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## Exchange of N<sub>2</sub>O and CH<sub>4</sub> between the atmosphere and soils in spruce-fir forests in the northeastern United States

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**Abstract.** We measured the exchange of N<sub>2</sub>O and CH<sub>4</sub> between the atmosphere and soils in 5 spruce-fir stands located along a transect from New York to Maine. Nitrous oxide emissions averaged over the 1990 growing season (May–September) ranged from 2.1 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr in New York to 0.4 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr in Maine. The westernmost sites, Whiteface Mtn., New York and Mt. Mansfield, Vermont, had the highest nitrogen-deposition, net nitrification and N<sub>2</sub>O emissions. Soils at all sites were net sinks for atmospheric CH<sub>4</sub>. Methane uptake averaged over the 1990 growing season ranged from 0.02 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr in Maine to 0.05 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr in Vermont. Regional differences in CH<sub>4</sub> uptake could not be explained by differences in nitrogen-deposition, soil nitrogen dynamics, soil moisture or soil temperature. We estimate that soils in spruce-fir forests at our study sites released ca. 0.02 to 0.08 kg N<sub>2</sub>O-N/ha and consumed ca. 0.74 to 1.85 kg CH<sub>4</sub>-C/ha in the 1990 growing season.

### Introduction

Nitrous oxide and CH<sub>4</sub> are important in atmospheric chemistry and in the radiation budget of the Earth. Nitrous oxide contributes to the destruction of stratospheric O<sub>3</sub>, and CH<sub>4</sub> affects the oxidation capacity of the troposphere and water budget of the stratosphere (Warneck 1988; Houghton et al. 1990). In addition, N<sub>2</sub>O and CH<sub>4</sub> are greenhouse gases whose atmospheric concentrations are increasing at annual rates of 0.25% and 0.9%, respectively (Houghton et al. 1990). Microbial processes and anthropogenic activities are the major sources of these gases. Microbial processes are also important sinks. We have a relatively good understanding of the emissions from anthropogenic sources, but biotic sources and sinks

are less well known. Consequently, we need to understand the factors that control biotic sources and sinks.

Empirical evidence suggests that the exchange of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  between the atmosphere and soils is affected by soil nitrogen cycling. Melillo et al. (1983) found a strong relationship between  $\text{N}_2\text{O}$  produced by denitrification and  $\text{NO}_3^-$  in soils of hardwood forests in New Hampshire. Matson & Vitousek (1990) reported that net nitrogen mineralization in tropical forest soils were highly correlated with  $\text{N}_2\text{O}$  emissions. In many ecosystems, nitrogen fertilization has been shown to increase soil nitrogen cycling (nitrification, mineralization, and denitrification) and to increase the emissions of  $\text{N}_2\text{O}$  from soils into the atmosphere (Hutchinson & Mosier 1979; McKenney et al. 1980; Seiler & Conrad 1981; Duxbury & McConnaughey 1986; Keller et al. 1988). Nitrogen fertilization also lowers the uptake of atmospheric  $\text{CH}_4$  by soils in temperate forests, grasslands and tropical forests (Mosier et al. 1991; Keller et al. 1990; Steudler et al. 1989; Melillo et al. 1989). The linkage between nitrogen fertilization and  $\text{CH}_4$  uptake may be caused by nitrogen-induced inhibition of microorganisms that oxidize  $\text{CH}_4$  (Jones & Morita 1983; Steudler et al. 1989) or by alterations in soil nitrogen cycling (Mosier et al. 1991). These results suggest that soils in ecosystems with high nitrogen-deposition are likely to have higher soil nitrogen cycling, higher  $\text{N}_2\text{O}$  emissions and lower  $\text{CH}_4$  uptake compared to soils in ecosystems with lower nitrogen-deposition.

In the northeastern US (New York and New England), nitrogen-deposition increases with elevation and from east to west. Forests dominated by red spruce (*Picea rubens*) and balsam fir (*Abies balsamea*) occupy high elevations (> 750-m) in the Adirondack, Green and White Mountains, and receive the highest nitrogen-deposition in the region. Lovett & Kinsman (1990) report that upper elevation forests in the Adirondack and White Mountains receive 3 to 7 times more nitrogen-deposition (1.3 to 2.7 kg N/ha-month) than adjacent low elevation sites (0.4 to 0.5 kg N/ha-month). Regional gradients in nitrogen-deposition also occur across this region. Spruce-fir forests on Whiteface Mtn., New York receive 3 times more nitrogen-deposition (16 kg N/ha-yr, Friedland et al. 1991; Ollinger et al. 1992) than spruce-fir forests in Acadia National Park, Maine (4–6 kg N/ha-yr, McNulty et al. 1991; Ollinger et al. 1992). Since nitrogen deposition affects soil nitrogen cycling and the soil fluxes of  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , there may be large regional variations in the fluxes of these gases in forests of the northeastern US. High elevation spruce-fir forests may make a larger contribution to regional trace gas budgets of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  than low elevation forests in this region.

The objectives of this study were to: (1) measure the exchange of  $\text{N}_2\text{O}$

and CH<sub>4</sub> between the atmosphere and soils in spruce-fir forests in New York and New England; and (2) to examine the environmental factors and processes controlling the fluxes.

