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Evening Methane Emission Pulses from a Boreal Wetland Correspond to Convective Mixing in Hollows

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Evening methane emission pulses from a boreal wetland correspond to convective mixing in hollows

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[1] Spatial and temporal heterogeneity of methane flux from boreal wetlands makes prediction and up-scaling challenging, both within and among wetland systems. Drivers of methane production and emissions are also highly variable, making empirical model development difficult and leading to uncertainty in methane emissions estimates from wetlands. Previous studies have examined this problem using point-scale (static chamber method) and ecosystem-scale (flux tower methods) measurements, but few studies have investigated whether different processes are observed at these scales. We analyzed methane emissions from a boreal fen, measured by both techniques, using data from the Boreal Ecosystem-Atmosphere Study. We sought to identify driving processes associated with methane emissions at two scales and explain diurnal patterns in emissions measured by the tower. The mean methane emission rates from flux chambers were greater than the daytime, daily mean rates measured by the tower, but the nighttime, daily mean emissions from the tower were often an order of magnitude greater than emissions recorded during the daytime. Thus, daytime measurements from either the tower or chambers would lead to a biased estimate of total methane emissions from the wetland. We found that the timing of nighttime emission events was coincident with the cooling and convective mixing within hollows, which occurred regularly during the growing season. We propose that diurnal thermal stratification in shallow pools traps methane by limiting turbulent transport. This methane stored during daytime heating is later released during evening cooling due to convective turbulent mixing.

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1. Introduction

[2] Methane emissions from natural wetlands are estimated to range from 100 to 231 Tg per year, which makes wetlands the largest natural source of methane [*Solomon et al.*, 2007]. Boreal wetlands are a major source of methane (CH_4) emissions [*Mikaloff-Fletcher et al.*, 2004b, *Harriss et al.*, 1985] and are expected to have a net warming effect on global climate [*Frolking et al.*, 2006]. Although total wetland area has been constrained for North America and Eurasia [*Bridgham et al.*, 2006], substantial uncertainty exists in the total emissions from these wetlands [*Mikaloff-Fletcher et al.*, 2004a; *Olivier et al.*, 2005; *Wuebbles and Hayhoe*, 2002]. Much of this uncertainty is due to the substantial variation in emission rates among wetlands [*Bubier and Moore*, 1994; *Moore and Knowles*, 1990; *Saarnio et al.*, 2007] and the difficulty of predicting emission rates from habitat classification and remote-sensing data [*Christensen et al.*, 1996; *Potter et al.*, 2006]. Estimates from a single wetland are affected by spatial [*Alm et al.*, 1999; *Dinsmore et al.*, 2009b] and temporal [*Dinsmore et al.*, 2009a; *Mikkela et al.*, 1995; *Windsor et al.*, 1992] variability. Locally, emission rates are often correlated with environmental parameters including soil temperature [*Hargreaves et al.*, 2001; *Høj et al.*, 2005; *Wille et al.*, 2008], water table position [*Bubier*, 1995; *Heikkinen et al.*, 2002; *Huttunen et al.*, 2003], soil moisture content [*Granberg et al.*, 1997; *Rhew et al.*, 2007], vegetation coverage [*Bartlett et al.*, 1992; *Joabsson and Christensen*, 2001], and interactions among several of these variables [*Christensen et al.*, 1995; *Nakano et al.*, 2000; *Rask et al.*, 2002]. Integrating flux rates across spatially variable landscapes improves emission estimates [*Christensen et al.*, 2007; *Dalva et al.*, 2001; *Flessa et al.*, 2008; *Huttunen et al.*, 2003], but this method of up-scaling requires fine-scale spatial models of parameters that drive CH_4 emission.

[3] Emissions of CH_4 from wetlands are commonly measured using the flux chamber method [*Moore and Roulet*, 1991]. In this method, a small area of wetland soil (typically $<1\ m^2$) is covered with an airtight chamber, and the flux is

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calculated from the change in headspace CH_4 concentration over time [*Levy et al.*, 2011]. These short-term measurements have high certainty for the area covered by the chamber, but many chambers are needed to describe spatial variability within a wetland. Data from manually operated chambers often have poor temporal resolution due to the amount of time required to sample the chambers and measure the headspace gas concentration. As a result, few studies using chambers attempt to characterize temporal dynamics at timescales shorter than weeks [*Mikkelä et al.*, 1995; *Waddington et al.*, 1996; *Whalen and Reeburgh*, 1988]. Furthermore, chamber sampling may have artifacts due to collar installation, differential heating [*Denmead*, 2008], headspace pressure, and lack of turbulence within the headspace [*Moore and Roulet*, 1991; *Pihlatie et al.*, 2013].

[4] Whereas the chamber method yields measurements that are spatially and temporally restricted, tower-based flux measurements integrate the flux over much larger spatial scales [*Fan et al.*, 1992; *Riutta et al.*, 2007] and have superior temporal resolution [*Laurila et al.*, 2012]. In both the flux gradient tower method and the eddy-covariance tower method, the footprint of the flux tower is proportional to the tower height, atmospheric boundary layer conditions [*Hargreaves et al.*, 2001], and surrounding topography [*Vesala et al.*, 2008]. These tower-based micrometeorological methods have the advantage of larger measurement area than chambers, which means that the tower measurements integrate across greater spatial variability. However, because tower measurements are sensitive to micrometeorological conditions, their effective footprint is variable depending upon wind direction, atmospheric stratification, and turbulence levels.

[5] Efforts to integrate CH_4 flux from plant-scale chamber measurements to wetland-scale tower-based measurements have shown reasonably good correspondence between the two methods. *Alm et al.* [1999] measured CH_4 flux from a bog using both chambers and a tower and found that the tower measurements were within the range of flux measured by chambers in different microhabitats. Others have shown correspondence between flux tower measurements and area-weighted estimates from chamber measurements based upon habitat classifications [*Schrier-Uijl et al.*, 2010], microtopography [*Clement et al.*, 1995], and plant communities [*Riutta et al.*, 2007]. *Forbrich et al.* [2011] showed that separate predictive models for three habitat classifications produced better correspondence with the tower than a single model for an entire wetland. However, a similar area-weighted model by *Hendriks et al.* [2010] overestimated the flux measured by a tower. Although these studies have shown encouraging results, there remains a critical need to reconcile chamber-based measurements with flux tower measurements, particularly with regard to driving forces at disparate scales including temporal dynamics in emissions that occur over timescales that are not readily resolved by the chamber method.

