

Winter Methane Dynamics Beneath Ice and in Snow in a Temperate Poor Fen

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ABSTRACT

Winter's influence on methane (CH₄) stored in pore water and emitted through snow was investigated in a temperate poor fen in New Hampshire over two winters (1993–94, 1994–95). CH₄ accumulated beneath ice layers (1 cm) deposited by freezing rain, resulting in snow pore air mixing ratios as high as 140 ppmv the first winter and 600 ppmv the second. An early winter snow crust of 300 kg m⁻³ caused no discontinuity in a linear mixing ratio profile and therefore was not observed to retard snowpack emissions. CH₄ concentration depth profiles in pore water steepen and concentrations increase by as much as 400 μM at the 10- and 20-cm depths as the ice cover forms. This suggests that the peat–ice cover plays an important role in CH₄ buildup in pore water by limiting transport of gases between the peat and the atmosphere. Pore water concentrations gradually decline through late winter. The seasonality of dissolved CH₄ in pore water over two winters and one summer shows an average annual amplitude of 1.3 g CH₄ m⁻² (25- to 75-cm depth range), with a winter maximum of 4.7 g CH₄ m⁻². Emissions during the winter with average snowfall accounted for a larger percentage (9.2% in 1993–94) of total annual emission than did the winter with below average snowfall and warmer air temperature (2% in 1994–95). Emissions averaged 56 and 26 mg m⁻² day⁻¹ during the first and second winter (December, January and February), respectively.

Key words: Methane, Pore water, Snowfall, Frozen peat

INTRODUCTION

Methane (CH₄) emissions from wetlands associated with temperate forests have played a role in atmospheric chemistry and radiative forcing of the Earth's climate. Between the preindustrial Holocene and the present, conversion of wetlands associated with midlatitude temperate forests to agricultural, residential, and other use may have reduced CH₄ emissions from 20 to

9 Tg yr⁻¹; this compares to a reduction from 17 to 15 Tg CH₄ yr⁻¹ from the boreal coniferous forests (Chappellaz et al. 1993). Year-round emission measurements in the field are relatively few (Whalen and Reeburgh 1988, 1992; Dise 1992, 1993; Yavitt et al. 1993; Frohling and Crill 1994) and are of persistently moist sites that attain peak emissions in midsummer. Warm season CH₄ effluxes from northern peatlands have been the subject of numerous investigations that together have identified water table position, peat temperature, and peat/plant type as the main factors controlling CH₄ emissions (Moore 1994). The observed seasonality of CH₄ emissions suggests that cold weather and ice formation may limit both CH₄ production and efflux to the atmosphere at middle and high latitudes. Only two previous site investigations have been known to address the contribution of CH₄ to the atmosphere during winter (Dise 1992, 1993, Whalen and Reeburgh 1988, 1992). At both sites CH₄ efflux in winter was found to be significant, but only for certain vegetation types. Whalen and Reeburgh (1988) in a study of subarctic tundra noted episodic release of CH₄, amounting to 41% of the annual flux from moss-covered areas. In the same study, winter CH₄ release was less than 10% of the annual flux for *Eriophorum* tussock, intertussock and *Carex* stands. Dise (1992) reported winter CH₄ fluxes at a site within the Marcell Experimental Forest in Minnesota from open bog, open poor fen, forested bog hollow and forested bog hummock of 4, 11, 15 and 21% of the annual flux, respectively. Current models of seasonal and annual CH₄ budgets make the assumption that emissions occur only during the warm season and vary the length of the warm season with latitude (Chappellaz et al. 1993, Fung et al. 1991). There is a need for additional site investigations to determine the dynamics of winter emissions and the physical factors that control cold-season exchange, so that conditions leading to significant winter emissions can be identified.

Our study focuses on winter conditions in an open poor fen associated with a broad-leaved temperate forest of southern New Hampshire. The seasonal storage of

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CH₄ in pore water beneath a peat–ice layer, conveyance or entrapment of CH₄ in snow, and emission of CH₄ through a frozen surface to the atmosphere are investigated. The field seasons were the winters of 1993–1994 and 1994–1995. The study has determined that winter conditions exert a significant influence on CH₄ storage and emission.

