

Available online at www.sciencedirect.com



JOURNAL OF ENVIRONMENTAL SCIENCES <u>ISSN 1001-0742</u> CN 11-2629/X www.jesc.ac.cn

Journal of Environmental Sciences 20(2008) 1312-1322

Nitrous oxide fluxes from upland soils in central Hokkaido, Japan

MU Zhijian^{1,2}, Sonoko D. KIMURA³, Yo TOMA², Ryusuke HATANO^{2,*}

1. Key Laboratory of Agricultural Resources and Environmental Research, College of Resources and Environment, Southwest University, Chongqing 400716, China. E-mail: muzj01@yahoo.com

2. Graduate School of Agriculture, Hokkaido University, Sapporo 060-8589, Japan

3. Graduate School of Agriculture, Tokyo University of Agriculture and Technology, Tokyo 183-8509, Japan

Received 9 November 2007; revised 5 December 2007; accepted 7 January 2008

Abstract

Nitrous oxide (N₂O) fluxes from soils were measured using the closed chamber method during the snow-free seasons (middle April to early November), for three years, in a total of 11 upland crop fields in central Hokkaido, Japan. The annual mean N₂O fluxes ranged from 2.95 to 164.17 μ gN/(m²·h), with the lowest observed in a grassland and the highest in an onion field. The instantaneous N₂O fluxes showed a large temporal variation with peak emissions generally occurring following fertilization and heavy rainfall events around harvesting in autumn. No clear common factor regulating instantaneous N₂O fluxes was found at any of the study sites. Instead, instantaneous N₂O fluxes at different sites were affected by different soil variables. The cumulative N₂O emissions during the study period within each year varied from 0.15 to 7.05 kgN/hm² for different sites, which accounted for 0.33% to 5.09% of the applied fertilizer N. No obvious relationship was observed between cumulative N₂O emission and applied fertilizer N rate (*P* > 0.4). However, the cumulative N₂O emission was significantly correlated with gross mineralized N as estimated by CO₂ emissions from bare soils divided by C/N ratios of each soil, and with soil mineral N pool (i.e., the sum of gross mineralized N and fertilizer N) (*P* < 0.001).

Key words: mineral N pool; N2O flux; soil; upland cropping system

Introduction

Nitrous oxide (N₂O) has a lifetime of about 120 years in the atmosphere and contributes to global warming and to the destruction of ozone in the stratosphere (IPCC, 2001). The atmospheric concentration of N₂O has increased from 270 to 314 ppbv during the past 250 years and continues to increase with an annual rate of 0.8 ppbv (IPCC, 2001). Emissions from agricultural soils are a key item in the global atmospheric N₂O budget, making up about half of the terrestrial biogenic emissions (Mosier *et al.*, 1998).

 N_2O is produced in the soil by both denitrification and nitrification (Conrad, 1996). Although denitrification is an anaerobic process and nitrification aerobic, the two processes can occur simultaneously at aerobic and anaerobic microsites, within the same aggregate (Azam *et al.*, 2002), and can be influenced by soil variables that influence microbial activity, such as concentrations of NO_3^- , NH_4^+ , O_2 , and labile organic carbon, which, in turn, are controlled by a combination of soil properties (soil moisture, texture, structure) and soil management practices (Li *et al.*, 1992; Davidson *et al.*, 2000). Without accounting for the complicated interactions among climate, soils, and cropping systems in controlling N₂O emission from agricultural soils, the emission factor-based approach recommended by IPCC (2001), has been widely employed for developing the relates direct N_2O emission to the amount of fertilizer N applied, on the basis of a fixed emission factor. This clearly provides uncertainties in inventory calculations (Mosier *et al.*, 1998). Since agricultural activities are gradually coming into focus in the GHG budget calculations, precise estimates of N_2O emissions from arable land are being sought, along with possible means of abatement. Researchers have developed empirical and process models for this purpose (Li, 2000). However, more field measurements are also required to validate and improve the models, and to build regional or global inventories of N_2O from soils. Thus, this study was carried out to quantify the magnitude of N_2O emissions from soils under different cropping systems in central Hokkaido, and to identify the environmental factors controlling these emissions.

national inventory of greenhouse gas (GHG) emissions. It

1 Materials and methods

1.1 Site description

This study was conducted at Mikasa City, central Hokkaido, Japan (43°14'N, 141°50'E). In this area, the snow cover usually begins in middle November and snowmelt completes around middle April. The mean annual air temperature and annual precipitation over 30 years (1971 to 2000) were, respectively, 7.4°C and 1,154 mm

*

^{*} Corresponding author. E-mail: hatano@chem.agr.hokudai.ac.jp.

(Iwamizawa Meteorological Station Database, 2005). The mean annual air temperatures in 2003, 2004, and 2005 were 7.4, 8.5, and 7.5°C, respectively; and the annual precipitations were 986, 1,295 and 1,398 mm, respectively.

Soil gas fluxes were measured at a total of 11 upland sites during the no snow cover seasons (middle April to early November) from 2003 to 2005. These sites were designated as A to G for 2003; A, D, E, and I for 2004; and A, D, P, Q, and R for 2005. These sites were distributed within an area of about 30 km² along the Ikushunbetsu River and had an altitude of 30 to 90 m a.s.l. These fields covered major upland soil-crop systems in the study area and were under the local farmers' conventional management. The sites A and D had been converted from forest to agricultural use for over 18 years at the initiation of this study. For other sites, the history of agricultural use ranged from about 40 to 100 years. The soil types were brown forest soils (Dystric Cambisols, FAO-UNESCO, 1988), at sites A, D, and R, gray lowland soils (Eutric Fluvisols) at sites B and P, brown lowland soils (Dystric Fluvisols) at sites C, E, F, G, and I, and pseudogleys (Dystric Gleysols) at site Q. The basic physical and chemical properties of these soils are shown in Table 1.

