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Methane fluxes measured by eddy covariance and static chamber techniques at a temperate forest in central ontario, Canada

J. M. Wang[1], J. G. Murphy[1], J. A. Geddes[1], C. L. Winsborough[2], N. Basiliko[2], and S. C. Thomas[3]

[1]University of Toronto, Department of Chemistry, Canada
[2]University of Toronto, Mississauga, Department of Geography, Canada
[3]University of Toronto, Faculty of Forestry, Canada

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Correspondence to: J. G. Murphy (jmurphy@chem.utoronto.ca)

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Abstract

Methane flux measurements were carried out at a temperate forest (Haliburton Forest and Wildlife Reserve) in central Ontario (45°17′11″ N, 78°32′19″ W) from June–October, 2011. Continuous measurements were made by an off-axis integrated cavity output spectrometer Fast Greenhouse Gas Analyzer (FGGA) from Los Gatos Research Inc. that measures methane (CH_4) at 10 Hz sampling rates. Fluxes were calculated from the gas measurements in conjunction with wind data collected by a 3-D sonic anemometer using the eddy covariance (EC) method. Observed methane fluxes showed net uptake of CH_4 over the measurement period with an average uptake flux (± standard deviation of the mean) of $-2.7 \pm 0.13\,\mathrm{nmol\,m^{-2}\,s^{-1}}$. Methane fluxes showed a seasonal progression with average rates of uptake increasing from June through September and remaining high in October. This pattern was consistent with a decreasing trend in soil moisture content at the monthly time scale. On the diurnal timescale, there was evidence of increased uptake during the day, when the mid-canopy wind speed was at a maximum. These patterns suggest that substrate supply of CH_4 and oxygen to methanotrophs, and in certain cases hypoxic soil conditions supporting methanogenesis in low-slope areas, drive the observed variability in fluxes. A network of soil static chambers used at the tower site showed close agreement with the eddy covariance flux measurements. This suggests that soil-level microbial processes, and not abiological leaf-level CH_4 production, drive overall CH_4 dynamics in temperate forest ecosystems such as Haliburton Forest.

1 Introduction

Methane (CH_4) is the predominant hydrocarbon in the atmosphere and the third most important greenhouse gas after water (H_2O) and carbon dioxide (CO_2); its atmospheric abundance has increased by 150 % since the pre-industrial era (Dlugokencky et al., 2009). As of 2010, the radiative forcing of CH_4 from anthropogenic emissions

BGD
9, 17743–17774, 2012

was 0.50 W m^{-2}, corresponding to approximately 30 % of the radiative forcing from CO_2 (Montzka et al., 2011). Estimates of the soil sink for CH_4 vary between 15–45 Tg CH_4 yr^{-1} (Curry, 2007), making it the second largest sink after tropospheric oxidation. The main losses of atmospheric CH_4 in the biosphere are oxidation in upland soils, however the overall magnitude of the soil sink and the factors governing its variability are not well understood. It is believed that between 30–50 % of the global CH_4 soil sink is in the temperate zone (Price et al., 2004). Natural CH_4 emissions are dominated by methanogenesis in wetlands, especially under conditions of high humidity and temperature. Recently, it has been suggested that plants themselves have the potential to emit CH_4 depending on environmental conditions (Keppler et al., 2006; Brüggemann et al., 2009; Wang et al., 2011) although the significance of these emissions in natural environments has been disputed (Nisbet et al., 2009). The uncertainties in the global CH_4budget result from limited observational data coverage and the large variability in the factors that influence CH_4 fluxes in natural environments (Heimann, 2011).

Methane surface-atmosphere exchange has been measured using a variety of different approaches, most commonly chamber enclosure techniques (Christensen et al., 1995; van Huissteden et al., 2005), eddy covariance techniques (Fan et al., 1992; Verma et al., 1992; Edwards et al., 1994; Hendriks et al., 2008; Detto et al., 2011), flux-gradient techniques (Simpson et al., 1997; Miyata et al., 2000; Edwards et al., 2001), or inferred by CH_4 mixing ratio measurements (e.g. Dlugokencky et al., 1994, 2009). Methane flux studies using chamber enclosures and eddy covariance have focused mainly on CH_4-emitting ecosystems such as peatlands and other wetlands (Bartlett and Harriss, 1993; Simpson et al., 1995; Meijide et al., 2011; Baldocchi et al., 2012; Hatala et al., 2012). In these ecosystems, CH_4-producing microbes (methanogens) are the main CH_4 source, and studies have shown that environmental factors such as soil moisture and temperature can control microbial activity (Satpathy et al., 1997). Physical and biological processes, such as ebullition and diffusive emissions through water, and plant-mediated transport of CH_4 through aerenchyma (Miyata et al., 2000), are known to impact CH_4 emission rates. The presence of CH_4-oxidizing microbes