## Methods

### *Study sites*

We measured trace gas fluxes and soil nitrogen dynamics in 5 spruce-fir stands in the northeastern US in 1990. These stands were located at: Whiteface Mtn., New York; Mt. Mansfield, Vermont; Mt. Ascutney, Vermont; Mt. Washington, New Hampshire; and Acadia, Maine (Fig. 1). Except for Acadia, all sites had thin (<10-cm) organic horizons underlain by thin (<1-m) soils of the spodosol group (Table 1). The Acadia site had thick organic (20-cm) and mineral horizons (>1-m).



<u>Site</u>	<u>Location</u>
1	Whiteface Mtn., NY
2	Mt. Mansfield, VT
3	Mt. Ascutney, VT
4	Mt. Washington, NH
5	Acadia, ME

Fig. 1. Location of sampling sites.

Table 1. Site descriptions.

Site	Elevation (m)	Vegetation (%) (spruce/fir/others)	Organic horizon			N deposition**			
			Thickness (cm)	Mass* (kg/ha)	pH	Cloud (kg N/ha-yr)	Wet	Dry	Total
Whiteface Mt, NY 44.4°, 73.9°	1000	40/60/0	3-9	$7.2 \times 10^4$	2.90	5.2	9.3	1.5	16
Mt Mansfield, VT 44.5°, 72.8°	985	45/45/10	2-7	$6.4 \times 10^4$	2.85	5.7	8.6	1.7	16
Mt Ascutney, VT 43.3°, 72.4°	762	83/12/5	8-10	$8.4 \times 10^4$	2.80	0.2	7.5	2.3	10
Mt Washington, NH 44.3°, 71.2°	1000	30/70/0	2-10	$7.4 \times 10^4$	2.71	5.5	7.8	1.7	15
Acadia, ME 44.3°, 68.4°	13	99/0/1	7-20	$1.7 \times 10^5$	2.55	0	3.8	1.7	5.5

\* These values are the averages of 60 samples collected at each site.

\*\* Calculated with the model presented in Ollinger et al. 1992.

### *Gas sampling*

We measured  $\text{N}_2\text{O}$  and  $\text{CH}_4$  fluxes at the surface of the forest floor by measuring changes in gas concentrations inside air-tight static chambers over a 30-minute period. Details of our sampling technique can be found in Steudler et al. (1989) and Bowden et al. (1990). Briefly, our polyvinyl chloride chambers (29-cm diameter) consisted of a 4.0-cm tall top with a luer lock sampling port and a 5.2-cm tall anchor. The sharpened portion of the anchor was inserted 1-cm into the soil at least 4 hours before each gas sampling. On each sampling date at each site, 6 chambers were randomly located in each spruce-fir stand. Samples were collected every 10 minutes for 30 minutes in 20-ml nylon syringes and stored (less than 2-weeks) until analysis at The Ecosystems Center of the Marine Biological Laboratory (Woods Hole, MA). We measured gas fluxes 4 times (0600–0630, 1400–1430, 1800–1830 and 2200–2230) in a 24-hr period to determine if there were diel variations. Since we did not find any consistent diel variations, fluxes measured at all 6 chambers on each sampling date were averaged to estimate the daily flux.

### *Lab analysis*

We used gas chromatography to measure  $\text{N}_2\text{O}$  and  $\text{CH}_4$  concentrations of samples collected inside our static chambers. We injected 20-ml gas samples into a 14-port pneumatic valve which directed 1-ml subsamples into 2 different Shimadzu 8A gas chromatographs. Nitrous oxide was quantified with a  $^{63}\text{Ni}$  electron capture detector at 250 °C. P-5 mixture (5%  $\text{CH}_4$  in Ar) was the carrier for a Porapak Q column maintained at 45 °C. Methane was quantified with a flame ionization detector at 130 °C. Ultra-high-purity nitrogen was the carrier for the Hayesep Q column maintained at 100 °C. Peak areas were recorded with a dual channel Shimadzu C-R5A integrator. Standards that bracketed the sample gas concentrations (Scott Specialty Gases, Inc.) were analyzed before and after every 50 samples. We used a linear interpolation procedure (based on analytical times) to calculate the gas concentrations.

### *Additional measurements*

#### *Temperatures*

We measured temperatures of ambient air (1-m above the forest floor) and the organic horizon at 2-cm and 5-cm depths with Omega dial thermometers. Measurements were made at only one chamber during all 30-minute sampling periods.

*Initial nitrogen pools, turnover rates and moisture contents*

We made monthly measurements of the  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  concentrations, net nitrogen mineralization and net nitrification in the organic horizons of each site from May–August 1990. After the 24-hr gas sampling periods, three soil cores (10-cm length and 5.4-cm diameter) were taken near each of the 6 sampling chambers: one within each anchor and the other two within 0.5-m of the anchor. All cores were divided into organic and mineral horizons, but only the organic horizons were analyzed. Cores collected within the anchors were returned to the lab for analysis of the initial  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  concentrations. The organic horizons from the other two cores were placed in polyethylene bags (0.1-mm thick), returned to their original locations, and incubated in the field for approximately one month. These bags were gas permeable and thereby prevented anaerobic conditions from occurring during field incubation. After field incubation, the organic horizons were brought to the lab for analysis of the  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  concentrations.