[6] We used an existing data set of chamber and tower measurements (previously not analyzed) from the Boreal Ecosystem-Atmosphere Study [*Bubier et al.*, 1998; *Crill and Varner*, 1998; *Sellers et al.*, 1997] to compare chamber-based measurements of CH_4 emissions to tower-based measurements for a single wetland. We sought to address three questions using this data set: (1) How do measurements of CH_4 flux differ between the chamber and tower measurement techniques? (2) Which drivers of CH_4 flux are important at these two measurement scales? and (3) Are episodic events in flux rate apparent when using the tower method?

2. Methods

2.1. Description of the Field Site

[7] The Boreal Ecosystem-Atmosphere Study (BOREAS) was an international collaborative project conducted from 1990 until 2000, with the purpose of quantifying the exchange of greenhouse gases between the boreal ecosystem and the atmosphere [*Sellers et al.*, 1997]. Substantial effort was made to measure the exchange of carbon dioxide (CO_2) and CH_4 at nested spatial scales using multiple methods. Previous publications provide detailed descriptions of the methods, data, and findings associated with the project [*Bellisario et al.*, 1999; *Bubier et al.*, 1995b; *Lafleur et al.*, 1997; *Sellers et al.*, 1997]. During the 1996 field season, BOREAS investigators conducted intensive sampling of CH_4 and CO_2 flux from a minerotrophic fen using static chambers and tower-based methods. The fen (tower fen) is located in the Northern Study Area (NSA), near Thompson, Manitoba Canada and is characterized by hummock-hollow microtopography [*Lafleur et al.*, 1997]. The fen is approximately 50 ha in area and is surrounded by boreal forest. *Lafleur et al.* [1997] describe the hydrology, plant composition, and climate of the fen.

2.2. Static Chamber Measurements

[8] Methane emissions were measured using the static chamber method [*Bubier et al*, 1998; *Bubier et al.*, 1995b; *Moore and Roulet*, 1991] from June to October 1996. Opaque chambers ($0.053\,m^2$) were used to collect samples of headspace gas from permanent collars embedded in the peat. Twelve chambers were sampled along spurs off of a boardwalk leading to the flux tower. The chambers were sampled during the day (P. Crill, personal communication, 2011) by collecting five samples of headspace gas at 2–4 min intervals and measuring the CH_4 concentration by gas chromatography [*Bubier et al.*, 1998]. The CH_4 flux from the chambers was calculated from the regression of CH_4 concentration in the chamber versus time. Uncertainty in the CH_4 flux measurements was estimated at less than 1%, with a minimum detectable flux of 0.07 nmoles $CH_4\,m^{-2}\,s^{-1}$ [*Bubier et al.*, 1998]. Chambers were sampled at approximately 7-day intervals for a total of 20 sampling dates. Data were excluded when ebullition was observed while manipulating the chambers [*Bubier et al.*, 1998]. Flux measurements were obtained from a minimum of six chambers on each date, with at least 10 chambers on 14 of the sampling dates. The CH_4 flux data from the chambers were included in a regional analysis by *Bubier et al.* [2005].

2.3. Tower Flux Measurements

[9] The tower-based CH_4 flux measurements from the BOREAS NSA fen tower have not been published previously. Methane flux was measured over the fen surface from May to November 1996 using the flux gradient technique from wind speeds recorded at heights of 2.5, 4.0, and 6.0 m [*McCaughey et al.*, 1999]. Half-hourly averaged concentration gradients of CH_4 were calculated from measurements every 6 min using a gas chromatograph with a flame ionization detector at heights of 3.59 m and 6.65 m [*Crill and*

Varner, 1998]. The gas chromatograph had an analytical precision of 0.2%.

[10] The CH_4 flux was measured using the flux gradient approach (equation (1)) where F_s is the mole flux density (nmoles $m^{-2} s^{-1}$) following Monin-Oboukhov similarity theory [*Oke*, 1987].

$$F_s = -K_s \frac{\Delta c}{\Delta z} \quad (1)$$

$K_s = kzu_*/\Phi_s$ is the eddy diffusivity ($m^2 s^{-1}$), c is the amount of CH_4 (nmoles m^{-3}), Δz is the distance between the two measurement heights z_1 and z_2 (m), $u_* = k\left(\frac{\Delta u}{\Delta \ln z}\right)\Phi_m^{-1}$ is the friction velocity ($m\,s^{-1}$), determined from the slope of the wind profile. k is the von Karman constant (=0.4), and u_* and K_s are corrected for atmospheric stability by Φ_m and Φ_s following *Businger et al.* [1971]. u_* and K_s were determined using momentum flux and heat flux measured based on log-law similarity in an adjusted surface layer.

2.4. Quality Control for Tower Data

[11] In general, micrometeorological techniques are limited to ideal sites where the flow is fully adjusted to the surface and where Monin-Obukov similarity theory holds [*Kaimal and Finnigan*, 1994]. Forest or short shrub cover surrounds the fen, which is rougher than the fen surface. Transitions from an upwind rough forested surface to a relatively smooth fen lead to a change in drag on the flow resulting in the flow accelerating at the transition and adjusting to the new surface. The flow equilibrates to the fen surface and adjusts vertically with downwind fetch from the transition. The resulting internal boundary layer grows downwind. The thickness of the equilibrium layer is about 30% of the fetch distance over surfaces like that of a sedge fen [*Raabe*, 1991]. Additionally, at the transition between the fen and the forest, the flow may be displaced from the ground surface by approximately the height of the forest *h*, often resulting in a separation and wake region to form downwind of a transition, and a long fetch is required (~100 h) for the flow to equilibrate [*Markfort et al.*, 2010]. The forest on the eastern boundary of the fen is about 150 m from the tower. Currently, methods do not exist to account for the effect of wakes behind forest canopies in the estimation of fluxes from wetlands. Therefore, due to relatively short fetch length downwind of the forest, fluxes cannot be determined downwind of the forest canopy using the flux gradient method.

[12] There are two main lobes of the fen with a sufficiently long fetch, each greater than 400 m (Figure 1). The narrowest lobe extends to the southeast while a broad region extends to the north and northwest of the tower. The longer fetch of these lobes allows for use of the flux gradient approach to measure CH_4 fluxes. Tower data were excluded when the wind direction was not parallel to the axes of the suitable fetches of the fen. Data were accepted for wind blowing from the following sectors: ESE (115°–145°), W (245°–297°), and NNW (315°–340°) (Figure 1). A total of 6725 half hour average CH_4 measurements were collected; however, 70% were eliminated based on wind direction.