BGD
9, 17743–17774, 2012

(methanotrophs) in aerated zones (Mikkelä et al., 1995) with oxic conditions can mediate the emissions of CH_4 in these systems.

Forest soils can transition between oxic and anoxic conditions depending on topographic position and environmental conditions, leading to significant variability in local microbial activity and potentially to methane surface flux (Ueyama et al., 2012). Itoh et al. (2009) found that the soil moisture patterns could greatly affect seasonal and spatial variations in CH_4 fluxes within a temperate forest: wetter sites exhibited large CH_4 emissions during rainy summers compared to dry areas, which were net sinks of CH_4. Soil moisture, forest type, temperature, and pH have been shown to affect microbial communities involved in CH_4 production and oxidation; however, much remains unknown about methanogens and CH_4-oxidizing bacteria in forests (Aronson et al., 2012). In past studies, CH_4 flux measurements in forests were conducted mainly using the chamber enclosure technique due to the simplicity and relatively inexpensive nature of the method. Chamber measurements are particularly suitable for process-level studies of components within the ecosystem, such as individual plants or the soil (Keller et al., 1990; Singh et al., 1997). Drawbacks to this technique are that it is labour-intensive, may lack spatial and temporal representativeness, and may be subject to biases arising from soil disturbance and inadequate gas mixing (Christiansen et al., 2011). Simpson et al. (1997) found that at an aspen site, chambers in the tower footprint measured net uptake of CH_4; however, flux-gradient measurements showed the forest as a net source due to large sources located in small areas in the forest. This discrepancy stresses the importance of spatial heterogeneity in sources and sinks, and the challenge in upscaling from point measurements within a forest ecosystem. In contrast, the eddy covariance (EC) method integrates fluxes over a larger area, leading to measurements that are more representative of the ecosystem as a whole (Clement et al., 1995). Recent technological advances providing high measurement precision at sampling frequencies of 10–20 Hz have allowed for the accessibility of the EC technique for measurements of CH_4 fluxes. The off-axis integrated output spectrometer (OA-ICOS) has been shown to provide a robust system that meets these requirements

BGD
9, 17743–17774, 2012

(Hendriks et al., 2008) and has been used in previous studies to measure CH_4 fluxes (Smeets et al., 2009; Eugster and Plüss, 2010; Parmentier et al., 2011; Querino et al., 2011).

In this paper, we describe growing season flux measurements of CH_4 in a mixed-deciduous forest in central Ontario, Canada using an OA-ICOS instrument operating at 10 Hz. Soil-level CH_4 exchange within the tower footprint was characterized using small flux chambers along seven toposequences. The flux estimates are also compared with diurnal cycles in the mixing ratio of CH_4 to assess the spatial representativeness of the flux values measured by eddy covariance.

2 Materials and methods

2.1 Site description

Measurements were made at the Haliburton Forest and Wildlife Reserve (45°17′11″ N, 78°32′19″ W) located in the Great Lakes/St. Lawrence forest region of Ontario. The forest is an uneven-aged forest managed under selection system silviculture, and the measurement site has not been harvested since 1997 resulting in a heterogeneous canopy structure. The forest region is dominated mainly by *Acer saccharum* Marsh., *Fagus grandifolia* Ehrh., *Tsuga canadensis* L., and *Betula alleghaniensis* Britt, and contains sandy and acidic soils with a pH of 3.6–5.7 (Peng and Thomas, 2006). The topography near the tower site contains a mix of highlands and valleys ranging from in elevation between 380–506 meters above sea level (masl). The tower itself is located in a higher elevation area at approximately 500 m a.s.l., and is surrounded mainly by forest with pockets of small fens, marshes, and lakes located at lower elevations. Measurements were carried out at the top of a 32 m tower, 8 m above the canopy. A diesel generator was located 100 m northeast (usually downwind) of the measurement tower. The generator showed negligible interference for CH_4 and CO_2 based on simultaneous measurements by a NO_x / NO_y chemiluminescence instrument (Air Quality Design Inc.,

BGD
9, 17743–17774, 2012

Wheat Ridge, Colorado) that recorded obvious spikes in nitrogen oxides during periods of generator influence.