In the lab, we removed roots and rocks from each sample. Ten gram subsamples were then extracted with 1N KCl for 48-hours (Aber et al. 1983). The extracts were filtered and frozen until wet chemical analysis for  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  (Technicon Methods 780-86T and 782-86T). Net mineralization was calculated as  $\text{NH}_4\text{-N}$  plus  $\text{NO}_3\text{-N}$  concentrations in the field incubated samples minus the initial values. Net nitrification was the difference between the incubated and initial  $\text{NO}_3\text{-N}$  concentrations. All rates were standardized to a 28-day period.

We measured the moisture content of the organic horizon gravimetrically. Ten gram subsamples were weighted, dried at 105 °C for 48-hours, and reweighed to determine the water content. Moisture is expressed as the ratio of water to dry soil multiplied by 100.

*Fertilization experiment*

In June 1988, McNulty & Aber (1992) established a fertilization experiment in a spruce-fir stand at Mt. Ascutney. Six times in the 1990 growing season, we measured  $\text{N}_2\text{O}$  and  $\text{CH}_4$  fluxes in one of their 15 × 15 m fertilized and control plots. The fertilized plot received 31.4 kg  $\text{NH}_4\text{Cl-N/ha-yr}$  in 1988, 1989 and 1990. The fertilizer was dissolved in 15 L of distilled water and applied to the forest floor with a backpack sprayer. The total annual amount of fertilizer was applied in 3 equal doses on June 15, July 15 and August 15.

*Data interpretation*

Correlations between gas fluxes and soil variables were determined by using Pearson's correlation matrices and forced entry multiple linear

regression analysis. Significant differences between the mean fluxes for each site were identified by analysis of variance and least significant difference tests (ANOVA-LSD). Individual comparisons of means were made using paired *t*-tests.

## Results

### *Trace gas fluxes*

#### *Nitrous oxide fluxes*

All of the soils studied functioned as sources and sinks of N<sub>2</sub>O at different times during the growing season (Fig. 2). In general, the highest N<sub>2</sub>O emissions were measured in May. Soils were typically sinks for N<sub>2</sub>O in July and/or August. Largest uptake rates (−6.7 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr) were measured at Mt. Ascutney in August.

Mean N<sub>2</sub>O fluxes at Whiteface Mtn. (2.11 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr) and Mt. Mansfield (1.95 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr), the sites with the highest nitrogen-deposition (Table 1), were significantly higher than the fluxes from all other sites (1-way ANOVA LSD *p* < 0.05). Mt. Ascutney (−1.12 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr) and Mt. Washington (−0.23 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr) were net sinks for N<sub>2</sub>O, and Acadia was a small net source (0.36 ug N<sub>2</sub>O-N/m<sup>2</sup>-hr). Using our mean N<sub>2</sub>O fluxes and assuming 154 frost-free days/yr, we estimate that N<sub>2</sub>O emissions from soils at Whiteface Mtn., Mt. Mansfield and Acadia released 0.02 to 0.08 kg N<sub>2</sub>O-N/ha during the 1990 growing season.

#### *Methane fluxes*

Soils at our sites were predominantly sinks for atmospheric CH<sub>4</sub>. Daily mean uptake rates ranged from 0.02 to 0.08 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr. Highest uptake rates were measured in either May or July (Fig. 2).

All soils released CH<sub>4</sub> into the atmosphere during approximately 10% of the total flux measurements. Emissions from these sites ranged from 0.005 to 0.096 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr.

The only significant difference in the net CH<sub>4</sub> flux averaged over the growing season was between Mt. Mansfield and all other sites (ANOVA-LSD *p* < 0.05). Methane uptake at Mt. Mansfield (0.053 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr) was 1.5 to 2.6 times higher than CH<sub>4</sub> uptake at the other sites (Whiteface Mtn., 0.035 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr; Mt. Ascutney, 0.033 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr; Mt. Washington, 0.035 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr and Acadia, 0.02 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr). Using our mean CH<sub>4</sub> uptake rates and assuming 154 frost-

free days/yr, we estimate that soils at our study sites consumed ca. 0.74 to 1.85 kg  $\text{CH}_4\text{-C}/\text{ha}$  during the 1990 growing season.

*Soil moisture, temperature, inorganic nitrogen, and nitrogen cycling*

Whiteface Mtn. and Mt. Mansfield had the highest soil moisture; mean moisture contents ranged from 220 to 317% (Fig. 2). Mt. Ascutney had the lowest soil moisture; means ranged from 81 to 220%. The times of lowest soil moisture at Mt. Ascutney coincided with the highest  $\text{CH}_4$  uptake rates.

Temperatures at all sites were usually lowest in May and highest in July (Fig. 2). Methane uptake was not correlated with temperature, but the highest  $\text{N}_2\text{O}$  emissions occurred at the lowest temperatures.

