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Phyton (Austria) Special issue: "Nitrogen emissions"	Vol. 41	Fasc. 3	(49)-(60)	1.11.2001

Source of N_2O Emission from a Soil During Freezing and Thawing

By

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Key words: N_2O flux, soil atmospheric concentration, freezing & thawing, biological production.

Summary

CHANG C. & HAO X. 2001. Source of N_2O emission from a soil during freezing and thawing. - Phyton (Horn, Austria) 41 (3): (49) - (60).

Agricultural soils are an important source of N_2O. These soils frequently emit large amounts of N_2O even when soil temperatures are near freezing, but the source of N_2O and the emission during thawing events are not clear. Our objective was to identify the source of N_2O in soil under field conditions during 'Chinook' (thawing) events in winter months. Nitrous oxide fluxes from agriculture soils were measured in three winters. Nitrous oxide, CO_2 and O_2 concentrations of the soil atmosphere, N_2O fluxes, air temperature and precipitation were measured periodically throughout the winter months. Soil atmospheric N_2O concentrations, especially at 7.5 to 45 cm depths, increased during the Chinook events in the winter of 1995-1996 when air temperatures increased above freezing. The accumulation of N_2O in soil was attributed to the biological activity because: i) the high concentration of soil atmospheric N_2O persisted for significant period time, ii) N_2O and CO_2 concentration increased simultaneously; iii) the soil temperature and moisture content changes had small effect on soil atmospheric N_2O concentration, physical release of dissolved N_2O upon temperature increase would have been small; and iv) the concentration of N_2O generally peaked at depths of 7.5 to 45 cm, so that the N_2O there could not have been diffused from lower depths. N_2O flux from soil to atmosphere during the Chinook events was highly correlated with N_2O concentration in the soil atmosphere near the surface. Our findings support the hypothesis that biological processes contribute to significant amount of N_2O emissions during thaw events, even when temperatures are at or below freezing point.

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Introduction

Agricultural soils are an important source of N_2O (MOSIER & al. 1998) largely through two microbial-mediated processes: nitrification and denitrification. These soils frequently emit large amounts of N_2O even when soil temperatures are near freezing. In northern Germany, KAISER & al. 1998 found that up to 46 % of annual N_2O emissions from agricultural soils occurred during the winter months. In Iowa, BREMNER & al. 1980 observed that 6 to 21 % of the annual N_2O emission occurred during spring thaw. Numerous other studies, in a wide range of conditions, have confirmed that a large proportion of annual N_2O emissions can occur during spring thaw (e.g., GOODROAD & KEENEY 1984, GOODROAD & al. 1984, CHANG & al. 1998, LEMKE & al. 1998a,b).

CHRISTENSEN & TIEDJE 1990 attributed the brief and vigorous N_2O release during spring thaw to biological production. ROVER & al. 1998, comparing N_2O emission from sterilized and a non-sterile soils exposing to freeze-thaw cycles, showed much higher N_2O emission from non-sterile soils than that from sterilized soils in a laboratory study. They concluded that the source of N_2O was from microbial N_2O production even in frozen soils under thawing conditions. High concentrations of N_2O already occur at depth before spring thaw were also reported (EGGINTON & SMITH 1986, CATES & KEENEY 1987) such that emissions may simply reflect physical release of this accumulated N_2O. GOODROAD & al. 1984 speculated that N_2O emission during thawing came from several sources: N_2O diffusing from sub-surface soil, physical release of N_2O from surface soil, as well as biological production of N_2O.

The source for the high rates of N_2O emission at low soil temperature is still unclear. Does the N_2O emissions during thawing period originate from recent biological activity, or is it simply a physical release of trapped N_2O produced in earlier the season? Southern Alberta is a unique environment to study N_2O emissions during freeze-thaw events. This area has frequent 'Chinook' events, typically 4 to 6 per year. Associated a Chinook event, temperature may increase by 22°C in ten minutes. This creates an ideal condition for studying the effects of freezing and thawing on N_2O emission. This paper summarizes three N_2O emission studies at Lethbridge Research Centre, Lethbridge, Alberta Canada to identify the source of N_2O and emission during freezing and thawing cycles. Two studies were conducted on a site with long-term manure application (CHANG & al. 1998) and one study was on a site with long-term straw management (HAO & al. accepted).