2.2 Eddy covariance flux measurements

Gas measurements were made between 2 June and 24 October 2011 using an OA-ICOS model #09-0033 Fast Greenhouse Gas Analyzer (FGGA) developed by Los Gatos Research Inc. (Mountain View, California, USA), which provided continuous simultaneous measurement of CH_4, CO_2, and H_2O at a response rate of 10 Hz. The FGGA was located in an environmentally-controlled building at the bottom of the tower and an external dry vacuum scroll pump (Varian TriScroll 300, Palo Alto, California, USA) was used to pull air at approximately 30 l per minute (lpm) through 38 m of PVC tubing with an internal diameter of 0.95 cm. In addition to the internal filter, an external filter (7 µm) was placed in the inlet line near the instrument, however the air stream was not dried. A correction factor for the cross-sensitivity to H_2O in the CO_2 and CH_4 measurements by the OA-ICOS was determined through external calibration (refering to the Supplement), which results in a calculated CH_4 mixing ratio in dry air. The mirror ringdown time remained above 10 µs during the measurement period, well above the lower limit of 3.5 µs defined by the manufacturer. Audits of the OA-ICOS with a gas standard showed good accuracy and stability in the measured mixing ratio (refer to supplementary information), although the measurement noise at 10 Hz was higher using the external pump ($1\sigma = 5\,\mathrm{nmol\,mol^{-1}}$) for CH_4 compared to $\pm 2\,\mathrm{nmol\,mol^{-1}}$ using the internal pump (1 lpm). This increased noise could be the result of the higher pumping speed leading to pressure fluctuations in the measurement cell (Smeets et al., 2009). The inlet of the FGGA was positioned 0.7 m away from the sonic anemometer (model CSAT3, Campbell Scientific Inc., Logan, Utah, USA) used for wind direction and wind speed measurements, and a LI-7500 (LI-COR Inc., Lincoln, Nebraska, USA) open-path (OP) infrared gas analyzer (IRGA) for additional CO_2 and H_2O measurements. These sensors were secured to a pole extended from the top of the tower and

BGD
9, 17743–17774, 2012

directed in a southwesterly direction (215°) to minimize the influence of flow distortions on the flux measurements.

2.3 Data processing and corrections

Data processing was carried out using an EC flux program written in Igor Pro (WaveMetrics). Covariances between the vertical wind component and fluctuations in gas concentrations were calculated in intervals of 30 min. Lag time due to sensor separation was calculated for each averaging period by cross-correlating the measured gas mixing ratio with the vertical wind. Maximum correlation between the FGGA and the anemometer was found with a lag of approximately four seconds. Non-horizontal terrain and tilt in the sonic anemometer was corrected for by a 3-D coordinate rotation using the planar fit technique (Wilczak et al., 2001). Non-stationarity was also tested for by splitting each 30-min interval of data into six periods of 5 minutes. The average covariance from the six 5-min periods were compared to the 30-min average, which provided a measure of relative non-stationarity for each flux data point (Foken and Wichura, 1996). Fluxes with a relative non-stationarity larger than 40 % were removed. High frequency noise present from the FGGA instrument was observed to influence the high frequency portion of the cospectrum above 0.2 Hz for CH_4 (refer to the Supplement). This was corrected for by assuming cospectral similarity compared to other scalars not influenced by high frequency noise, such as open-path CO_2 and temperature, with vertical wind. The high frequency correction resulted in an average reduction in the flux magnitude of ∼21 % for CH_4. In post-processing, flux data were then despiked for significant outliers by removing values more than eight standard deviations from the average. In addition, flux data from a wind direction of 0°–93° were also removed due to flow distortion that resulted from the tower scaffolding (this also removed a significant fraction of airmasses influenced by the generator). The limit of detection for an individual half hour CH_4 flux measurement was calculated to be $\sim 5\,\mathrm{nmol\,m^{-2}\,s^{-1}}$ (refer to the Supplement). The quality control procedures resulted in the removal of

BGD
9, 17743–17774, 2012

approximately 35 % of the CH_4 flux data: non-stationarity test (~ 22 % removed), despiking (4 points total, < 0.1 %), flow distortion data removal (~ 13 %).

