Thick ice layers in snow and frozen soil affecting gas emissions from agricultural soils during winter

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Abstract. We investigated soil and snow cover gas concentrations at two agricultural sites (St-Lambert; Chapais) in Quebec, Canada, during winter 1998–1999. Both sites showed frozen and unfrozen soils and complex snow cover structure. At St-Lambert we measured higher average concentrations of N_2O (35 to 62 μL L^{-1}) and CO_2 (3 to 19 mL L^{-1}) below the frozen soil surface of plots subjected to a treatment of pig slurry than in the control plot (N_2O, 9 to 30 μL L^{-1}; CO_2, 3 to 7.5 mL L^{-1}). The lack of vertical gaseous concentration gradients in the snowpack was due to the trapping of accumulating gas below the impermeable frozen soil layer. Soil gas concentrations decreased sharply when soil warmed to the freezing point. At the same time, the snow cover was isothermal. N_2O could have been lost at spring thaw through gaseous emissions and/or dissolved in meltwaters and leached to the drainage system. High N_2O fluxes were measured using closed chambers (215 ng m^{-2} s^{-1}, slurry treatment; 55 ng m^{-2} s^{-1}, control) as soon as snow ablation was completed, but became negligible 2 days later, suggesting that emissions were the result of passive degassing rather than of increased biological activity. At Chapais, N_2O and CO_2 accumulated in the unfrozen soil surface below a thick (0.1 m) basal ice layer. The basal ice layer and the continuous ice layer above it were impermeable to gas diffusion, as demonstrated by the accumulation of a tracer gas (Ar, >50 mL L^{-1}) introduced by a diffuser into the soil. The existence of a basal ice layer is uncommon in eastern Canada. The occurrence of such a phenomenon may increase with climate change due to more frequent rain events during the cold season and affect the dynamics of winter gas emissions from soils.

1. Introduction

Soils in a wide range of ecosystems around the world are sources or sinks of three of the main greenhouse gases: nitrous oxide (N_2O), carbon dioxide (CO_2), and methane (CH_4) [Bouwman, 1990]. Cultivated soils are thought to be a major source of anthropogenic N_2O [Duxbury et al., 1993], which has a greenhouse warming potential 310 times greater than CO_2 and contributes to the depletion of ozone from the stratosphere [Crutzen, 1970]. The amount of N_2O transferred from agricultural soils to the atmosphere constitutes a significant part of the annual N budget and affects the energy balance of agriculture through losses of fossil fuel from N synthetic fertilizer production [Janzen et al., 1999]. Climate and agricultural practices are responsible for variations in soil environmental parameters, which have a large impact on the magnitude of N_2O emissions [van Bochove et al., 2000; Wagner-Riddle and Thurtell, 1998]. Several winter studies of natural ecosystems at both high latitudes and high altitudes have highlighted the physical role of snow cover in gas emissions from soil to the

atmosphere [Sommerfeld et al., 1993, 1996; Hardy et al., 1995; Brooks et al., 1996; Winston et al., 1997; Oechel et al., 1997; Mast et al., 1998]. In cold temperate climates of Canada and northern Europe, significant N_2O losses have been reported from cultivated soils following freeze-thaw cycles in spring [Nyborg et al., 1997; Wagner-Riddle and Thurtell, 1998; Röver et al., 1998] or from unfrozen soils during winter and snowmelt [van Bochove et al., 1996, 2000].

It is known that microbiological denitrification is active at temperature as low as −2°C [Dorland and Beauchamp, 1991] and significant denitrification potential occurs at 2°C during the coldest months of the year [Pelletier et al., 1999]. However, gaseous emissions are highly variable [Jones et al., 1999], and the processes for gas production and transport are difficult to study and remain poorly understood. The exchange of gas between the snow-covered soil and the atmosphere may be subjected to physical constraints due to conditions at the soil-snow interface and the structure of the snow cover. Of particular significance are basal ice layers, which form at the soil-snow interface. These ice layers are generally thicker than ice lenses within the snow cover, and their permeability is relatively low [Marsh and Woo, 1984].

In Canada, wintertime conditions lead to the frequent freez-

ing of surface soils due to low temperatures and thin snow cover on the midcontinental prairies [Pomeroy and Brun, 2001]. Basal ice layers can also form on the frozen soil layer during melt and/or rain-on-snow episodes. In the Canadian subarctic, basal ice is a common characteristic of snow-soil systems [Marsh and Woo, 1984]. Frozen soil and/or basal ice formation is not frequent in much of eastern Canada as the snow cover is relatively deep and insulates the soil from subzero temperatures [van Bochove et al., 2000]. Exceptions occur in sites which experience high rates of snow erosion by wind leading to thin snow cover and where thermal losses to the atmosphere are high. However, eastern Canadian snowpacks are sensitive to the formation of ice lenses and crusts within the snow cover due to frequent melts and rain-on-snow events.

During a study of soil gas emissions during the winter of 1998–1999 at two agricultural sites in eastern Canada, we encountered soil and snow-structural characteristics that were representative of frozen surface soils, in-pack formation of ice lenses, and the formation of basal ice layers. The experimental protocol permitted the distinction between the effects that these different conditions had on gas emissions. This paper reports the results of that study.

2. Methods

2.1. Study Sites

Two experimental sites approximately 20 km apart were instrumented during the fall of 1998 to cover the sampling period of winter and spring 1998–1999. Both sites were agricultural fields and had initially different experimental designs because each one was part of a distinct research project.

The first field site was located on the experimental farm of the Institut de Recherche et de Développement en Agroenvironnement (IRDA) at Saint-Lambert-de-Lévis, Quebec, Canada (46°36′N, 71°10′W). The soil was a loam, Le Bras series (frigid Aeric Haplaquept) with 0.31 g of sand, 0.42 g of silt, and 0.27 g of clay per g of soil. Plot treatment consisted of a fall application of pig slurry at 60 Mg ha^{-1} repeated three times in randomized blocks; control plots were not treated. The slurry was from a commercial hog and sow operation and contained 15.8 kg m^{-3} dry matter, 2.52 kg m^{-3} total N, 2.03 kg m^{-3} NH_4^+-N, 0 kg m^{-3} NO_3^--N, and 5.06 kg m^{-3} C. The second field site was located on the Chapais Research Farm of Agriculture and Agri-Food Canada at Lévis, Quebec, Canada (46°46′N, 71°12′W). The soil type was a sandy loam, St-Pacome series (Umbric Dystrochrept) [Gagné, 1985]. Barley (Hordeum vulgare L.) was grown under conventional tillage during summer 1998 and fertilized with ammonium nitrate during seeding at a rate of 70 kg N ha^{-1}. Straw residues were left on the ground after crop harvest in September.