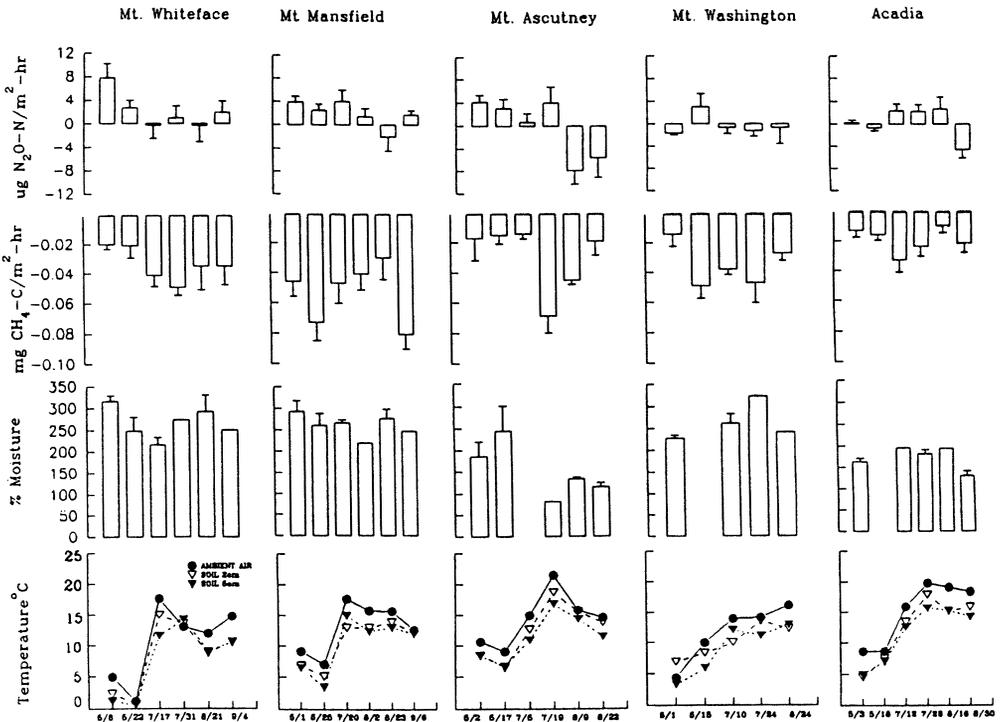


Fig. 2. Nitrous oxide and  $\text{CH}_4$  fluxes, soil moisture and temperature at our study sites.

Ammonium was the dominant form of inorganic nitrogen in the organic horizon at all sites (Fig. 3). In general,  $\text{NH}_4\text{-N}$  concentrations were lowest in May and highest in either July or August. Mt. Mansfield had approximately 2 times higher  $\text{NH}_4\text{-N}$  concentrations (33 to 57 mg  $\text{NH}_4\text{-N}/\text{kg}$  dry soil) than all other sites. Ammonium concentrations at Whiteface Mtn.,

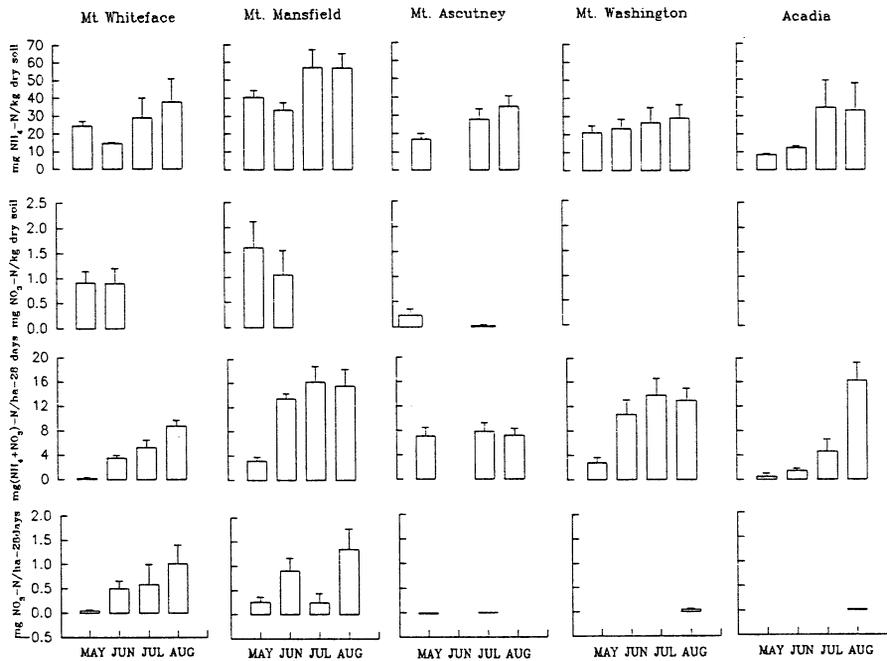


Fig. 3. Ammonium and  $\text{NO}_3$  concentrations, net nitrogen mineralization and net nitrification rates at our study sites.

Mt. Ascutney, Mt. Washington and Acadia ranged from 8 to 35 mg  $\text{NH}_4\text{-N}$ /kg dry soil.

Only 3 to 5 sites had detectable  $\text{NO}_3\text{-N}$  concentrations. Whiteface Mtn. and Mt. Mansfield had detectable  $\text{NO}_3\text{-N}$  concentrations (0.9 to 1.6 mg  $\text{NO}_3\text{-N}$ /kg dry soil) in May and June 1990. Mt. Ascutney had detectable  $\text{NO}_3\text{-N}$  concentrations (0.2 mg  $\text{NO}_3\text{-N}$ /kg dry soil) in May 1990.

Net nitrogen mineralization was lowest at all sites in May and highest in either July or August (Fig. 3). Mt. Mansfield and Mt. Washington had the highest net nitrogen mineralization rates. Monthly rates at these sites ranged from 3 to 16 kg N/ha-28 days, and the annual rate (sum of monthly rates) was 48 kg N/ha at Mt. Mansfield and 40 kg N/ha at Mt. Washington. Whiteface Mtn., Mt. Ascutney, and Acadia had the lowest net nitrogen mineralization rates; monthly rates usually ranged from 0.1 to 9 kg N/ha-28 days, and the annual rates ranged from 17 to 22 kg N/ha.