Webb-Pearman-Leuning (WPL) corrections were not applied during the calculation of the fluxes because dry mixing ratios could be calculated from the FGGA data at high frequency, and because variations in temperature were dampened by the closed-path system. Friction velocity (u^*) cut-offs are typically used in the EC technique to remove the influence of aerodynamic and boundary layer dynamics on fluxes that are expected to be dominated mainly by biologically-controlled surfaces. For CO_2 fluxes, which are strongly controlled by biological activity of plants in the ecosystem, fluxes affected by low u^* are considered artefacts of the EC technique. However, for CH_4 fluxes, the influence of turbulence could potentially be an environmental driver in determining variability in CH_4 fluxes, as has been proposed in previous studies (Sachs et al., 2008; Wille et al., 2008). Emission fluxes can be missed during periods of low turbulence due to the competing influences of horizontal advection or from storage effects within the canopy; however, uptake is unlikely to be as strongly influenced.

2.4 Soil chamber measurements

Static chamber measurements of CH_4 were conducted using a distributed network of soil flux chamber stations established within the footprint of the tower approximately every 2.5 weeks (on eight separate days) throughout the EC measurement period following Basiliko et al. (2009). An additional intensive measurement campaign was carried out, in which fluxes were measured every four hours over 20 h on 22–23 August 2011. A permanent collection of 10 cm PVC collars were installed over seven topographical gradients including five sites each: low slope, toe slope, mid-slope, high slope, and ridge slope. On measurement days, PVC chambers were secured to each collar sealed with a closed cell foam gasket, and 30 mL was taken four times over a 90-min period using airtight syringes. The samples were analyzed in the laboratory using gas chromatograph/flame ionization detector (GC-FID) and a packed column (SRI Instruments, Menlo Park, California, USA). The changes in mixing ratio (relative to

BGD
9, 17743–17774, 2012

calibration standards) were used to infer a flux for each site, corrected for chamber temperature and ambient atmospheric pressure at each sampling time. Soil temperature and moisture measurements were also made adjacent to the chamber measurement sites throughout the year using automated DS1921G Thermochron iButtons (Maxim Integrated Products, San Jose, California, USA) and Odyssey Soil Moisture Loggers (Dataflow Systems PTY Ltd., Christchurch, New Zealand). Sensors were located at a 5 cm soil depth and each logger recorded hourly average temperatures and volumetric soil moisture measurements respectively. The soil temperature data used in the analysis is the average of measurements from six different sites. Soil moisture data was collected at two sites, but only data from the drier site was used in the analysis.

2.5 Ancillary measurements

Additional sensors (Onset Computer Corporation, Bourne, Massachusetts) measured temperature, wind direction and speed within the canopy, and amount of rain, which was measured at intervals of 5 minutes throughout the measurement period. A canister study was also conducted at the tower site 22–24 August 2011 using 6 L stainless steel summa polished canisters (Scientific Instruments Specialists, Moscow, Idaho) with a passive flow controller integrated over two hour sampling periods during the day, and between 18:00–06:00 h over the nighttime. Samples were collected at three different levels; near ground (5 m), mid-canopy (20 m), and above-canopy (32 m). The samples were brought to the Environment Canada Analysis and Air Quality Section laboratory (AAQS) where the samples were diluted with air and analyzed with a GC-FID 6890 (Agilent, Santa Clara, California) equipped with a 1 mL sample loop. A Valco Bond (VICI, Gig Harbor, Washington) capillary column (VP-molsieve 5A, 30 m, 0.53 mm ID, 15 µm film thickness) was used to separate CH_4. A four point calibration curve using a CH_4 certified reference standard was used to determine the concentration of the sample from the peak area of the chromatograms. The dilution factor was then applied to give the proper mixing ratio of CH_4.