2.2. Experimental Plots and Gas Sampling

2.2.1. Saint-Lambert site.
Three multilevel gas sampling probes were installed per plot. The probes [van Bochove et al., 2000] consisted of a 1.8 m long polyvinyl chloride tubing (watering microtubes; ID, 0.127 cm; Chapin Watermatics Inc., New York) topped with a double-ended needle (18G, Vacutainer Brand, Becton Dickinson and Co, Rutherford, New Jersey) and inserted at different levels in a PVC pipe (length, 2 m; OD, 5.1 cm). The PVC pipes were installed vertically to a depth of 1 m in the soil through a soil auger hole. Gases were sampled from the soil at 0, 10, 35, and 70 cm below ground level and from within the snowpack at 5, 10, 30, and 50 cm

above the soil, depending on snow cover depth. The sampling dates were February 2 and 26; March 12 and 29; April 6, 12, 19, and 22.

2.2.2. Chapais site.
Two diffusion plots were chosen in the field. The diffusion plot method and theory are described by van Bochove et al. [1998]. The diffusion plot consisted of a gas diffusion system composed of a mass flowmeter and controller and a diffusion chamber. An argon (Ar) gas standard bottle (UHP Argon, regulated at 6.9 × 10^4 Pa outlet pressure) was connected to an electronic mass flowmeter and controller (EMF) (FMA761, Omega, Stamford, Connecticut). The EMF employs thermodynamic principles to measure the true mass flow, without compensation for environmental pressure and temperature. The outlet of the EMF was set at a pressure of 0 kPa to maintain a constant diffusive flow of gas through the system. The EMF was connected to a sealed copper diffusion chamber (OD, 20 cm; thickness, 4.5 cm) by a tygon tube (length, 1.6 m). The chamber was welded to nine pipes of equal length (ID, 1.9 cm; length, 20 cm) radiating outward from the chamber over an angle of 100°. Diffusion holes (ID, 0.28 cm; spacing, 0.5 cm) were made all along the upper side of the pipes. The diffusion system was designed to provide a gas diffusion as uniform as possible through a soil area of 0.485 m^2 (91.5 × 53 cm). The diffusion chamber was placed horizontally 40 cm below the soil surface in a trench next to the soil profile. The perimeter surrounding the area of diffusion was delimited and sealed from the surface down to a 50-cm depth by a polythene membrane and by a plate below the diffusion system to obtain a one-dimensional flow regime. The artificial Ar flow rate was kept constant (2 mL min^{-1}) by the EMF. The artificial flux level of Ar was adjusted within the validation limit of a diffusive process model [van Bochove et al., 1998]. In order to obtain the same one-dimensional flow regime into the snow cover above the diffusion plot, a modular enclosure made of 10 rectangular rings (100 × 60 × 15 cm) was assembled or disassembled to follow the surface of the snow cover through accumulation and ablation periods.

To sample soil atmosphere, soil gas sampling probes were inserted in copper pipes (ID, 0.635 cm) in triplicate at four depths (5, 15, 25, and 35 cm) through the polythene membrane. Each probe consisted of the polyvinyl chloride tubing (length, 1.8 m) topped with a double-ended needle (18G, Vacutainer Brand, Becton Dickinson and Co, Rutherford, New Jersey).

To sample snow atmosphere, snow gas sampling probes were inserted in copper pipes (ID, 0.635 cm) with six repetitions at five heights above soil surface (5, 10, 30, 60, and 70 cm). Copper pipes were inserted through a wooden tutor set inside the modular enclosure above the plots. Each probe consisted of the same polyvinyl chloride tubing as described above which was topped with a double-ended needle (22G, Vacutainer Brand, Becton Dickinson and Co, Rutherford, New Jersey).

At sampling time (February 24 and 25; March 2, 10, 11, 15, 23, and 30; April 9, 12, 20, and 27; May 21) the probes of both experimental sites were individually purged with a syringe. The purged volume corresponded to the in-line volume of each probe. The samples were then drawn directly from the probes into evacuated vials (7.5 mL).

2.3. Gas Analysis

The N_2O and CO_2 concentrations were analyzed by gas chromatography (Hewlett Packard 5890 series II, Wilmington,

Delaware) equipped with an automatic sample injector system (CTC Analytics, Zwingen, Switzerland). The operating setup and conditions for CO_2 were as follows: a Porapak Q column (3.66 m by 0.32 cm ID) used with N_2 as carrier gas (20 mL min^{-1}), oven temperature at 50°C, injection port temperature at 250°C, and CO_2 was reduced to CH_4 in a Ni catalyst tube (375°C) coupled to the flame ionization detector (FID, 250°C). The operating setup and conditions for N_2O were as follows: a Porapak Q precolumn (0.91 m by 0.32 cm ID) coupled to an analytical Porapak Q column (1.83 m by 0.32 cm ID) through a switching valve system, Ar/CH_4 as carrier gas (0.95:0.05 m^3 m^{-3}, 25 mL min^{-1}), oven temperature at 50°C, injection port temperature at 250°C, and an electron capture detector (ECD, 250°C). The detection limit and precision were 0.077 and 0.016 mL L^{-1}, respectively, for CO_2 and 0.070 and 0.014 μL L^{-1} for N_2O.

The Ar and O_2 concentrations were analyzed by gas chromatography (Hewlett Packard 5890, Wilmington, Delaware). The operating conditions for Ar and O_2 were as follows: a Hayesep A column (11 m by 0.32 cm ID), oven temperature maintained at 20°C with a cryogenic valve device, injection port temperature at 165°C, carrier gas He (30 mL min^{-1}), and a thermal conductivity detector (TCD, 110°C). The detection limit and precision were 0.49×10^{-2} and 0.12×10^{-2} L L^{-1}, respectively, for argon, and 1.11×10^{-2} and 0.27×10^{-2} L L^{-1} for oxygen.

2.4. Flux Measurements

Soil-surface N_2O fluxes during the snow-free season were measured (April 15, 19, 22, and 29; May 6 and 13) by the static chamber method detailed by *Lessard et al.* [1994] and briefly described as follows. Two acrylic frames (0.60 × 0.60 m; height, 0.14 m; wall thickness, 6.35 mm) were inserted to a depth of 0.10 m in each plot on October 7, 1998. The frames completely covered the width of the slurry application band between maize rows. They were left at the same locations for the duration of the experiment. The height of the frames was measured (48 measuring points per frame) at regular intervals during the experiment to account for variations in headspace due to soil settling. At sampling time the frames were covered with a lid, and air samples were taken through a rubber septum at regular intervals (0, 10, 20, and 30 min) with 10 mL Plastipack syringes (Becton Dickinson and Co., Rutherford, New Jersey). Flux measurements were always made between 0900 and 1200 LT.