CH₄ PRODUCTION, DESTRUCTION AND TRANSPORT IN WINTER

The concentration of CH₄ in the pore water of peat depends on the balance of processes that produce, destroy and transport CH₄. The potential for CH₄ production is high in winter due to the presence of labile organic materials, but production is constrained by low temperatures (Yavitt and Lang 1988). In peat samples from a subarctic mire, Svensson and Rosswall (1984) noted CH₄ production at just above 0°C, with a sevenfold increase between 2° and 12°C.

Transport mechanisms of CH₄ through peat differ in winter compared to the warm season. Plant-associated transport was the dominant (90%) mechanism, producing emissions during the growing season in *Carex* spp.-dominated fens in subarctic Canada (Whiting and Chanton 1992). Winter transport may depend more on diffusion through pore water, concentration at the freezing front, and release through cracks and fissures in the peat–ice and along or through plant stems. Winter has low ratios of surface efflux to pore water concentration, while the opposite is true in summer (Dise 1992), signifying seasonal change in net production and transport processes or rates. A discussion of the fate of decayed plant matter buried in peat bogs (Clymo 1984) suggests that winter fluxes could be partly due to upward transport by diffusion, mass flow in solution and possibly ebullition of CH₄ produced at depth.

Effects of ice on the transport of CH₄ have been observed. Whalen and Reeburgh (1988) suggest that episodic releases occur through voids in a frozen moss matrix in subarctic Fairbanks, Alaska, as the freezing front progresses toward impermeable permafrost. Svensson and Rosswall (1984) note thaw–season CH₄ concentration maxima lower in the peat profile than the zone of maximal potential CH₄ production, suggesting that downward freezing may concentrate CH₄ at depth. CH₄ may be excluded from peat–ice until the water becomes saturated in dissolved gases, and then entrained in bubbles in the ice; when the ice melts a release of the gas bubbles may occur (Svensson and Rosswall 1984). Moore et al. (1990a, 1990b) and Windsor et al. (1993) observed high flux rates at the time of thaw in subarctic fens near Schefferville, Quebec. The high fluxes were thought to be associated with the release of methane

produced during the winter and trapped within and beneath the ice.

SITE DESCRIPTION

Sallie's Fen is a midlatitude poor fen in southeastern New Hampshire (43°12.5'N, 71°03.5'W) at an altitude of 110 m. This peat-forming wetland has low nutrient input and a vegetation cover more like a precipitation-fed bog than a groundwater-fed marsh. The fen is situated in an upland depression surrounded by forest. The average thickness of the peat is 2 m, with depths up to 4.5 m. The surface area of the fen is 1.7 ha. A lagg occurs around the entire perimeter and is the only significant open water. The outflow elevation maintains water levels within a few centimeters of the sphagnum moss hollows except during the late growing season, when evapotranspiration lowers the water table. Sallie's Fen is a bryophyte, especially *Sphagnum* spp., dominated wetland. The vascular plant community consists of both sedges and shrubs. Sedge (*Carex rostrata* Stokes), leather-leaf (*Chamaedaphne calyculata* L.), and small cranberry (*Vaccinium oxycoccus*) are prevalent throughout the fen. The surrounding upland is dominated by eastern hemlock (*Tsuga canadensis* L.), white pine (*Pinus strobus*), red maple (*Acer rubrum* L.), sugar maple (*Acer saccharum* L.) and white birch (*Betula papyrifera* Marsh). The pH at 10 cm beneath the water level ranges between 6 and 4 across the fen, with slightly lower acidity (higher pH) near an intermittent inlet stream.

CH₄ efflux from the fen surface in summer is two orders of magnitude greater than in the winter and is influenced by peat temperature and time-proximity to heavy rainfall events (Frolking and Crill 1994). Winter emission measurements over a five-year period indicate no positive relationship between thaw-season emission rates and emissions during the following winter (Melloh and Crill, in prep.). Thus, storage of CH₄ during highly productive and emitting summers leading to increased degassing during the following winter, is not indicated. Total snowfall during months with average temperatures below 0°C (December, January, February) may be the best climatologic indicator of flux rates from midlatitude (43°N) poor fens (Melloh and Crill, in prep.). Emissions over five winters (December, January and February) at this fen have varied between 2 and 9% of the total annual emissions. The three winters with both moderate temperatures and snowfall had lower emissions while the two winters with lower temperatures and more snowfall had higher emissions.