The field management practices at the study sites are presented in Table 2. The grassland site A was renovated in middle September 2002. *Dactylis glomerata* and *Phleum* pratense were the dominant species, and the aboveground biomass was harvested twice each year. The crop fields had been continuously managed under conventional tillage. Each year, the fields were generally plowed to a depth of roughly 25 cm before planting and after harvesting, to incorporate crop residues. Winter wheat was generally sown in September of the previous year and harvested in late July or early August of the following year. Other crops were planted around May and harvested around September of the same year. Site D was planted with wheat in 2003, but was left fallow in 2004 and 2005, and ploughed frequently to control weed growth. Both the sites E and P were under continuous onion (Allium cepa L.) growth for more than 10 years. Site R was subjected to a long-term pumpkin-potato rotation system. The total amount of chemical N application ranged from 0 kgN/hm² at the fallow site D to 300 kgN/hm² at the wheat site Q. Ammonium sulfate was the form of chemical N fertilizer used, except for the site Q to which 20% of N fertilizer was applied as urea. At site Q, a compost made from cow excreta mixed with wheat straw was also utilized at a rate of 100 Mg/hm² on a wet basis, and incorporated into the soil after the wheat was harvested in late August. The organic carbon and total N applied with the compost were at the rates of 6,543 kgC/hm² and 339 kgN/hm², respectively.

Table 1 Basic characteristics of the topsoil (0-20 cm) at the study sites

Site	А	В	С	D	Е	F	G	Ι	Р	Q	R
Soil type	BF	GL	BL	BF	BL	BL	BL	BL	GL	PG	BF
pH (H_2O)	5.42	5.08	5.76	5.32	5.30	5.94	5.73	5.61	6.02	6.51	5.53
Total C (g/kg)	17.2	19.8	24.1	16.2	23.3	26.2	16.0	17.6	27.0	17.0	38.7
Total N (g/kg)	1.36	1.82	2.20	1.34	1.82	2.15	1.54	1.43	2.07	1.67	3.14
C/N ratio	12.6	10.9	11.0	12.1	12.8	12.2	10.4	12.3	13.0	10.2	12.3
CEC (cmol c/kg)	17.2	28.5	26.1	19.7	21.8	27.9	24.1	23.0	28.8	23.6	34.4
Sand (%)	44.9	27.4	47.0	38.4	51.6	43.3	36.2	34.1	18.4	26.1	27.9
Silt (%)	38.3	37.9	32.6	40.6	30.8	38.1	40.1	40.0	47.7	40.5	47.6
Clay (%)	16.8	34.7	20.4	21.0	17.6	18.6	23.7	25.9	33.9	33.4	24.5

BF: brown forest soil; BL: brown lowland soil; GL: gray lowland soil; PG: pseudogley; CEC: cation exchange capacity.

 Table 2
 Field management practices at the study sites

Site	Land use	Date of sowing	Inorganic N	application	Date of harvest
			Rate (kgN/hm ²)	Date	
A	Grass	15 Sep., 2002	32	20 Apr., 2003	15 Jun./1 Sep., 2003
В	Wheat	16 Sep., 2002	48	15 Sep., 2002	23 Jul., 2003
С	Maize	5 May 2003	196	30 Apr., 2003	30 Aug., 2003
D	Wheat	6 Sep., 2002	40	5 Sep., 2002	26 Jul., 2003
		-	100	18 Apr., 2003	
Е	Onion	26 Apr., 2003	220	23 Apr., 2003	5 Sep., 2003
F	Wheat	8 Sep., 2002	112	5 Sep., 2002	26 Jul., 2003
G	Soybean	1 Jun., 2003	36	29 May 2003	16 Oct., 2003
А	Grass		50	1 May 2004	30 Jun./1 Sep., 2004
D	Fallow		0	-	
Е	Onion	26 Apr., 2004	228	20 Apr., 2004	25 Aug., 2004
Ι	Wheat	26 Sep., 2003	30	17 Sep., 2003	18 July 2004
		x ·	90	19 Apr., 2004	2
А	Grass		100	25 Apr., 2005	30 Jun./10 Sep., 2005
D	Fallow		0		, ,
Р	Onion	3 May 2005	200	27 Apr., 2005	26 Sep., 2005
Q	Wheat	15 Sep., 2004	240	24 Apr., 2005	
		x ·	30	16 May 2005	1 Aug., 2005
			30	23 Jun., 2005	
R	Potato	15 May 2005	94	14 May 2005	20 Sep., 2005

1.2 Measurement of gas fluxes

The gas fluxes from the cultivated soils were measured using a closed chamber technique as described in the previous article (Mu et al., 2006). In 2003, three openended stainless steel cylinders with an internal diameter of 20 cm and a height of 25 cm were pushed into the soil between growing plants (without covering living plants) at each site to a depth of 3 cm to prevent gas leakage from the bottom of the chamber. The open cylinders were left standing for 30 min to obtain an equilibrium state before gas sampling (Norman et al., 1997). When equilibrium was reached the cylinders were sealed using white-colored acrylic lids with two ports, one for gas sampling and the other for the attachment of a sampling bag to equilibrate the chamber pressure with the atmospheric pressure. To determine N₂O fluxes, headspace samples of 20 ml were withdrawn from the chamber at 0, 10, and 20 min, using a polypropylene syringe with a three-way stopcock, and transferred into a 10-ml vacuumed vial. In 2004 and 2005, three chamber bases made of stainless steel were inserted into soils at each site, to a depth of 3 cm, two days before the first sampling and remained there till the last sampling within each year, except for temporary removal when plowing, planting, or harvesting were practiced. At each sampling event, the chamber bottom was fitted into the base groove, in which water was filled to make the system air-tight. Sampling was conducted twice or thrice per month between 09:00 and 13:00 on each sampling date. Gas samples were analyzed for N2O concentrations using a gas chromatographic system equipped with an ⁶³Ni electron capture detector (ECD, GC-14B, Shimadzu, Japan).

A bare plot $(3 \text{ m} \times 2 \text{ m})$ was established at each site (except for site A) for the crop growth periods of each year. These plots were kept bare through regular cutting and removal of green vegetation for excluding root respiration (Hanson et al., 2000). The CO₂ fluxes from these bare plots were determined on the same dates as the measurement of N₂O fluxes, using the closed chamber method as described earlier, and were taken as an indicator of the rate of soil organic matter decomposition. Headspace gas samples were withdrawn at times of 0 and 6 min, since some studies had shown that reducing the time between gas measurements to less than 10 min could minimize the underestimation of CO₂ fluxes during the closed chamber method (Nakano et al., 2004). The CO₂ concentrations in gas samples were measured using an infrared gas analyzer (ZFP-9, Fuji Electric Co., Ltd., Japan). After the crop was harvested, CO₂ fluxes were only determined from previously cropped plots from which N₂O fluxes were also measured from the beginning of the study. Thus the crop residue decomposition could be included. At grassland site A, the CO₂ fluxes were measured from planted plots and half of them were taken as the rate of soil organic matter decomposition.