Soil-surface N_2O fluxes F_{N2O} were calculated using the following equation [*Hutchinson and Livingston*, 1993]:

$$F_{N_2O} = \frac{dC}{dt} \frac{VM_{mol}}{AV_{mol}},$$

where dC/dt is the rate of change of N_2O concentration (mmol mol^{-1} s^{-1}), V is the chamber headspace volume, M_{mol} is the molecular weight of N_2O (44 g mol^{-1}), A is the surface area covered by the chamber (0.36 m^2), and V_{mol} is the volume of a mole of gas at 20°C and 1 atm (0.024 m^3 mol^{-1}). Nitrous oxide concentrations in closed chambers varied linearly with time in 82% of cases. For these observations the slope (dC/dt) obtained from simple linear regression between concentration and time was used to calculate the fluxes. The relationship between concentration and time was tested with the Student-t test for $n = 4$ and $\alpha = 0.10$ [*Hutchinson and Livingston*, 1993]. Second-order polynomial equations were fitted to con-

centrations when the rate of change decreased with time, and the Student-t test was applied to the initial slope of the curve (time equal to 0). In both cases, if the calculated t was greater than the critical value of t, the rate of change in concentrations was considered significantly different from zero.

2.5. Snowpack Properties

Average snow depth H (m) was measured in each plot from 10 stakes. The snow water equivalent SWE (mm) of the snowpack was determined by the use of a snow corer (Adirondack corer, Gamma Instrument Company, Hempstead, New York). The bulk density d (kg m^{-3}) for any core was then calculated from the SWE (mm or kg m^{-2}) and the core height of each sample H' (m) as SWE/H'. The bulk porosity of the snowpack (ϕ) was calculated from the expression:

$$\phi = 1 - (d/d_i),$$

where d_i is the ice density (917 kg m^{-3}).

2.6. Soil Temperatures and Volumetric Water Contents at Saint-Lambert

Soil temperatures were monitored with copper-constantan thermocouples (Type T, Omega, Stamford, Connecticut) at 7.5 and 15 cm depths. Soil water content was measured for the top 15 cm using time domain reflectometry probes (TDR). Both parameters were monitored inside the experimental plots. A data logger (CR-10, Campbell Scientific, Logan) recorded temperatures on an hourly basis that were afterward converted into daily mean values.

2.7. Soil and Snow Temperatures and Meteorology at Chapais

Soil temperatures were monitored with copper-constantan thermocouples (105T-L, Campbell Scientific, Logan) at 1, 5, and 25 cm depths. Water content reflectometry probes (CS615, Campbell Scientific, Logan) were installed horizontally at 2, 10, and 20 cm depths. Both parameters were monitored inside the area of diffusion. A data logger (CR-10, Campbell Scientific, Logan) recorded on an hourly basis the soil volumetric water content and temperature that were converted into daily mean values.

Snow temperatures were measured with an array of fine Type E hypodermic needle thermocouples (Omega, Stamford, Connecticut) controlled by a Campbell 21X data logger for hourly measurements. Hypodermic needle thermocouples have a fine needle measurement surface that undergoes little net absorption of solar radiation, is robust, and provides a minimal disturbance to snow structure and porosity. Thermocouples were placed at heights of 0, 5, 15, and 40 cm above the soil surface before the snowpack formed.

Supplementary air temperature, humidity, and wind speed measurements were made using calibrated Vaisala HMP35CF hygrothermometers (1.2 and 2.2 m above soil), and wind tunnel calibrated Weathertronics 2032 cup anemometers (1.4 and 2.25 m above soil). These measurements provided an indication of conditions in which snow could melt rapidly.

3. Results and Discussion

3.1. Snow Cover and Soil Temperatures

The total snowfall in the Quebec region from November 1998 to April 1999 was 294.7 mm SWE (Environment Canada, Quebec airport), which was below normal (337 mm SWE; s.d.,

Table 1. Average of Soil Temperature (Daily Mean, $n = 24$), Method of Measurement, Snow Porosity ($n = 3$), Snow Depth ($n = 3$), and N_2O Fluxes Measured Immediately After Snowmelt Using Closed Chambers ($n = 2$ to 6, Standard Deviation in Parentheses) on Agricultural Plots at St-Lambert Experimental Research Station

Date	Day of Year	Soil Temperature at 15 cm Depth, °C	Method[a]	Snow Porosity, $m^3\ m^{-3}$	Snow Depth, m	N_2O Flux, ng m^{-2} s^{-1} Slurry	Control
1998							
Dec. 15	349	−0.2	NA[b]	NA[b]	NA[b]	NA[b]	NA[b]
1999							
Jan. 1	1	−4.4	NA[b]	NA[b]	NA[b]	NA[b]	NA[b]
Jan. 15	15	−2.1	NA[b]	NA[b]	NA[b]	NA[b]	NA[b]
Feb. 2	33	−2.4	Cg	0.67	0.45	NA[c]	NA[c]
Feb. 26	57	−2.0	Cg	0.63	0.50	NA[c]	NA[c]
March 12	71	−1.0	Cg	0.64	0.54	NA[c]	NA[c]
March 29	88	−0.1	Cg	0.56	0.34	NA[c]	NA[c]
April 6	96	−0.1	Cg	0.64	0.29	NA[c]	NA[c]
April 12	102	0.1	Cg	NA[b]	trace	NA[c]	NA[c]
April 15	105	1.3	Cc	NA[d]	NA[d]	23 (14)	17 (11)
April 19	109	4.4	Cg, Cc	NA[d]	NA[d]	215 (82)	55 (55)
April 22	112	6.9	Cg, Cc	NA[d]	NA[d]	106 (51)	13 (10)
April 29	119	6.2	Cc	NA[d]	NA[d]	9 (14)	9 (14)
May 6	126	17.5	Cc	NA[d]	NA[d]	5 (6)	7 (7)
May 13	133	7.9	Cc	NA[d]	NA[d]	2 (3)	1 (1)

[a]Cg, concentration gradient measurement; Cc, closed chamber flux measurement.
[b]Not applicable; no measurements.
[c]Not available; no suitable flux calculation method.
[d]Not applicable; spring, complete ablation of snow.

34 mm: 1961 to 1990). The Saint-Lambert clay loam soil, which had a high water content in late Autumn 1998 (October, 0.38 $m^3\ m^{-3}$), remained frozen throughout most of the winter to a depth of 15 cm (Table 1). Conversely, the Chapais sandy loam soil never froze during the winter (Table 2), despite its initial lower soil water content (plot 1, 0.12 $m^3\ m^{-3}$; plot 2, 0.08 m^3 m^{-3}). The absence of freezing at the surface of the sandy loam is primarily due to the deeper snow accumulation at this site, the difference in snow depth being the result of greater exposure of snow cover to wind [*Li and Pomeroy*, 1997] at Saint-Lambert than at Chapais. The thermal implications of the

deeper snow at Chapais are significant. Using the midwinter depths and snow densities (Tables 1 and 2) as an example, the heat flux through snow [*Sturm et al.*, 1997] at Saint-Lambert was approximately twice that at Chapais. Although the difference in soil water content required 50% greater energy loss at Saint-Lambert to account for the latent heat of fusion upon freezing, the greater heat loss through the snow cover at this site permitted freezing of the soil, while the soil at Chapais remained unfrozen.

