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Abstract

Nitric oxide (NO) fluxes from soils were measured using the closed chamber method during the snow-free seasons (middle April to early November) for 3 years in a total of 11 upland crop fields in central Hokkaido, Japan. The annual mean NO fluxes ranged from 0.44 to 127 μg N m^{-2} hr^{-1} with the lowest observed in a grassland and the highest in a potato field. The instantaneous NO fluxes showed a large temporal variation with peak emissions generally occurring following fertilization and heavy rainfall events around harvesting in autumn. There was no clear common factor regulating instantaneous NO fluxes at all the study sites. Instead, instantaneous NO fluxes at different sites were affected by different soil variables. The cumulative NO emissions during the study period within each year varied from 0.02 to 5.11 kg N ha^{-1} for different sites, which accounted for 0.02% to 5.44% of the applied fertilizer N.

1. Introduction

Agricultural soils have been recognized as an important source of NO, making up 10% to 23% of the global atmospheric NO_x budget (Davidson and Kingerlee, 1997; Delmas et al., 1997). NO is produced in soil by both nitrification and denitrification (Conrad, 1996). Although nitrification is an aerobic process and denitrification is anaerobic, the two processes can occur simultaneously at aerobic and anaerobic microsites within the same soil aggregates (Azam *et al.*, 2002), and can be regulated by soil variables that influence microbial activity, such as the availability of NO_3^-, NH_4^+, O_2, and labile organic carbon, which, in turn, are controlled by a combination of soil properties (soil moisture, temperature, texture, structure) and soil management practices (Li *et al.*, 1992; Davidson *et al.*, 2000). Due to the complexity of interactions among soils, climate and land management driving the exchanges of NO between soils and the atmosphere, estimates of biogenic sources remain highly uncertain at regional and global scales (Rolland et al., 2010). National inventory of NO emissions from croplands are currently developed mainly by employing sets of emission factors derived from field-scale monitoring campaigns, assuming NO emission to be a fixed fraction of N inputs to soils. This clearly provides uncertainties in inventory calculations. Since agricultural activities play a significant role in the regional and global NO_x budget, precise estimates of NO 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 still required to validate and improve the models, and to build regional or global inventories of NO from soils. Thus this study was carried out to quantify the magnitude of NO emissions from soils under different cropping systems in central Hokkaido.

2. Materials and methods

2.1 Site description

This study was conducted at Mikasa city, central Hokkaido, Japan (43˚14′N, 141˚50′E). In this area, 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 1154 mm (Iwamizawa Meteorological Station Database, 2005). The mean annual air temperature in 2003, 2004, and 2005 was 7.4, 8.5, and 7.5˚C, respectively; the annual precipitation was 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) across 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 are distributed within an area of about 30 km^2 along with Ikushunbetsu River and have an altitude of 30 to 90 m a.s.l. The selected fields covered major upland soil-crop systems in the study area and were under local farmers' conventional management. The sites A and D have been converted from forest to agricultural use 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 soils and field management practices at the study sites have been detailed in the previous paper (Mu et al., 2008). 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 the previous September and harvested in the next late July or early August. Other crops were planted around May and harvested around September in the same year. The site D was planted with wheat in 2003, but was left fallow in 2004 and 2005 and ploughed frequently to control weed growth. Both of the sites E and P were under continuous onion (*Allium cepa* L.) for more than 10 years. The site R was subjected to a long-term pumpkin-potato rotation system. The total amount of chemical N application ranged from 0 kg N ha^{-1} at fallow site D to 300 kg N ha^{-1} at wheat site Q. Ammonium sulfate was the form of chemical N fertilizer except for the site Q to which 20% of N fertilizer was applied as urea. At site Q, the compost made from cow excreta mixed with wheat straw was also broadcast at a rate of 100 Mg ha^{-1} by wet basis and incorporated into the soil after wheat was harvested in late August. The organic carbon and total N applied with the compost were at the rates of 6543 kg C ha^{-1} and 339 kg N ha^{-1}, respectively.