The instantaneous gas fluxes from soils were calculated from the changes in gas concentrations in the chambers, with time, using linear regression, and expressed as arithmetic means (n = 3). The cumulative gas emissions during the sampling period within each year were calculated assuming the existence of linear changes in gas fluxes between two successive sampling dates. The cumulative number of days was 200 for 2003 (i.e., from 10 April to 26 October), 213 for 2004 (i.e., from 10 April to 8 November), and 184 for 2005 (i.e., from 4 May to 3 November). There might be some uncertainties in the estimates of cumulative N₂O emission due to the lower sampling frequency that might have caught or missed some peaks of N₂O flux.

1.3 Soil physical and chemical analyses

Triplicate measurements of soil temperature at 5 cm were recorded next to each chamber by a handy digital thermometer (CT-220, Custom Corp., Japan) at the time the gas flux was measured. The nine point soil temperature measurements at a site were averaged to produce a mean value for the site on each sampling date.

Three disturbed soil samples were collected from 0-5 cm depth adjacent to each chamber, at each site, on each gas sampling date. In the laboratory soil samples from the same sites were mixed and sieved (2 mm). Subsamples were used to determine soil moisture by oven drying at 105°C for 24 h. Soil moisture was expressed as the percentage of water-filled pore space (WFPS) using the measured soil bulk density (average of three measuring dates in May, July, and September of each year) and assuming a particle density of 2.65 g/cm³. Subsamples were extracted with distilled water (dry soil to water ratio of 1:5, W/V), shaken in a reciprocating shaker for 1 h, centrifuged at 8,000 r/min for 10 min, and filtered through 0.2-µm membrane filters. The filtration solution was stored at 4°C until analysis for dissolved nutrients, which would be within one month. Water-soluble organic carbon (DOC) content in the extract solution was analyzed using a TOC analyzer (TOC 5000A, Shimadzu, Japan). The content of NO₂⁻ and NO₃⁻ was analyzed by using a Dionex ion chromatograph equipped with a conductivity detector. The content of NH₄⁺ was determined by using the indophenol blue colorimetric method. The total nitrogen content in the extract solution (TDN) was determined by using persulfate oxidation-UV spectrophotometric analysis at a wavelength of 220 nm (UVmini-1240, Shimadzu, Japan). The content of water-soluble organic nitrogen (DON) was calculated as the difference between the content of TDN and that of dissolved inorganic nitrogen (DIN) (i.e., $NO_3^- + NH_4^+ +$ NO_{2}^{-}).

1.4 Statistical analysis

One-way ANOVA procedure with Fisher LSD test (SAS 8e for Windows, SAS Institute, Cary, NC) was used to compare the mean difference (P < 0.05) of a given variable between sites within each year. Relationships between variables were assessed using stepwise multiple regression analysis (P < 0.05). Gas fluxes and soil chemical parameters, such as the contents of nitrate, ammonium, DOC, and DON, and the derivative variables were usually log normally distributed and, if needed, were naturally log-transformed before statistical analysis.

2 Results

2.1 Soil temperature and moisture

Soil temperature generally increased from April through August and decreased after September (Fig.1). Soil temperature was usually above 10°C, but rarely exceeded 30°C from May through early November. The lowest temperatures were always measured from site A across the three years. Site D was left fallow in 2004 and 2005, hence the surface soil was directly exposed to sunlight, resulting in higher soil temperature than other sites (Table 3).

Soil moisture also responded to rainfall patterns over years (Fig.1). Soil moisture usually ranged from 30% to 75% WFPS during April to May and September to November. Soil dry conditions prolonged from June through July

in 2003. Some soil dry events also occurred during June to early August in 2004 and 2005, but each dry event was rapidly ameliorated by precipitation. Soil WFPS averaged for the sampling period varied from 38.0% at maize site C to 47.6% at grassland site A for 2003, from 40.6% at fallow site D to 62.0% at wheat site I for 2004, and from 36.4% at potato site R to 63.1% at grassland site A for 2005 (Table 3).

2.2 Dynamics of soil NH₄⁺, NO₃⁻, DON, and DOC

Chemical fertilizer was applied mainly in the form of ammonium sulfate to the study sites. However, the concentrations of soil NH4⁺ at most sites rarely exceeded 10.0 mgN/kg soil for the study period (Fig.2). On the other hand, the concentrations of soil NO₃⁻ increased to high levels (> 20 mgN/kg soil) following fertilization as a result



Fig. 1 Seasonal patterns of local weather conditions (a), soil temperatute (b), and WFPS (c) at cropped plots.

MU Zhijian et al.

plots
1

Year	Site	Land use	Soil temperature (°C)	Soil WFPS (%)	NH4 ⁺ (mgN/kg)	NO ₃ ⁻ (mgN/kg)	DON (mgN/kg)	DOC (mgC/kg)
2003	А	Grass	15.3 a	47.6 ab	0.94 a	11.90 a	3.45 a	26.89 c
	В	Wheat	17.5 ab	46.7 ab	3.18 a	14.55 a	2.36 a	9.26 a
	С	Maize	20.3 b	38.0 a	1.99 a	42.04 b	7.10 a	14.98 b
	D	Wheat	19.7 b	48.6 b	3.87 a	13.97 a	22.73 b	9.60 a
	Е	Onion	19.9 b	38.4 a	0.96 a	39.35 b	4.98 a	15.99 b
	F	Wheat	18.5 ab	40.9 ab	1.52 a	23.17 a	5.48 a	15.54 b
	G	Soybean	21.1 b	39.2 ab	0.53 a	15.49 a	2.78 a	15.35 b
2004	А	Grass	16.4 a	54.1 b	1.43 a	14.14 a	3.82 a	29.57 b
	D	Fallow	21.0 a	40.6 a	0.14 a	10.66 a	2.57 a	12.93 a
	Е	Onion	18.3 a	42.4 a	0.46 a	17.01 a	4.25 a	18.09 a
	Ι	Wheat	17.7 a	62.0 c	1.82 a	19.23 a	3.35 a	9.85 a
2005	А	Grass	17.7 a	63.1 c	2.51 a	6.81 a	3.64 a	31.74 b
	D	Fallow	21.9 a	37.7 a	0.17 a	7.49 a	4.44 a	19.92 a
	Р	Onion	20.3 a	52.7 b	1.82 a	47.75 b	22.88 ab	25.67 ab
	Q	Wheat	18.8 a	55.8 bc	1.50 a	10.36 a	6.20 a	27.42 ab
	R	Potato	20.0 a	36.4 a	33.14 b	39.31 b	30.27 b	32.86 b

