



Spatiotemporal variability in peatland subsurface methane dynamics

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Received 25 April 2007; revised 10 December 2007; accepted 21 December 2007; published 15 April 2008.

[1] Peatlands are large natural sources of atmospheric methane (CH₄). While many studies have measured CH₄ emissions to the atmosphere, less is known about the stock and residence time of subsurface CH₄. In this study we examined dissolved CH₄ concentration in near-surface peatland pore waters of a poor fen near Québec City, Canada, in order to (1) investigate the variability in and potential controls on these concentrations and (2) combine measured dissolved CH₄ concentration with estimated bubble CH₄ stock and measured CH₄ fluxes to estimate the mean residence time of subsurface CH₄. Concentrations ranged from 1 to 450 μM during both study seasons. Depth profiles were generally consistent at one location within the peatland throughout the sampling period but varied between locations. Patterns with depth were not well correlated to pore water pH or EC; however, changes in CH₄ concentration through time in the upper 30 cm were related to temperature and water table at some locations. Depth profiles taken at 2- to 5-cm intervals revealed discrete concentration “spikes” which were often maintained throughout the season and are likely related to bubble CH₄ dynamics. Estimated subsurface CH₄ stocks indicate that even when relatively low bubble volume (5% of peat volume) is assumed, bubble CH₄ accounted for greater than half of total stocks. Calculated mean residence times were 28–120 days. This implies that CH₄ flux may lag changes in water table and temperature which happen on shorter timescales (hours or days). To improve our description of subsurface CH₄ stocks, links between dissolved and bubble CH₄ stocks and peatland CH₄ residence time, coincident measurement of pore water CH₄ concentrations, entrapped gas content and composition, diffusive CH₄ flux, and ebullition are required.

Citation: Strack, M., and J. M. Waddington (2008), Spatiotemporal variability in peatland subsurface methane dynamics, *J. Geophys. Res.*, 113, G02010, doi:10.1029/2007JG000472.

1. Introduction

[2] Peatlands play an important role in the global carbon cycle storing at least 25% of world soil carbon and emitting significant quantities of atmospheric methane (CH₄) [Gorham, 1991; Bridgham *et al.*, 2006]. While field and laboratory data have revealed that CH₄ emissions are related to water table position [e.g., Moore and Dalva, 1993; Roulet *et al.*, 1992], peat temperature [e.g., Moore and Dalva, 1993], vegetation community type [Bubier, 1995] and ecosystem productivity [Waddington *et al.*, 1996; Joabsson *et al.*, 1999], much of the variability in CH₄ efflux often remains unexplained [e.g., Daulat and Clymo, 1998].

[3] In addition to the aforementioned controls, some of this variability may also be related to localized differences in the rate of CH₄ production, oxidation and transport which could vary owing to differences in peat substrate quality and/or

quantity [e.g., Rothfuss and Conrad, 1992; Bridgham *et al.*, 1998], availability of nutrients [Keller *et al.*, 2006], metals and/or cations [Basiliko and Yavitt, 2001], vegetation community [e.g., Bellisario *et al.*, 1999; Popp *et al.*, 1999] or peat physical properties [e.g., Strack *et al.*, 2006a]. Difficulty in relating CH₄ fluxes to environmental variables may also be caused by incompatibility of the temporal scale of flux measurements and the residence time of CH₄ in peatlands.

[4] We are unaware of any study which has estimated the mean residence time of subsurface CH₄ in peat. This is likely due to the difficulty in estimating the stock of stored CH₄ in the soil. Liblik *et al.* [1997] calculated a turnover rate for CH₄ in the saturated zone of various wetlands in the discontinuous permafrost zone, but did not consider the potential stock of bubble CH₄ or emission of CH₄ via ebullition. Recently, it has been determined that bubble stocks and ebullition efflux of CH₄ are likely of equal or greater importance than dissolved stocks and diffusive fluxes. For example, Tokida *et al.* [2005] determined that CH₄ stored in bubbles accounted for 33–83% of total subsurface CH₄ stocks. Also, several studies have reported ebullition fluxes equal to [e.g., Strack *et al.*, 2005] or much greater than [e.g., Glaser *et al.*, 2004] diffusive flux measured from the same peatland. Despite this, only a few studies have measured the relative size of dissolved

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Table 1. Description of Sampling Sites, Depths, and Dates

Sampling Site	Vegetation	Dates Sampled	Sampling Interval	Depths Sampled (cm)
MA	hepatics, ^a <i>S. majus</i> , <i>R. alba</i> , <i>C. limosa</i>	24 May to 03 Sep 2003 12 May to 16 Aug 2004	weekly	15, 30, 60
MB	<i>R. alba</i>	24 May to 03 Sep 2003 04 May to 16 Aug 2004	weekly	15, 30, 60 (2003) 2, 4, 6, 8, 10, 12, 15, 18, 22, 26, 30, 45, 60 (2004)
MC	<i>S. magellanicum</i> , <i>S. majus</i> , <i>R. alba</i>	25 May to 16 Aug 2004	weekly	8, 15, 30, 45, 60
M1, M2, M3	hepatics, <i>D. intermedia</i> , <i>R. alba</i> <i>S. majus</i> , <i>S. magellanicum</i> , <i>R. alba</i> , <i>C. limosa</i> <i>S. majus</i> , <i>S. cuspidatum</i> , <i>R. alba</i>	24 May to 03 Sep 2003	biweekly	30
Mi, Mii, Miii, Miv, Mv	hepatics, <i>U. cornuta</i> , <i>U. cornuta</i> , <i>R. alba</i> hepatics, <i>D. intermedia</i> , <i>R. alba</i> hepatics, <i>S. cuspidatum</i> , <i>U. cornuta</i> , <i>D. intermedia</i> hepatics, <i>S. majus</i> , <i>D. intermedia</i> hepatics, <i>U. cornuta</i> , <i>D. intermedia</i>	06 Aug 2003	once	15, 30, 60, 90

^aHepatics include *Gymnocolea inflata* and *Cladopodiella fluitans*.

and bubble CH₄ stocks [e.g., Tokida et al., 2005]. Even studies investigating only dissolved CH₄ often cover a limited spatial and temporal extent [Clymo and Pearce, 1995; Chasar et al., 2000]. Measurements are generally taken in only a few locations varying greatly in biophysical conditions [e.g., Chasar et al., 2000]. Not only does this eliminate the opportunity to estimate the total stock of subsurface CH₄, but also the investigation of potential small-scale variability in these stocks is difficult. This small-scale variability could play a role in the variable patterns of emissions commonly observed [e.g., Dise et al., 1993; Bellisario et al., 1999; Kettunen, 2003; Strack et al., 2004]. Finally, bubbles tend to build up and then persist throughout the growing season [Kellner et al., 2004] and may redissolve owing to changes in local conditions of temperature and pressure [e.g., Kellner et al., 2006]. Therefore, the dissolved and bubble CH₄ pools are in dynamic equilibrium, with the bubble phase providing a potential disconnect between CH₄ production and efflux.

[5] The objectives of this study were to (1) investigate pore water CH₄ concentration at sites within a peatland with similar water table positions and vegetation communities, (2) determine whether variability in measured CH₄ concentration could be explained by easily measurable parameters such as water table, temperature, pH, and specific conductivity, and (3) combine measured concentrations with estimates of subsurface bubble CH₄ stocks and measured CH₄ fluxes to determine the mean residence time of CH₄ within the peatland.