Materials and Methods

Field site description

Both experimental sites were adjacent to each other and at the Agriculture and Agri-Food Canada Research Centre in Lethbridge, Canada (SOMMERFELDT & CHANG 1985, CAREFOOT & al. 1994). The first site was located at the long-term manure application site. Solid cattle feedlot manure has been applied annually since 1973 at four rates to a well-drained Dark Brown Chernozemic (Typic Haploboroll). The applications rates (0, 60, 120, and 180 Mg ha^{-1} y^{-1}, wet weight) are equivalent to zero, one, two, and three times the recommended maximum rates (ALBERTA AGRICULTURE

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1980). The 2nd site was located at a long-term straw management site and the details on the experimental design were reported by CAREFOOT & al. 1994. Briefly, the long-term irrigated wheat-wheat-oats rotation study was initiated in 1986. The factorial experiment included with five straw-tillage treatments, four N fertilizer (NH_4NO_3) rates (0, 50, 100 and 200 kg N ha^{-1}) applied either in spring or fall using a randomized complete block design with 4 replications. For this study, N_2O emissions were collected from four treatments: no fertilizer + no crop residue + fall tillage (T1), 100 kg N ha^{-1} fertilizer in fall + no crop residue + fall tillage (T4), 100 kg N ha^{-1} in fall + no crop residue + spring tillage (T6) and 100 kg N ha^{-1} in fall + 5000 kg straw ha^{-1} + fall tillage (T7).

In the fall, soil nitrate content of manured soil (after 20 year annual application) was high (CHANG & al. submitted). The high nitrate content of the manured soil created a potential condition for denitrification. At the long-term straw management experimental site, CAREFOOT & al. 1994 reported that the nitrate content of the surface 60 cm depth of soil was higher in fall N-fertilizer application at 100 kg N ha^{-1} than in without N fertilizer application. The high nitrate concentrations would favour N_2O over N_2 as an end product of denitrification (KHDYER & CHO 1983).

In the manured plots, CHANG & al. (submitted) showed that the changes in soil temperature were smaller than changes in air temperature. During winter months, soil temperature normally increased with depth. However, during Chinook events, the surface temperature increased and was briefly higher than the depth below.

Long-term experimental sites were under irrigation, the soil water content of the surface soil was low after crop harvest and the surface soil moisture was recharged during the thawing periods. The surface soil moisture was relatively high and maintained at a fairly constant level throughout the 2nd half of the winter (CHANG & al. submitted). The crop at the long-term manure application site (1993/94 and 1995/96) was barley (*Hordeum vulgare* L. 'Galt') and at the long-term irrigated wheat-wheat-oats rotation site (1996/97) was white soft spring wheat (*Triticum aestivum*). However, all the N_2O emissions were measured after the crops were harvested.

N_2O flux measurement

Nitrous oxide fluxes from soil were measured using a vented chamber technique (HUTCHINSON & MOSIER 1981), modified to permit separation of the chamber from the base. The volume of the chamber was 1604 cm^{-3} (9.2 cm H x 14.9 cm ID) and the cross-sectional area was 174 cm^{-2}. Measurements of N_2O flux were initiated in November at various time intervals until next April when the air temperature was consistently above zero.