The mean annual precipitation at a meteorological station 15 km to the southeast in Durham, New Hampshire, is 109.8 cm. Precipitation is fairly evenly distributed throughout the year, though it is heaviest in No-

ember and December when 20% of the annual total falls. The average annual temperature is 8.3°C, with the average winter minimum of -4.9°C occurring in January. Midwinter thaws may occur in any winter month. Average temperatures are below freezing only for the months of December through February. The average of the three monthly temperatures is -3.7°C for the 1951 to 1980 climatic period (NCDC 1985). In these winter months, air temperatures measured on the fen have averaged 1.6°C lower than those reported for Durham.

METHODS

Pore water sampling stations were deployed at three locations in the fen: near the surface inlet, near the outlet and near the fen center. The stations allowed repeated sampling of unfrozen peat layers throughout the winter at 10-cm depth intervals to a maximum depth of 70 cm, without disturbing the ice. Freeze-up between sampling dates was avoided by injecting a measured amount of air into the tubes to clear water from the Ty-

gon tubes to below frost depth. Pore water was drawn through each tube using a 3-psi/stroke vacuum, pressure hand pump. Initial pore water and gas were expelled into an in-line stoppered flask until the pore water flow was bubble-free. Thirty-milliliter samples of bubble-free water were then drawn into 60-mL plastic syringes. Dissolved CH₄ was stripped into a 30-mL headspace of room air or N₂ (McAuliffe 1971), then analyzed by gas chromatography. The pore water sampling stations and the University of New Hampshire laboratory procedures are described in more detail elsewhere (Melloh and Crill, in prep.). The amount of CH₄ stored in the peat was estimated by multiplying the concentrations by peat porosity and integrating over depth.

Snow pore air was sampled through a 1-m length of 0.32-cm-OD stainless steel tubing inserted into the snow horizontally through a side access pit. The pit was deepened to a given sampling horizon, pore air was drawn promptly and the pit was deepened to the next sample horizon. Snow layering, density and temperature were measured in snow pits (Fig. 1). Snow