2. Study Site

[6] The study was conducted at a poor fen near St. Charles-de-Bellechasse (SCB), Québec, Canada (46°40'N, 71°10'W). The fen has gentle pool-ridge topography with pool bottoms and ridge tops varying in elevation by ~50 cm and standing water in pools generally less than 30 cm deep. Peat depth was 1.5 m on average. At the edge of most pools the peat has a highly mobile, "floating" surface that rises and falls with the water table [Strack et al., 2006a]. Vegetation in these zones generally consists of a surface cover of liver-

worts *Gymnocolea inflata* and *Cladopodiella fluitans*, sedges *Rynchospora alba* and *Carex limosa*, and herbs *Utricularia cornuta* and *Drosera intermedia*. Some areas may have patchy or complete cover of *Sphagnum* mosses with *Sphagnum majus* and *Sphagnum magellanicum* being most common. All measurements were made in these low-lying, floating mats.

3. Methods

3.1. Pore Water CH₄ Concentration

[7] Pore water CH₄ concentration was determined at 11 locations across the study site between May and September in 2003 and May and August in 2004 (Table 1). In 2003, two permanent profiles were investigated with samples taken weekly at 15, 30 and 60 cm below the surface. The first profile, MA, had liverworts (*Gymnocolea inflata* and *Cladopodiella fluitans*) and *R. alba* as the dominant vegetation with sparse *Sphagnum* cover, while the second, MB, was predominantly bare peat with a few sedges (*Carex limosa*, *Carex oligosperma*, *R. alba*). MB was often flooded. Three additional locations (M1-M3) were sampled every two weeks at 30 cm depth. On 6 August 2003 additional profiles were sampled at five locations (Mi-Mv) with pore water collected from 15, 30, 60 and 90 cm below the surface. One week later water was collected from 15, 30, 45, 60, 75 and 90 cm below the surface for determination of conductivity and pH. In 2004, one additional permanent profile, MC, was installed at a low-lying area dominated by *Sphagnum* moss. Samples were collected from 8, 15, 30, 45 and 60 cm below the surface. Additional samplers were also installed in the MB profile allowing measurement at 2, 4, 6, 8, 10, 12, 18, 22, 26, 30, 45, and 60 cm depth.

[8] Pore water was collected using samplers constructed of 0.6 cm diameter PVC pipe. At one end of the pipe, the end was blocked and small holes were drilled to create a sampling window. Samplers installed above 12 cm depth had 1 cm windows, those between 18 and 26 had 2 cm windows, and 30 cm and below had 5 cm windows. The window was centered at the desired sampling depth. The window was covered with Nytex screening to prevent clogging of the

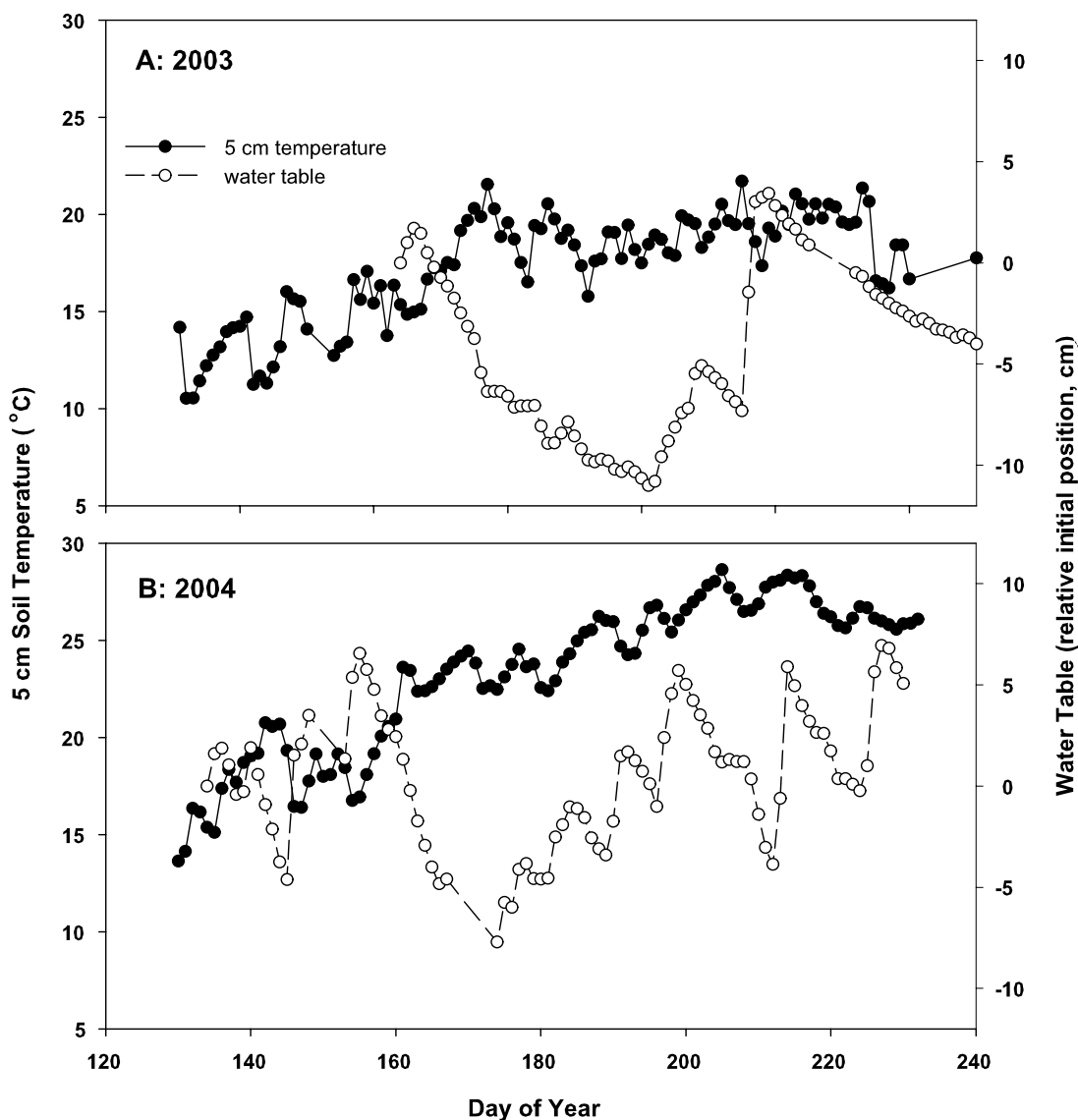


Figure 1. Water table position relative to its initial position (open symbols) and peat temperature at 5 cm below the surface (solid symbols) in (a) 2003 and (b) 2004.

sampler with peat. At the other end of the sampler a three-way valve was inserted and sealed with household adhesive. Samplers were inserted to the appropriate depth in May and left in place throughout the study; however, for the profiles taken on 6 August 2003 the sampler was inserted to the shallowest depth, water collected and then inserted deeper until the profile was complete. After insertion, water was pulled into the sampler and the valve was closed holding the water within the sampler and preventing air from traveling to the sampling depth. Before sampling, 60 mL of water was removed from the sampler to flush it and then a sample of 20 to 30 mL was collected.