2.2 Measurement of gas fluxes

The gas fluxes from the cultivated soils were measured using a closed chamber technique as described in the previous paper (Mu *et al.*, 2006). To determine NO fluxes, headspace samples of 200 ml were withdrawn from the chamber at 0 and 20 min, using a polypropylene syringe with a three-way stopcock, and transferred into a 500 ml Tedlar® bag. Concentrations of NO were analyzed using a chemiluminescence nitrogen-oxide analyzer (Model 265P, Kimoto Electric Co. Ltd., Japan). Sampling was conducted at a frequency of two to three times per month between 09:00 and 13:00 on each sampling date. The instantaneous gas fluxes from soils were calculated from the changes in gas concentrations in the chambers with time using a 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).

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. Soil samples from the same sites were mixed and sieved (2 mm) to produce a homogeneous sample. Subsamples were used to determine soil moisture by oven drying at 105˚C for 24 hr. 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 in each year) and assuming a particle density of 2.65 g/cm^3.

3. Results and Discussion

3.1 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 3 years. The site D was left fallow in 2004 and 2005 and thus surface soil was directly exposed to sunlight, resulting in higher soil temperatures than other sites.

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 across 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.

3.2 Soil NO flux

All soils were sources of NO, except for site A with several measurements showing a little uptake (-0.35 to -0.001 μg N m^{-2} hr^{-1}) (Fig. 2). In 2003, a pulse of higher NO emissions were observed in wheat soil D. On the first sampling date after snow melting (10 April), the NO fluxes in wheat soil D were 0.65 μg N m^{-2} hr^{-1}. Afterwards, the fluxes increased rapidly to higher emission levels (148 to 403 μg N m^{-2} hr^{-1}) with fertilization and lasted about 20 days. Through June to November 2003 the NO emissions in wheat soil D were lower than 21 μg N m^{-2} hr^{-1} (0.34 to 20.4). The seasonal pattern of NO emissions from other soils was characterized by low flux rates on most sampling dates and a few dates on which flux rates were higher than 20 μg N m^{-2} hr^{-1}. The lower rates were measured in grassland soil A (-0.35 to 2.25 μg N m^{-2} hr^{-1}). A slight increase in NO emissions followed fertilization or heavy rainfall during June to August for wheat soil B and F, maize soil C, onion soil E and soybean soil G, but few rates exceeded 50 μg N m^{-2} hr^{-1}. The emissions of NO from all soils were towards decrease in autumn in 2003. Seasonal patterns of soil NO fluxes in 2004 and 2005 were similar to that in 2003, with a pulse of higher emissions following fertilization observed in wheat soil I (41 to 117 μg N m^{-2} hr^{-1}),