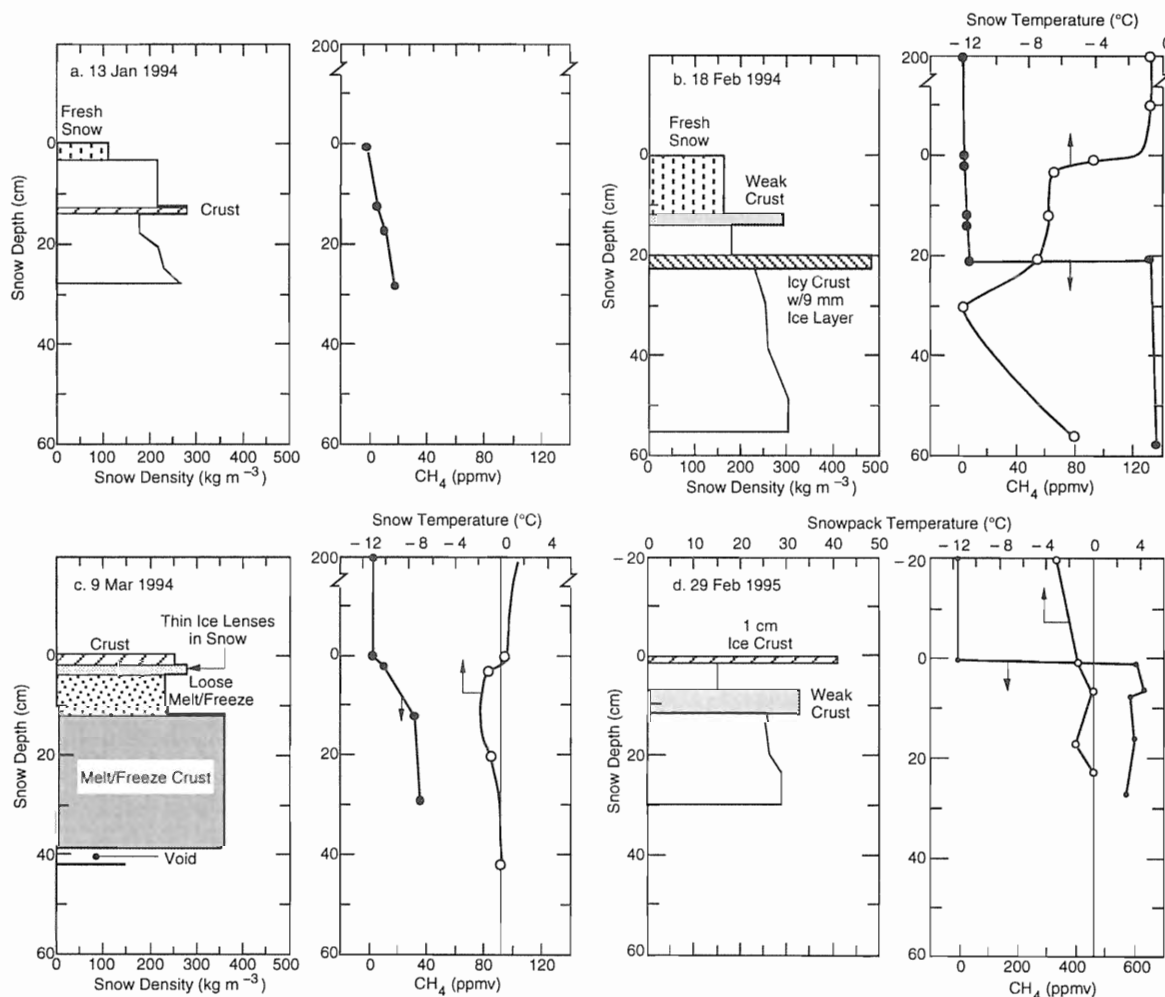


Figure 1. Snow density, stratigraphy, pore air mixing ratios (solid circles) and snow temperatures (open circles) on four dates.

density was measured using a 100-cm³ snow sampler and hanging scale accurate to 1 g. Snow temperatures were taken with long-stemmed (20-cm) coil-actuated thermometers accurate to $\pm 0.75^\circ\text{C}$. Ice cover initiation and progression of the freezing front were observed at three pore water sampling stations and a thermistor probe at the meteorology station. A temperature of 0°C was used to estimate frost depth from thermocouple and thermistor readings, and the onset of thaw was interpreted from above-freezing temperatures immediately above the 0°C isothermal peat–ice layer.

Flux measurements were made using a static chamber technique (Crill et al. 1988) described in more detail by Frolking and Crill (1994). A portable aluminum chamber (150 L) was placed into pre-installed collars (0.4 m²) and sealed from the atmosphere with water. During the snow-covered period the chamber was placed on top of the snow. Five, 30-mL air samples were drawn at 4-minute intervals into 60-mL plastic syringes. The rate of surface efflux was determined from the slope of CH₄ mixing ratios over time.

RESULTS

CH₄ mixing ratio profiles in snow pore air

In 1993–94, a 30-cm snowcover was established by 28 December 1993 and 4 January 1993 snowfalls. Early-winter crusts in the snowpack were composed of bonded snow crystals and had densities less than 300 kg m⁻³. Mixing ratios were near 20 ppmv at the base of the snow with a linear gradient of 70 ppmv m⁻¹ through the snowpack (Fig. 1a).

Frequent minor snowfalls throughout the winter maintained and thickened the snowpack to a maximum of around 50 cm in mid-February. Freezing rain on 18 and 19 January deposited a thick layer of ice on the surface that was subsequently buried by fresh snow. The ice layer, observed in snowpits on 3 February (at 10-cm depth) and on 18 February (at 20-cm depth), was 5 to 9 mm of clear ice within an icy crust (Fig. 1b). The combined density of the ice layer and surrounding crust, in a 100-cm³ sampler, was nearly 500 kg m⁻³. This ice layer was remarkably continuous, occurring consistently along the snow depth transect. The development of the ice layer and its later decay controlled CH₄ mixing ratios in the snow pore air. Following the freezing rain, pore air mixing ratios increased beneath the ice layer to around 90 ppmv on 3 February and 130 ppmv CH₄ on 18 February (Fig. 1b), indicating that gas was being trapped beneath the ice layer. Above the ice layer, mixing ratios were near ambient. A three-day midwinter thaw occurred 19 through 21 February. Overnight air temperatures measured at 2-m height were above freezing, and daytime tempera-

tures rose to 17.5°C at midday on 20 February. The ice layer in the snowpack was decayed by this midwinter thaw and was not observed in a 9 March snow pit (Fig. 1c); snow pore air CH₄ mixing ratios decreased.

