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Accumulation of nitrous oxide and depletion of oxygen in seasonally frozen soils in northern Japan – Snow cover manipulation experiments

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ABSTRACT

It has been suggested that soil-thawing and snow-melting are critical triggers for vigorous emissions of nitrous oxide (N_2O) from soils in cold regions. However, because soil freezing is affected by air temperature and snow cover, accurate predictions that estimate subsequent emissions of this important greenhouse gas are difficult to make. In this study, we measured *in situ* soil gas N_2O and oxygen (O_2) concentrations at two experimental sites in northern Japan over the period of a year, from November 2008 to October 2009, to clarify the factors stimulating N_2O production in soil at low temperatures. The sites were N-fertilized bare arable lands with different soil frost depths and snowmelt rates, according to the snow cover management imposed. Winter-to-spring net N_2O fluxes, ranging from −0.10 to 1.95 kg N_2O–N ha^{-1}, were positively correlated with the annual maximum soil frost depth (ranging from 0.03 to 0.41 m; $r = 0.951^{***}$). In the plots with deeper maximum soil frost, winter-to-spring N_2O fluxes represented 58% to 85% of the annual values. Soil N_2O production was stimulated when the soil frost depth was greater than 0.15 m or the daily mean soil temperature at 0.05-m depth was below −2.0 °C. In the soil with the greatest frost depth, soil gas N_2O concentrations at the depth of 0.10 m peaked at 46 ppm when soil gas O_2 concentrations fell down to 0.12 m^3 m^{-3} under soil temperature below 0.0 °C. Snowmelt acceleration had no stimulating effect on N_2O production in the soil during the winter-to-spring period.

1. Introduction

Nitrous oxide (N_2O) is a trace greenhouse gas that contributes to 6% of global warming (Solomon et al., 2007) and is a dominant substance related to the depletion of the stratospheric ozone layer (Ravishankara et al., 2009). The atmospheric N_2O concentration has increased from less than 280 ppb in the pre-industrial era to about 320 ppb in 2006 and is higher in the northern than in the southern hemisphere (Ishijima et al., 2009). In northern high-latitude regions, large N_2O emissions from soils are found not only in the summer (Martikainen et al., 1993; Repo et al., 2009) but also in the winter-to-spring periods. Because N_2O fluxes during winter-to-spring periods sometimes account for more than 50% to 70% of the annual values (Duxbury et al., 1982; Flessa et al., 1995; Röver et al., 1998; Regina et al., 2004; Wagner-Riddle et al., 2007), significant attention has been paid to N_2O production in soil under cold conditions (Phillips, 2008).

The freezing and thawing of soils are important factors related to large N_2O fluxes in cold regions. Soil freezing is primarily affected by air temperature. However, the timing of snowfall and depth of the snowpack greatly affect the soil frost formation as well (Hirota et al., 2002, 2006). Wagner-Riddle et al. (2007) first demonstrated a positive correlation between the cumulative N_2O emissions from November to April and the soil freezing intensity estimated by the accumulated degree-hours below 0 °C at a 0.05 m depth of soil (referred to as $ADH_{0.05}$). They reported that no-tillage management, one of the recommended cultivation practices in Canada, reduced the N_2O emissions as a result of suppressing soil frost formation due to the accumulated snowpack that was entrapped by crop residues on the fields. On the other hand, in northern Japan, removal of the snowpack and an acceleration of the snowmelt are locally conducted as part of the agricultural management from

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winter to spring for the purpose of weed control (Hirota, 2008) and to extend the growing season for crops (Hirota et al., 2008). These management practices can affect soil freezing-thawing dynamics and thus change N_2O fluxes in agricultural lands in cold regions.

Both laboratory incubation (Christensen and Tiedje, 1990; Röver et al., 1998; Sharma et al., 2006; Maljanen et al., 2009) and *in situ* stable isotope tracer studies (Müller et al., 2002; Wagner-Riddle et al., 2008), conducted to clarify the mechanisms of N_2O production in cold soil, have indicated that soil freezing triggers denitrification-derived N_2O production. This production causes the N_2O to accumulate in the surface soil layer. Because denitrification is mediated by facultative anaerobic microorganisms (Zumft, 1997), several studies have speculated the presence of anaerobic microsites in soil under freeze-thaw conditions (Teepe et al., 2001, 2004; Koponen et al., 2004; Öquist et al., 2004; Phillips, 2008). We suggest that a decrease in oxygen (O_2) concentration in soil gas in addition to an increase in the N_2O concentration would support the hypothesis that the soil freeze-thaw conditions enhance denitrification activity in soil. Therefore, we conducted snow cover manipulation experiments in which we created different snow cover, soil frost and snowmelt rate conditions *in situ* and followed simultaneously the changes in the net N_2O fluxes and soil gas concentrations of N_2O and O_2.

2. Materials and methods

2.1. Experimental setup

The two study sites were located at the western and eastern part of Hokkaido prefecture, northern Japan (Supplementary Fig. 1a). Both sites were experimental research fields of the National Agricultural Research Center for Hokkaido Region, National Agriculture and Food Research Organization. The western site (141° 25′ E, 43° 05′ N: "932 field" in the Sapporo site) was characterized by heavy snowfall with shallow soil-freezing, whereas the eastern site (143° 04′ E, 42° 53′ N: "A3 field" in the Memuro site), some 200 km distant, was characterized by light snowfall with deep soil-freezing. The 30-year (1979–2008) mean annual temperature and precipitation for the Sapporo site were 8.8 °C and 1022 mm, respectively, compared to 6.0 °C and 941 mm, respectively, for the Memuro site. As described below, the experimental plots were mostly established in September 2008. At both sites, volcanic ash-derived Andosols were present. The physico-chemical properties of surface soils, from 0 to 0.1 m deep, are shown in Table 1 (see also Supplementary Table 1).