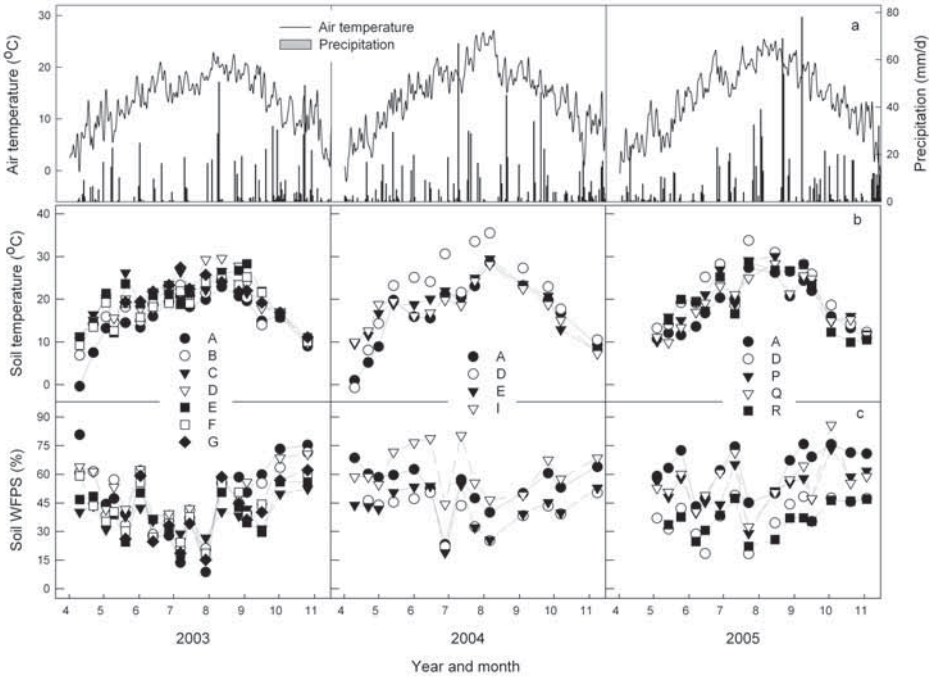

Fig. 1. Seasonal pattern of (a) local weather conditions, (b) soil temperature and (c) moisture at different fields. WFPS is water-filled pore space.

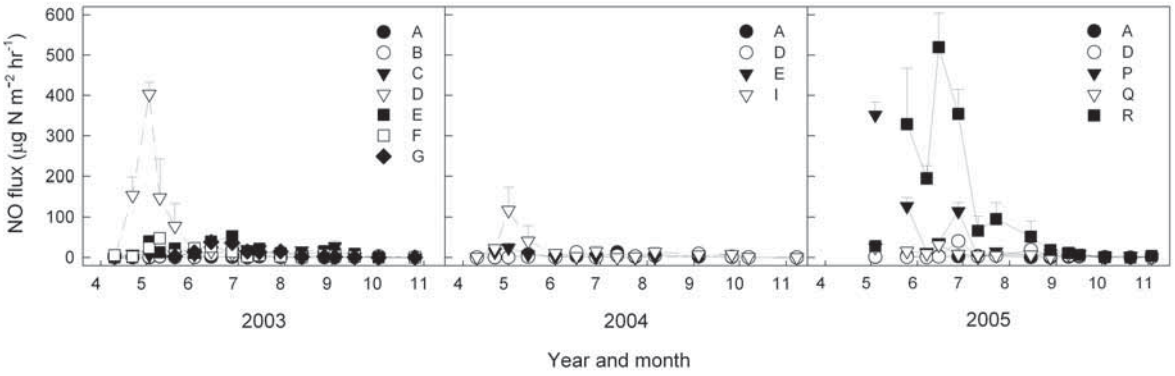

Fig. 2. Seasonal pattern of the NO fluxes from soils at different study sites.

onion soil P (127 to 352 μg N m^{-2} hr^{-1}) and potato soil R (195 to 520 μg N m^{-2} hr^{-1}) (Fig. 2). The lasting duration of the peak emissions for these sites were approximately 3 to 7 weeks. Previous studies have shown a strong stimulation of NO emission from agricultural soils just after nitrogen fertilization, lasting from a few days (Akiyama *et al.*, 2002) to 5 weeks (Akiyama and Tsuruta, 2003; Laville et al., 2009; Venterea et al., 2005). The reported peak values of NO fluxes ranged from 100 to 3960 μg N m^{-2} hr^{-1} (Akiyama and Tsuruta, 2002; Zhang et al., 2011). For this study, the maximum NO fluxes after fertilizer application ranged from 127 to 520 μg N m^{-2} hr^{-1}, which were well within the reported values.