The structure of the snow in late winter was dominated by thick meltfreeze crusts with densities from 360 to 420 kg m⁻³. The snow profile of 9 March consisted of a 27-cm-thick meltfreeze crust below an 8-cm layer of melt-freeze polycrystals (Colbeck et al. 1990) and a surface crust that included several thin ice lenses, each less than 1 mm thick. Mixing ratio gradients were nonlinear in spring. On 9 March (Fig. 1c), CH₄ mixing ratios of 36 ppmv were measured 29 cm below the snow surface and the gradient between the surface and 29 cm decreased from 310, 205 and 35 ppmv CH₄ m⁻¹ in the 0- to 2-cm, 2- to 12-cm, and 12- to 29-cm depths, respectively. On 24 March, the snow was melting and mixing ratios were near ambient at 0- and 2-cm depths but averaged 39 ppmv in the 8- to 14-cm depth range. The high gradients occurred when surface fluxes were relatively low (Fig. 4) and imply low diffusivities through wet, densified layers near the surface of the snowpack. CH₄ concentrations in water-saturated snow at the base of the pack on 24 March 1994, averaged 70 μM (1500 ppmv).

In 1994–1995, there was no persistent snow cover and no significant snow on the ground until February when a total of 35.6 cm fell. On 28 February, snow depths on the fen averaged 24.9 cm. Freezing rain on 27 February deposited 1 cm of ice on top of the snow (Fig. 1d). Pore air mixing ratios on 28 February were 600 ppmv beneath the ice layer. The snow melted in early March.

Pore water concentrations

Steepening of the near-surface pore water concentration profiles (Fig. 2) beneath newly formed peat–ice occurred at the boardwalk and outlet site during the winter of 1993–1994 and at all three sites during the winter of 1994–95. Concentrations increased by as much as 400 μM at the 10- and 20-cm depths as the peat–ice covers formed.

Pore water concentrations deeper in the profiles exhibit general increases in concentrations into February (1993–1994) or January (1994–1995), then decline through late winter. Integrated methane stores between 25- and 75-cm depths (Fig. 3) readily show this seasonal trend. The average amplitude in stored CH₄ for the three sites was 1.3 g CH₄ m⁻² between the average of the two winter peaks (4.7 g CH₄ m⁻²) and the intervening summer low in August (3.4 g CH₄ m⁻²). This deeper layer of peat generally does not freeze in winter, nor does it become unsaturated during the growing season. For the 0- to 70-cm layer, the average annual