[13] Data were also excluded when the friction velocity (u_*) was less than 0.1 $m\,s^{-1}$ or the atmospheric stability was not near neutral ($Ri > 0.2$). These criteria ensured that the boundary layer flow over the surface of the fen was fully turbulent, and the flow was shear dominated and fully interacting with the surface. The choice of a threshold u_* and Ri can be rather arbitrary. In practice, the lowest threshold for u_* has been found to vary from 0.1 to 0.5 $m\,s^{-1}$, but this is highly dependent upon site characteristics [*Aubinet et al.*, 2012; *Laurila et al.*, 2012]. The friction velocity u_* was tested for the site-specific flux data to determine the threshold of dependence (Figure 2). No clear u_* dependence was found, except possibly near zero, so a conservative value ($u_* = 0.1$) was chosen to minimize artifacts due to limited shear. The Ri threshold is set to the established critical value (0.25) where turbulence may not fully interact with the surface due to negative buoyancy [*Baker and Griffis*, 2005]. Only 22% of collected data met these strict criteria, therefore no attempt was made to quantify a seasonal CH_4 budget. This resulted in a semi-continuous record of CH_4 flux. Most of the data excluded from analysis from the tower were during nighttime and periods of weak winds. Data from the tower were separated into daytime measurements between 08:00 and 17:00 h (n = 625) and nighttime measurements between 17:00 and 08:00 h (n = 869). Seasonal, monthly, and diurnal mean flux rates were computed as the mean of multiple flux measurements during a specific time period. These averages are not equivalent to fluxes integrated over time (e.g., monthly flux) or budgets, both of which require more complete continuous records of flux.

[14] An advantage of the flux gradient approach is that it is not sensitive to many of the limitations of the eddy covariance method, namely sensor alignment and flow deflection. Both methods are based on the assumption of stationary and homogeneous flow and require a long fetch to limit advection effects. Therefore, for long-term measurements of trace gas flux, the flux gradient approach may not be better or worse than the more commonly employed eddy covariance method. *Pattey et al.* [2006] present a modern technique for measuring CH_4 with a tunable diode laser in conjunction with the eddy covariance method. In their study, they found that eddy covariance and flux gradient methods show good correspondence. An important limitation of the flux gradient technique is that significant gradients in the scalar quantity must be measured to accurately resolve fluxes; however, this may not be the case over forests and under highly convective conditions in the atmosphere. The measurements presented here do not consider fluxes over the forest but over short vegetation covering the fen. The flux gradient technique was developed for such a case. The effect of convection in the atmosphere does contribute to small gradients during the day; however, since our focus is on capturing the large pulses during the evening transition when the atmospheric stability is nearly neutral and turbulence is shear derived, the accuracy of the measured gradient in CH_4 is optimal. The footprint of the flux tower is limited by the selected wind sectors to ensure that the flux measurements are derived from the fen. Additionally, due to the criteria excluding data from times when the atmosphere is stable or during weak-wind conditions, the extent of the footprint is not expected to vary significantly.

2.5. Auxiliary Data and Analyses

[15] Various other environmental, meteorological, and ecological data were measured in the fen and were available in the BOREAS data set. Additional data included air

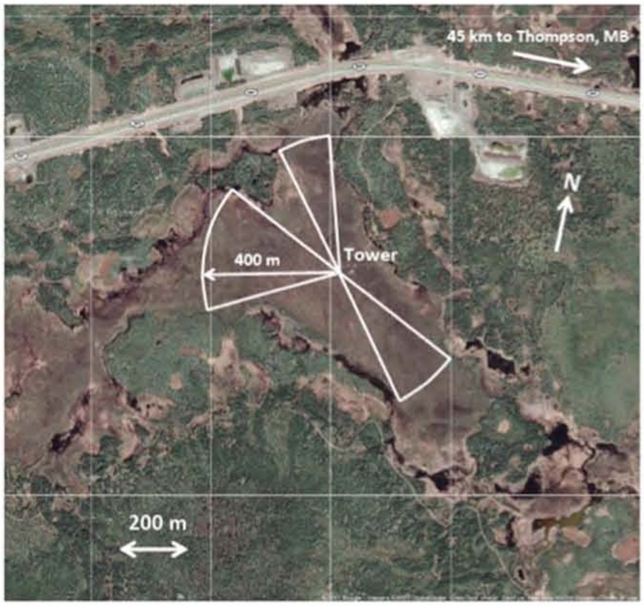

Figure 1. Layout of the BOREAS NSA fen site, after *Lafleur et al.* [1997]. Sectors identifying acceptable wind directions and approximate source area represented in tower-based flux measurement. Image copyright GeoEye, obtained through Google Earth (www.google.com).

temperature, water table height, and soil temperature profiles adjacent to the flux tower at 30 min intervals over the sampling period. Temperature measurements in the hollows were partitioned into three depths representing overlying water or pools (1, 5, and 10 cm) and six depths representing the underlying peat (25, 50, 75, 100, 150, and 200 cm). We performed this classification using the diurnal variability in temperature, which was much greater near the surface (1–10 cm) than below 25 cm. This result indicates that the peat-water interface was 10 to 25 cm below the surface. We represent the strength of thermal stratification as the temperature gradient between 1 and 5 cm depth in the water ($\Delta T/\Delta z$). We performed Spearman's rank correlation analyses for both the chambers and tower to determine if commonly measured parameters explain variability in CH_4 flux. For each chamber sampling date, the chamber data describe only spatial variance but the tower data describe both temporal and spatial variance. Because the spatial and temporal components of the tower data cannot be distinguished, we chose to compare the chambers and tower without using statistical hypothesis tests about the means.

3. Results

3.1. Spatial and Temporal Variability in Chamber Flux Measurements

[16] The 12 chamber locations produced mean seasonal fluxes between 22.4 and 318 nmoles $CH_4\,m^{-2}\,s^{-1}$ (range of measurements 1–1389 nmoles $m^{-2}\,s^{-1}$). Although chambers differed in their seasonal mean flux, each chamber showed substantial temporal variability. The majority of the chambers showed a seasonal pattern of CH_4 flux, reaching a maximum during August (Figure 3). The mean of chamber flux measurements taken in each 24 h span was positively correlated with daily water table level (Spearman's $r^2 = 0.42$, $n = 9$), whereas no correlation was observed between methane flux and daily mean air temperature ($r^2 < 0.01$, $n = 20$), minimum air temperature ($r^2 = 0.05$, $n = 20$), or peat temperature at 20 cm ($r^2 < 0.01$, $n = 20$).