A chamber base (collar) was installed in each of 4 replicated plots to a depth of about 3 cm. On each sampling date, the chamber was attached to the collar, and 5 mL of air was drawn from the headspace with a syringe after 0, 5, 10, 15, and 30 min. The air samples were taken to the laboratory and N_2O was analyzed immediately using a gas chromatograph as described previously. Fluxes were calculated from the N_2O concentrations in the headspace by assuming a steady state gradient of N_2O in the underlying soil (ANTHONY & al. 1995, HUTCHINSON & LIVINGSTON 1993). The N_2O concentration versus time relationships was fitted with second order polynomial equations for each sampling date, and the flux of N_2O at time 0 at each sampling time was calculated from the derivatives of the equations. The flux was corrected to the field temperature using the ideal gas state law (PV=nRT, where P is the air pressure, V is the ideal gas volume, n is the molar concentration of N_2O gas, R is the gas constant, and T is the temperature at the time when gas sample was taken). The flux was then converted to kg N ha^{-1} d^{-1}.

Soil atmospheric measurements

The concentrations of N_2O, CO_2, O_2 and N_2 in the soil atmosphere at 0, 7.5, 22.5, 45, 75, 105 and 135 cm depths were measured only in 1995-96 (November 1995 to April 1996) using a multilevel sampling probe installed in each of 4 replicated plots (BURTON & BEAUCHAMP 1994).

Analytical measurements of N_2O

N_2O was analyzed using a GC equipped with an ECD (Varian 3600) and CO_2 and O_2 were determined on a GC equipped with a concentric column (inner column packed with chromosorb 101

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for CO_2 and outer column packed with active molecular sieve 13 for O_2) and a thermal conductivity detector (Varian 3400).

N_2O solubility and partition coefficient for soil atmosphere and water

CHANG & al. (submitted) used regression analysis based on published data (MORAGHAN & BURESH 1977, HANDBOOK OF CHEMISTRY AND PHYSICS 1955) to define the ratio of N_2O in water to that in air as a function of temperature and volume (water:atmosphere):

$$y_w = 7.70 \times 10^{-2} - 1.148 \times 10^{-2}\, T + 1.13\, R + 3.279 \times 10^{-4}\, T^2 + 1.157 \times 10^{-3}\, R^2 - 2.133 \times 10^{-2}\, T \times R \qquad [1]$$

$r^2 = 0.9984$

where: y_w = ratio of N_2O in water to N_2O in air

T = temperature (°C)

R = volume ratio (water : soil atmosphere)

Based on the assumption that each soil interval is a closed system, we could calculate the net release of N_2O from soil water in response to changes in soil temperature and moisture content from equation 1.

Results and Discussion

Freezing and thawing cycles

Several freezing and thawing cycles occurred during each of three winters (Fig. 1). The frequency and duration of the cycles varied from year to year. The duration of the cycle could last from 1 to 18 days. Temperature changed very quickly at the onset of a freezing and thawing (Chinook) event. For example, air temperature increased by 33°C (from -29 to 4°C) within two days, then fell by 27°C (from 11 to -16°C) two days later in January 1996 (Fig. 1b). In contrast, the temperature changes were more gradual in early fall and late spring. Total precipitation (most in snow) for the winter months was 70 mm for 1993-94, 137 mm for 1995-96 and 111 mm for 1996-97 and winter of 1995-96 was the wettest of the three winters. Daily snow depth varied from 0 to as high as 226 mm (1996/97). The average snow cover depths were 16, 62 and 41 mm for each winter respectively.

N_2O flux from soils

The rates of N_2O emission were a function of manure rates (Fig. 1a, 1b) and tillage, straw and fertilizer treatments (Fig. 1c). N_2O fluxes increased during the thawing periods, then decreased again as the temperature fell below freezing (Fig. 1). Nitrous oxide fluxes during the 1st half of winter period were smaller than that during the 2nd half. This could be due to higher soil moisture content in the 2nd half (CHANG & al submitted). The N_2O flux was sustained for the duration of the warm period, suggesting that the N_2O emission during thawing be from biological processes rather than from a sudden physical release.