* The data followed by the same letters within a column and a soil parameter are not significantly different (p = 0.05) among the sites in the same year.



of microbial transformation of NH4⁺ to NO3⁻ through nitrification. Later, cropping effectively diminished the amount of NO₃⁻ in the soil, hence after July or middle August all sites other than maize C and onion E in 2003, showed less amount of NO_3^- (< 20 mgN/kg). These maize C and onion E soils maintained a high level of NO₃⁻ (around 60 mgN/kg) from May through August in 2003 (Fig.2). Site D was left fallow and did not receive fertilizer in 2004 and 2005, so the concentration of NO_3^- was usually lower than 20 mgN/kg across the two years. In 2003, the mean concentrations of NO_3^- in maize soil C and onion soil E were significantly higher than those measured at other soils (Table 3). In 2004, the mean concentrations of NO3⁻ were not significantly different among sites, ranging from 10.66 to 19.23 mgN/kg. In 2005, the mean concentrations of NO_3^- at sites A, D, and Q were significantly lower than those measured at onion P and potato R.

Soil DON content was no more than 20 mgN/kg on most of the sampling dates for most of the sites (Fig.2). The mean contents of DON were 22.73, 22.88, and 30.27 mgN/kg for wheat D in 2003, and onion P and potato R in 2005, respectively (Table 3). For the other sites, the mean contents of DON ranged from 2.36 to 7.10 mgN/kg. Soil dissolved organic carbon (DOC) was relatively high just after snow melting (Fig.2), probably due to soil freezing and thawing, which could have damaged the soil aggregates and promoted the release of water extractable organic carbon. It was then decreasing rapidly, but later increased with time. The mean contents of DOC at grassland A, onion P, wheat Q, and potato R were about 30.0 mgC/kg. For other sites, the mean contents of DOC were lower than 20.0 mgC/kg (Table 3).

2.3 Soil N₂O flux

All soils were sources of N₂O, except for several measurements showing a little uptake (-6.60 to -3.12 μ gN/(m²·h)) (Fig.3). In 2003, high N₂O fluxes were generally observed in wheat soil D and onion soil E. On the first sampling date after snow melting (10 April), the N₂O fluxes were 36.7 and 0.4 μ gN/(m²·h) in wheat soil D and onion soil E, respectively. The fluxes increased rapidly to higher emission levels (242 to 433 μ gN/(m²·h) in wheat soil D and 47.2 to 157 μ gN/(m²·h) in onion soil E) with

Table 4Mean and cumulative fluxes of N_2O from different cultivated
soils*

Year	Site	Land	Mean flux (ugN/	Cumulative flux	Percentage of ferti-
		use	$(m^2 \cdot h))$	(kgN/hm ²)	lized N (%)
2003	А	Grass	2.95 a	0.15 a	0.48
	В	Wheat	13.34 ab	0.83 a	1.74
	С	Maize	20.35 ab	0.96 a	0.49
	D	Wheat	129.57 c	6.04 c	4.31
	Е	Onion	56.31 b	2.75 b	1.25
	F	Wheat	7.37 a	0.37 a	0.33
	G	Soybean	15.66 ab	0.60 a	1.67
2004	А	Grass	32.43 a	1.54 a	3.08
	D	Fallow	127.11 b	6.44 b	_
	Е	Onion	28.12 a	1.34 a	0.59
	Ι	Wheat	115.97 b	6.11 b	5.09
2005	А	Grass	10.16 a	0.42 a	0.42
	D	Fallow	48.88 ab	2.33 b	_
	Р	Onion	164.17 c	7.05 d	3.53
	Q	Wheat	80.02 b	3.62 c	1.21
	R	Potato	49.31 ab	2.05 b	2.18

* The data followed by the same letters within a column and a parameter are not significantly different (p > 0.05) among sites in the same year. -: data not available.

fertilization and lasted for about three weeks. From June to November 2003, the N₂O emissions in wheat soil D were lower than 40 μ gN/(m²·h) (4.3 to 36.9), except for the measurements on three sampling dates (2 June, 12 and 27 August 2003). The highest emission in wheat soil D $(476 \ \mu gN/(m^2 \cdot h))$ was recorded on 12 August, 2003, with a heavy precipitation in the previous week (amounting to 60 mm, Fig.1). The emissions of N₂O from onion soil E maintained a relatively high level (generally above 50 $\mu gN/(m^2 \cdot h)$) from June to September, but decreased after October 2003. The seasonal pattern of N₂O emissions from other soils was characterized by low flux rates (< 20 $\mu gN/(m^2 \cdot h)$) on most sampling dates. The lower rates were measured in grassland soil A (-3.31 to $8.60 \,\mu gN/(m^2 \cdot h)$). N₂O emissions from wheat soil B, maize soil C, and soybean soil G were increasing toward autumn in 2003, but few rates exceeded 90 μ gN/(m²·h).

Seasonal patterns of soil N_2O fluxes in 2004 and 2005 were similar to those in 2003, with peak emissions generally occurring following fertilization and heavy rainfall events (Fig.3).

The mean N₂O fluxes in 2003, varied greatly from 2.95



 μ gN/(m²·h) at the grassland site A to 129.57 μ gN/(m²·h) at the wheat site D (Table 4). The mean N₂O fluxes from other soils ranged from 7.37 to 56.31 μ gN/(m²·h), but were not statistically different from grassland site A or onion site E. In 2004, the mean N₂O fluxes ranged from 28.12 μ gN/(m²·h) at onion site E to 127.11 μ gN/(m²·h) at fallow site D. In 2005, the mean N₂O fluxes ranged from 10.16 μ gN/(m²·h) at grassland site A to 164.17 μ gN/(m²·h) at onion site P.