Each snow profile was characterized in detail once during the winter (Figure 1). The Saint-Lambert snow profile showed

Table 2. Snow Depth, Snow Porosity, Soil Temperature, and Soil Volumetric Water Content on Agricultural Plots at Chapais Experimental Research Farm

Date	Day of Year	Snow Depth,[a] m	Snow Porosity,[a] $m^3\ m^{-3}$	Soil Temperature,[a] °C −5 cm	−25 cm	Soil Water Content, $m^3\ m^{-3}$ −2 cm Plot 1	Plot 2	−10 cm Plot 1	Plot 2	−20 cm Plot 1	Plot 2
1998											
Dec. 30	364	0.35		0.55	1.46	0.12	0.08	0.28	0.41	0.15	0.37
1999											
Jan. 6	6	0.50	NA[b]	0.33	1.23	0.11	0.07	0.25	0.38	0.13	0.35
Jan. 11	11	0.60	NA[b]	0.34	1.16	0.11	0.08	0.21	0.32	0.12	0.34
Jan. 18	18	0.80	NA[b]	0.36	1.08	0.11	0.08	0.17	0.25	0.11	0.33
Feb. 24	55	NA[b]	NA[b]	0.47	1.16	0.14	0.10	0.28	0.40	0.14	0.35
March 2	61	0.80	NA[b]	0.59	1.20	0.14	0.11	0.27	0.40	0.13	0.35
March 9	68	0.75	NA[b]	0.55	1.18	0.19	0.16	0.29	0.40	0.15	0.36
March 15	74	0.91	0.65	0.61	1.22	0.20	0.15	0.29	0.40	0.15	0.36
March 23	82	0.92	0.64	0.61	1.18	0.23	0.20	0.29	0.41	0.16	0.37
March 30	89	0.77	0.64	0.55	1.09	0.27	0.26	0.32	0.43	0.17	0.38
April 9	99	0.50	NA[b]	0.63	1.07	0.28	0.26	0.34	0.45	0.19	0.39
April 12	102	0.43	0.61	0.66	1.11	0.27	0.25	0.33	0.44	0.18	0.39
April 20	110	no snow	NA[c]	4.79	3.22	0.23	0.22	0.30	0.42	0.16	0.38

[a]Mean values, $n = 2$, one measurement by plot.
[b]Not available; no measurements.
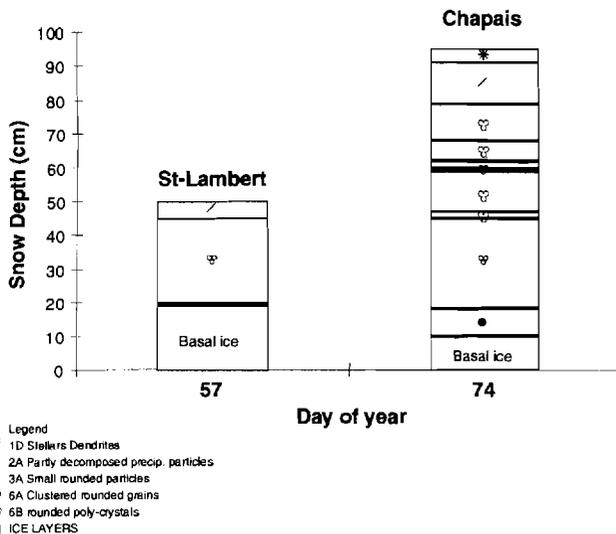[c]Not applicable; no measurement possible.

Figure 1. Characterization of snow profile structure for Saint-Lambert site on day 57 and for Chapais site on day 74 during winter 1998–1999. The figure was drawn using Snow Pit 98, an automated procedure for graphing snow pit stratigraphy, by T. Shultz and M. Albert, Cold Regions Research and Engineering Laboratory, Hanover, New Hampshire.

the existence of a thick basal ice layer (20 cm) on the soil surface. The basal ice layer was the result of the 17-mm rain event of December 20, 1998 (Figure 2), which infiltrated through the snow cover and froze on contact with the ground. An analysis of the meteorological record after December 20 and of heat fluxes through the snow cover indicated that the basal ice layer could have persisted until the onset of spring thaw. A basal ice layer (10 cm) was also present at Chapais on March 15. However, the higher temperatures of the unfrozen soils and the lower heat losses through the deeper snow cover

at this site could have lead to the rapid structural degradation of this ice layer.

The profiles of the snow cover at both Chapais and St. Lambert (Figure 1) also show the presence of layers of ice lenses of varying thickness above the basal ice layers. These ice lenses originated from meltwater or rain freezing on the surface or within the snow cover.

The existence of these basal ice layers at the soil-snow interface and of ice lenses within the snow cover are of prime importance in the measurement of gaseous fluxes as these layers may be impermeable to gaseous diffusion. The evolution of ice structures within snow cover is poorly understood, but it is known that the permeability of the ice layers and lenses can vary considerably depending on their formation and temperature [*Langham*, 1975]. Thick basal ice layers are highly impermeable to water infiltration, while ice lenses are more permeable due to structural factors, e.g., crust structure, vertical flow fingers, and horizontal discontinuity [*Marsh and Woo*, 1984]. We would expect the same behavior for gaseous diffusion. If the basal layers are impermeable, then no gas will diffuse from the soil to snow, and no concentration gradients will occur in the snow cover. If there is no basal layer, but an ice lens within the snow cover is impermeable, then gaseous diffusion from the soil into the snow cover will take place. However, concentrations will tend to an equilibrium between the soil and snow below the ice lens, and there will be no gradient above the ice lens itself.

3.2. Soil and Snow N_2O and CO_2 Concentration Gradients

3.2.1. Saint-Lambert site. Soil and snow concentrations of N_2O and CO_2 in both control and slurry treatments are plotted against depth in Figures 3a–3c for three dates during winter 1998–1999: days 57, 109, and 112. Only traces of N_2O and CO_2 were found in snow cover during the winter as shown on day 57. Although most of the profiles of the three dates had
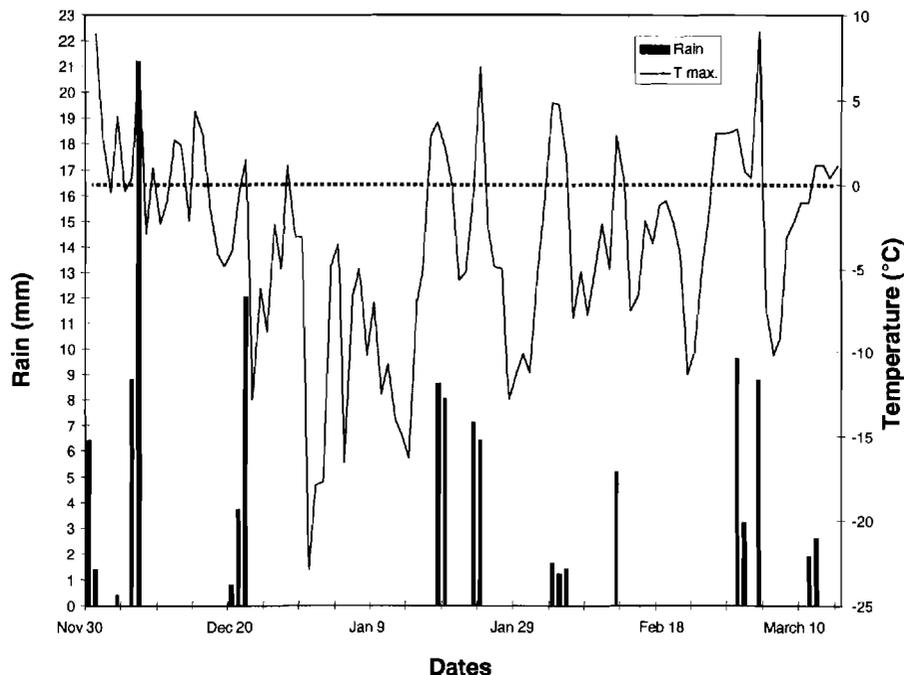