We measured detectable net nitrification during all samplings at Mt. Mansfield and Whiteface Mtn. (Fig. 3). Monthly rates ranged from 0.05 to 1.4 kg N/ha-28 days, and the annual rate was 2.8 kg N/ha-yr at Mt. Mansfield and 2.2 kg N/ha at Whiteface Mtn. The other sites had either very low (< 0.10 kg N/ha-28 days) or undetectable nitrification rates.

*Fertilization experiment*

In general, daily mean  $N_2O$  flux from the fertilized plot was higher than the daily mean  $N_2O$  flux from the control (Fig. 4). The 1990 growing season mean  $N_2O$  flux from the fertilized plot ( $3.11 \text{ ug } N_2O\text{-N}/m^2\text{-hr}$ ) was significantly higher than the mean from the control ( $-1.12 \text{ ug } N_2O\text{-N}/m^2\text{-hr}$ ;  $p < 0.05$ ).

In only 2 out of 6 gas samplings (7/19 and 8/9), daily mean  $CH_4$  uptake rates by the fertilized plot were significantly lower than the control.

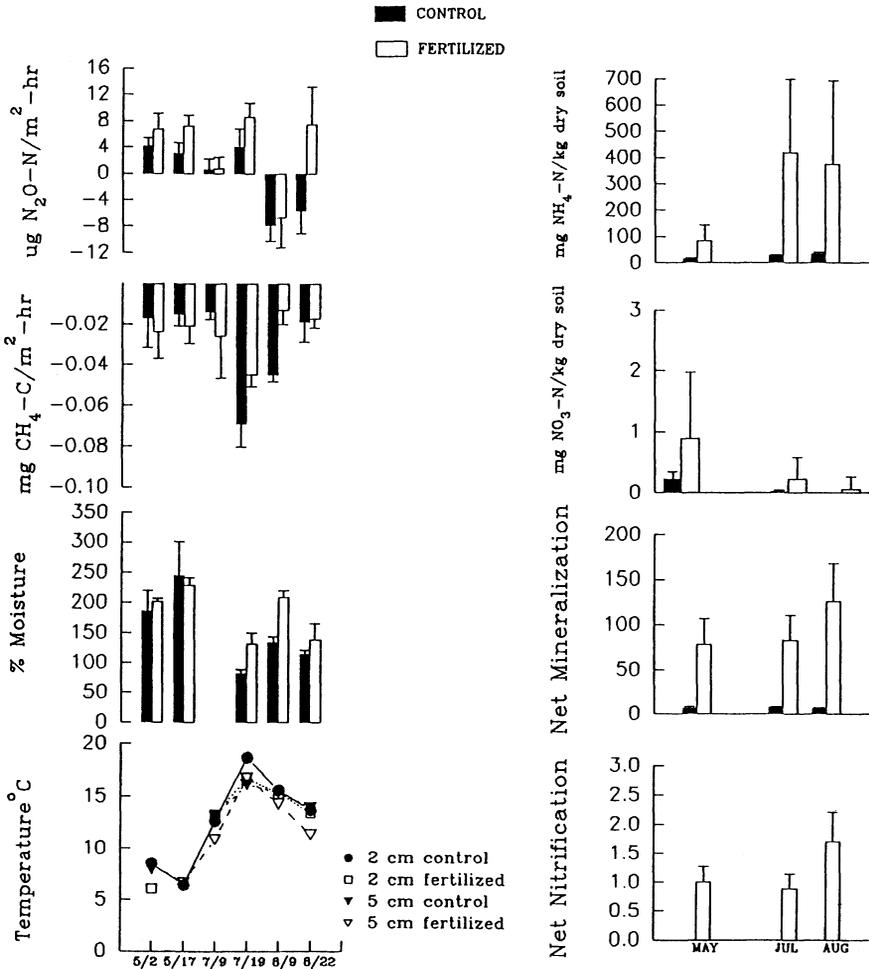


Fig. 4. Nitrous oxide and  $CH_4$  fluxes, soil nitrogen concentrations, net nitrogen mineralization ( $\text{mg } (NH_3 + NO_3)\text{-N}/ha\text{-28 days}$ ) and net nitrification ( $\text{mg } NO_3\text{-N}/ha\text{ 28-days}$ ) at Mt. Ascutney.

The 1990 growing season mean  $\text{CH}_4$  uptake rates by the fertilized ( $0.033 \text{ mg CH}_4\text{-C/m}^2\text{-hr}$ ) and control plots ( $0.020 \text{ mg CH}_4\text{-C/m}^2\text{-hr}$ ;  $p < 0.05$ ) were not significantly different ( $p < 0.05$ ).

The fertilized organic horizon had ca. 5 to 15 times higher  $\text{NH}_4\text{-N}$  concentrations and ca. 2 to 6 times higher net nitrogen mineralization than the control organic horizon (Fig. 4). Net nitrification rates ( $< 0.2 \text{ mg NO}_3\text{-N/ha-28-days}$ ) and  $\text{NO}_3\text{-N}$  concentrations ( $< 1 \text{ NO}_3\text{-N mg/kg dry soil}$ ) in the organic horizons from both the control and fertilized plots were not significantly different.