The timing of peak emission of N_2O varied over the three winters. However, the peak emission generally occurred during the spring thaw. The N_2O fluxes were measured at same site for 1993-94 and 1995-96. However, the daily N_2O fluxes for those two winters were similar even though, the precipitation was higher in 1995-96 than in 1993-94. Nitrous oxide fluxes from the straw management site

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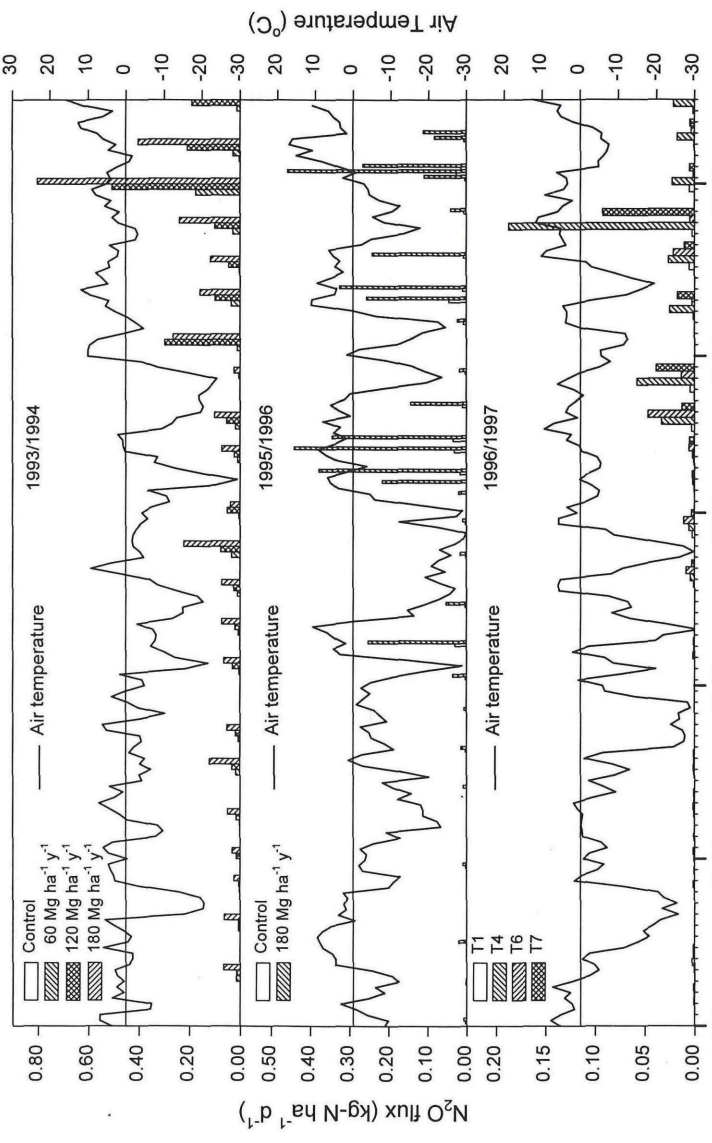

Fig. 1. Air temperature and N_2O flux from (a) manured soil from November 1, 1993 to April 15, 1994, (b) manured soil from November 1, 1995 to April 15, 1996 and (c) straw management site from November 1996 to April 15, 1997.