BGD
9, 17743–17774, 2012

3 Results

3.1 Methane mixing ratio measurements

The average methane mixing ratio (± one standard deviation) was $1920 \pm 40\,\mathrm{nmol\,mol^{-1}}$ over the measurement period. The mixing ratio had synoptic scale variations through the measurement period ranging between 1860–2080 $\mathrm{nmol\,mol^{-1}}$. Many of these increases in the methane mixing ratio were observed to match with a southerly wind direction. A back trajectory analysis (not shown here) indicated that the highest mixing ratios corresponded to airmasses that had passed over the Greater Toronto Area, approximately 200 km to the southwest. Mixing ratio measurements from the FGGA agreed with canister measurements at the top of the tower to within $5\,\mathrm{nmol\,mol^{-1}}$ (0.3 %) when averaged over the two-day measurement period.

3.2 Methane fluxes and environmental measurements

Over the measurement period, the average flux value (± one standard deviation of the mean) was $-2.7 \pm 0.13\,\mathrm{nmol\,m^{-2}\,s^{-1}}$ with highly variable fluxes, as shown in Fig. 1. Methane fluxes are expected to be more variable than fluxes of other gases, such as CO_2, due to the episodic nature of many of the processes affecting CH_4 fluxes (Eugster and Plüss, 2010). For an ecosystem that contains both potential sources and sinks of CH_4, this variability is likely to be higher compared than that of an ecosystem that is predominantly a source. Noise in each individual half hour measurement is another important contributor to the variability seen in Fig. 1. The right hand-panel indicates the distribution of calculated fluxes when the covariance analysis is carried out by substituting a half hour measurement of calibration gas at a constant mixing ratio, used to estimate the detection limit (see Fig. S5). While a significant number of individual flux measurements are within the detection limit, calculation of the 48 h running average (black line in Fig. 1) demonstrates that there are coherent signatures in

BGD
9, 17743–17774, 2012

the flux time series that emerge through time averaging. Further support for net uptake in the vicinity of the tower comes from the canister measurements at the three levels on the tower, which showed a decrease in mixing ratio from above the forest canopy to the near-ground level, with a vertical gradient ranging from 0–100 nmol mol^{-1}.

The average of the soil-level fluxes from the multiple sites along the five different topographical gradient positions over the eight measurement dates was −0.19 nmol m^{-2} s^{-1}, also indicating net uptake, but of a smaller magnitude. This calculation was performed assuming equal contribution to the flux footprint from each elevation represented in the topographical gradients. However, using LiDAR data, the low-slope areas within the tower footprint are estimated to have a percent area contribution of 6.1 %. The soil-level fluxes measured from these low-slope areas in June and July represented high emission rates, skewing the averages measured from the chambers in these months. After July, chamber fluxes were more consistent, with a net flux of approximately −2 nmol m^{-2} s^{-1} across all topographical gradients.

The average ambient air and soil temperatures (± standard deviation) were 11.8 ± 6.8 °C and 14.0 ± 2.8 °C, respectively, during the measurement period of 145 days from June 2 to October 24, 2011. The soil temperature, averaged over six sites, remained relatively constant compared to ambient temperature, even during a cold period lasting over 4 days in mid-September where ambient temperatures dropped to −9.0 °C, but soil temperatures remained above 5.0 °C. The soil percent moisture by mass was measured at two mid-elevation sites with average values of 60.0 ± 14 % and 72.1 ± 7.9 %, with ranges of 8–100 % and 55–100 %, respectively. Soil moisture was influenced by rain events that dried in subsequent days. Data from the wetter site is not included in the statistical comparisons, because that range of soil moisture did not appear representative of the tower footprint. However, measurements at the two sites varied in consistent ways, and both lacked evidence of a diurnal cycle.

Monthly averages of CH_4 fluxes (Table 1) showed an increase in uptake from June at −1.28 nmol m^{-2} s^{-1} to a peak of −3.99 nmol m^{-1} s^{-1} in September. Subsequently, in October, rates dropped to an average of −2.95 nmol m^{-2} s^{-1}. Both air and soil

BGD
9, 17743–17774, 2012

temperatures peaked in July and decreased as the season progressed. Soil moisture also had the highest average value in July at 69 % and decreased soil moisture in September and August. Wind speed remained relatively constant from June through September with higher average wind speeds in October. The wind direction was predominantly northwesterly, however in September, southeasterly conditions were often experienced.