3.2. Comparison of Daytime Flux Measurements by the Chamber and Tower Methods

[17] Due to equipment failures and prevailing wind patterns, only 10 sampling dates had at least one daytime CH_4 flux measurement from both the chambers and the tower. Mean flux measurements from chambers exceeded the mean of flux measurements from the tower during the daytime for all dates except 22 July (Figure 3), but the minimum chamber flux was less than the mean of flux measurements from the tower on six of the dates. On dates where the tower recorded a positive flux of CH_4 to the atmosphere, the mean of flux measurements from the chambers was 28–420% higher than the mean of flux measurements from the tower recorded during the daytime. Across sampling dates, the mean of daytime

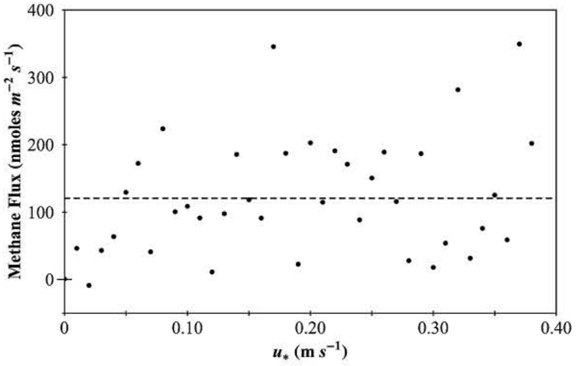

Figure 2. Dependence of methane flux on friction velocity (u_*). Data points are mean flux, binned by levels of u_*, the mean methane flux is shown as a dashed line.

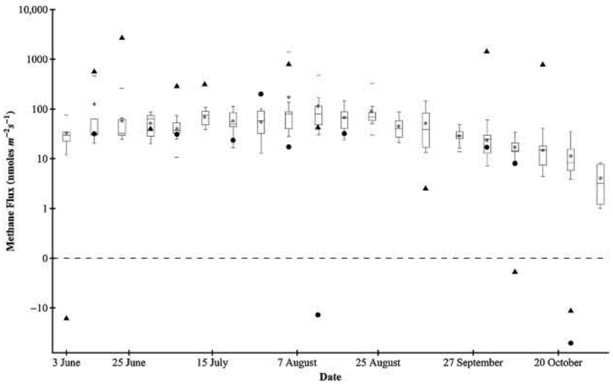

Figure 3. Seasonal trend in methane emission from the fen as measured by the chambers and the flux tower. The chamber data are displayed as boxplots for each date, with the centerline representing the median flux, the edges of the box representing the 25% and 75% quantiles, and the whiskers representing the maximum and minimum values. The mean chamber flux is denoted as a star and outliers greater than 1.5 times the interquartile range are denoted by horizontal dashes. Mean tower measurements during the daytime (08:00–17:00) are represented by circles and mean measurements during the following nighttime period are represented by triangles. For each measurement date, at least six chamber measurements were included (n = 10). The number of half-hour mean measurement represented in each point for the daytime tower flux was n = 7, 3, 6, 6, 3, 7, 1, 1, 4, and 1, respectively. The number of half-hour mean measurement represented in each point for the nighttime tower flux was n = 5, 11, 11, 3, 9, 3, 2, 9, 3, 1, 11, 1, and 26, respectively.

tower measurements was weakly correlated with the mean chamber measurements (Spearman's $r^2 = 0.15$, n = 10).

3.3. Temporal Variability in Tower Flux Measurements

[18] Similar to the chamber measurements, the daytime (08:00–17:00) tower measurements show a strong seasonal pattern. Daytime flux measurements from the fen were mostly negative during the spring, but flux became positive and reached a plateau during the growing season from early June until early October (Figure 4a). The means of daytime flux measurements in each month were the following: −90 nmoles $CH_4\ m^{-2}\ s^{-1}$ in May, 19 nmoles $CH_4\ m^{-2}\ s^{-1}$ in June, 27 nmoles $CH_4\ m^{-2}\ s^{-1}$ in July, 12 nmoles $CH_4\ m^{-2}\ s^{-1}$ in August, 9.5 nmoles $CH_4\ m^{-2}\ s^{-1}$ in September, and −8.5 nmoles $CH_4\ m^{-2}\ s^{-1}$ in October. The nighttime emissions from the fen showed a different seasonal pattern than the daytime measurements with consistently positive flux (Figure 4b). The means of nighttime flux measurements in each month were the following: 298 nmoles $CH_4\ m^{-2}\ s^{-1}$ in May, 322 nmoles $CH_4\ m^{-2}\ s^{-1}$ in June, 891 nmoles $CH_4\ m^{-2}\ s^{-1}$ in July, 597 nmoles $CH_4\ m^{-2}\ s^{-1}$ in August, 93 nmoles $CH_4\ m^{-2}\ s^{-1}$ in September, and 28.7 nmoles $CH_4\ m^{-2}\ s^{-1}$ in October. The maximum emission rate of 24,008 nmoles $CH_4\ m^{-2}\ s^{-1}$ occurred on 1 July at 21:38. The micrometeorological data indicated near-neutral atmospheric stability ($Ri \cong 0$) and a high gradient of CH_4 near the surface (0.84 ppm m^{-1}). Across the entire season, the mean of nighttime flux measurements was 325 nmoles $CH_4\ m^{-2}\ s^{-1}$ (n = 869, standard error = 42), compared to 53 nmoles $CH_4\ m^{-2}\ s^{-1}$ (n = 625, standard error = 10) for daytime flux. The mean of nighttime emission rates was often an order of magnitude greater than the mean of positive daytime emission rates on the same date (n = 50, mean 11-fold, max 138-fold). These elevated nighttime emissions were highest during July (mean ± standard error, 24 ± 10-fold, n = 15) and August (17 ± 10-fold, n = 6) and lower during June (4.5 ± 1.7-fold, n = 16), September (1.5 ± 0.57-fold, n = 8), and October (1.1 ± 0.45-fold, n = 4).

[19] Daily mean CH_4 flux measurements from the tower were weakly correlated with other measured variables (including temperature in hummocks or hollows, wind direction, water table height, photosynthetic activity, and solar radiation) during the entire measurement period and within each month (Table A1, all $r^2 < 0.50$). Daily mean flux rates during daytime were weakly correlated with air temperature and peat temperature at 10 cm over the measurement period ($r^2 = 0.25$–0.28). Daily mean flux rates during the nighttime were weakly correlated with nighttime maximum air temperature ($r^2 = 0.23$), peat temperature at 10 cm ($r^2 = 0.16$–0.17), daily mean moisture flux ($r^2 = 0.24$), and CO_2 flux ($r^2 = 0.21$) from the fen. Methane flux was poorly explained by all measured variables at half-hour intervals throughout the measurement period and within each month (Table A2). The strongest predictors of flux rates averaged at half-hour intervals were air temperature ($r^2 = 0.15$, n = 1455) and peat temperature at 10 cm ($r^2 = 0.21$–22, n = 1455). Daytime flux rates averaged half-hourly showed weak correlation with air temperature ($r^2 = 0.15$, n = 610) and peat temperature at 10 cm ($r^2 = 0.22$, n = 610). Nighttime flux rates averaged at half-hour intervals over the measurement period were weakly correlated with peat temperature at 10 cm ($r^2 = 0.23$–0.25, n = 845). Overall, explanatory power of any of these known drivers of flux was low ($r^2 < 0.25$).