Table 1
Physico-chemical properties of surface soils (0.0–0.1 m deep).

		Sapporo	Memuro
pH (H_2O)	[b]	5.2 (0.1)	5.5 (0.1)
pH (KCl)	[b]	4.3 (0.0)	4.9 (0.1)
Total C	g C kg^{-1} [a]	57.8 (7.6)	36.8 (5.1)
C/N ratio	[b]	15.2 (0.3)	12.2 (0.7)
NO_3^- content	mg N kg^{-1} [a]	28.5 (9.0)	22.8 (3.1)
NH_4^+ content		0.4 (0.5)	2.1 (1.0)
Available P	mg P_2O_5 kg^{-1} [a]	90 (7.8)	58 (24)
Exchangeable K	$cmol_c$ kg^{-1} [a]	0.57 (0.06)	0.33 (0.09)
Exchangeable Mg		1.35 (0.09)	0.54 (0.10)
Exchangeable Ca		11.6 (0.8)	6.1 (1.4)
Cation exchangeable capacity		39.4 (2.2)	21.1 (1.0)
Phosphate sorption coefficient	g P_2O_5 kg^{-1} [a]	1632 (56)	1680 (83)
Bulk density	Mg m^{-3}	0.88 (0.00)	0.74 (0.03)
Texture[c]	[b]	Clay loam	Loam

The standard deviation of three experimental plots (CO, SR, SM) is shown in parentheses except for bulk density, where a standard deviation of three replicates collected from the CO plot are presented.
CO: untreated control, SR: snow removal, SM: snowmelt acceleration.
[a] Oven dry basis.
[b] Dimensionless.
[c] Examined by touch, in detail in Supplementary Table 1.

Three snow cover management treatments (SR, removal of snow cover to enhance soil frost depth; SM, acceleration of snow cover melting; and CO, untreated control) were implemented on three 10 m × 10 to 15 m non-cultivated bare experimental plots for both sites (Supplementary Fig. 1a). Considering that the predominant wind direction during the winter is NNW to NW (Hayashi et al., 2005), these treatment plots were arranged to avoid cross-contamination of snow due to the snowdrift. Snow cover management activities were contingent on the weather conditions from January to March. On SR plots, snow removal was performed seven times at the Sapporo site but only two times at the Memuro site. Snow cover was removed using a combination of heavy machinery and manual shoveling. On SM plots, the snowmelt rate was accelerated by spreading 2 Mg ha^{-1} of charcoal (dark 4 mm-granular, 250 g kg^{-1} of moisture content, 922 g C kg^{-1}) on the snowpack surface, thereby lowering its albedo below 0.3 (Hirota et al., 2008). Charcoal application was performed once on March 9, 2009 (67th day-of-year in 2009: DOY 2009) at both sites because snowmelt had started on March 6 (64 DOY 2009). Although it snowed occasionally after the application of the charcoal in the Sapporo site, we did not perform any supplemental applications. The soil frost depth was determined by either the frost tubes containing 0.03% methylene blue solution or a soil temperature below 0.0 °C. Micrometeorological conditions were measured, including air temperature, precipitation, soil temperature, height of the snowpack and the albedo of the ground surface at each plot throughout the observation period according to the methods as described in Iwata et al. (2008). The ADH (°C h) of soil was calculated as follows:

$$\mathrm{ADH}_z = \left|\sum (t_z \cdot 1)\right|$$

where, z and t, respectively, stand for a given soil depth (m) and soil temperature (hourly mean value, °C) less than or equal to 0.0 °C. Micrometeorological measurements at the Memuro CO and SR plots were established in October 2005 (Iwata et al., in press). A subplot for gas monitoring was added in November 2007 and then rebuilt in September 2008 as described below. For the other plots, all the measurements were newly established in September 2008.

Within each treatment plot, there was a subplot for monitoring gas dynamics (2 m × 2 m; Supplementary Fig. 1b). Undisturbed 1 m × 1 m space was set at the center of the subplot, and a pit (1 m wide, 1.5 m long, 0.6 m deep) was dug at one side of the space. Across the belowground part of the space, horizontal holes (18 mm OD) were bored to insert diffusion chamber type soil gas samplers (Kammann et al., 2001) without disturbing the soil. In order to collect long range-averaged soil gas samples, a 1.0-m-long probe consisting of a silicone tube (10 mm ID; 13 mm OD) was inserted at each depth of 0.10, 0.20, 0.30 and 0.40 m for N_2O concentration measurements, whereas a 0.50-m-long probe was inserted at each depth of 0.10 and 0.30 m for O_2 concentration measurements. To avoid deformation, the silicone tube was covered with a perforated polyvinylchloride (referred to as PVC) pipe (13 mm ID; 16 mm OD with 8-mm OD holes at 20-mm intervals; Supplementary Fig. 1c). One end of the silicone tube was sealed with a silicone rubber septum, and the other was connected to 2-m-long nylon tubing equipped with a two-way cock to allow gas sampling from above ground. The pit was carefully backfilled and, a month later, 144 g of potassium nitrate was applied to the surface of the subplot (equivalent to 50 kg N ha^{-1}) to mimic post-cultivation conditions in local farmlands. Then, a circular stainless steel collar (275 mm OD) with a water groove (20 mm in height) was embedded at the center of the subplot to monitor net soil N_2O emission rates based on the closed-chamber technique.