2.4 Relationships between instantaneous N_2O fluxes and soil variables

Stepwise multiple regression analysis was used to identify key factors regulating instantaneous N₂O fluxes from soils. The results, however, showed that there was large variability in the model forms (Table 5). A single soil variable or a combination of soil variables could explain 40% to 94% of the temporal variability in site-specific soil instantaneous N2O fluxes, except for several sites where no soil variables met the significance level of P < 0.05, for entry into the model. Soil NO3⁻ content, soil temperature, and WFPS together explained 46% of the inter-site variation in instantaneous N₂O flux in 2003. In 2004, soil instantaneous CO2 flux and soil WFPS together explained 56% of the inter-site variation in instantaneous N_2O flux. In 2005, 44% of inter-site variability in soil instantaneous N₂O flux was explained by soil fine particle (clay plus silt) content and instantaneous CO2 flux. Most of the measured soil variables entered the model of instantaneous N2O flux when all data across the three years were pooled together.

2.5 Cumulative N₂O emission

Cumulative N_2O flux from soils during the sampling period within each year ranged from 0.15 kgN/hm² at

grassland site A to 6.04 kgN/hm² at wheat site D in 2003, from 1.34 kgN/hm² at onion site E to 6.44 kgN/hm² at fallow site D in 2004, and from 0.42 kgN/hm² at grassland site A to 7.05 kgN/hm² at onion site P in 2005 (Table 4). The seasonal distribution of soil N₂O emission was distinct across sites and years. A large proportion of the total N₂O emission was released during the crop growth period (from April through August) at most of the study sites. For example, the N2O emission during this period in 2003, accounted for 69% (grassland site A) to 93% (wheat site D) of the total emission, except for wheat site B and maize site C from which only 40% of the annual total emission occurred during this period. The N2O emission from April through August, at site D, decreased to 66% and 52% of the total emission in 2004 and 2005, respectively. The N₂O emission at site E during this period increased from 76% in 2003 to 99% in 2004. The N_2O emission from April through August also contributed to 97% of the total emission at site I in 2004.

3 Discussion

3.1 Seasonal pattern of N₂O emission

Previous studies have shown that large N₂O emission could be expected from agricultural soils just after nitrogen fertilization, lasting from 10 d (Yoh *et al.*, 1997) to six weeks (Freney, 1997). The reported values of N₂O fluxes ranged from 100 to 1,900 μ gN/(m²·h) (Smith *et al.*, 1998; Akiyama and Tsuruta, 2003; Kusa *et al.*, 2002; Sehy *et al.*, 2003). Peak emissions just after fertilization were also observed in this study, particularly distinct at sites D and E in 2003, site I in 2004, and site P in 2005 (Fig.3). At these study sites, the maximum N₂O fluxes within one

Table 5 Stepwise regression analysis of the correlation of instantaneous soil N2O fluxes with environmental factors

Year	Site	Model	Model R ²	Р
2003	А	$\ln(F_{N_2O}) = -1.075\ln(C_{DOC}) + 0.666\ln(F_{CO_2}) + 0.175\ln(C_{NH^+_{+}}) + 1.734$	0.89	< 0.003
	В	$\ln(F_{N_2O}) = -2.347 \ln(C_{TDN}) + 1.509$	0.40	< 0.01
	С	$\ln(F_{N_2O}) = -0.413\ln(C_{NH^+}) + 2.225$	0.74	< 0.006
	D	$\ln(F_{N_2O}) = -4.188 \ln(C_{DOC}) + 13.196$	0.54	< 0.007
	Е	$\ln(F_{N_2O}) = 0.704 \ln(C_{NO_3}/C_{NH^+}) + 0.856$	0.61	< 0.0006
	F	$\ln(F_{\rm N_2O}) = 0.846 \ln(C_{\rm NO_3}/C_{\rm NH^+}) + 1.139 \ln(F_{\rm CO_2}) - 6.214$	0.69	< 0.009
	G	$\ln(F_{N_2O}) = 1.414 \ln(C_{TDN}) + 0.682 \ln(F_{CO_2}) - 4.315$	0.87	< 0.002
	All sites	$\ln(F_{N_2O}) = 0.607 \ln(C_{NO_3}) + 0.142 T_{soil} + 0.039 W_{soil} - 3.492$	0.46	< 0.0001
2004	А	No variable met at the 0.05 significance level for entry into the model		
	D	No variable met at the 0.05 significance level for entry into the model		
	E	$\ln(F_{\rm N_2O}) = 1.35 \ln(F_{\rm CO_2}) + 0.043 W_{\rm soil} - 3.967$	0.94	< 0.0001
	Ι	$\ln(F_{N_2O}) = 1.226\ln(F_{CO_2}) - 1.56\ln(C_{DOC}) + 1.719$	0.92	< 0.0001
	All sites	$\ln(F_{N_2O}) = 1.026\ln(F_{CO_2}) - 0.937\ln(C_{DOC}) + 1.291$	0.56	< 0.0001
2005	А	$\ln(F_{N_2O}) = 3.361 \ln(C_{DOC}) + 0.079 W_{soil} - 14.881$	0.62	< 0.003
	D	$\ln(F_{N_2O}) = 0.147 W_{soil} + 0.192 T_{soil} - 6.668$	0.89	< 0.0001
	Р	No variable met at the 0.05 significance level for entry into the model		
	Q	No variable met at the 0.05 significance level for entry into the model		
	R	No variable met at the 0.05 significance level for entry into the model		
	All sites	$\ln(F_{N_2O}) = 0.133 Pcs + 0.72\ln(F_{CO_2}) - 8.858$	0.44	< 0.0001
2003-2005	All data	$\ln(F_{N_2O}) = 0.204 \ln(C_{NO_3^-}/C_{NH_4^+}) + 0.388 \ln(F_{CO_2}) + 0.038 P_{cs} +$		
		$0.379 \ln(C_{\text{DIN}}) + 0.029 \ \vec{W}_{\text{soil}} + 0.063 \ T_{\text{soil}} - 14.881$	0.41	< 0.0001