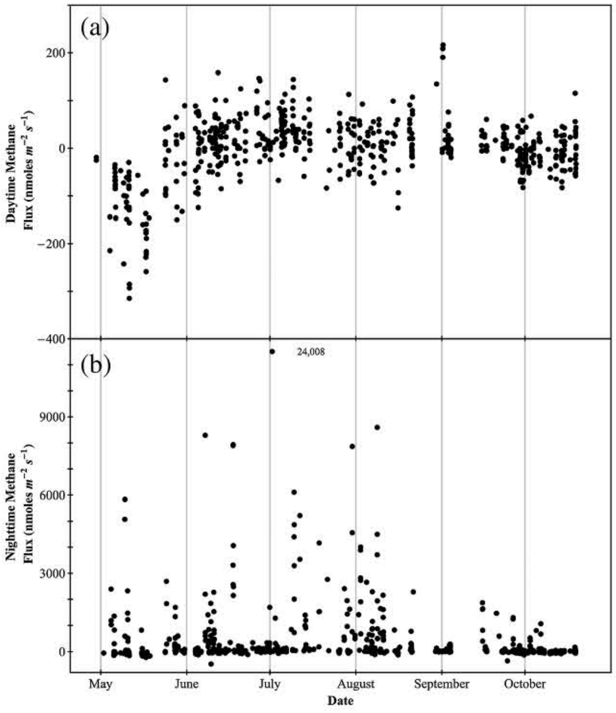

Figure 4. (a) Seasonal pattern of daytime (08:00–17:00 CST) and (b) nighttime methane fluxes during the growing season.

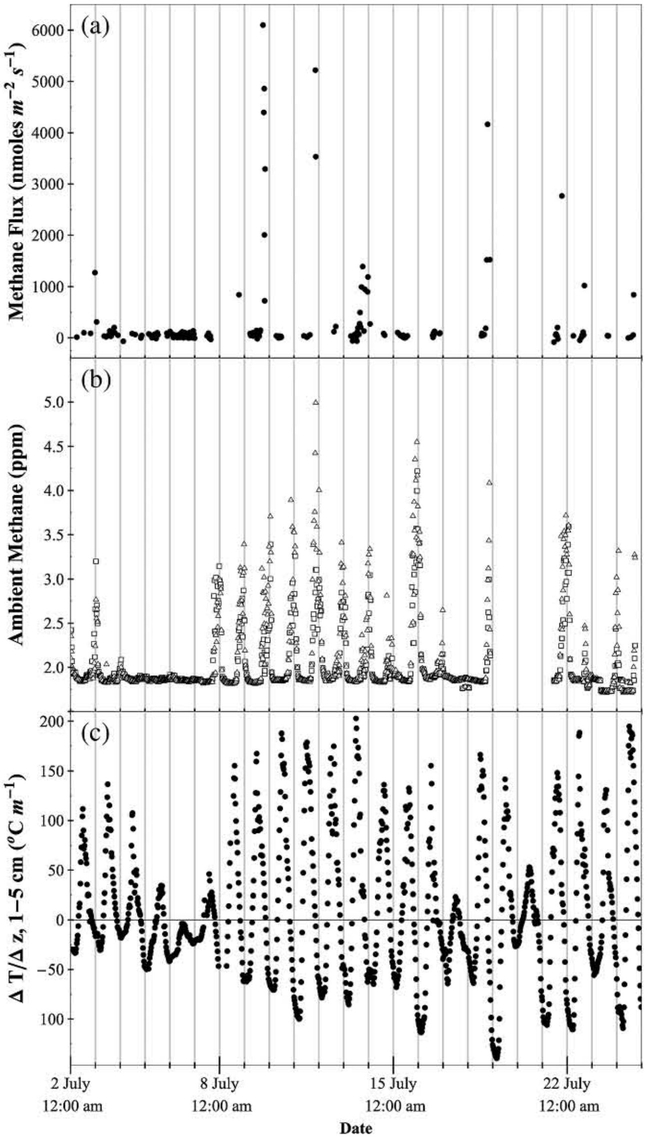

Figure 5. (a) Semi-continuous time series of methane flux as measured by the tower during the dates 2–25 July. (b) Ambient methane concentrations measured at 3.59 m (open triangles) and 6.65 m (open squares). (c) Thermal gradient ($\Delta T/\Delta z$) in the upper 5 cm of a hollow.

[20] Two periods are apparent in the semi-continuous flux record. During the first period (early morning until early afternoon), fluxes are nearly zero. During the second period (15:00 and 24:00), the largest fluxes of CH_4 occur. Unfortunately around 23:00 to 01:00, the shear stress and wind speed are unacceptably low, so we cannot identify the end of the event (Figure 5a). Evidence that high flux continues after the wind decreases can be seen in the comparison between the flux time series and the ambient CH_4 concentration measured at the two heights (Figure 5b). Although the flux time series is discontinuous due to the stringent quality control restrictions, and it cannot be shown that high flux rates occur every day, ambient concentrations were measured continuously and suggested high nighttime methane emissions. Unlike the flux measurements from the tower, concentrations are less sensitive to wind speed, wind direction, or atmospheric stability.

[21] The thermal gradient ($\Delta T/\Delta z$) in the hollows (between 1 and 5 cm) showed a strong diurnal pattern (Figure 5c). The surface of the standing water in the hollows was heated during the day due to solar input and cooled at night. Throughout the measurement record, cooling of the water in the hollows was found to be consistently coincident with the peaks in CH_4 concentration and flux measured by the tower (Figure 5). Although data on the spatial coverage of hollows are not available for the fen, *Lafleur et al.* [1997] indicate that the fen is characterized by hummock-hollow structure. On dates when thermal stratification of hollows was absent (e.g., 6–7 July), the nighttime emission events were not observed (Figure 5). Periods without thermal stratification ($n = 17$ days) were observed from June through October and were characterized by low irradiance, cooler air temperatures, some precipitation, and low ambient methane concentrations (supporting information).