Figure 2. Precipitation (rain) and maximum temperature at Quebec airport meteorological station (46°47'N, 71°23'W, Environment Canada) during winter 1998–1999.
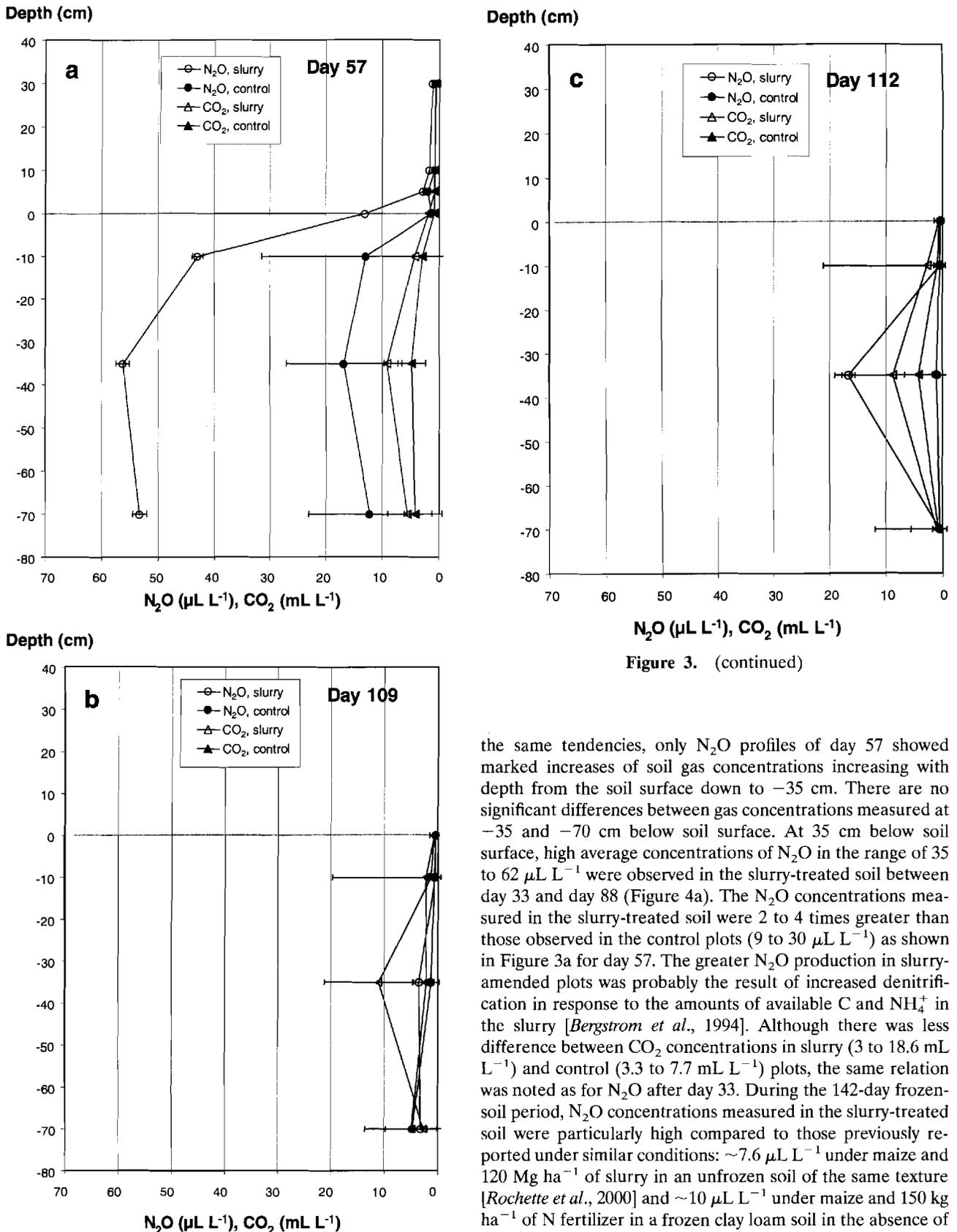
Depth (cm)

Depth (cm)

Depth (cm)

Figure 3. (continued)

Figure 3. Average concentrations of N_2O and CO_2 in the snow cover and the soil on days (a) 57, (b) 109, and (c) 112 for Saint-Lambert agricultural plots during winter 1998–1999. Solid symbols are for slurry treatment, and open symbols are for control. Data are means; slurry treatment, $n = 6$; control, $n = 12$. Error bars equal to standard errors.