[9] CH₄ concentration in the pore water sample was determined using head space equilibration [Ioffe and Vitenberg, 1982] with an equal amount of nitrogen. The concentration of CH₄ in the headspace was determined on a Varian 3800 gas chromatograph (Varian Canada Inc., Ontario, Canada). Conductivity was determined with a Checkmate 2 Conductivity/TDS probe (Corning, New York, USA) and pH with an Oakton model 300 pH probe

(Oakton Instruments, Illinois, USA) both of which were calibrated the day before measurement with manufacturer standards.

3.2. Environmental Variables

[10] Water table and soil temperature were measured at a central meteorological site within the SCB poor fen. Water table was measured using a counterbalanced float, in a stilling well, on a pulley attached to a potentiometer. Temperature was measured at 5 cm below the surface with a thermocouple. Both the thermocouple and water table recorder were attached to a CR21X data logger (Campbell Scientific, Utah, USA), measured every minute and averaged every 20 min. To determine whether pore water CH₄ concentrations were related to water table or temperature, concentration data were compared with daily averages of soil temperature or water table for the sampling dates using Pearson correlation analysis in Minitab 14.1 (Minitab Inc.). At sites where Pearson correlations were significant, stepwise regression with soil temperature and water table as potential

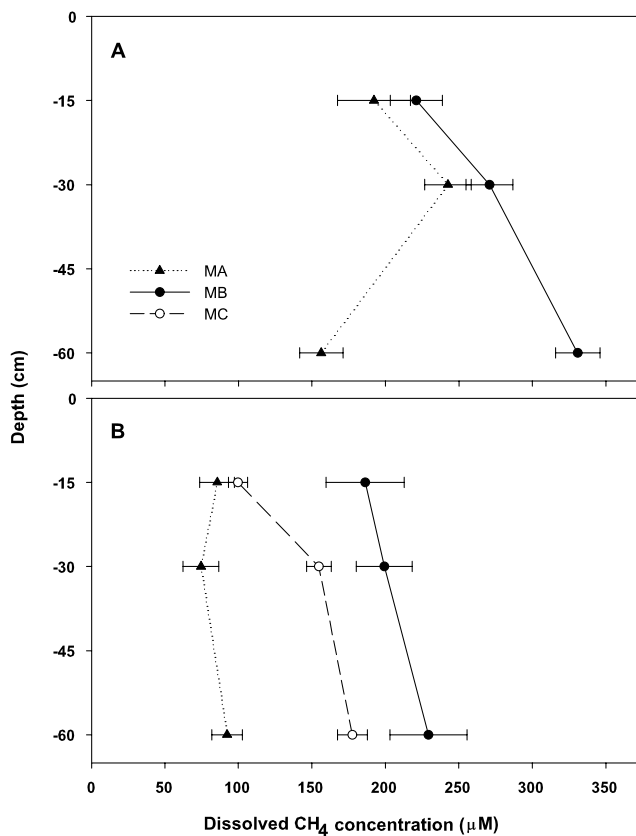


Figure 2. Coarse depth profiles (–15, –30, –60 cm only) at permanent sampling locations for (a) 2003 and (b) 2004. Values plotted are the mean seasonal concentration at that depth. Error bars give standard error.

predictors was performed with alpha value less than 0.15 required for the predictor to enter the model.

3.3. Subsurface CH₄ Stock and Mean Residence Time

[11] The mean residence time (MRT) of CH₄ within the 150 cm deep peat profile was calculated according to:

$$\text{MRT} = \text{stock}/\text{flux}, \quad (1)$$

where stock is the total amount of CH₄ present in dissolved and bubble forms within the peat profile and flux is the daily surface-atmosphere flux of CH₄ averaged on an annual basis. Because most pore water data was collected in the upper 60 cm of the peat profile, MRT was calculated for this layer of peat. However, for completeness we also estimated MRT for the entire peat profile using limited data from below this depth. All calculations were computed using 2004 data except CH₄ concentration in entrapped bubbles which was measured comprehensively only in 2003.

[12] The stock of dissolved CH₄ was calculated by weighting measured concentrations at the permanent profiles by the depth between samplers. For example, at MA the concentration measured at 2 cm was considered to apply to the depths from the surface to halfway to the next sampler at 4 cm, whose concentration applied from halfway between the 2 and 4 cm sampler to halfway between the 4 and 6 cm sampler. So, the 2 cm concentration was multiplied by 3 cm added to the 4 cm concentration multiplied by 2 cm and so

on until 60 cm depth was reached. Since limited pore water data was collected below 60 cm, average concentration determined from 90 cm samples collected at Mi-Mv was assumed to apply to the remaining profile from 60 to 150 cm deep. Because the pore water concentrations were observed to vary spatially and temporally, the stock of dissolved CH₄ was determined at all three permanent profiles (MA, MB, and MC) on 25 May and 16 August 2004, representing the beginning and end of the growing season. To determine total mass of CH₄ stored from the concentrations, the depth integrated average concentration was multiplied by the total amount of water stored in a 1 m² column in the depth of peat considered (60 or 150 cm), assuming a water table at the peat surface and a porosity of 0.95 based on previous bulk density measurements [Strack *et al.*, 2006a], and assuming a particle density of 1.4 g cm⁻³ [King and Smith, 1987].

[13] The stock of bubble CH₄ was estimated on the basis of previous measurements of entrapped bubbles varying between 3 and 20% of peat volume at this sampling site [Strack *et al.*, 2005, 2006a]. Briefly, gas content was determined in the field from changes in volumetric moisture content over the growing season as measured by Campbell Scientific CS615 moisture probes. In both cases, measured gas contents represent a change in gas content from the start of the measurement period and not absolute gas contents, and thus are conservative estimates of the bubble stock. To incorporate this variability we determined MRT for a “low” scenario in which bubble content was 5% and a “high” scenario in which bubble content was 15% of peat volume.

[14] In order to determine the proportion of CH₄ contained in subsurface bubbles and ebullition fluxes gas traps were inserted into the peat and placed at the surface, respectively [see Strack *et al.*, 2005]. The traps were constructed from inverted funnels with flexible plastic tubing fitted over the narrow end of the funnel. The opposite end of the tubing was fitted with a septum. The entire trap was filled with water and gas release determined by measuring displacement of water in the tubing by gas. Subsurface funnels were installed by cutting out a column of peat, inserting the funnel to the desired depth and replacing the peat column. Funnels were inserted at 25, 40, 60, 85 and 100 cm depth and the flexible tubing was long enough to reach from the buried funnel to the surface. The volume of gas was measured weekly, samples collected with a syringe when at least 3 mL was present, and CH₄ concentration determined on the Varian 3800 GC. The concentration of CH₄ in bubbles was computed by weighting the seasonal average concentration determined from each gas trap by the depth between samplers.