3.3 Diurnal variability in CH_4 mixing ratio and fluxes and in environmental variables

Methane fluxes averaged over the measurement period showed a diurnal trend (Fig. 2) where fluxes decreased in magnitude between 03:00–08:00 with a minimum uptake at $-0.6\,\mathrm{nmol\,m^{-2}\,s^{-1}}$, and increased in uptake mid-day between 10:00–16:00 at $-4.6\,\mathrm{nmol\,m^{-2}\,s^{-1}}$. The diurnal trend for CH_4 fluxes showed little seasonal variability in terms of timing, though on average tended towards higher overall uptake as the summer season progressed. Mixing ratios also exhibited a diurnal trend reaching a minimum in the mid-afternoon around 15:00 and with a gradual increase starting at 18:00 that peaked at 06:00 in the early morning. This is shown in Fig. 3, which displays the diurnal cycle in the difference from the daily average, dCH_4 ($\equiv CH_4(t)$–CH_4(average). Figure 4 indicates that the average slope in mixing ratio change each night between 0:00 and 05:00 depends on the average wind direction over that time period. It is evident that CH_4 was more likely to accumulate overnight under southerly flow conditions, indicating that important emission areas may be found to the south of the tower.

The diurnal variability in CH_4 fluxes and several environmental variables is displayed in Fig. 5. Of the environmental measurements made at the tower; ambient and soil surface temperature (Fig. 5b and c), and wind speed above the canopy and within the canopy (mid-canopy) (Fig. 5e and f) showed the strongest diurnal cycles. Soil moisture (Fig. 5d) did not exhibit a significant and consistent diurnal pattern. To investigate if these environmental variables could explain the variability in CH_4 flux, scatter plots against soil moisture, and above-canopy and mid-canopy wind speed were made

BGD
9, 17743–17774, 2012

(Fig. 6). Each panel is overlaid with the average CH_4 flux for narrow bins of the independent variable, and with a line of best fit through the entire dataset. The relationship is strongest for CH_4 flux with the mid-canopy wind speed (similar diurnal pattern) and with soil moisture, though over longer timescales.

3.4 Comparison of eddy covariance and chamber flux measurements

Measurements from the static chambers can help to shed light on the observed seasonal progression fluxes measured from the tower, with the lowest net uptake observed in June and July (Table 1). Figure 7 compares the weighted average (6 % influence from low-slope areas, equivalent contributions from all other topographic positions) of the chamber flux measurements with eddy covariance measurements made during the nighttime (22:00–07:00) and daytime (08:00–19:00) of the same day. This is effectively comparing a spatial average of the chamber measurements with a time average of the eddy covariance measurements. Based on the EC measurements alone, it was not possible to determine whether the lower net uptake in June and July resulted from reduced methanotrophy or increased methanogenesis, however the chambers indicate that methanogenesis played a role in limited parts of the tower footprint. For example, on 21 June, all seven chambers at low slope positions showed evidence of emission, averaging to 163 nmol m^{-2} s^{-1}. After the low-slope soils began to dry (August to October), the chamber fluxes at all five topographical gradient positions showed net uptake.

4 Discussion

4.1 Methane mixing ratio and fluxes

The diurnal amplitude of the mean CH_4 mixing ratio is approximately 15 nmol mol^{-1} corresponding to an increase of $\sim$1.25 nmol mol^{-1} h^{-1} between sunset and sunrise (Fig. 3). One explanation for the observed diurnal trend is the accumulation of surface

BGD
9, 17743–17774, 2012

emissions within the shallow stable boundary layer during the night, with the subsequent breakup of the nocturnal boundary layer resulting in the dilution of accumulated CH_4 (Culf et al., 1997). If the observed diurnal cycle is the result of a local surface flux, then assuming an arbitrary nocturnal boundary layer height of 100 m, this corresponds to a nighttime *emission* of 1.4 nmol m^{-2} s^{-1}. While this appears at odds with the net uptake calculated by eddy covariance in the flux footprint, the concentration footprint of a tower is much larger than the flux footprint (Vesala et al., 2008), suggesting that emissions may dominate in the areas further upwind of the tower. This is not unexpected, as the study area is a mosaic of lakes, wetlands, and upland forests, with the tower situated at a relatively high elevation.