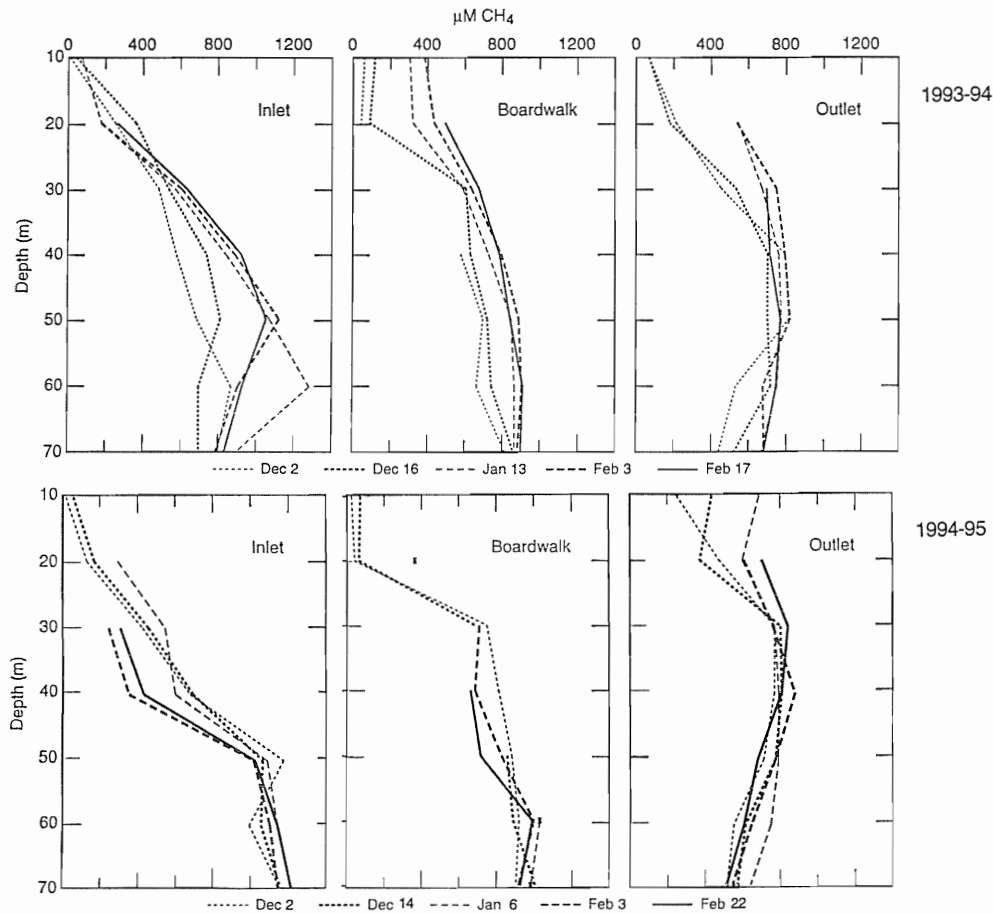


Figure 2. Methane pore water concentrations at 0- to 70-cm depths at the three in-situ pore water sampling sites, for two winters.

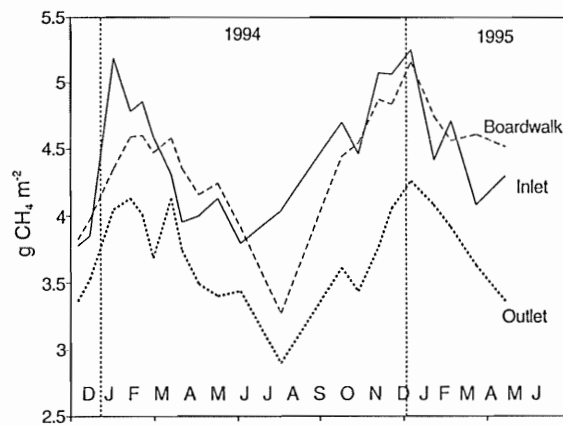


Figure 3. Integrated CH_4 stores in the 25- to 75-cm depth range. (Assumed peat porosity 0.7.)

amplitude may be as high as $2.2 \text{ g CH}_4 \text{ m}^{-2}$ with a $5.7\text{-g CH}_4 \text{ m}^{-2}$ maximum in winter and $3.5\text{-g CH}_4 \text{ m}^{-2}$ minimum in August. Frozen layer concentrations for the near surface layer are estimated by interpolation between pre- and post-frozen concentrations, and unsaturated zone concentrations are assumed nil.

Surface emissions

Total monthly flux averaged about 1.7 and $0.8 \text{ g CH}_4 \text{ m}^{-2}$ during the winters of 1993–94 and 1994–95, respectively (Fig. 4). Seasonally low fluxes coincide with the presence of a peat–ice cover and decreasing peat temperature. The lowest flux rates occurred in

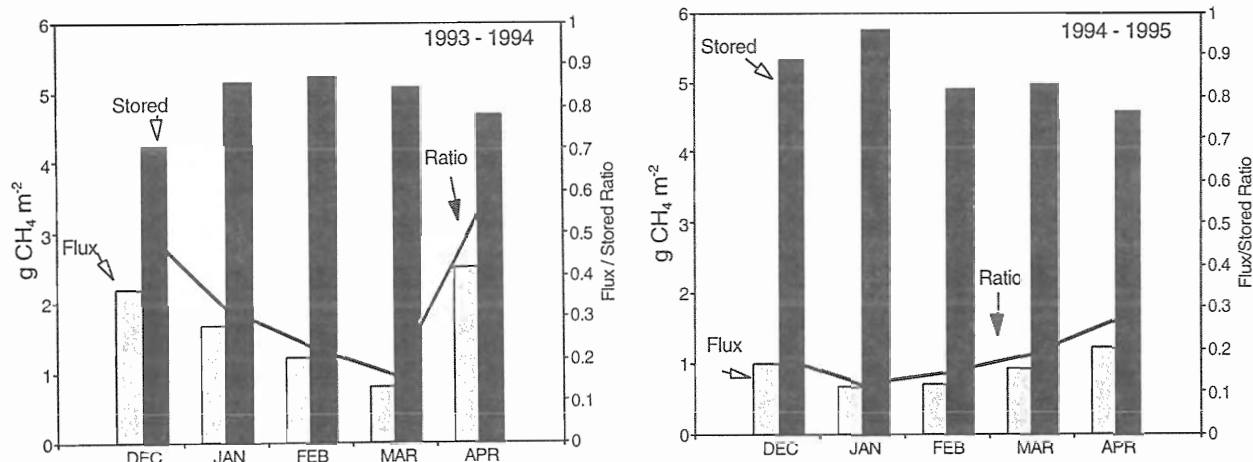


Figure 4. Ratios of $(\text{CH}_4 \text{ flux})/(\text{stored CH}_4)$ on a monthly basis from December through April, for two winters. Stored values represent average CH_4 resident in pore water during the month (0 to 70 cm). Flux values are total flux, computed from average measured flux multiplied by days in the month.