[15] The diffusive flux of CH₄ from the surface, including CH₄ released through the stem of vascular vegetation, was based on static chamber flux measurements [see Tuittila *et al.*, 2000]. Flux measurements were made weekly at nine plots located adjacent to MA, MB and MC. Average annual release was calculated by weighting flux by the number of days between measurements and determining a sum of total CH₄ emitted between May and August. This was then divided by the number of days during the sampling period and multiplied by a 180 day growing season [Gorham, 1991]. Non-growing-season

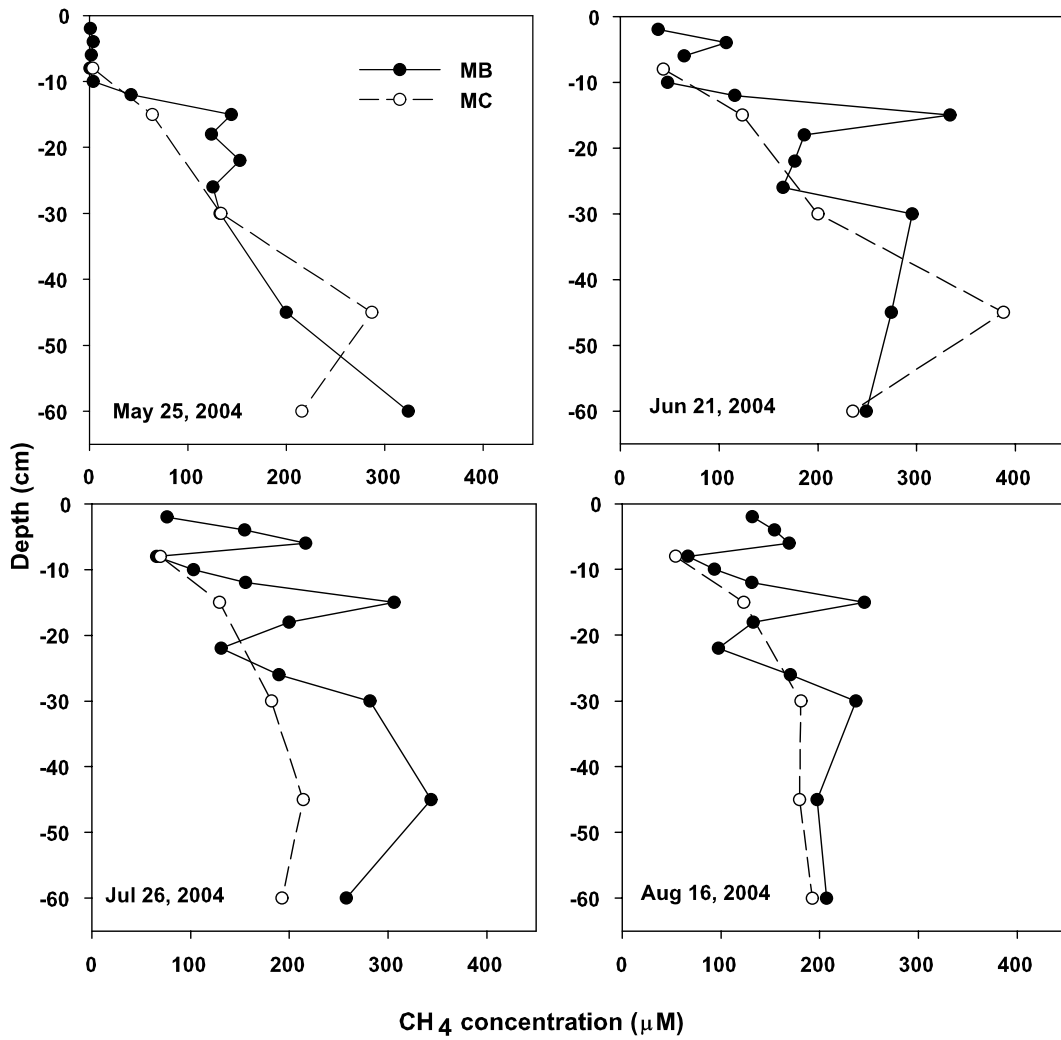


Figure 3. Detailed depth profiles at MB and MC in 2004 on several sampling days.

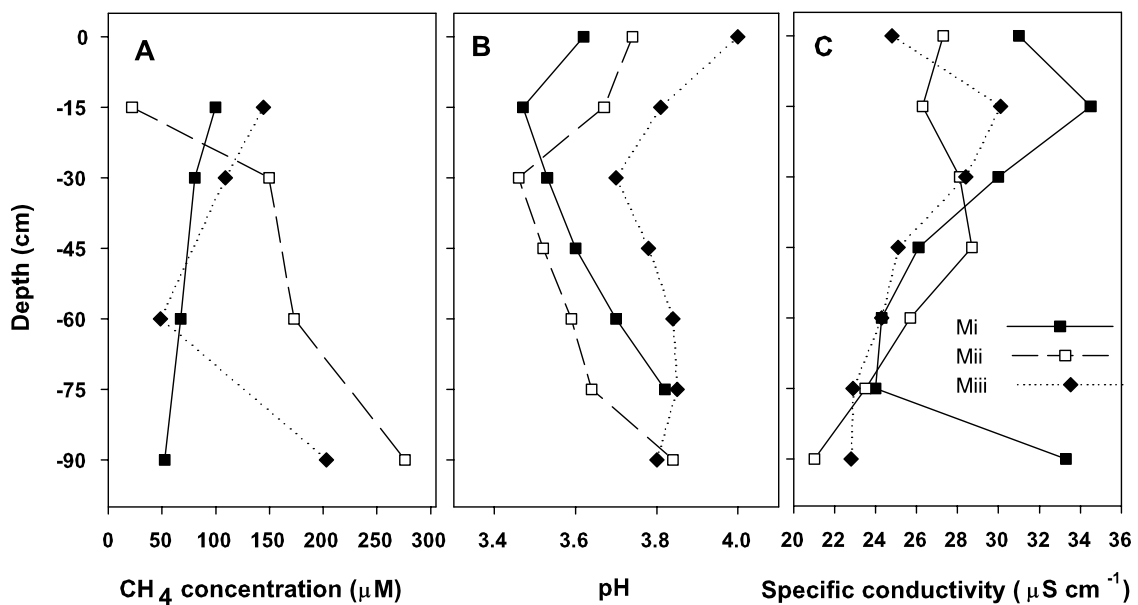


Figure 4. Depth profiles of pore water (a) CH₄ concentration, (b) pH, and (c) conductivity collected during the spatial survey in 2003. For clarity only data from Mi, Mii, and Miii are shown.

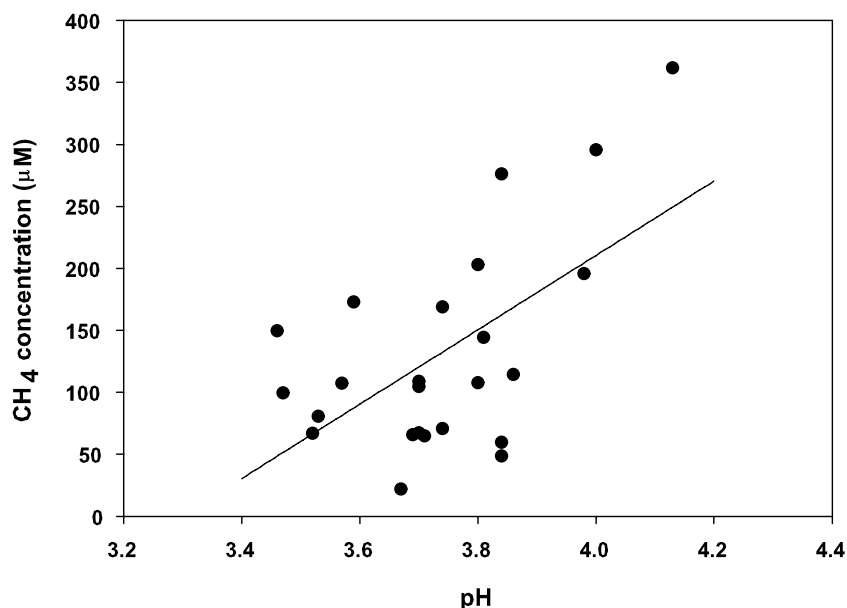


Figure 5. Pore water CH₄ concentration versus pH based on data collected from spatial survey sites and permanent profile plots 6–14 August 2003. The regression line shown is significant ($R^2 = 0.35$, $p = 0.002$).

fluxes were estimated on the basis of winter fluxes from hollows given by *Strack et al.* [2004] for the SCB poor fen.