## Discussion

### *Nitrous oxide fluxes*

Nitrous oxide fluxes ( $-1.12$  to  $2.11 \text{ ug N}_2\text{O-N/m}^2\text{-hr}$ ) from the spruce-fir forests examined in this study were similar to those measured in a red pine plantation and hardwood forests in the northeastern US, which ranged from  $0.17$  to  $1.10 \text{ ug N}_2\text{O-N/m}^2\text{-hr}$  (Keller et al. 1983; Bowden et al. 1991). In addition, Steudler (personal communication) measured daily mean  $\text{N}_2\text{O}$  fluxes that ranged from  $0.04$  to  $0.29 \text{ ug N}_2\text{O-N/m}^2\text{-hr}$  from soils in a red spruce stand at Camels Hump, Vermont in June and September 1988. Nitrous oxide fluxes from our study sites were lower than  $\text{N}_2\text{O}$  fluxes measured in other temperate forests. Annual mean  $\text{N}_2\text{O}$  emissions from a white pine plantation ( $36.4 \text{ ug N}_2\text{O/m}^2\text{-hr}$ ) and a black oak forest ( $15.5 \text{ ug N}_2\text{O/m}^2\text{-hr}$ ) in Wisconsin and from many hardwood forests ( $3$  to  $11 \text{ ug N}_2\text{O-N/m}^2\text{-hr}$ ) in Germany were 1.3 to 17 times higher than the emissions from our sites (Goodroad & Keeney 1984; Schmidt et al. 1988).

The low  $\text{N}_2\text{O}$  emissions from our study sites are consistent with the low net nitrification rates. In nitrification,  $\text{N}_2\text{O}$  is a byproduct of the oxidation of  $\text{NH}_4$  to  $\text{NO}_3^-$ . Although  $\text{NH}_4$  was readily available in the organic horizon at all of our sites, the net nitrification rates were very low (Fig. 3;  $< 2.8 \text{ kg N/ha-yr}$ ) and may have resulted in low  $\text{N}_2\text{O}$  production and to low  $\text{NO}_3^-$  availability for denitrifiers to convert to  $\text{N}_2\text{O}$ . The nitrification rates at our sites are similar to the net nitrification rates measured in the organic horizon of many New England forests (Federer 1983; Bowden et al. 1991; McNulty et al. 1990). Low net nitrification rates in these forests may have been caused by many factors including pH, redox potential, moisture content, absence of nitrifying bacteria, alleopathic inhibition, and/or high rates of microbial assimilation of  $\text{NO}_3^-$ .

Soils at our sites were sometimes sinks for atmospheric  $\text{N}_2\text{O}$  (Fig. 2).

Keller et al. (1983) and Bowden et al. (1990) also reported that soils in hardwood forests of the northeastern US were sometimes sinks for  $N_2O$ , and Ryden (1981) reported that grasslands were sinks for  $N_2O$  when conditions were conducive to microbial reduction of  $N_2O$ . Uptake in grasslands occurred when  $NO_3-N$  available for reduction by denitrification was essentially exhausted ( $< 1$  mg  $NO_3-N/kg$  soil) and the soils had high moisture contents ( $> 20\%$  wet/wet), and high temperatures ( $> 5$  °C). Soils at our study sites had both high soil moistures and low  $NO_3-N$  concentrations and may have been suitable for the reduction of  $N_2O$  (Figs. 2 and 3). Thus, denitrification may have been responsible for the uptake of  $N_2O$  at our sites.

Seasonal trends in  $N_2O$  fluxes at the spruce-fir stands examined in this study were similar to those observed in other temperate forests (Goodroad and Keeney 1984; Schmidt et al. 1988; Bowden et al. 1990). In general, maximum  $N_2O$  emissions were usually observed in Spring and coincide with low temperature and high soil moisture. This spring pulse may have been caused by the release of  $N_2O$  trapped beneath frozen surface soils, rapid onset of microbial processes that produce  $N_2O$ , and/or the lack of competitive processes (e.g. plant nitrogen uptake).

There were small regional differences in the  $N_2O$  emissions, nitrification rates and the ratio of annual net nitrification to annual net mineralization (% nitrified). The westernmost sites, Whiteface Mtn., New York and Mt. Mansfield, Vermont, had the highest  $N_2O$  emissions (approximately 2  $\mu g$   $N_2O-N/m^2-hr$ ), net nitrification rates (2–3 kg  $NO_3-N/ha-yr$ ), and % nitrified (Whiteface Mtn. 13% and Mt. Mansfield 7%). All other sites had very low  $N_2O$  fluxes ( $-1.12$  to  $0.36$   $\mu g$   $N_2O-N/m^2-hr$ ), net nitrification ( $< 0.06$  kg  $N/ha-yr$ ), and less than 1% of the annual net nitrogen mineralization nitrified. The westernmost sites, Whiteface Mtn. and Mt. Mansfield, have received higher cumulative doses of nitrogen than the other sites in our transect, suggesting that chronic nitrogen-deposition may be starting to increase soil nitrogen cycling and  $N_2O$  emissions.