The mean NO fluxes in 2003 varied greatly from 0.44 μg N m^{-2} hr^{-1} at grassland site A to 50.4 μg N m^{-2} hr^{-1} at wheat site D (Table 1). The mean NO fluxes from other soils ranged from 3.42 to 15.7 μg N m^{-2} hr^{-1}, and was not statistically different from grassland site A, but significantly lower than that from wheat

Table 1 Mean and cumulative fluxes of NO from different cultivated soils*

Year	Site	Land use	Mean flux	Cumulative emission	Fertilizer N rate	Percentage of fertilized N
			(μg N m^{-2} hr^{-1})	(kg N ha^{-1})	(kg N ha^{-1})	(%)
2003	A	Grass	0.44 a	0.02 a	32	0.06
	B	Wheat	3.42 a	0.17 a	48	0.35
	C	Maize	10.61 a	0.48 a	196	0.24
	D	Wheat	50.44 b	2.04 b	140	1.46
	E	Onion	15.72 a	0.72 a	220	0.33
	F	Wheat	10.89 a	0.44 a	112	0.40
	G	Soybean	10.89 a	0.40 a	36	1.12
2004	A	Grass	2.86 a	0.15 a	50	0.30
	D	Fallow	2.85 a	0.17 a	0	-
	E	Onion	4.39 a	0.20 a	228	0.09
	I	Wheat	17.22 a	0.77 a	120	0.64
2005	A	Grass	1.83 a	0.08 a	100	0.08
	D	Fallow	6.01 a	0.28 a	0	-
	P	Onion	50.14 b	2.14 b	200	1.07
	Q	Wheat	9.57 a	0.41 a	300	0.14
	R	Potato	127.16 b	5.11 b	94	5.44

* 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.

soil D. In 2004, the mean NO fluxes ranged from 2.85 μg N m^{-2} hr^{-1} at fallow site D to 17.2 μg N m^{-2} hr^{-1} at wheat site I. In 2005, potato site R showed the highest mean NO flux (127 μg N m^{-2} hr^{-1}), and the mean flux for other soils ranged from 1.83 μg N m^{-2} hr^{-1} at grassland site A to 50.1 μg N m^{-2} hr^{-1} at onion site P.

3.3 Cumulative NO emission

Cumulative NO flux from soils during the sampling period within each year ranged from 0.02 kg N ha^{-1} at grassland site A in 2003 to 5.11 kg N ha^{-1} at potato site R in 2005 (Table 1). The pulse of higher NO fluxes following fertilization accounted for a large proportion of the total NO emission for wheat soil D (71%) and I (56%), onion soil P (59%) and potato soil R (71%). For other sites, no distinct peak emissions occurred after fertilization and the total NO emissions were evenly distributed to different cropping seasons.

3.4 Fertilizer N loss as NO

A few studies have reported a positive correlation between cumulative soil NO emission and fertilizer N rate (Liu et al., 2005; Mei et al., 2009; Veldkamp and Keller, 1997). Many other field studies, however, have failed to link soil NO emission directly to the application rate of fertilizer N. Similarly, no significant relationship existed between soil NO emission and fertilizer N rate in our study ($p > 0.6$). As an alternate approach, the emission factor is generally recommended to use for estimating regional NO emissions due to fertilizer application at annual or sub-decadal scales (IPCC, 2006). In this study, unfertilized or control plots were not established, therefore the fertilizer-induced NO emission factor could not be calculated as defined by IPCC (2006). Assuming that the NO emissions entirely came from the applied fertilizers, the fertilizer N loss rates as NO were estimated to be 0.06% to 5.44%, with an average of 0.84 ± 1.39% for all sites (Table 1). Our observed NO loss rates were mainly within the range of 0.01% to 4.0% reported for the agricultural soils over the world (Mei et al., 2009; Stehfest and Bouwman, 2006; Yao et al., 2010). The sites with peak emissions occurring after fertilization showed a consistently high NO loss rates when compared to other sites without distinct peak emissions. There might be some uncertainties in the estimates of NO loss rates due to the lower sampling frequency that might miss some peaks of NO flux. Soybean site G in 2003 was not observed for peak emissions after fertilization, but its NO loss rate was approximately 3 to 20 times higher than grassland site A and wheat site B to which a comparable amount of fertilizer N was applied. Leguminous crops such as soybean can transfer N from the atmosphere to soil, so the NO emissions from site G might partly be from fixed N and accordingly the fertilizer loss rates be exaggerated.

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