the same tendencies, only N_2O profiles of day 57 showed marked increases of soil gas concentrations increasing with depth from the soil surface down to -35 cm. There are no significant differences between gas concentrations measured at -35 and -70 cm below soil surface. At 35 cm below soil surface, high average concentrations of N_2O in the range of 35 to 62 μL L^{-1} were observed in the slurry-treated soil between day 33 and day 88 (Figure 4a). The N_2O concentrations measured in the slurry-treated soil were 2 to 4 times greater than those observed in the control plots (9 to 30 μL L^{-1}) as shown in Figure 3a for day 57. The greater N_2O production in slurry-amended plots was probably the result of increased denitrification in response to the amounts of available C and NH_4^+ in the slurry [*Bergstrom et al.*, 1994]. Although there was less difference between CO_2 concentrations in slurry (3 to 18.6 mL L^{-1}) and control (3.3 to 7.7 mL L^{-1}) plots, the same relation was noted as for N_2O after day 33. During the 142-day frozen-soil period, N_2O concentrations measured in the slurry-treated soil were particularly high compared to those previously reported under similar conditions: $\sim$7.6 μL L^{-1} under maize and 120 Mg ha^{-1} of slurry in an unfrozen soil of the same texture [*Rochette et al.*, 2000] and $\sim$10 μL L^{-1} under maize and 150 kg ha^{-1} of N fertilizer in a frozen clay loam soil in the absence of significant snow accumulation [*Burton and Beauchamp*, 1994]. However, extremely high N_2O concentrations $\sim$1900 μL L^{-1} were measured in a soil surface submitted to freeze and thaw [*Cates and Keeney*, 1987]. Figure 4a also shows that little N_2O was produced in the upper soil horizon above 10 cm and that the gas produced by denitrifiers below the frozen soil layer did
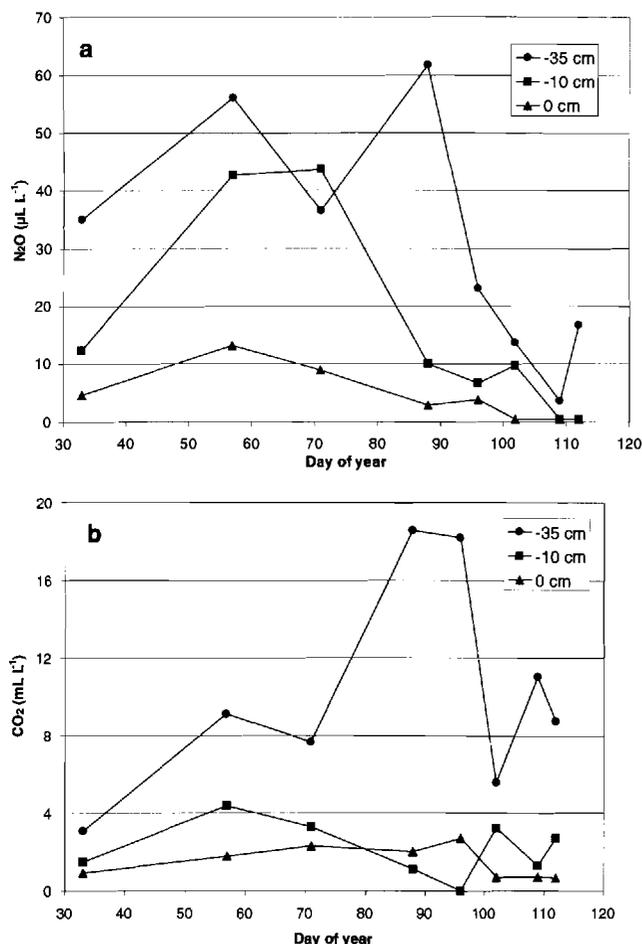
Figure 4. Average concentrations of (a) N_2O and (b) CO_2 in the soil at 0-, 10-, and 35-cm depths during winter 1998–1999, slurry treatment, Saint-Lambert agricultural site.

not diffuse to the surface horizon. The same observation can be made from CO_2 profile concentrations (Figure 4b).

The situation seems to be more complex when analyzed from the temporal progression of both N_2O and CO_2 concentrations measured at -10 cm during the winter because both gases do not have the same gradient pattern (Figures 3a–3c). In the case of N_2O the evolution of gas concentrations at -10 cm followed those measured at -35 cm during the winter (Figure 4a), indicating that some N_2O was produced at the temperature of $-1°C$ prevailing at 10 cm below ground surface. *Dorland and Beauchamp* [1991] reported that denitrification may occur at $-2°C$. If N_2O diffusion had occurred through the frozen soil of Saint-Lambert, it would have been corroborated by CO_2 diffusion as well which was not observed (Figure 4b). An impermeable barrier to effective gas diffusion may have been created by water freezing into the soil pore space between the ground surface and -15 cm depth. *Winston et al.* [1997] observed a similar phenomenon that restricted gas diffusion through the soil pores and leads to increases of CO_2 concentrations at depth in a clay soil.

At the beginning of snowmelt (day 88, Table 1) when the snow cover was isothermal, soil temperatures rose close to 0°C, and N_2O concentrations at 35 cm below ground surface decreased sharply from 60 to 20 μL L^{-1} (day 96), 10 μL L^{-1} (day 102), and 2 μL L^{-1} (day 109). We believe that the significant and simultaneous decreases in N_2O and CO_2 concentrations

resulted partly in gas transfer from the soil to the atmosphere when some soil pore space became free of barriers to gas transport. This phenomenon was also noted by *Cates and Keeney* [1987], *Burton and Beauchamp* [1994], and *Lemke et al.* [1998]. Some amount of both gases could also have been lost through their high solubility in meltwaters at low temperature and subsequent leaching to the drainage system. N_2O and CO_2 fluxes were not monitored at the snow-atmosphere interface when gas concentrations began to decrease in the soil profile because no satisfactory method exists to measure gas fluxes from a water-saturated snow cover in spring [*Winston et al.*, 1997; *van Bochove et al.*, 2000], with the exception of micrometeorological methods using trace gas analyzers [*Wagner-Riddel and Thurtell*, 1998; *Grant and Pattey*, 1999]. However, these latter methods were not applicable to our study.

At the end of snowmelt when the soil surface was free of snow, closed chambers were used to quantify N_2O fluxes (Table 1) from the soil to the atmosphere. On day 109, when the soil temperature increased to 4.4°C, high fluxes (215 ng m^{-2} s^{-1}, slurry; 55 ng m^{-2} s^{-1}, control) were measured and were followed by high fluxes on day 112 in the slurry-treated soil (106 ng m^{-2} s^{-1}). Following soil thaw and concomitant emission peaks, N_2O fluxes decreased from day 119 to low levels in both the treated and control plots. The N_2O fluxes estimated on days 109 and 112 for the two treatments can be interpreted by a significant but late increase in gaseous N_2O soil concentrations at different depths, which could have been a combination of degassing and biological production. In fact, during day 109 to day 112, soil N_2O concentrations varied from 3.2 μL L^{-1} at 70 cm depth and 16.6 μL L^{-1} at 35 cm depth in the slurry-treated plots, and between 4.5 μL L^{-1} at 70 cm depth and 1.0 μL L^{-1} at 35 cm depth in the control plots (Figures 3b and 3c). N_2O fluxes of the same magnitude were also reported from corn or barley plots during winter and spring [*van Bochove et al.*, 1996, 2000; *Grant and Pattey*, 1999; *Wagner-Riddle et al.*, 1997].

3.2.2. Chapais site. Figures 5a–5d illustrate that the soil and snow mean concentration profiles of four gases (N_2O, CO_2, Ar, and O_2) for four dates during winter 1998–1999 (days 55, 82, 99, and 110) approximately matched those shown for the Saint-Lambert site (Figures 3a–3d). In addition to gas measurements made on air samples collected at Saint-Lambert, Ar (tracer gas) and O_2 were analyzed in the diffusion plots established at the Chapais experimental farm.