4. Discussion

4.1. Comparison of Tower and Chamber Measurements

[22] The discrepancy between the chamber measurements and daytime flux tower measurements is likely attributable to spatial heterogeneity in CH_4 emission, which has been observed within other wetlands [*Alm et al.*, 1999; *Bubier et al.*, 2005; *Dinsmore et al.*, 2009b]. Variation in topography [*Waddington and Roulet*, 1996], plant distribution [*Moosavi and Crill*, 1997; *Riutta et al.*, 2007], soil moisture or water table position [*Bellisario et al.*, 1999], and oxygen availability in the soil [*Askaer et al.*, 2010] lead to patchiness in emissions within a wetland. Given this heterogeneity, a small number of chambers located adjacent to the flux tower is likely inadequate to characterize the flux across the footprint area of the tower and therefore the entire ecosystem. Wetlands with more homogeneous structure would be expected to have similar flux estimates as measured by the chambers and tower. In a heterogeneous wetland, chamber-based estimates may be biased due to chamber locations and up-scaling the flux measurements across the area of representative habitat. The BOREAS fen has a moisture gradient and the tower was located in a wetter area near the edge of the fen [*Lafleur et al.*, 1997], both of which suggest that the chamber locations are likely to have higher flux rates than other areas within the footprint of the tower. Due to quality control criteria, the comparisons in Figure 3 include only a few half-hourly tower measurements. A more continuous record of flux might provide a more robust comparison with the chambers and would allow integration of a daily flux. However, since the flux estimates were based upon 30 min averages of measurements recorded every 6 min, these estimates are sufficiently supported for comparison with thechambers that were sampled once each day over approximately 30 min.

[23] Sampling artifacts from the chambers (such as heating or ebullition) are typically small in magnitude [*Denmead*, 2008; *Moore and Roulet*, 1991], but may be sufficient to account for a portion of the difference in daytime CH_4 flux observed between the chambers and the tower.

4.2. Temporal Patterns in Flux

[24] The nighttime emissions measured by the flux tower were greater than the daytime emissions. This phenomenon has been observed in other studies utilizing chamber sampling and soil gradient methods, although the amplitude of the nighttime or evening increases were small (nighttime magnitude <150% of daytime) [*Nakano et al.*, 2000; *Whiting and Chanton*, 1992] compared to those presented here. *Yavitt et al.* [1990] used chambers to document increased nighttime emissions from a sedge meadow during the summer (magnitude 200%), but this pattern was absent at the same sites during the spring and reversed in the fall. Similarly, *Whalen and Reeburgh* [1988] recorded elevated nighttime and evening emissions at two tundra sites using chambers (magnitude and 150–200%), but the diurnal pattern was absent or reversed at other sites. In contrast, the elevated nighttime emission rates presented here were observed throughout the growing season. *Mikkelä et al.* [1995] documented elevated nighttime emissions in a boreal mire using chambers, but this difference was not consistently observed in lower areas of the wetland. Nighttime emission rates in drier communities were elevated (2 to 20-fold) relative to daytime, but this pattern was absent or reversed in more moist communities, including standing pools. The authors proposed that the elevated nighttime emissions were attributable to decreased methanotrophy due to lower temperatures at night or to the delayed release of substrates by plants. Although we are unable to determine if drier areas such as hummocks contributed to elevated CH_4 fluxes in our analysis, there is strong evidence that drier regions of the wetland have lower CH_4 flux [*Bellisario*, 1999; *Moosavi and Crill*, 1997], suggesting that the substantial nighttime emission events were not localized to drier regions.

[25] Nighttime emissions peaks of comparable magnitudes have not been found in other studies utilizing the flux tower method [*Harazono et al.*, 2006; *Zona et al.*, 2009]. Previous studies using tower-based measurements show no evidence of diurnal patterns in CH_4 emissions in wetlands lacking appreciable surface water [*Forbrich et al.*, 2011; *Rinne et al.*, 2007; *Shurpali et al.*, 1993]. Elevated daytime CH_4 emissions have been described in a wet tundra meadow adjacent to a lake [*Fan et al.*, 1992] and from a managed peat meadow where the pattern corresponded to peaks in CO_2 uptake and latent heat flux [*Hendriks et al.*, 2010]. Higher flux rates in daytime compared to nighttime were recorded by eddy correlation measurements from the BOREAS southern study area fen [*Suyker et al.*, 1996], which included inundated hollows during the growing season [*Suyker et al.*, 1997]. *Jackowicz-Korczyński et al.* [2010] found little diurnal variation in CH_4 flux from a Swedish mire, but did document elevated nighttime emissions from areas of the wetland adjacent to a lake (magnitude <150%). *Kroon et al.* [2010] documented a consistent diurnal pattern in CH_4 flux from a peatland with a substantial area of surface water in ditches. Emission rates were elevated (magnitude <130%) during the afternoon and early evening, closely matching the diurnal pattern in soil temperature. In comparison to all other published studies of CH_4 flux over daily timescales, the BOREAS fen shows a distinct diurnal pattern with the majority of the flux from the ecosystem occurring during the night. It remains possible that nighttime emission events occur in other wetlands, but have been missed due to a lack of nighttime sampling. Also, wind velocity and shear stress were often reduced at night relative to daytime, which prevented reliable tower-based measurements. This shortcoming of the flux tower approach resulted in exclusion of the majority of nighttime measurements in the BOREAS data set, but the acceptable data show that the nighttime pulses are regular.

[26] Despite the consistency and large magnitude of the nighttime peaks observed in the BOREAS fen, the flux was poorly correlated with commonly associated variables including peat temperature [*Bartlett et al.*, 1992; *Bubier et al.*, 1995a; *Heikkinen et al.*, 2002], water table height [*Alm et al.*, 1999; *Bellisario et al.*, 1999; *Hendriks et al.*, 2010], and net ecosystem exchange [*Christensen et al.*, 2000]. The strength of the correlations for the fen data set showed little improvement when performed separately by month or by daytime and nighttime. This lack of strong dependence upon any single driver might be explained by significant spatial heterogeneity within the tower footprint, or a less-studied driver.

[27] The flux rates observed by the tower during the nighttime were higher and had a greater range than previously

Table 1. Summary of Methane Flux Measurements in Northern Wetlands Using Eddy Covariance and Flux Gradient Methods