[16] CH₄ flux via ebullition was determined on the basis of gas collected in gas traps at the surface of open water pools and saturated hollows. In total two and six gas traps were deployed in 2003 and 2004, respectively. For MRT calculations, an average flux was computed from all 2004 gas traps and extended over the 180 day growing season. Ebullitive fluxes during the winter were assumed to be negligible. The total annual diffusive and ebullitive CH₄ release was expressed as an average daily flux per square meter.

[17] MRT was determined on the basis of the stock of dissolved CH₄ at MA, MB and MC for the upper 60 cm and the entire 150 cm peat profile considering both the high and low bubble CH₄ stock scenarios.

4. Results

[18] Water table and soil temperature varied over the sampling season with 2004 having generally higher soil temperatures than 2003 (Figure 1). In 2003 water table was drawn down ~11 cm relative to its initial position by day 200 whereas in 2004 water table draw down occurred earlier (Figure 1).

[19] Considering all depths and sampling locations, dissolved CH₄ concentration varied between 3 and 440 µM in 2003 and less than 1 and 430 µM in 2004. Concentrations varied with depth and between sampling locations, with the shape of the depth profile also varying between locations. At any particular depth the concentration also varied through time over the sampling period.

4.1. Variability With Depth

[20] In 2003 only three depths were investigated at the permanent profiles limiting our ability to make observations of fine-scale variability in pore water CH₄ concentration. However, at both MA and MB the pattern of changes in concentration with depth remained fairly consistent

throughout the study period. For example, at MA the highest concentrations were generally found at 30 cm depth while at MB highest concentrations were consistently found at 60 cm depth (Figure 2).

[21] In 2004, more detailed profiles at MB and MC revealed complex patterns of CH₄ concentration (Figure 3). Despite this complexity, consistency of the depth profile through the season was still apparent (Figures 2 and 3).

[22] While it appears that CH₄ concentration generally increased with depth at MB and MC, this pattern is not apparent at MA (Figure 2) and also does not appear at several of the spatial profiles sampled (Mi, Mii and Miii shown in Figure 4). There is a clear increase of CH₄ concentration with depth at Mii, a small decrease with depth at Mi, but no clear depth related pattern at any of other profiles. Comparison of CH₄ concentration with depth profiles of pH and specific conductivity do not reveal any correlation between these variables at any particular location (Figure 4). When the data was pooled there was a significant positive relationship ($p = 0.002$, $R^2 = 0.35$) between CH₄ concentration and pH (Figure 5), however, this is driven by two points with both high pH and CH₄ concentration, again suggesting limited correlation between these variables.

4.2. Spatial Variability

[23] In 2003 the standard error of the five measurements made at 30 cm depth on a given day (MA, MB, M1–M3) was similar to the standard error at a particular location over the season. CH₄ concentrations at this depth varied between locations from approximately 100 to 300 µM, although the pattern of concentrations changed between sampling dates. For example, on July 5, 2003 M2 had the highest concentration at 298 µM while M3 had the lowest concentration, 149 µM. Then on 16 July 2003, M3 had the highest concentration at 316 µM while M2 had one of the lowest concentrations measured that day (Figure 6).

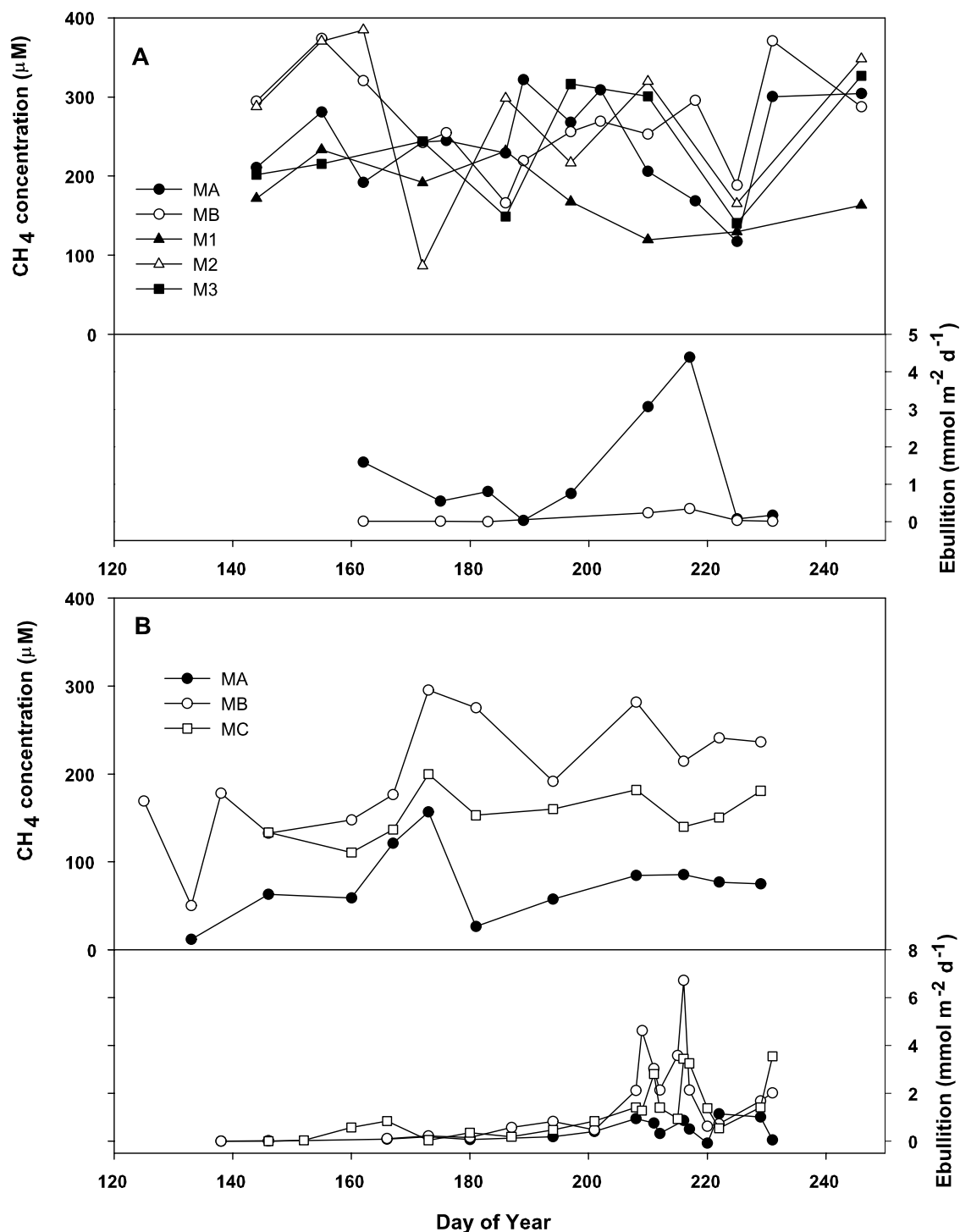


Figure 6. Temporal patterns of dissolved CH₄ concentration at -30 cm (top portion of plot) and CH₄ released via ebullition (bottom portion) in (a) 2003 and (b) 2004.