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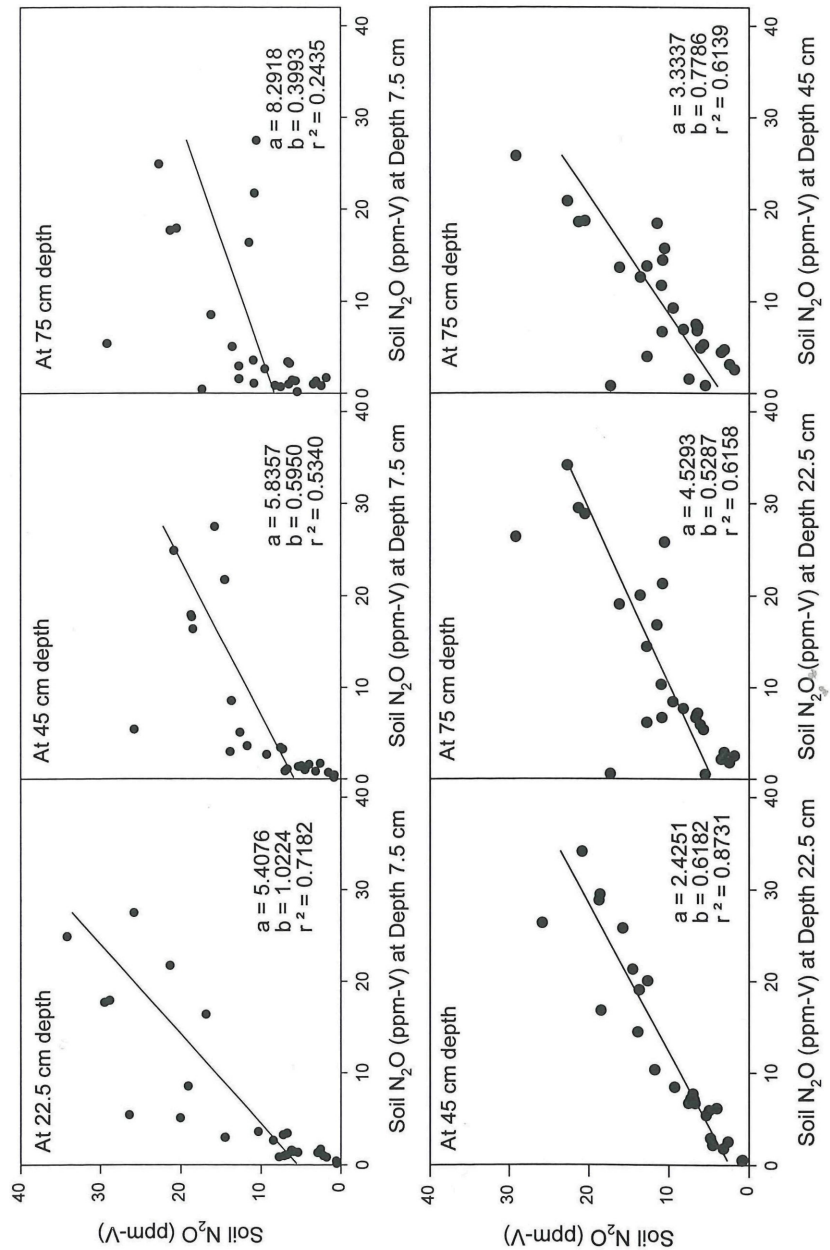

Fig. 2. Correlation among soil atmospheric N_2O at depth 7.5, 22.5 and 45 cm.

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could be not statistically compared to that from manured treated soil. However, the patterns of N_2O emission from straw management site with fall N-fertilizer treated plots regardless straw and tillage management during the freezing and thaw cycles were similar to that from manure treatment site.

Soil atmospheric N_2O concentration profile

CHANG & al. (submitted) reported that the N_2O concentration in the soil atmosphere was highly variable, depending on the sampling times and depths. The coefficient of variation for the four replicates, at a given time and depth, ranged from 1 to 197 %. The N_2O concentrations at depth 7.5, 22.5, 45 and 75 cm depths were positively correlated with each other indicating that N_2O concentrations at these depths increased or decreased simultaneously in proportion at the same time (Fig. 2). The N_2O concentrations at 22.5 cm were often the highest and declined toward the surface, reflecting diffusion and emission of N_2O to the atmosphere. Although Concentration of N_2O in soil atmosphere was very responsive to the air temperature, especially at the 22.5 cm depth, increased sharply during thawing at surface soil and the concentration stayed high through the thawing period (Fig. 3). Although the soil temperatures at 22.5 to 75 cm depths were still below zero (unpublished data).

