1989].

Soil moisture and temperature generally exert strong controls over rates of N_2O flux [Bremner, 1978; Rolston, 1981; Knowles, 1981; Payne, 1981a,b; Firestone, 1982; Freney et al., 1978; Sahrawat and Keeney, 1986], but because N_2O emissions at Harvard Forest appear to be limited by low rates of net nitrification, relationships with moisture or temperature were not found. Intense rainfall following dry periods did not produce dramatic increases in N_2O effluxes, in contrast to other studies [Binstock, 1984; Sexstone et al., 1985].

The seasonal trends of N_2O fluxes in the hardwood forest stands are comparable to other temperate forest ecosystems in that maximum efflux rates were observed in spring or early summer, with increases occurring again in the fall [Schmidt et al., 1988; Duxbury et al., 1982; Goodroad and Keeney, 1984a]. They differ, however, in that long periods of net N_2O uptake from the atmosphere by the soils were observed in both stands. Short periods of nocturnal uptake by grass-covered soil [Slemr et al., 1984] and slightly longer periods (days to weeks) by agricultural soils [Bremner et al., 1980] have been measured, but reports of extended uptake by forest soils have not been reported previously.

We have used our pine forest flux rates (0.17 μg N/m^2/h) to estimate global temperate coniferous N_2O fluxes, and our hardwood rates (0.27 μg N/m^2/h) to estimate global temperate deciduous forest fluxes. Areal values were 4.88×10^6 km^2 for temperate coniferous forests and 4.41×10^6 km^2 for temperate deciduous forests [Steudler et al., 1989]. We assumed that fluxes within all temperate forests occurred for the same number of days as in the Harvard Forest (256 days). Based on these assumptions, we estimate that the annual global N_2O emission from temperate forests is 0.012 Tg N/yr. The estimated N_2O source from the remaining nonagricultural terrestrial ecosystems, including tropical forests, ranges from 3.8 to 14.9 Tg N/yr [Bowden, 1986; Schmidt et al., 1988]. Based on these estimates, temperate forest soils contribute less than 1% of the terrestrial N_2O source.

The wide range of flux rates reported for temperate forests has resulted in a wide range of estimates for the contribution of the temperate forest ecozone to global N_2O emissions. Our estimate is among the lowest estimates of global N_2O emissions from temperate forests. For example, Bowden [1986] estimated a contribution ranging from trace amounts to 2.5 Tg N/yr, and Schmidt et al. [1988] estimated that global N_2O emissions from temperate forests are 0.7-1.5 Tg N/yr. The breadth of flux estimates from temperate forest ecozones leads to uncertainty in the importance of this ecozone in contributing to atmospheric N_2O. It is probable that the actual global emission rate lies between our estimate and the upper values reported elsewhere. Linkages between N_2O effluxes and nitrogen cycling processes (e.g., nitrification as suggested in this study) imply that better prediction of flux rates in temperate forests will result from identification of processes important in controlling flux rates. As suggested by Matson and Vitousek [1987] for tropical forests, increased accuracy of global estimates for the temperate forest ecozone will then result from more detailed stratification and application of the most applicable flux estimates.

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