 F_{N_2O} (µgN/(m²·h)) and F_{CO_2} (mgC/(m²·h)): instantaneous fluxes. C_{NO_2} (mgN/kg), $C_{NH_4^+}$ (mgN/kg), C_{DIN} (mgN/kg), C_{TDN} (mgN/kg) and C_{DOC} (mgC/kg): concentrations of nitrate, ammonium, DIN (dissolved inorganic nitrogen), TDN (total dissolved nitrogen), and DOC (dissolved organic carbon) in soils, respectively. P_{cs} (%), W_{soil} (%), and T_{soil} (°C): sum of soil clay and silt contents, water-filled pore space (WFPS), and soil temperature, respectively.

month after fertilizer application ranged from 157 to 792 $\mu g N/(m^2 \cdot h)$, which were well within the reported values. Besides the fertilizer-induced peak emissions, several researchers have reported peak emissions of N₂O during the harvest season just after heavy rainfall. For example, Koga *et al.* (2004), observed the highest N_2O flux of 300 $\mu g N/(m^2 \cdot h)$ from Andisols in eastern Hokkaido during the harvest season, just after a heavy rainfall. Kusa et al. (2006), reported that N₂O fluxes during the harvest season, just after a heavy rainfall, ranged from 1,730 to 6,420 μ gN/(m²·h) in an Andisol cropped with maize in central Hokkaido, which were much higher than the fertilizer-induced peak emissions. In the present study, the maximum N₂O fluxes during the period from July to October, just after a heavy rainfall (amounted to around 80 mm within the previous week), were measured at site D in 2003 and 2004, and ranged from 308 to 525 $\mu gN/(m^2 \cdot h)$ (Fig.1). The N₂O fluxes, just after a heavy rainfall, were lower than 300 $\mu gN/(m^2 \cdot h)$ for other sites. The climate during the harvest season in Hokkaido was characterized with heavy or frequent rainfall together with warmer temperature, which could make soil moisture and temperature regimes favorable for N₂O production by denitrification (Kusa et al., 2006).

3.2 Factors regulating instantaneous N₂O fluxes

N₂O is produced by both denitrification and nitrification (Conrad, 1996; Davidson et al., 2000). Any factor affecting these processes could alter N₂O emission. Multiple regression analysis showed that a series of soil factors could affect N₂O fluxes from soils (Table 5). However, there was no clear common factor regulating N₂O fluxes at all the study sites. Instead, N₂O fluxes at different sites were affected by different factors. This was not surprising, since there were large differences in soil properties, land use, and management, among the study sites. The positive relationship of N₂O flux with soil moisture and temperature has been widely reported (Smith et al., 1998; Dobbie et al., 1999; Sehy et al., 2003). Previous studies have shown that nitrification activity increased with soil moisture at WFPS values less than 60% and that it drastically decreased when the WFPS values exceeded 60%. In contrast, the denitrification activity was limited when the WFPS values were less than 60%, but increased dramatically when the values exceeded 60% (Linn and Doran, 1984; Aulakh et al., 1992). In the present study, soil WFPS values were less than 60% on most sampling dates, for most study sites (Fig.3). This could indicate that nitrification might be the primary mechanism of N₂O emission in the present study sites. N₂O emission rate usually increased with increasing soil available nitrogen (Skiba et al., 1998; Weitz et al., 2001; Sehy et al., 2003). A positive relationship between N₂O flux and the ratio of NO₃⁻-N to NH₄⁺-N was further found at sites C, E, and F in 2003. This further suggested that nitrification could have been responsible for N₂O production at these sites. The dominance of NO_3^- in the soil mineral N pool, in spite of ammonium sulfate as the main form of applied fertilizer N, also demonstrated that nitrification was active in these

soils (Firestone *et al.*, 1980; Šimek *et al.*, 2000). Soil NH_4^+ could become an important factor limiting nitrification rates. Mineralization of soil organic carbon could supply NH_4^+ for nitrifying bacteria. As a specific fraction of soil organic carbon, DOC is easily degradable and available to microorganisms (Boyer and Groffman, 1996). Under such cases, N₂O flux may be positively related to soil CO₂ flux, but negatively related to DOC. Mineralization of organic carbon also depletes oxygen and therefore helps create anoxic conditions conducive for denitrification (Weier *et al.*, 1993; Skiba *et al.*, 1998; Azam *et al.*, 2002).

3.3 Cumulative N₂O emission and soil mineral nitrogen pool

The cumulative N₂O emission during no snow cover seasons, within 2003 and 2004, accounted for 0.33% to 5.09% of the applied fertilizer N for wheat land F and I, respectively (Table 4). This percentage was not the emission factor as defined by the IPCC (2001) because the background emission was not subtracted. Both the wheat sites F and I were amended with similar amounts of fertilizer N (Table 2), but the cumulative N₂O emission at site I was about 17 times higher than that at site F (Table 4), which could be due to heavier soil texture and higher soil moisture at site I than at site F (Tables 1 and 3). As a result, the lowest relative loss was found for wheat F and the highest for wheat I (Table 4). In the main island of Japan, the N₂O losses were below 1% of the applied N-fertilizer, and large N₂O emissions occurred just after fertilization (Tsuruta, 1997). In this study, the percentage, on an average, for all sites (except for fallow D in 2004 and 2005), was 1.88%, and a large proportion of N_2O emissions occurred in the early crop growing period and/or around harvesting in autumn.

N₂O is derived from the soil inorganic N pool (NO₃⁻ and NH4⁺) by denitrification and nitrification. Mineralization of organic nitrogen is one of the main sources of soil inorganic N. Nitrogen mineralization must be closely linked with C cycling in the soil. Tietema (1998) and Bengtsson et al. (2003) found gross and net N mineralization rates to be highly dependent on respiration rates. In a 7-d incubation experiment with six soils from different land uses, Erickson et al. (2001), found the soil C/N ratio to be positively correlated with respiration rate, and negatively with the net N mineralization rate. Similarly, Ross et al. (2004), showed that higher gross N mineralization rates were generally observed at lower C/N ratios. Hence CO₂ evolution, together with the C/N ratio, might be a valuable indicator of the magnitude of gross N mineralization rates in the soil. In the present study, the authors divided the CO2 emission from bare soils by C/N ratios of each soil and took the quotients as estimates of gross N mineralization rates. As a result, the total amount of mineralized N throughout the study period within each year was in a range of 95 to 607 kgN/hm² (Table 6). Mineralized N and fertilized N constitute the soil mineral N pool.