February and March during both winters, coinciding with the presence of ice layers deposited by freezing rain within or on top of the snow.

Winter CH_4 emissions decline from near $100 \text{ mg CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ in October and November to cold season minimums near $25 \text{ mg CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ (Melloh and Crill, in prep.). Dwindling emissions in fall and early winter coincide with increased CH_4 stores.

DISCUSSION

CH_4 mixing ratio profiles in snow pore air

CH_4 mixing ratio profiles, and thus gas transport in snow pore air, are directly affected by snowpack density and stratigraphy. CH_4 concentrations measured here varied from near ambient just beneath the snow surface to as high as 600 ppmv (29 February 1995) when entrapment occurred beneath a 1-cm ice layer deposited by freezing rain (Fig. 1). A snow crust (300 kg m^{-3}) in early winter caused no discontinuity in the mixing ratio profile, and therefore was not observed to significantly retard snowpack emissions. Early in winter when the ice layer was not present a linear mixing ratio profile occurred in the snowpack with a measured gradient of 70 ppmv m^{-1} . The diffusive flux (J) supported by this gradient (dC/dz), estimated using Fick's relationship

$$J = -\phi^n D_s \frac{dC}{dz},$$

is $74 \text{ mg m}^{-2} \text{ day}^{-1}$, assuming a diffusivity of CH_4 in air (D_s) of $0.22 \text{ cm}^2 \text{ s}^{-1}$ (Sommerfeld et al. 1993) and no reduction for tortuosity or pore size. A porosity (ϕ) of 0.78 was estimated from the pack density of 200 kg m^{-3} and ice density of 917 kg m^{-3} . The exponent (n) is

not well known for snow (E. Arons, CRREL, pers. comm., 1995). The calculated flux value for the 12 January gradient is in excellent agreement with the flux estimated from chamber measurements on 13 January ($75 \text{ mg m}^{-2} \text{ day}^{-1}$), assuming an n of 1. Late-winter profiles after the ice layer decayed were nonlinear, with high gradients near the surface suggesting lower diffusivities through melting snow. The amount of data collected here on snow pore air mixing ratios is very limited; more data are needed to assess the spatial variability and change in gradient with metamorphism of the winter snowpack and ripening of the snowpack in spring.

Concentrations in pore water

CH_4 concentration profiles were steepened, and there were substantial increases in concentrations at the 10- and 20-cm depths as the ice cover formed and the freezing front progressed into the peat (Fig. 2). This near-surface change and its timing strongly suggest that ice cover plays an important role in CH_4 buildup in the pore water by limiting transport of gases between the peat and the atmosphere. The concentration gradients in deeper parts of the profiles (50 to 70 cm) suggest that upward diffusion from below occurred only in the early winter of 1993–1994 at the boardwalk site and throughout the winter of 1994–1995 at the inlet site.

CH_4 concentration maxima occurred at 40- to 70-cm depths in the profiles and ranged from 800 to $1200 \mu\text{M}$, somewhat higher than the $700 \mu\text{M}$ maximums reported in winter profiles in a Minnesota fen (Dise 1992). Both the Minnesota site and Sallie's Fen increased CH_4 stores in winter and decreased stores in spring. A probable source of increased pore water CH_4 is a continued low rate of production, combined with restricted transport through the ice to the atmosphere. The ice cover separates the fen surface from the atmosphere, restrict-

ing oxygen diffusion into the near-surface peat at the same time that continued decomposition increases anaerobic conditions favorable to methanogenesis. The increase in CH₄ solubility as the peat cools may contribute to the seasonal trend of increased pore water concentrations in winter. The solubility of CH₄ in water is low, but increases with decreasing temperature from 1.8 mM at 10°C to 2.5 mM near 0°C (Yamamoto et al 1976) representative of midwinter deep (3 m) and surface peat temperatures, respectively. The higher winter solubilities may allow newly produced CH₄ to remain

those of Dise (1993) in a Minnesota fen. Flux ratios indicate the balance between net production and transport and, particularly in the winter of 1993–94, follow the trend of increasingly restricted transport as the ice layers form. This suggests the low flux/storage ratios may in part be due to restricted transport. Peat temperatures also decrease, suggesting production slows progressively through the winter (Fig. 5). The flux/storage ratios increased in April, probably indicating a combination of increased production and increased transport as the ice thaws.