Location	Sampling Period	Range of Flux (nmoles $m^{-2}\,s^{-1}$)	Mean Flux (nmoles $m^{-2}\,s^{-1}$)	Source
Mire, Sweden	2 years	0 to 346	107 (midseason)	*Jackowicz-Korczyński et al.* [2010]
Mire, Finland	Discontinuous	<0 to 75	10.8 (annual)	*Hargreaves et al.* [2001]
Fen, Finland	1 year	−35 to 173	24.9 (annual)	*Rinne et al.* [2007]
Peatland, Scotland	2 years	-	118 (annual)	*Dinsmore et al.* [2010]
Peatland, MN, USA	Discontinuous	87 to 195		*Verma et al.* [1992]
Tundra floodplain, Russia	Growing season	4.1 to 25	13.5 (seasonal)	*Sachs et al.* [2008]
Bog, Finland	Growing season	0 to 87	5.3 to 37 (seasonal)	*Alm et al.* [1999]
Managed fen, Netherlands	3 years	<0 to 113	23 (annual)	*Kroon et al.* [2010]
Peatland, MN, USA	Growing season	0 to 121	11.5 to 14.4 (annual)	*Clement et al.* [1995] and *Shurpali et al.* [1993]
Fen, Finland	2 Growing seasons	−0.5 to 409	15.0 to 16.4 (seasonal)	*Riutta et al.* [2007]
Peatlands, Netherlands	3 years		0 to 69 (annual)	*Hendriks et al.* [2010]
Mire, Finland	Growing season	0 to 142	13.4	*Forbrich et al.* [2011]
Fen, SK, Canada	Growing season	0 to 337	140	*Suyker et al.* [1996]
Fen, MB, Canada	Growing season (nighttime)	−474 to 24,008	325	This study
Fen, MB, Canada	Growing season (daytime)	−442 to 2,999	53	This study

published measurements from flux towers (Table 1). However, previous studies using the chamber method in northern wetlands have reported mean fluxes greater than 250 nmoles CH_4 $m^{-2}\,s^{-1}$ [*Harriss et al.*, 1985; *Moosavi and Crill*, 1997; *van Huissteden et al.*, 2005; *Vourlitis et al.*, 1993] and maximum rates greater than 1000 nmoles CH_4 $m^{-2}\,s^{-1}$ [*Harriss et al.*, 1985; *Moosavi and Crill*, 1997; *Roulet et al.*, 1994]. The chamber measurements of CH_4 flux from the BOREAS NSA fen were high relative to many northern wetlands and indicate substantial capacity for CH_4 production within the fen. Methane production from the fen may be supported by comparatively high net carbon uptake documented during the 1996 growing season [*Bubier et al.*, 1999] and increased precipitation [*Bubier et al.*, 2005].

4.3. Possible Mechanisms for Nighttime Emission Events

[28] The nighttime methane pulses could be the result of several driving forces. In this section, we evaluate a number of documented mechanisms by using the available data and by comparing the magnitudes of pulses observed elsewhere to those presented in this paper. First, we propose a novel mechanism whereby CH_4 produced during the daytime is trapped in thermally stratified hollows and is released as pulses during evening cooling and convective mixing of the water. The magnitude and timing of nighttime methane emission pulses in our data set could be readily explained by this mechanism alone, as detailed below. The second group of mechanisms involves the role of vascular plants. Methane emission is commonly augmented by transport through vascular tissues and by the substrates that are exuded by plants. Vascular plants may also inhibit methane emission by transporting oxygen into the peat. Finally, effects of diurnal temperature fluctuations on the production and consumption of CH_4 are discussed.

4.3.1. Stratification in Hollows

[29] The periodic nighttime CH_4 emission events observed in the tower data set were not explained by hourly regressions against forcing variables (temperature in hummocks or hollows, wind direction, water table height, photosynthetic activity, and solar radiation, see Tables A1 and A2). However, the episodic evening emission events and increased CH_4 concentrations just above the fen showed coincident timing with thermal destratification and convective cooling within the upper 10 cm of hollows (Figure 5). Stratification within wetland pools and hollows has been documented previously [*Van der Molen and Wijmstra*, 1994]. Methane produced beneath the hollows may be effectively trapped by thermal stratification, accumulating within the lower (cooler) layers of water or at the peat-water interface. Under thermal stratification, emission of CH_4 occurs primarily through molecular diffusion. Molecular diffusion is substantially slower than turbulent diffusion and is likely the dominant transport process in the pools [*Fischer et al.*, 1979]. Ebullition has also been found to occur in stratified water bodies and wetlands, but could not be detected in this study. The strength of the thermal gradient should not affect the size of the emission event, and thus $\Delta T/\Delta z$ was not used as a predictive variable for regressions. Although it is not possible with a discontinuous record of half-hourly flux measurements, this mechanism could be evaluated by comparing the rate of destratification with the onset of emission events in a data set with finer temporal resolution (e.g., eddy covariance).

[30] Although the solubility of CH_4 in water is low at the temperatures recorded in the hollows [*Duan and Mao*, 2006], this mechanism is capable of producing emission events of the same magnitude as those observed by the tower. For instance, we assume that if the hollows covered 30% of the fen surface at a mean depth of 20 cm, the cooler layer of water near the peat could store the equivalent of 45 mmoles m^{-2} across the area of the fen. If this stored methane were to be released over a 6 h time period with a linear rise and fall, the equivalent peak emission rate would be 2074 nmoles CH_4 $m^{-2}\,s^{-1}$. This rate represents a hypothetical maximum storage capacity for the defined hollows, and only 1% of the measurements from the tower exceeded this emission rate. Thus, the storage capacity within pools can account for the released methane during the evening transition, and the feasible emission rates via this mechanism are within the observed rates in this study.

[31] Other studies have documented diurnal accumulation of dissolved CH_4 due to thermal stratification in shallow aquatic systems [*Crill et al.*, 1988; *Ford et al.*, 2002]. Hollows have been shown to act as hotspots for CH_4 production and emission in wetlands [*Alm et al.*, 1999; *Bubier et al.*, 1993; *Clement et al.*, 1995; *Waddington and Roulet*, 1996]. In addition to destratification releasing trapped CH_4, cooling at the surface dramatically increases the flux of gas to the

atmosphere [*MacIntyre et al.*, 2002]. Studies in stratified lakes show that the flux attributable to cooling (buoyancy flux) at night exceeds the flux that may be attributed to wind-driven flux [*MacIntyre et al.*, 2010]. The effect of destratification and heat flux on gas emissions from wetland hollows has not been identified previously, but these physical processes may impact the flux of CH_4 from wetlands with standing water.

[32] Studies have identified terrestrial freshwater bodies as major contributors of CH_4 to the atmosphere [*Bastviken et al.*, 2011; *Roulet et al.*, 1997]. Convective mixing has been identified as a control of CH_4 and CO_2 release, especially from small water bodies [*Eugster et al.*, 2003; *Read et al.*, 2012]. Recent work on the abundance and distribution of lakes has revealed that the majority of water bodies are smaller than 0.01 km^2 [*Downing et al.*, 2006; *McDonald et al.*, 2012]. Although the role of convective mixing in gas flux has been described at a range of spatial scales from small lakes [*Read et al.*, 2012] to the ocean [*Rutgersson et al.*, 2011], convective mixing of inundated wetlands could represent a substantial and previously unrecognized component of methane flux.