No obvious relationship was observed between cumulative N₂O emission and applied fertilizer N rate (P > 0.4). However, cumulative N₂O emission was significantly correlated with gross mineralized N rate (P < 0.001) and with the total soil mineral N pool (fertilized N + mineralized N) (P < 0.001) if the site P was excluded (Fig.4). The highest content of clay and silt in the soil at site P (Table 1) might partly explain the reason why the N₂O emission at this site appeared scattered from those at other sites, as shown in Fig.4. On the one hand, the clay and silt particles might absorb organic matter and inhibit it from decomposition by microorganisms (Hassink et al., 1993), resulting in lower CO_2 emission and thus a lower estimate of mineralized N. On the other hand, the heavy soil texture could facilitate the formation of anaerobic microsites, which might enhance N₂O production through denitrification (Weier et al., 1993; Skiba et al., 1998). The variation in cumulative N₂O emission among different sites could not be predicted by other variables such as soil temperature, WFPS, NH_4^+ , or NO_3^- .

It has been widely recognized that CO₂ emission can well be predicted using the parameters of soil temperature and moisture (Lloyd and Taylor, 1994; Davidson et al., 1998). Thus, the regression model (Fig.4) can be reconstructed as follows: first, using the data of soil temperature and moisture to estimate CO2 emission; second, using C/N ratio to convert CO₂ emission into gross mineralized N; and finally, connecting the total mineral N pool with N2O emission. Soil temperature, moisture, and availability of C and N substrates are generally the key controlling factors for N₂O production. It is encouraging that all these factors are directly or indirectly embedded into the regression model. It must be noted, however, that the regression model is derived from limited data sets, and more data sets from different land uses in different regions are needed to test its suitability and, if promising, to improve it.

Table 6	Estimate	of soil	mineral N	pool a	t different sites
---------	----------	---------	-----------	--------	-------------------

Year	Site	Land	Soil C/N	Cumulative CO ₂	N (kgN/hm ²)			
		use	ratio	flux ^a (kgC/hm ²)	Gross mineralized ^b	Fertilized	Soil mineral ^c	
2003	А	Grass	12.6	2,691	214	32	246	
	В	Wheat	10.9	2,867	263	48	311	
	С	Maize	11.0	2,800	255	196	451	
	D	Wheat	12.1	5,282	437	140	577	
	Е	Onion	12.8	3,302	258	220	478	
	F	Wheat	12.2	4,310	353	112	465	
	G	Soybean	10.4	4,370	420	36	456	
2004	А	Grass	12.6	3,821	303	50	353	
	D	Fallow	12.1	7,349	607	0	607	
	Е	Onion	12.8	1,617	126	228	354	
	Ι	Wheat	12.3	6,916	562	120	682	
2005	А	Grass	12.6	3,636	289	100	389	
	D	Fallow	12.1	4,301	355	0	355	
	Р	Onion	13.0	1,159	89	200	289	
	Q	Wheat	10.2	4,352	427	300	727	
	R	Potato	12.3	2,854	232	94	326	

^a Measured from bare soils, except for site A for which half of the total soil respiration from planted soil was used. ^b Calculated from cumulative CO₂ fluxes divided by soil C/N ratios. ^c Sum of gross mineralized N and fertilized N.



Fig. 4 Correlations of cumulative N₂O fluxes with fertilized N (a), grossly mineralized N (b) and soil mineral N pool (c), with site P not included.

4 Conclusions

All soils were sources of N₂O. Seasonal patterns of soil N₂O fluxes were characterized with peak emissions, generally occurring following fertilization and heavy rainfall events around harvesting in autumn. A series of soil factors could affect N₂O fluxes from soils. However, there was no clear common factor regulating instantaneous N₂O fluxes at all the study sites. Instead, instantaneous N₂O fluxes at different sites were affected by different soil variables. No obvious relationship was observed between cumulative N₂O emission and applied fertilizer N rate (P >0.4). However, cumulative N₂O emission was significantly correlated with the gross mineralized N rate (P < 0.001) and the total soil mineral N pool (i.e., chemical fertilizer N + gross mineralized N) (P < 0.001) if one scatter point (the highest N₂O flux) was removed.

Acknowledgments

This work was supported by the Global Environmental Research Program of the Ministry of the Environment of Japan (No. S-2).

References

- Akiyama H, Tsuruta H, 2003. Effect of organic matter application on N₂O, NO, and NO₂ fluxes from an Andisol field. *Global Biogeochem Cycles*, 17(4): 1100.
- Aulakh A, Doran J W, Mosier A R, 1992. Soil denitrificationsignificance, measurement, and effect of management. Adv Soil Sci, 18: 1–57.
- Azam F, Müller C, Weiske A, Benckiser G, Ottow J C G, 2002. Nitrification and denitrification as sources of atmospheric nitrous oxide-role of oxidizable carbon and applied nitrogen. *Biol Fertil Soils*, 35: 54–61.
- Bengtsson G, Bengtson P, Månsson K F, 2003. Gross nitrogen mineralization-, immobilization-, and nitrification rates as a function of soil C/N ratio and microbial activity. *Soil Biol Biochem*, 35: 143–154.
- Boyer J N, Groffman P M, 1996. Bioavailability of water extractable organic carbon fractions in forest and agricultural soil profiles. *Soil Biol Biochem*, 28: 783–790.
- Conrad R, 1996. Soil microorganisms as controllers of atmospheric trace gases (H₂, CO, CH₄, OCS, N₂O and NO). *Microbial Rev*, 60: 609–640.
- Davidson E A, Belk E, Boone R D, 1998. Soil water content and temperature as independent or confounded factors controlling soil respiration in a temperate mixed hardwood forest. *Global Change Biol*, 4: 217–227.
- Davidson E A, Keller M, Erickson H E, Verchot L V, Veldkamp E, 2000. Testing a conceptual model of soil emissions of nitrous and nitric oxides. *Bioscience*, 50: 667–680.
- Dobbie K E, McTaggart I P, Smith K A, 1999. Nitrous oxide emissions from intensive agricultural systems: variations between crops and seasons; key driving variables; and mean emission factors. *J Geophys Res*, 104: 26891–26899.
- Erickson H, Keller M, Davidson E A, 2001. Nitrogen oxide fluxes and nitrogen cycling during postagricultural succession and forest fertilization in the humid tropics. *Ecosystems*, 4: 67– 84.
- FAO-UNESCO, 1988. Soil Map of the World, Revised Legend. World Soil Resources Report, No. 60. Roma: FAO.