[24] An examination of the permanent concentration profiles in 2004 reveals that CH₄ concentration profiles averaged over the sampling season varied between MA, MB and MC. MB had significantly higher concentrations at 15 cm depth while MA had significantly lower concentrations at both 30 and 60 cm depths (ANOVA, $p < 0.05$; Figure 2). The extent of this variability was also shown in the spatially distributed profiles collected in 2003 (Figure 4). These data revealed that not only were concentrations at a particular depth variable

across space, but that depth concentration profiles also differed between sampling locations.

4.3. Temporal Variability

[25] Considering the depths at MA and MB which were installed in both 2003 and 2004, seasonal average CH₄ concentrations were slightly higher in 2003 at both sites. At MA, concentrations averaged 160–240 μM in 2003 and 70–90 μM in 2004, while at MB concentrations averaged

Table 2. Pearson Correlation Between CH₄ Concentration, Water Table, and Temperature for All Locations Sampled in 2004^a

Site	Depth (cm)	Water Table	Temperature
MA	15	-0.09	0.29
	30	0.27	0.39
	60	0.37	0.57
MB	2	0.83**	0.80**
	4	0.80**	0.76**
	6	0.82**	0.74**
	8	0.82**	0.73**
	10	0.79**	0.77**
	12	0.76**	0.68*
	15	0.49	0.73**
	18	0.58	0.63*
	22	0.12	0.05
	26	0.48	0.38
	30	0.34	0.68*
	45	0.16	0.46
	60	-0.16	-0.49
MC	8	0.39	0.58
	15	0.66*	0.62
	60	0.43	0.36
	45	0.25	-0.26
	60	0.14	-0.37

^aSignificant correlations are indicated (*p < 0.05, **p < 0.01).

220–330 μM in 2003 and 190–230 μM in 2004 (Figure 2). Despite the change, concentrations at MB were higher than MA in both years suggesting that at least on a seasonal average basis, spatial differences are consistent through time. As discussed above, however, on the timescale of days to weeks, patterns of concentration can change, resulting in inconsistency in the relative CH₄ concentration between sites.

[26] Across the season there is an indication of an increase in CH₄ concentration at depths less than 30 cm, but no pattern of concentration seasonality exists deeper in the profile. Even at the shallow depths this increasing pattern is often obscured by large declines in concentration throughout the sampling period. Within a particular profile, rises and falls in concentration generally occur coincidentally at all depths, resulting in similar patterns of concentration profiles throughout the season (Figure 3). In contrast, on many sampling days, concentration changes did not occur coincidentally between sampling locations and this resulted in the observed changes in the relative concentration pattern between sites (Figure 6).

[27] Because large-scale changes in temperature and water table occur across the sampling sites, it appears that these are not the only controllers of the concentration changes, or at least that individual sampling sites respond to different extents to these controllers. On the basis of Pearson correlation, in 2003 CH₄ concentration was only significantly correlated to water table at MA and temperature at MB at 30 cm depth. In 2004, correlations were observed between CH₄ concentration and water table at MB at depths 12 cm below the surface and shallower and at MC at 15 cm depth. Concentrations were also correlated to temperature at MB at 18 cm below the surface and shallower and at 30 cm depth (Table 2). In this case it is difficult to identify whether water table or temperature is the more dominant control on pore water CH₄ concentration as they are weakly correlated (Pearson correlation = 0.564, p = 0.07) to each other. Both are also significantly positively correlated to day of year, providing further evidence that CH₄ concen-

tration increases throughout the growing season (Figure 6). Stepwise regression revealed that in cases where significant correlations were observed with both water table and soil temperature (FB above -15 cm), water table always entered the regression in the first step, yet the further inclusion of temperature in the model resulted in higher adjusted R-squared in all cases. Overall, it appears that these controls are generally only important at depths shallower than 30 cm.

[28] Pore water CH₄ concentration may also be related to presence and size of stored bubble CH₄ [Rothfuss and Conrad, 1998; Strack et al., 2005]. Large reductions in pore water CH₄ concentrations occurred coincidentally at the majority of sites toward the end of the season (Figure 6). These reductions in concentration occurred during periods of higher measured ebullition (Figure 6), indicating further evidence of a link between entrapped gas dynamics and dissolved CH₄ concentration.

4.4. Mean Residence Time of Subsurface CH₄

[29] The total stock of dissolved CH₄ in the upper 60 cm of the peat profile was between 0.03 at MA and 0.1 mol m⁻² at MB (MC was intermediate); in the total 150 cm peat profile estimates varied between 0.15 at MA and 0.22 mol m⁻² at MB. At all three permanent profiles the stock of dissolved CH₄ was greater at the end of the growing season than at the beginning. The proportion of CH₄ in subsurface gas bubbles generally decreased with depth from 28% at 25 cm depth to <1% at 100 cm depth. The weighted average proportion of CH₄ in subsurface bubbles was 20% in the upper 60 cm and 10% for the full 150 cm profile. In the upper 60 cm bubble CH₄ stock was 0.3 and 0.8 mol m⁻² for the low and high bubble content scenarios, respectively. Total peat profile bubble CH₄ stock was 0.3 mol m⁻² for the low scenario, and 1.0 mol m⁻² for the high scenario.

[30] Diffusive CH₄ flux was highly variable between the sampling plots. Total CH₄ flux over the 110 days of the sampling period was between 0.5 and 10 mol m⁻² with an average flux of 2.0 mol m⁻². This rate of CH₄ emission integrated over 180 days resulted in a CH₄ release of 3.3 mol m⁻². Non-growing-season CH₄ flux from hollows was assumed to be 6 μmol m⁻² d⁻¹ [Strack et al., 2004] for a total non-growing-season release of 0.012 mol m⁻².

[31] CH₄ flux via ebullition was also spatially and temporally variable with a range of CH₄ release over a 100 day sampling period from 0.05 to 0.6 mol m⁻². The average ebullitive CH₄ flux was 0.3 mol m⁻² resulting in an estimate of total emissions over the 180 day growing season of 0.5 mol m⁻². Combining estimates of diffusive and ebullitive CH₄ flux resulted in a total CH₄ release of 3.8 mol m⁻², or 0.01 mol m⁻² d⁻¹ averaged on an annual basis.

[32] On the basis of the calculated stock and flux of CH₄ MRT was estimated as 28 to 87 days for the upper 60 cm. For the full 150 cm poor fen peat profile, the MRT for CH₄ was between 46 and 120 days.