4.3.2. CH_4 Transport Through Plants

[33] Diurnal patterns in CH_4 emission from wetlands have been attributed to diffusion of CH_4 through aerenchymatous tissues and stomatal conductance [*Joabsson et al.*, 1999]. In many wetland plant species, these tissues transport atmospheric oxygen to roots and stems in anoxic sediments, but may also be an important pathway for CH_4 flux as well [*Hargreaves et al.*, 2001; *Morrissey et al.*, 1993]. However, unlike the elevated nighttime CH_4 emissions observed in the BOREAS fen, aerenchymatous transport of CH_4 produces diurnal patterns in which flux is highest during the period of peak photosynthetic activity [*Lloyd et al.*, 1998; *Mikkelä et al.*, 1995; *Thomas et al.*, 1996], though this correlation may be weak [*Askaer et al.*, 2011]. Although aerenchymatous transport of CH_4 may have occurred in the fen, the timing and magnitude of this mechanism are inconsistent with the nighttime emission events observed here.

4.3.3. Control by Plant Exudates and Oxygen

[34] Oxygen transport through aerenchymatous tissue may lead to diurnal fluctuations in the rate of methanotrophy. However, unlike the diurnal patterns observed in CH_4 transport, decreased transport of oxygen at night due to stomatal closure would serve to decrease CH_4 oxidation, leading to increased emission rates. Studies have documented decreased soil oxygen content at night [*Lloyd et al.*, 1998; *Thomas et al.*, 1996] and seasonal patterns in CH_4 oxidation [*King*, 1996; *Roslev and King*, 1996], but it is not clear that plant-mediated cycles in oxygen availability within the soil could affect emission rates over diurnal timescales. Plants play another important role in CH_4 dynamics by supplying carbon substrates for methanogenesis. This coupling is evidenced by vegetation clipping studies [*Waddington et al.*, 1996; *Whiting and Chanton*, 1992]. Isotope analysis and assays of methanogenesis and methanotrophy performed in the BOREAS NSA fen in 1993 indicated that the carbon in CH_4 was recently sequestered, and oxidation within the soil did not control CH_4 emission rates [*Bellisario et al.*, 1999]. Since the availability of oxygen is closely coupled to the water table depth [*Granberg et al.*, 1997], it is hypothesized that CH_4 oxidation most likely occurred in the hummocks rather than in the hollows. Diurnal fluctuations in methanogenesis may also be attributed to a time lag between CO_2 fixation by plants and the release and consumption of substrate by soil microbes [*Waddington et al.*, 1996; *Whiting and Chanton*, 1992]. Although the diurnal pattern of CO_2 flux from the BOREAS fen during the 1994 growing season indicated peak photosynthetic activity around noon [*Lafleur et al.*, 1997] and a similar pattern was documented in 1996 [*McCaughey et al.*, 1999], it is not clear if the timing and magnitude of documented lag effects are consistent with the nighttime emission events described here.

4.3.4. Control by Peat Temperature

[35] While CH_4 emission peaks commonly occur during daytime [*Long et al.*, 2010], peak emissions have been observed during nighttime when the water table was 0–40 cm below the surface [*Mikkelä et al.*, 1995]. These authors suggested that diurnal temperature fluctuations caused methanotrophic activity to decline during nighttime. Under favorable conditions, methanotrophs can consume CH_4 at rates greater than 3500 nmoles CH_4 $m^{-2}\,s^{-1}$ [*Gupta et al.*, 2012; *Popp et al.*, 2000], although these rates are extreme and might not be representative of the complexity found in a wetland. Granberg *et al.* [1997] demonstrated that water table depth controls the effect of temperature on net CH_4 emission (production-oxidation) from wetland soils. Increasing temperature above the water table leads to higher rates of methanotrophy and decreased net flux, whereas warmer temperatures at and below the water table lead to higher rates of methanogenesis and increased net flux.

[36] While these studies demonstrate that it is feasible for methanotrophs to consume CH_4 at a rate similar to that of nighttime emission events, the magnitude of diurnal temperature changes is not sufficient to explain the magnitude of the emission events. The parameter Q_{10} is the proportional increase in the rate of methanogenesis or methanotrophy attributed to a 10°C increase in temperature and is used to describe the sensitivity of methanogenesis to temperature [*Whalen*, 2005]. Estimates of the Q_{10} for methanogenesis in wetlands range from <1 to 35 [*Whalen*, 2005] and the Q_{10} for methanotrophy is approximately 2 [*Segers*, 1998; *Whalen*, 2005]. During the measurement period, the maximum diurnal temperature range of peat beneath the hummocks was 26.4°C at 1 cm, 15.5°C at 10 cm, 12.1°C at 25 cm, and less than 1.4°C below 50 cm. In the hollows, the maximum diurnal temperature change was 27.6°C at 1 cm, 21.7°C at 5 cm, 12.7°C at 10 cm, 3.0°C at 25 cm, and less than 1.4°C below 50 cm. The temperature maxima in the shallow peat (1–10 cm) typically occurred during daytime, but the maxima in deeper layers occurred later, between 18:00 and 24:00. The effect of diurnal temperature fluctuation on methanogenesis is clearly insufficient to explain the large nighttime emission events measured by the tower. Similarly, the temperature fluctuations in the shallow peat indicate a maximum change of 550% in the rate of methanotrophy. Although diurnal patterns in methanotrophy due to temperature may occur, the potential rates do not appear sufficient to explain the nighttime emission events during the warmest months. Furthermore, the lack of consistent correlation between flux and peat temperature in hummocks and hollows at daily or half-hourly timescales suggests that the nighttime peaks in emission are likely not the result of temperature fluctuations.

4.4. Summary

[37] This study compared previously unpublished flux tower measurements of CH_4 flux with chamber measurements from the BOREAS NSA fen. The spatial extent of the chambers was much smaller than the footprint of the flux tower, which might explain the apparent discrepancy between the chamber data and the daytime measurements by the tower. Additionally, regular nighttime CH_4 emission events were found that were not previously detected using chambers. The substantial nighttime CH_4 emissions observed from the fen exceed the magnitude of diurnal fluctuations observed in other studies using flux tower methods. We attribute these emission events to short-term storage of CH_4 in thermally stratified hollows and subsequent release through destratification and buoyancy flux. The flux rates derived from the chambers are compatible with the estimates of CH_4 production required to produce these emission events. Other previously identified (or classical) drivers could not explain the magnitude of CH_4 emissions observed in the fen. The large emission events are unlikely to be captured using discrete samples from chambers, but nevertheless may represent a substantial portion of the daily flux from the ecosystem. The results of this study illustrate that relatively short-term physical controls can have a significant influence on ecosystem-atmosphere exchange and must be captured in measurement strategies. However, biogeochemical processes leading to methane production must coincide with surface water thermal stratification for this phenomenon to be present. Future work should determine what physical conditions must be present for such dynamics to exist, and if indicators can be identified to help modelers include these processes in biogeochemical models.

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