It should be noted that, as described above, the sandy loam soil of Chapais was not frozen during the snow-covered period at 5 cm, nor at 25 cm depth (Table 2). Consequently, no restrictions to gas diffusion may be attributed to frozen water in soil pore spaces. Although the soil remained unfrozen during winter, we observed increased mean concentrations of N_2O (0.55 to 3.9 μL L^{-1}) and CO_2 (0.75 to 7.56 mL L^{-1}) in the soil atmosphere with depth from the beginning of measurements at day 55 until day 74 (Figures 5a and 5b). Because N_2O and CO_2 concentrations in snow remained at atmospheric levels all winter and no gradients were measured in the snow cover as observed in other studies [*Sommerfeld et al.*, 1996; *van Bochove et al.*, 2000], we believe that no gas diffusion occurred through the snowpack from the soil to the atmosphere. An ice layer, or layers, may have created the barrier to gas exchanges with the snow cover. The ice layer must have been below the gas probe at 5 cm above the soil surface, i.e., probably a basal layer. If there had been no basal ice, and impermeable ice layers had been present in the pack above the 5 cm level, then diffusion
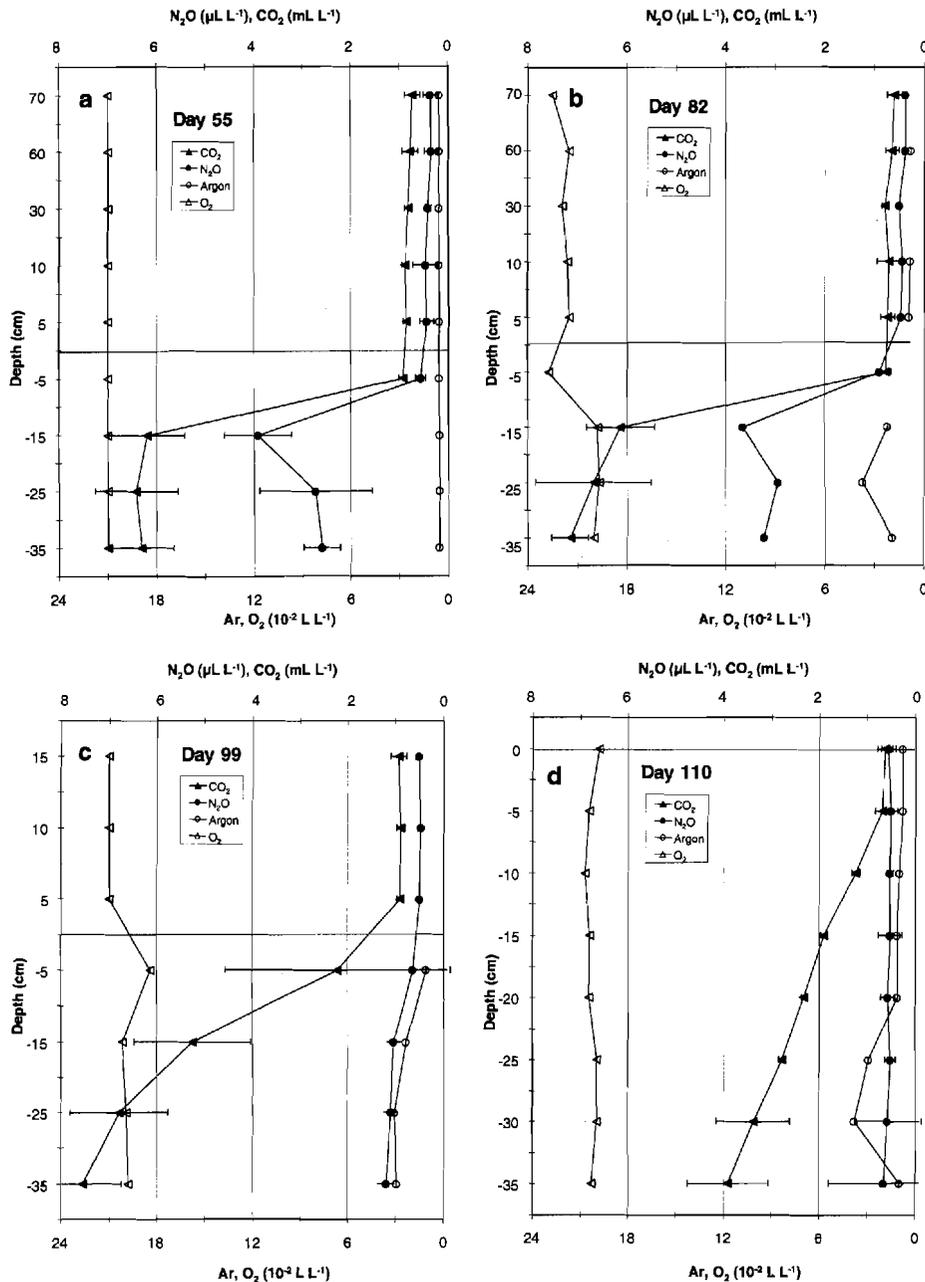
Figure 5. Average concentrations of N_2O, CO_2, Ar, and O_2 in the snow cover and the soil on days (a) 55, (b) 82, (c) 99, and (d) 110 for Chapais diffusion plots during winter 1998–1999. Data are means, n = 6. Error bars equal to standard errors.

from the soil to the snow would have given concentrations higher than those in the atmosphere at the 5 cm probe (Figures 4a–4c).

N_2O and CO_2 mean concentrations decreased sharply in the soil profile on day 99 (Figure 5c). This means that the gastight barrier due to ice lenses in snowpack disappeared completely during snowmelt, when snow depth was still 50 cm. There is evidence for homogeneous snowmelt by day 99, when air temperatures peaked at 5°C. Isothermal conditions had developed 3 days before this measurement and existed over days 83–90 before this. Through isothermal metamorphism, the basal ice layer (Figure 1) would have slowly reformed to a more porous structure and possibly degraded [*Pomeroy and Brun*, 2001]. The primary mechanism by which basal ice layers can rapidly

degrade, however, is due to heterogeneous meltwater flow. Meltwater tends to follow preferential flow paths to the snowpack base and is delivered most rapidly near these zones, often before isothermal conditions develop [*Marsh and Woo*, 1984; *Marsh and Pomeroy*, 1999]. At the base, ponding and some refreezing release latent heat in patches near to preferential flow paths. This would effect to break up the continuous basal ice layer in some areas while possibly augmenting it in others. Isothermal conditions developing after the arrival of this meltwater would then further degrade the basal ice. The deterioration of both the basal ice layer observed near the ground surface and the ice layers in the snow cover allowed for better gas diffusivity when the impermeable barrier became discontinuous. This phenomenon is corroborated in our study by
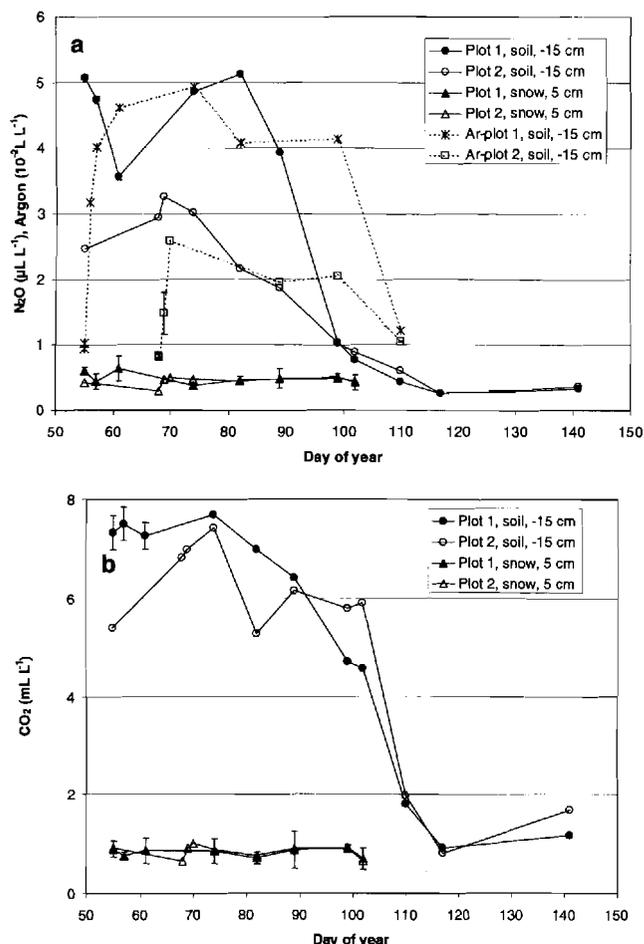
Figure 6. Average concentrations of (a) N_2O and Ar and (b) CO_2 in the soil at 15-cm depth and in the snow cover at 5-cm above ground surface for Chapais diffusion plots during winter 1998–1999. Solid symbols are for plot 1, and open symbols are for plot 2. Data are means; soil, $n = 3$; snow, $n = 6$.