5. Discussion

[33] Pore water CH₄ concentrations up to 440 μM observed in this study were similar to those reported elsewhere in literature [Clymo and Pearce, 1995; Waddington and Roulet, 1997; Rothfuss and Conrad, 1998; Chasar et al.,

2000; *Blodau et al.*, 2004, 2007]. Increasing concentrations with depth, up to 1800 μM , have been reported below a fen and bog in Minnesota where a discrete spike of 7000 μM was observed at 1.6 m depth [*Chasar et al.* 2000]; however, these higher values were observed at depths close to 3 m, much greater than those considered in this study.

5.1. Spatiotemporal Variability of Dissolved CH₄

[34] Spatial patterns of dissolved CH₄ stock changed throughout the growing season. The pore water CH₄ concentration at a particular point in the peatland will be controlled by the difference between the rate of CH₄ addition to that point, via production and translocation, and the rate of CH₄ loss via oxidation, translocation or emission. Methane in peatlands is produced microbially under highly reduced conditions which occur in water saturated peat. Because all sampling sites were located in hollows with highly mobile surfaces, water table remained within the top 5–10 cm throughout the study. However, samplers close to the surface at MA and MC in 2004 may have experienced short periods above the water table in which CH₄ production would have been reduced. Also, as the water table drops, distance from each point to the unsaturated zone is reduced. Since CH₄ concentrations are expected to be lower in unsaturated peat, this will increase concentration gradients, thereby enhancing the diffusive efflux of CH₄ and reducing concentration if production remained constant. Finally, a lower water table position reduces hydrostatic pressure. According to Henry's law, the concentration of dissolved gas is inversely related to total pressure. Thus, reductions in pressure would encourage CH₄ to migrate from a dissolved to gaseous (bubble) phase. All of these factors likely interact to result in the positive relationship between water table position and CH₄ concentration observed at several of the surface sampling sites. That this effect is limited to the near-surface peat is expected, as the potential for air entry and significant enhancement of diffusion gradients would be minor at depth.

[35] Positive relationships were also observed between concentration and temperature. CH₄ production is positively related to temperature [*Dunfield et al.*, 1993], indicating that higher temperatures should result in higher pore water CH₄ concentrations if losses remain constant. Temperature can also affect concentrations according to Henry's law since more gas can be dissolved at lower temperatures. This would suggest a negative relationship between concentration and temperature. This likely plays a minor role in controlling concentrations on the timescale investigated as no negative temperature-concentration relationships were observed. Finally, since temperature was related to the day of year, it may represent a seasonal accumulation of dissolved CH₄ as is evident in Figure 6. Several studies have observed a time lag between CH₄ production and emissions [e.g., *Baird et al.*, 2004; *Blodau et al.*, 2004; *Strack et al.*, 2005], indicating that a pool of stored CH₄ must be developed before a sufficient concentration gradient, or pressure for gas release, is achieved to result in measurable emissions. Similar to water table, relationships with temperature were limited to depths above 30 cm. This is expected since peat temperature at depth varies little seasonally, and other studies have observed little temporal variability in concentrations of CH₄ deeper in peat [*Clymo and Pearce*, 1995]. Since temperature was not measured at depth in this

study, the lack of a temperature-CH₄ concentration relationship should be interpreted carefully. CH₄ pore water concentration may be related to temperature at the same depth; however, detailed temperature and pore water CH₄ profiles would be needed to address this question.

[36] Factors controlling the spatial variability in dissolved CH₄ concentration are not as clear. There is a weak positive relationship across sampling sites between pH and CH₄ concentration. This is consistent with laboratory incubations that have observed reduced production rates at low pH [*Valentine et al.*, 1994]. However, the correlation observed in this study is driven by a few samples at high concentration and pH, with a lot of scatter under more acidic conditions. Thus, pH is not an important control on dissolved CH₄ concentration at this site and cannot explain small-scale spatial variability observed in depth profiles (Figure 4).

[37] Differences in the pore water CH₄ pool could be related to local differences in peat and pore water chemistry potentially leading to variable rates of CH₄ production and oxidation. It is unclear how substrate variability and peat chemistry varies at a fine scale across a peatland, particularly between zones that are hydrologically similar. Moreover, it is likely that a large portion of uncertainty in rates of formation and flux of CH₄ is due to variations in decomposition pathways in the peat [e.g., *Duddlestone et al.*, 2002]. More research is required to determine the spatial scale of this variability and whether it is an important source of variability for subsurface CH₄ stock and CH₄ flux. The presence of vascular vegetation may play a role in this small-scale variability as roots provide substrate via root litter and exudates and also transport oxygen below the water table [e.g., *Popp et al.*, 1999]. Thus, understanding the variability of, and controls on, substrate availability for methanogenesis is particularly important as peatland vegetation communities are expected to shift in response to disturbance such as climate change [e.g., *Strack et al.*, 2006b].

[38] As suggested elsewhere, [*Rothfuss and Conrad*, 1998; *Chasar et al.*, 2000] spatial variability of pore water CH₄ concentrations may also be related to entrapped gas bubbles. Laboratory studies [*Baird et al.*, 2004] have shown that pore water CH₄ concentrations must build up before the release of entrapped gas via ebullition is observed. These higher dissolved CH₄ concentrations drive the equilibrium toward bubble CH₄; however, the partitioning between the phases depends on factors such as absolute pressure, temperature, bubble radius, and the partial pressure of other dissolved gases [e.g., *Kellner et al.*, 2006]. Therefore, the presence of entrapped bubbles is likely to be associated with high concentrations of dissolved CH₄ because these high concentrations are necessary to build entrapped bubble volume and also because, if these bubbles remains entrapped and increase in volume, it will drive equilibrium toward higher dissolved CH₄ concentrations.

[39] The release of CH₄ via ebullition should result in a decrease in pore water CH₄ concentrations [*Harrison*, 2006]. Large-scale releases of CH₄ have been associated with reductions in dissolved CH₄ concentrations [*Romanowicz et al.*, 1995; *Waddington and Roulet*, 1997]; however the relationship with ebullition on a small spatial scale is less clear [*Strack et al.*, 2005] likely because measurements of pore water concentrations and ebullition are not made at identical locations. *Strack et al.* [2006a] demonstrated that

entrapped gas volume and release varied between peatland locations with similar hydrology and vegetation. It is probable that much of the small-scale spatial variability observed in the present study relates to local differences in gas trapping ability between the studied peatland hollows. It remains unclear what peat properties best describe its ability to entrap bubbles, although bulk density and pore size distribution are the most likely candidates. Further research regarding the variability of these parameters within and between peatlands is necessary to improve our processed-based understanding of peatland function.