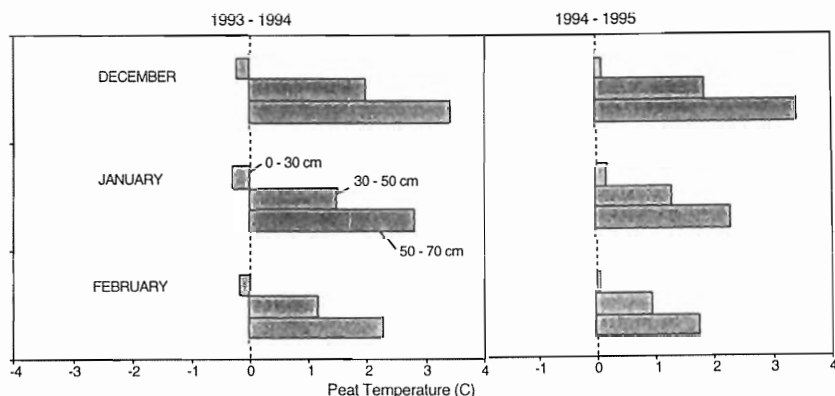


Figure 5. Peat temperatures during two winters (1993–94, and 1994–95) in three depth ranges (0 to 30 cm, 30 to 50 cm, 50 to 70 cm).

dissolved in the pore water, and it seems plausible that some CH₄ could redissolve from bubbles in the peat.

Further studies designed to determine the relative importance of processes responsible for increased pore water concentrations in winter should try to quantify upward diffusion from deep peat, exclusion of CH₄ at the freezing front or into bubbles in the ice, ebullition from deep peat, solubility changes and net production at cold temperatures.

CH₄ flux

The data suggest that snow and peat–ice affect winter flux rates. The highest winter fluxes occur in early winter when freezing of the saturated layer is incomplete. Low flux rates in February and March occur when the peat–ice cover is continuous. The seasonal decrease in flux coincides with increased near-surface (sub-ice) pore water concentrations, suggesting entrapment of CH₄ beneath peat–ice. Continuous ice layers in the snow formed by freezing rain act as flux barriers that persist until a significant melt event. Sommerfeld et al. (1993) reported that discontinuous ice lenses in a Wyoming snowpack did not have a significant effect on flux gradients.

Observations of the ratios of surface flux to CH₄ stored in the 0- to 70-cm layer (Fig. 4) are similar to

Of the two winters observed, the one with more snowfall and colder air temperatures (1993–1994) had higher winter emissions. Average winter emissions, for the five winters ending in 1994–1995, were 20, 39, 53, 56 and 26 mg CH₄ m⁻² day⁻¹, which amounted to 2.0, 5.2, 6.6, 9.2 and 2.0% of the total annual fluxes, respectively (Melloh and Crill, in prep.). Total winter snowfalls (December, January and February) were 63, 53, 93, 112 and 46 cm, respectively, compared to a long term average of 117 cm during the 1951–1980 climatologic period (NCDC 1985). Warmer peat beneath an insulating snowpack might enhance winter production of CH₄. Peat temperatures during the high emission winter (1993–1994) are slightly warmer than the low emission winter (1994–1995) (Fig. 5).

CONCLUSIONS

Snow and ice exert a significant influence on cold season CH₄ emissions. A cold winter with more snowfall had greater emissions than did a warm winter with less snowfall. Higher peat temperatures and less peat–ice beneath deeper, insulating snow covers may result in more CH₄ production and less transport restriction. Ice layers in snow deposited by freezing rain acted as flux barriers, increasing snow pore air mixing ratios. CH₄

concentrations in peat pore water increased in winter, probably due to continued low level production, reduced transport and increased solubility of CH₄ at cold temperatures. Winter season CH₄ emissions in this temperate poor fen can be significant. In a year with normal snowfall (1993–1994), winter emissions were 9.2% of the total annual emission and flux rates averaged 56 mg m⁻² day⁻¹ over the winter months (December, January and February).

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