### *Methane*

Our  $CH_4$  uptake rates (0.02 to 0.08 mg  $CH_4-C/m^2-hr$ ) are within the range of those reported for most studies conducted in the northeastern US. Steudler (personal communication) measured daily mean  $CH_4$  uptake rates that ranged from 0.04 to 0.07 mg  $CH_4-C/m^2-hr$  at 6 sites (three chambers per site) in a red spruce stand at Camels Hump, Vermont in June and September 1988. Crill (1991) reported on annual mean uptake of 0.07 mg  $CH_4-C/m^2-hr$  for a mixed deciduous-coniferous forest in New Hampshire, and Keller et al. (1983) reported an annual mean uptake of

0.01 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr for a hardwood stand in New Hampshire. Our uptake rates, however, are 3 to 6 times lower than the uptake rates measured at a mixed oak stand (0.13 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr) and a red pine plantation (0.11 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr) in central Massachusetts (Steudler et al. 1989).

There were no strong seasonal variations in CH<sub>4</sub> uptake at our sites. This is consistent with results reported by other investigators. Keller et al. (1983), Born et al. (1990) and Crill (1991) found no seasonal trends in CH<sub>4</sub> uptake by temperate forest soils. Seasonal trends are difficult to detect when the fluxes are low. At sites with higher CH<sub>4</sub> uptake rates, pronounced seasonal patterns have been observed (Steudler et al. 1989).

At our sites, air and soil temperatures (2 and 5-cm depths) were not correlated with CH<sub>4</sub> uptake. Keller et al. (1983) and Steudler et al. (1989) also reported that CH<sub>4</sub> uptake by temperate forest soils was not correlated with soil temperature. Crill (1991), however, reported that CH<sub>4</sub> uptake by soils in a mixed deciduous-coniferous forest in New Hampshire was correlated with monthly averaged air temperatures ( $r^2 = 0.563$  to  $0.837$ ;  $p < 0.05$ ) and with the previous 24-hour mean 8-cm soil temperature ( $r^2 = 0.141$ ;  $p < 0.05$ ), suggesting that there may be a time lag between changing temperatures and net CH<sub>4</sub> flux. Unfortunately, we can not evaluate this time lag with our data because temperatures were measured only during our gas samplings.

There was no apparent correlation between CH<sub>4</sub> uptake and soil moisture over our sites. This is consistent with results reported by Crill (1991). At Mt. Ascutney, however, soil moisture dropped from 220% in May to 81% in July, which coincided with a factor of 7 increase (0.07 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr compared to 0.01 mg CH<sub>4</sub>-C/m<sup>2</sup>-hr) in CH<sub>4</sub> uptake. Steudler et al. (1989) reported that CH<sub>4</sub> uptake by soils at low soil moisture (68%) was significantly higher than uptake by the same soils at higher soil moisture (113%). Our results suggest that soils in spruce-fir forests may become more important sinks for atmospheric CH<sub>4</sub> under drier conditions.

Methane uptake by soils in several ecosystems is influenced by nitrogen fertilization and/or soil nitrogen cycling. Steudler et al. (1989) reported a significant reduction in CH<sub>4</sub> uptake by forest soils fertilized with 37 and 120 kg NH<sub>4</sub>NO<sub>3</sub>-N/ha-yr. Mosier et al. (1991) reported reductions in CH<sub>4</sub> uptake by fertilized grasslands that received 450 kg Urea-N/ha-yr and 22 kg NH<sub>4</sub>NO<sub>3</sub>-N/ha-yr. In addition, the data of Keller et al. (1990) suggest that the high inorganic nitrogen concentrations in tropical agricultural soils were responsible for the lower CH<sub>4</sub> uptake by these soils compared to undisturbed tropical forest soils. At our study sites, however, CH<sub>4</sub> uptake was not affected by nitrogen-deposition and/or nitrogen

cycling. Excluding Mt. Mansfield,  $\text{CH}_4$  uptake by soils along our transect was not significantly different. Indeed, Mt. Mansfield had the highest net nitrification and nitrogen-deposition, yet it also had the highest  $\text{CH}_4$  uptake. High soil moistures at our sites could have prevented the nitrogen-induced reduction of  $\text{CH}_4$  uptake by limiting gas transport to the zones of  $\text{CH}_4$  oxidation. Our results are consistent with those of Whalen et al. (1991) who reported that nitrogen fertilization (1:1 mixture of urea and  $(\text{NH}_4)_2\text{SO}_4$  at 50 and 200 kg N/ha-yr) did not affect  $\text{CH}_4$  uptake by soils in upland taiga forests.

Measurements from individual chambers suggest that soils in spruce-fir forests are sometimes sources for  $\text{CH}_4$ . This result is consistent with measurements made in high elevation spruce-fir forests in West Virginia (Yavitt et al. 1990). Methane is produced by anaerobic microbial processes in the organic horizon of spruce-fir forests (Sextone & Mains 1990). High moisture conditions at our sites are likely to have created anaerobic microsites suitable for the production of  $\text{CH}_4$ . The production of  $\text{CH}_4$ , however, does not guarantee its release into the atmosphere. The direction and magnitude of the flux measured with static chambers reflects the balance between production and consumption. Since our sites were usually sinks for  $\text{CH}_4$ , consumption is the dominant process.