simultaneous decreases in N_2O and CO_2 concentrations on day 74 (Figures 6a and 6b).

We were able to demonstrate the dynamics of impermeable ice barriers on gas exchanges by using pure Ar gas from the diffuser as a tracer. Argon accumulated rapidly at 15 cm below soil surface (Figure 6a) from the natural background level (0.934×10^{-2} L L^{-1}) up to 5×10^{-2} L L^{-1} and 2.7×10^{-2} L L^{-1} in plots 1 and plot 2, respectively, after having activated the artificial flow on day 55 in plot 1 and on day 68 in plot 2. Activation of the mass flow controller in plot 2 was delayed because of instrument malfunction. Argon concentrations decreased in both plots by 20% at day 82 and by 100% after day 99. The accumulation of Ar at 15 cm below the soil surface in both diffusion plots (Figure 6a), and the absence of a gradient of Ar in the snowpack (Figures 5a–5c), further demonstrated that ice lenses in the snowpack (<5 cm) or basal ice could have constituted a barrier to gas transfer during a part of the winter. This period of approximately 50 days was not long enough to observe a decrease in O_2 soil concentrations, which never fell below 18×10^{-2} L L^{-1} (day 82, Figure 5b). The high O_2 soil concentrations and the low soil water content rendered O_2 sufficiently available as the terminal electron acceptor for denitrifiers, which explains the relatively low N_2O concentrations observed in the Chapais sandy loam soil. Generally, O_2 con-

centration of 5×10^{-2} L L^{-1} in the pore space is considered to be critical to produce high N_2O and N_2 amounts through denitrification [Arah et al., 1991]. If we consider that the maximum N_2O concentration of 3.9 μL L^{-1} measured during winter 1998–1999 was built up under an impermeable ice barrier, we suggest that N_2O production was relatively low compared to that of Saint-Lambert's plots, which accumulated under the frozen soil surface. In comparison, the significant amounts of N_2O emitted from another agricultural field at the Chapais experimental farm during winters of 1995, 1996, and 1997 were calculated from snow cover N_2O concentration gradients in the range of 0.5 to 12 μL L^{-1} [van Bochove et al., 2000].

3.3. Gas Solubility Factor

All points of discussion referring to N_2O gaseous concentrations must be set in the perspective of gas solubility in water. In fact, the solubility of N_2O, which is relatively high at low temperature, varied from 1.275 to 1.085 mL mL^{-1} in soil water at the depth of 5 cm during the experiment of Chapais. Soil water contents were significantly higher at -10 and -20 cm in plot 2 (Table 2); this was probably due to the fact that lysimeter 1 drained more efficiently than lysimeter 2. Consequently, we would have expected a higher total amount of N_2O in plot 2 due to the amount of gas dissolved in soil water. However, solubility calculations [Moraghan and Buresh, 1977] showed that on day 82, when maximum N_2O concentrations were reached at -15 cm in soil atmosphere (plot 1, 5.13 μL L^{-1}; plot 2, 2.16 μL L^{-1}), the total amount of gaseous and dissolved N_2O in plot 1 (depth 10 to 20 cm below soil surface) was 185 μg N compared to 8 μg N in plot 2. Thus plot 2 which had the lowest air-filled porosity showed the lowest soil N_2O concentrations. The estimated data obtained by solubility calculations emphasized the importance to study the availability of O_2 to denitrifiers that may have a significant impact on microbiological production of N_2O in agricultural soils [Hénault et al., 1998]. Also, in a study conducted at the landscape scale in Saskatchewan, van Kessel et al. [1993] concluded that hydrological processes were of primary importance in the control of the intensity of denitrification.

No sufficient data on soil air-filled porosity, temperature, and volumetric water content were available from the Saint-Lambert plots (e.g., different depths, space locations, and sampling dates) to calculate soluble N_2O or estimate gas fluxes at soil thaw. In addition, high N_2O solubility due to low soil temperature and high soil water content in winter may have resulted in significant N_2O losses to the drainage system. This phenomenon can be important as mentioned by McMahon and Dennewy [1999] and merits further investigation.

4. Summary

Monitoring soil temperature and gas concentrations in soil and snow have demonstrated that large amounts of N_2O can be produced by denitrification in agricultural soils over winter. Even though significant fluxes have already been measured from unfrozen soils through the snow cover, we demonstrated that particular situations, such as a frozen soil layer at Saint-Lambert or ice layers in the snowpack at Chapais, may delay gas emissions to the atmosphere. Such winter situations are difficult to study but merit attention because they can generate important N losses to the environment. Changes in N_2O concentrations over short periods of time underline the necessity of following critical soil and snow characteristics both rapidly

and extensively; otherwise, N cycling can be misinterpreted. Emissions of N_2O and CO_2 during thaw at Saint-Lambert could be attributed to gases produced throughout winter and accumulated under the frozen soil layer. The significant differences in N_2O concentrations between the slurry-treated soil and the control represent an undesirable N loss mechanism from residual slurry N that had been first nitrified after fall application.

The absence of gas concentration gradients in snow cover, due to impermeable barriers to gas diffusion between soil and atmosphere, did not allow the estimation of N_2O emissions at Saint-Lambert and Chapais sites during winter. Although gaseous N_2O emissions to the atmosphere are significant, soluble N_2O fluxes should also be considered due to its high solubility at low temperature.

Finally, basal ice layers do not normally develop in moderate to deep (>50 cm) eastern North American agricultural snow covers as frozen soils cannot persist under such depths of snow in such relatively temperate, moist climates. However, the occurrence of ice lenses and crusts are widespread due to the rainfall and occasional melts that occur in winter thaws. As most of these lenses are structurally weak (porous) and/or discontinuous, they may not eliminate diffusion processes. They can, however, significantly alter gas concentration gradients and should always be considered as complicating factors to flux estimates when evaluating gas fluxes from disjointed linear or nonlinear gradient concentration gradients.

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