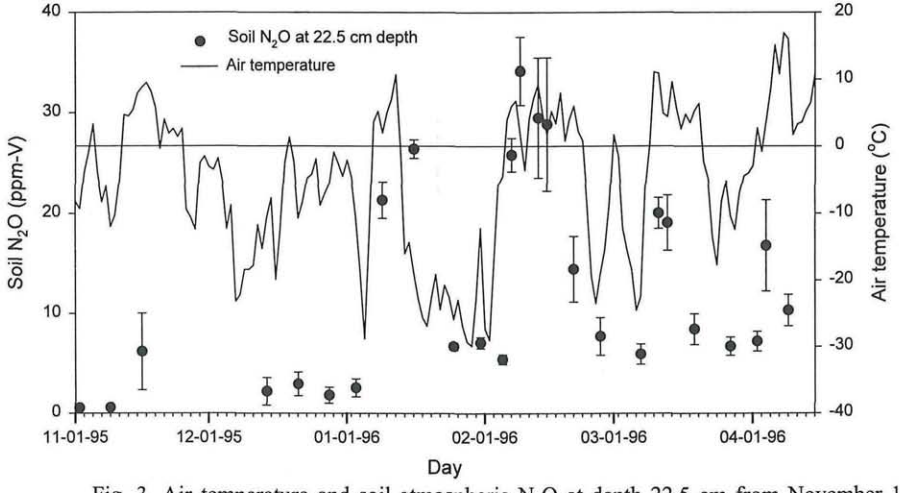

Fig. 3. Air temperature and soil atmospheric N_2O at depth 22.5 cm from November 1, 1995 to April 15, 1996 and the error bar representing standard error of mean of 4 replicates.

Concentrations of N_2O and CO_2 in soil atmosphere were highly correlated (Fig. 4). The N_2O concentration increases as a power function of CO_2 concentration (Fig. 4). The increase in CO_2 was the result of increase in microbiological activities as the temperature increased. This suggesting that the elevated N_2O concentrations were at least, partially derived from biological activity. There was no apparent relationship between N_2O concentration and soil moisture content or air filled porosity,

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suggesting that aeration was a net result of O_2 diffusion and consumption. Although the liquid water content of surface soil increased during thawing, O_2 in the soil profile was consumed by microorganisms and O_2 diffusion into the soil profile was reduced, resulting in anaerobic conditions. Therefore, the N_2O emission in the field may not always directly relate to soil moisture or air filled porosity.

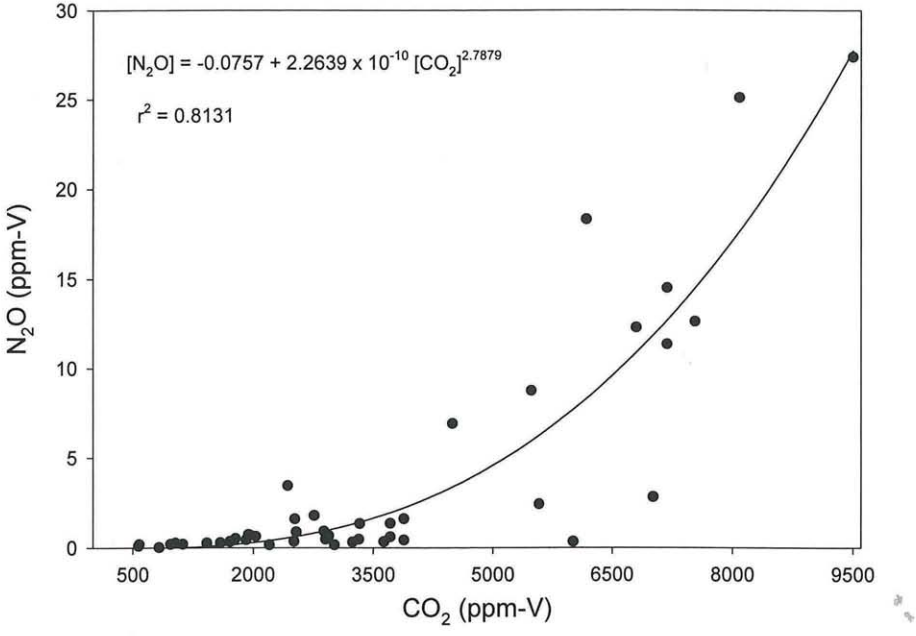

Fig. 4. Relation between soil atmospheric N_2O content and soil atmospheric CO_2 content at 22.5 and 45 cm depths.