[40] Temporally, it is expected that dissolved CH₄ should build up throughout the season as the soil warms and production rates increase. Depending upon temperature, pressure, presence of existing bubbles, etc. the concentration may become high enough that some of the dissolved CH₄ will partition into bubbles. When the volume of the entrapped gas bubbles, governed by temperature and pressure, is large enough to exceed the peat's gas-holding capacity, it may be released [Strack *et al.*, 2005]. The release results in localized reductions in pressure [e.g., Glaser *et al.*, 2004; Strack *et al.*, 2006a], and favors the transfer of dissolved CH₄ into any remaining entrapped bubbles, resulting in lower pore water CH₄ concentrations. Sudden reductions in CH₄ concentration observed in this study were often coincident with or just followed a period of low atmospheric pressure, a known trigger for peatland ebullition [Glaser *et al.*, 2004; Strack *et al.*, 2005; Tokida *et al.*, 2005]. However, at sampling locations where concentrations were consistently low throughout the season (Figure 6, M1 in 2003) sudden concentration changes were not observed, supporting the idea that a threshold concentration must be exceeded before bubbles will build up and be released [Baird *et al.*, 2004]. Despite this evidence, predictive links between pore water CH₄ concentrations, ebullition and atmospheric pressure remain unclear because of infrequent sampling and lack of knowledge of spatial variability in controlling factors such as peat physical properties, substrate availability, local pressure, and temperature. Controlled laboratory experiments and detailed field sampling is required to quantify these relationships.

[41] In the present study we estimated diffusive and ebullitive CH₄ efflux as 2.0 mol m⁻² and 0.5 mol m⁻², respectively over the 180 day growing season. As in previous studies, the low number of gas traps and frequency of sampling limited certainty in our estimate of ebullition. Since accurate measurements can only be made where disturbance is limited, gas trap location was limited to six locations near the boardwalk. Also, since measurements were made manually, temporal resolution was limited. While the estimated ebullition flux is similar to that determined at this research site in previous studies [e.g., Strack *et al.*, 2005], the flux is much lower than some estimates of ebullition from other peatlands. Comas *et al.* [2007] estimate that 1.25 mol m⁻² of CH₄ were lost in a five day ebullition event and Glaser *et al.* [2004] estimated that over 2 mol m⁻² of CH₄ may be lost in single large-scale ebullition event. Owing to our small number of gas traps, it is possible that we underestimated ebullition flux by missing hot spot areas of preferential bubble release. It is also likely, as suggested by Strack *et al.* [2005] that our chamber estimates of diffusive fluxes include some "constant"

ebullition flux as measured fluxes were often higher than potential diffusive fluxes determined from concentration gradients. Since it is not possible to separate diffusion and CH₄ release via regular bubble release in chamber measurements they are grouped here as diffusive fluxes. The presence of vascular vegetation can also enhance CH₄ efflux [e.g., Waddington *et al.*, 1996]; in this study CH₄ efflux through vegetation would be captured by chamber measurements and is thus also included under diffusive fluxes.

[42] Improved ecosystem-scale estimates of ebullition are unlikely using chambers and gas traps as these methods are labor intensive and spatially limited. The use of eddy covariance techniques in peatlands may improve our estimate of total ecosystem CH₄ efflux; however, Tokida *et al.* [2007] outline several limitations of eddy covariance for accurately quantifying episodic releases of CH₄ from peatlands.

5.2. Stock and Mean Residence Time of Subsurface CH₄

[43] Calculations of the subsurface stock of CH₄ in the peat profile indicate that, even assuming a relatively small pool of entrapped gas at 5% of peat volume with 10% CH₄, bubble CH₄ accounts for more than half of the total subsurface stock. This implies that understanding controls on entrapped bubble CH₄ is important for understanding peatland CH₄ dynamics [see also Rosenberry *et al.*, 2006]. Also, calculated MRT of length shorter than the growing season suggest that, as observed in previous studies, shifts in water table position or temperature during this time should be reflected in CH₄ efflux. However, it also suggests that response to changing environmental conditions will not be immediate, as stored subsurface CH₄ may continue to be released despite reduced production rates [e.g., Blodau *et al.*, 2004], or subsurface stock may need to be "rebuilt" before increased flux is observed [Moosavi *et al.*, 1996; Baird *et al.*, 2004]. This is likely responsible for the observation that a better relationship between peatland CH₄ flux and environmental variables is found when monthly or seasonal averages are considered than when daily observations are used [e.g., Treat *et al.*, 2007].

[44] In calculating MRT we have not considered loss of CH₄ via oxidation. This likely plays a limited role in the current study as study plots had water tables within 5 cm of the surface throughout the growing season. However, if significant oxidation of CH₄ occurs at inundated site in the rhizosphere, ignoring oxidation would result in an overestimation of MRT. Since vascular vegetation cover was sparse in the study locations, rhizospheric oxidation is likely minimal.

[45] The calculated turnover times of Liblik *et al.* [1997] are limited because they did not consider stored bubble CH₄, or loss of CH₄ via ebullition or oxidation. They calculated turnover times of 1–4 days at poor fen sites and sites with abundant graminoid vegetation, whereas at drier sites turnover times were up to 16 years. Poor fen turnover times likely represent underestimates because entrapped bubble CH₄ was not considered in the subsurface CH₄ stock, but likely was important given measured dissolved CH₄ concentrations of 125–500 μM. At drier locations, water tables were up to 40 cm below the surface. Thus, at these locations, oxidation probably represented an

important loss of CH₄ from the subsurface stock [Liblik *et al.*, 1997], leading to overestimation of turnover time. Therefore, quantification of CH₄ oxidation will be important for MRT calculation in drier microforms. In order to better constrain future MRT estimates we argue that coincident measurement of dissolved and bubble CH₄ profiles, and CH₄ flux via diffusion, ebullition and plant mediated transport is required.

6. Conclusions

[46] The MRT of CH₄ in a poor fen was 28–120 days. This suggests that shifts in CH₄ flux may lag changes in environmental controls such as water table and temperature which occur on daily or weekly timescales. This may explain why seasonal averages of water table and temperature explain variability in CH₄ better than daily measurements. Bubble CH₄ is an important component of the total peat CH₄ stock.

[47] The concentration of CH₄ dissolved in peatland pore water varied throughout the season and between hydrologically similar hollows within a poor fen. Variability across space and with depth was not related to conductivity and only weakly related to pH suggesting that these easily measurable parameters are not major controllers or appropriate predictors for dissolved CH₄ concentration.

[48] At depths above –30 cm, CH₄ concentration was related to seasonal shifts in water table and temperature. There were, however, rapid shifts in CH₄ concentration that could not be explained by these relationships, and are more likely linked to dynamics of entrapped bubble CH₄.

[49] In order to improve our understanding of the links between entrapped gas dynamics and dissolved CH₄ concentration, and constrain MRT estimates, controlled laboratory experiments and fine-scale field sampling with coincident measurement of entrapped gas, pore water CH₄ concentrations, diffusive CH₄ flux and ebullition are required. We also require an improved understanding of the relationship of peat properties, such as pore size distribution, vegetation composition, and bulk density, to bubble trapping ability, and the spatial scale over which these properties vary. Additionally, there is a need to understand the scale of spatial variability of potential CH₄ production, peat/pore water chemistry, and sources of substrate and links to variability in CH₄ pool size.

[50] **Acknowledgments.** We thank J. R. van Haarlem, Erik Kellner, Monique Waller, Pete Whittington, Bronwyn Findlay, Katy Shaw, Liz Buckton, Scott Ketcheson, and Jason Cagampan for assistance in the field and laboratory. We also kindly thank Nirom Peat Moss for site access. Comments by the associate editor and two anonymous reviewers greatly improved this manuscript. This research was funded by a Canadian Foundation for Climate and Atmospheric Sciences (CFCAS) research grant and a NSERC Discovery Grant to J. M. W.

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