- Firestone M K, Firestone R B, Tiedje J M, 1980. Nitrous oxide from soil denitrification: factors controlling its biological production. *Science*, 208: 749–751.
- Freney J R, 1997. Emission of N₂O from soils used for agriculture. *Nutri Cycl Agroecosyst*, 49: 1–6.
- Hanson P J, Edwards N T, Garten C T, Andrews J A, 2000. Separating root and soil microbial contributions to soil respiration: A review of methods and observations. *Biogeochemistry*, 48: 115–146.
- Hassink J, Bouwman L A, Zwart K B, Bloem J, Brussaard L, 1993. Relationships between soil texture, physical protection of soil organic matter, soil biota and C and N mineralization in grassland soils. *Geoderm*, 57: 105–128.
- IPCC (Intergovernmental Panel on Climate Change), 2001. The Scientific Basis. Cambridge: Cambridge University Press.
- Iwamizawa Meteorological Station Database, 2005. http://www.data.kishou.go.jp/.
- Koga N, Tsuruta H, Sawamoto T, Nishimura S, Yagi K, 2004. N₂O emission and CH₄ uptake in arable fields managed under conventional and reduced tillage cropping systems in northern Japan. *Global Biogeochem Cycles*, 18: GB4025, doi:10.1029/2004GB002260.
- Kusa K, Sawamoto T, Hatano R, 2002. Nitrous oxide emissions for 6 years from a gray lowland soil cultivated with onions in Hokkaido, Japan. *Nutri Cycl Agroecosyst*, 63: 239–247.
- Kusa K, Hu R G, Sawamoto T, Hatano R, 2006. Three years of nitrous oxide and nitric oxide emissions from silandic andosols cultivated with maize in Hokkaido, Japan. *Soil Sci Plant Nutr*, 52: 103–113.
- Li C S, Frolking S, Frolking T A, 992. A model of nitrous oxide evolution from soil driven by rainfall events. 1. Model structure and sensitivity. *J Geophys Res*, 97: 9759–9776.
- Li C, 2000. Modeling trace gas emissions from agricultural ecosystems. *Nutri Cycl Agroecosyst*, 58: 259–276.
- Linn D M, Doran J W, 1984. Effect of water-filled pore space on carbon dioxide and nitrous oxide production in tilled and no-tilled soils. *Soil Sci Soc Am J*, 48: 1267–1272.
- Lloyd J, Taylor J A, 1994. On the temperature dependence of soil respiration. *Funct Ecol*, 8: 315–323.
- Mosier A, Kroeze C, Nevison C, Oenema O, Seitsinger S, van Cleemput O, 1998. Closing the global N₂O budget: nitrous oxide emissions through the agricultural nitrogen cycle. *Nutri Cycl Agroecosyst*, 52: 225–248.
- Mu Z J, Kimura S D, Hatano R, 2006. Estimation of global warming potential from upland cropping systems in central Hokkaido, Japan. *Soil Sci Plant Nutr*, 52: 371–377.
- Nakano T, Sawamoto T, Morishita T, Inoue G, Hatano R, 2004. A comparison of regression methods for estimating soil-atmosphere diffusion gas fluxes by a closed chamber technique. *Soil Biol Biochem*, 36: 107–113.
- Norman J M, Kucharik C J, Gower S T, Baldocchi D D, Crill P M, Rayment M, Savage K, Striegl R G, 1997. A comparison of six methods for measuring soil-surface carbon dioxide fluxes. J Geophys Res, 102: 28771–28777.
- Ross D S, Lawrence G B, Fredriksen G, 2004. Mineralization and nitrification patterns at eight northeastern USA forested research sites. *For Ecol Manage*, 188: 317–335.
- Sehy U, Ruser R, Munch J C, 2003. Nitrous oxide fluxes from maize fields: relationship to yield, site-specific fertilization, and soil conditions. *Agri Ecosyst Environ*, 99: 97–111.
- Šimek M, Cooper J E, Picek T, Šantručková, H, 2000. Denitrification in arable soils in relation to their physico-chemical properties and fertilization practice. *Soil Biol Biochem*, 32: 101–110.

- Skiba U M, Sheppard L J, Macdonald J, Fowler D, 1998. Some key environmental variables controlling nitrous oxide emissions from agricultural and semi-natural soils in Scotland. *Atmos Environ*, 32: 3311–3320.
- Smith K A, Thomson P E, Clayton H, McTaggart I P, Conen F, 1998. Effects of temperature, water content and nitrogen fertilization on emissions of nitrous oxide by soils. *Atmos Environ*, 32: 3301–3309.
- Tietema A, 1998. Microbial carbon and nitrogen dynamics in coniferous forest floor material collected along a European nitrogen deposition gradient. *For Ecol Manage*, 101: 29–36.
- Tsuruta H, 1997. Emission rates of methane from rice paddy fields and of N₂O from fertilized upland fields estimated from intensive field measurement for three years (1992– 1994) all over Japan. *Res Rep Div Environ Planning NIAES*,

13: 101–130.

- Weier K L, Doran J W, Power J F, Walters D T, 1993. Denitrification and the dinitrogen/nitrous oxide ratio as affected by soil water, available carbon, and nitrate. *Soil Sci Soc Am J*, 57: 66–72.
- Weitz A M, Linder E, Frolking S, Crill P M, Keller M, 2001. N₂O emissions from humid tropical agricultural soils: effects of soil moisture, texture and nitrogen availability. *Soil Biol Biochem*, 33: 1077–1093.
- Williams D L, Ineson P, Coward P A, 1999. Temporal variations in nitrous oxide fluxes from urine-affected grassland. *Soil Biol Biochem*, 31: 779–788.
- Yoh M, Toda H, Kanda K, Tsuruta H, 1997. Diffusion analysis of N₂O cycling in a fertilized soil. *Nutr Cycl Agroecosyst*, 49: 29–33.