Source of soil atmospheric N_2O during thawing

Nitrous oxide is highly soluble in water. Dissolved N_2O could be released during thawing as the soil solution temperature increases. Therefore, the emission N_2O during thawing could be attributed to the physical release of N_2O rather than biological production of N_2O. It also suggests that the N_2O produced at deeper and warmer soil zones and diffused to upper soil profile during the thawing. These processes might contribute part of total N_2O emission. However, the emission of N_2O from all three studies indicated a substantial amount of N_2O emission during the thawing period but dropped off very quickly at the onset of freezing cycle. This suggested that the increasing N_2O in the soil atmosphere was most likely due to production rather than physical release. Further more, if we assume that Total-N_2O = 30 ppm-V in a unit soil volume and the soil porosity is 0.5 with 60 % filled by soil moisture (0.30 m^3m^{-3} volumetric water content), then the concentration of N_2O in the soil pore space will be 60 ppm-V since only half of soil volume is occupied by air and water. At 0.3 m^3 m^{-3} water content, the water:air ratio (R) = 1.5 and y_w =

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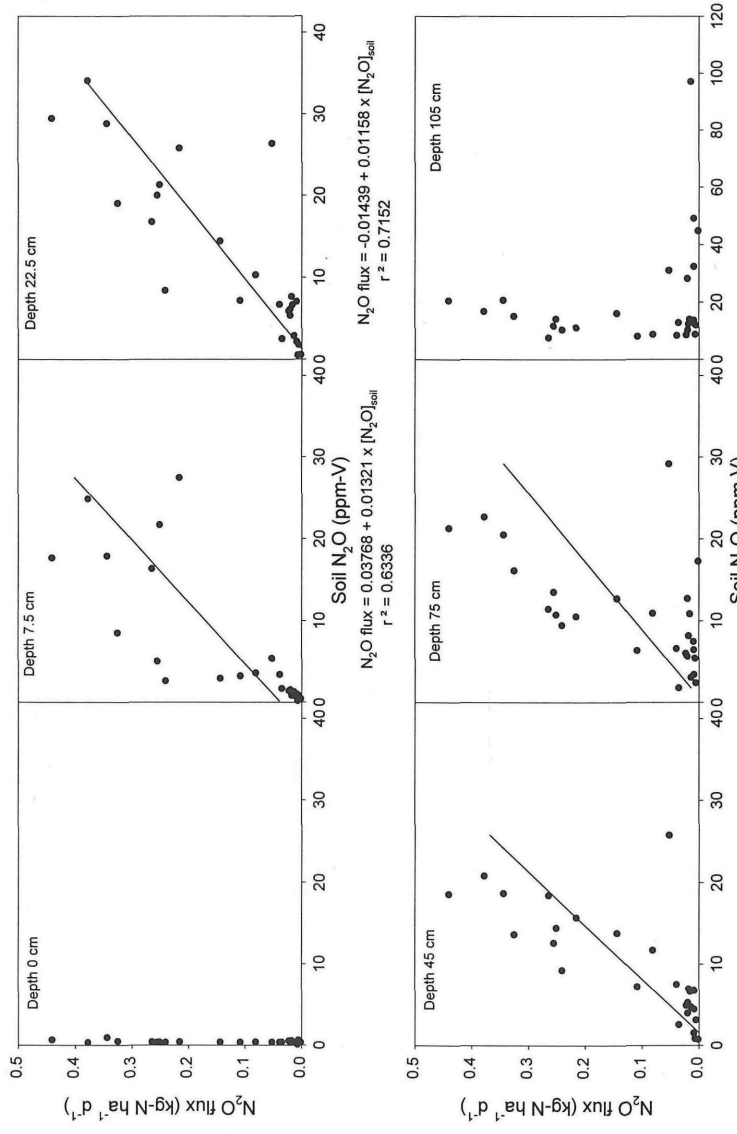