#### *Nitrogen fertilization experiment*

The purpose of this experiment was to determine if high rates of  $\text{NH}_4\text{-N}$  deposition would alter soil nitrogen cycling and soil gas fluxes of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  in a high elevation spruce-fir forest. In 1988, 1989 and 1990, approximately 3 times (31.4 kg  $\text{NH}_4\text{-N}$ /ha-yr) the total annual nitrogen deposition was added to the forest floor at Mt. Ascutney. We expected this fertilization treatment to: (1) increase net nitrogen mineralization and net nitrification, (2) increase the concentrations of  $\text{NH}_4$  and  $\text{NO}_3$  in the organic horizon, (3) increase  $\text{N}_2\text{O}$  emissions, and (4) lower the uptake of atmospheric  $\text{CH}_4$ .

The  $\text{NH}_4\text{-N}$  fertilization increased both the net nitrogen mineralization rate and  $\text{NH}_4\text{-N}$  concentrations, but had no effect on the net nitrification rate and  $\text{NO}_3\text{-N}$  concentrations. Higher  $\text{NH}_4\text{-N}$  concentrations (ca. 5 to 15 times) were caused by both direct addition of  $\text{NH}_4\text{-N}$  and through increased (ca. 2 to 6 times) net nitrogen mineralization. Other studies have also reported increases in net nitrogen mineralization following nitrogen fertilization (Adams & Atwill 1984; Bowden et al. 1991; McNulty & Aber 1992). Net nitrification in the control and fertilized organic horizons was less than 0.08 kg N/ha-28 days and the  $\text{NO}_3\text{-N}$  concentrations ranged from below the detection limit to 0.9 mg  $\text{NO}_3\text{-N}$ /kg dry soil.

Fertilized soil had higher  $\text{N}_2\text{O}$  emissions than the control soil, but the annual mean  $\text{N}_2\text{O}$  flux from the fertilized soils was only a small percentage of the applied fertilizer. Using 154 frost-free days/yr and our growing season mean  $\text{N}_2\text{O}$  flux for the fertilized soils ( $3.11 \text{ ug N}_2\text{O-N/m}^2\text{-hr}$ ), we estimate an annual  $\text{N}_2\text{O}$  flux of  $0.11 \text{ kg N/ha-yr}$  which is 0.35% of the total  $\text{NH}_4\text{-N}$  applied in 1990. This annual  $\text{N}_2\text{O}$  flux is about 10 times higher than the annual  $\text{N}_2\text{O}$  flux measured at a low elevation mixed hardwood and red pine stand in Massachusetts (Bowden et al. 1990) and ca. 180 times lower than the annual  $\text{N}_2\text{O}$  flux ( $20 \text{ kg N}_2\text{O-N/ha-yr}$ ) from a mixed oak-beech forest in the Netherlands that received 24–41  $\text{kg NH}_4\text{-N/ha-yr}$  from atmospheric deposition in 1979–1987 (Tietema et al. 1991; Tietema & Verstraten 1991). Unlike our study site, this mixed oak-beech forest had very high net nitrification rates (22–42  $\text{kg N/ha-yr}$ ) and soil  $\text{NO}_3$  concentrations because of long-term chronic nitrogen additions from the atmosphere.

The fertilized and control soils did not have consistently different  $\text{CH}_4$  uptake rates. In 4 of the 6 gas samplings (May 2 & 17, July 5 and August 22), daily mean  $\text{CH}_4$  uptake by the fertilized and control soils was not significantly different ( $p < 0.05$ ). In the other 2 gas samplings (July 19 and August 9),  $\text{CH}_4$  uptake was significantly higher in the control plots. This may have been caused by differences in soil moisture (Fig. 4). Soil moisture in the control on both July 19 and August 9 (81% on July 19 and 132% on August 9) was lower than the fertilized plot (132% on July 19 and 209% on August 9) suggesting that the higher  $\text{CH}_4$  uptake by control soils may have been caused by more rapid gas transport of  $\text{CH}_4$  to the zone of  $\text{CH}_4$  oxidation. These results suggest that soil moisture may be the dominant factor controlling  $\text{CH}_4$  uptake by soils in high elevation spruce-fir forests in the northeastern US.

### *Importance of spruce-fir forests in trace gas exchange*

Although atmospheric nitrogen-deposition has the potential to alter soil nitrogen dynamics and the fluxes of both  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , the high elevation spruce-fir forests examined in this study that have some of the highest rates of nitrogen deposition (10–16  $\text{kg N/ha-yr}$ ) in the northeastern US had low  $\text{N}_2\text{O}$  emissions and  $\text{CH}_4$  uptake rates that were not different from those reported for many other ecosystems. The low  $\text{N}_2\text{O}$  flux is consistent with the low net nitrification rates and  $\text{NO}_3\text{-N}$  concentrations in the organic horizons at our study sites. Unlike other studies,  $\text{CH}_4$  uptake by soils at our study sites did not appear to be affected by either soil nitrogen concentrations or nitrogen dynamics. Results from our fertilization experiment were consistent with results from our regional transect. Although our

fertilized plot received ca. 3 times the normal nitrogen deposition in 1988, 1989 and 1990, the fluxes of  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , during the 1990 growing season, were not dramatically altered. If results from our fertilization experiment can be directly extrapolated to other spruce-fir forests along our transect, then many more years of high ( $> 10 \text{ kg N/ha-yr}$ ) nitrogen-deposition may be required before dramatic changes in the  $\text{N}_2\text{O}$  and  $\text{CH}_4$  fluxes are observed.

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