Fig. 5. N_2O flux from November 2, 1995 to April 9, 1996 and (b) N_2O flux vs soil atmospheric N_2O concentration at depths (a) 0, (b) 7.5, (c) 22.5, (d) 45, (e) 75 and (f) 105 cm

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1.73 at T = 1°C (equation [1]). The N_2O concentration in the air-filled pore space is then 54.9 ppm-V. If temperature increases to 5°C, then y_w = 1.57 and Air-N_2O = 58.4 ppm-V (equation [1]). If soil water content is 40 % by volumetric (80 % of pores filled with water, R=4)) and at 1°C, the N_2O concentration in the air pore is 27.2 ppm-V. The N_2O concentration in soil atmosphere will increase from 54.9 ppm-V at 1°C to 58.4 ppm-V at 5°C and decrease from 58.4 ppm-V at 30% moisture content to 29.2 ppm-V at 40% moisture content at 1°C. These two hypothetical examples demonstrate the effects of soil moisture and soil temperature changes on on the redistribution of N_2O from the liquid phase to the gaseous phase. Without N_2O production, the effect of temperature changes on redistribution of N_2O was small and increasing soil moisture decreased N_2O concentration in gas phase.

Furthermore, because concentrations at 7.5 to 45 cm depths were usually higher than other depths in the profile, the appearance of N_2O could not be explained by diffusion from other depth. Consequently, the elevated concentrations of N_2O at those depths are probably caused by biological production.

Relationship between N_2O flux and soil atmospheric N_2O

The N_2O flux from soil surface to the atmosphere was highly correlated to the N_2O concentration in the soil atmosphere at 7.5, 22.5 or 45 cm (Fig 5). The intercept of the regression equation was not significantly different from zero, suggesting that N_2O flux becomes negligible as N_2O concentration in the soil atmosphere at these depths approaches zero. The soil atmospheric N_2O concentration below 105 cm depth had no influence on N_2O flux at the soil surface. Nearly 63 and 72 % of N_2O flux variation could be explained by the soil atmospheric N_2O concentration at 7.5 and 22.5 cm depths, respectively. The results suggested that N_2O produced at these depths and diffused upward to the atmosphere.

Conclusions

Nitrous oxide emissions were observed during the thawing periods of three winters. When soil NO_3^- (OR nitrate) was not a limiting factor, soil atmospheric N_2O increased during the thaw periods in the winter of 1995-1996 as air temperatures increased to above the freezing. N_2O concentration in the soil atmosphere increased rapidly, especially at 7.5 to 45 cm depths. The increases were attributed to the biological activity because: i) the high concentration of soil atmospheric N_2O persisted for a significant period time, ii) N_2O and CO_2 concentrations increased simultaneously; iii) the soil temperature and moisture content changes had no effect on soil atmospheric N_2O concentration, the physical release of dissolved N_2O upon temperature increase would have been small; and iv) the concentration of N_2O generally peaked at depths of 7.5 to 45 cm, so that the N_2O there could not have been diffused from lower depths. N_2O flux was highly correlated with N_2O concentration in the soil atmosphere near the surface during Chinook events. Our findings support the hypothesis that biological processes can contribute to significant

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amount of N_2O emissions during thawing, even when temperatures are at or below freezing point.

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Zeitschrift/Journal: Phyton, Annales Rei Botanicae, Horn

Jahr/Year: 2001

Band/Volume: 41_3

Autor(en)/Author(s): Chang C., Hao X.

Artikel/Article: Source of N2O Emission from a Soil During Freezing and Thawing . 49-60