A rough estimation of the flux footprint was calculated using a footprint parameterization by Kljun et al. (2004) for a 90 % limit of integration using averaged values for surface friction velocity and the standard deviation of vertical velocity fluctuations (σ_w) and with an estimate for the planetary boundary layer height (h). Different model inputs were used for daytime (σ_w = 0.47, u^* = 0.32, h = 100 m) and nighttime (σ_w = 0.78, u^* = 0.60, h = 1500 m). The footprint estimate was 574 m for nighttime and 516 m for daytime average values. However, nighttime values did extend further past ~800 m with low range u^* and σ_w values. With an average flux footprint of ~550 m, this area is much smaller than the expected mixing ratio footprint of the tower (Vesala et al., 2008). The discrepancy between the flux and mixing ratio footprint highlights the difficulty in capturing important land-surface emission and uptake processes from canopy-scale observations. Footprint considerations also make it difficult to compare direct observations of CH_4 fluxes with model predictions when the footprint is not known precisely (Riley et al., 2011).

4.2 Dependence of fluxes on environmental variables

Previous studies in CH_4-emitting ecosystems have found that physical processes such as ebullition or diffusive emissions, and biological processes such as plant-mediated transport have been the main controlling variables in CH_4 emissions (Mikkelä et

BGD
9, 17743–17774, 2012

al., 1995; Miyata et al., 2000), resulting in increased emissions during the daytime and decreased emissions at night (Satpathy et al., 1997; Miyata et al., 2000; Baldocchi et al., 2012; Hatala et al., 2012). However, the same controlling processes cannot explain the diurnal cycle in CH_4 fluxes at Haliburton Forest, given that uptake appears highest during the day. Soil temperature has been shown to affect methanotrophic activity (Reay et al., 2007), though at Haliburton Forest the CH_4 fluxes showed a weak relationship with soil temperature.

During the measurement period, soil moisture increased steeply during rain events and decreased in subsequent days, with minimal evidence of a diurnal cycle. An increase in soil moisture decreases air-filled pore space and hence the diffusion of relatively CH_4-rich air through the soil to the methanotrophs, decreasing net uptake. If soil moisture levels are high enough to support methanogens, the surface can act as a net emission source of CH_4 (Reay et al., 2007). From the scatterplot (Fig. 6a), CH_4 fluxes on average increased as soil moisture increased, with a change from an average uptake flux of approximately $-5\,\mathrm{nmol\,m^{-2}\,s^{-1}}$ below a soil moisture of 40 % to lower uptake fluxes of around $-2\,\mathrm{nmol\,m^{-2}\,s^{-1}}$ above 68 % soil moisture. From the chamber measurements, there is clear evidence that some low elevation areas in the tower footprint were methane sources, particularly in June and July. While the two separate soil moisture probes did not detect evidence of a diurnal variation in soil moisture measured at 5 cm, it is possible that in the rhizosphere, diurnal variations were more pronounced due to plant activity. For example, Werban et al. (2008) found that soil moisture decreased in the daytime and increased during the nighttime. Similarly, Mikkelä et al. (1995) observed an effect of soil moisture on methanotrophic activity, where oxygen was transported from the atmosphere to the rhizosphere, increasing CH_4 microbial oxidation. The position of the soil moisture probes may not have captured the rhizosphere-driven soil water changes, potentially underestimating the diurnal variation in the soil moisture and our interpretation of its influence on microbial CH_4 fluxes.

Ambient temperature (Fig. 5b) increased during the day after 08:00 and decreased slowly after 17:00. A similar trend was observed for soil surface temperatures (Fig. 5c);

BGD
9, 17743–17774, 2012

however, the variation in the soil temperature was much smaller and the daytime increase was delayed until 12:00. The influence of temperature variations on CH_4 fluxes at the soil level would greatly depend on the depth of microbial community influencing the fluxes. There was no measurement of the vertical distribution of the microbial population in the soil at the Haliburton site, although previous studies have shown that methanotroph habitat is typically near the surface at 3–15 cm in depth (Curry, 2007). Given that the soil layer at Haliburton Forest is very shallow, commonly only 35–65 cm (Peng and Thomas, 2006), the methanotrophs are likely to be quite close to the surface. A scatterplot of CH_4 flux with both soil and ambient temperatures (not shown) demonstrated minimal correlation compared to other environmental variables, suggesting that the temperature variations during the measurement period had little effect on CH_4 fluxes.

The diurnal cycle of wind speed is most similar to that of CH_4 fluxes, with both the wind speed above the canopy (Fig. 5e) and mid-canopy (Fig. 5f) maximizing between 08:00 and 20:00, similar to CH_4 uptake. Although at much lower magnitudes, the mid-canopy wind speed showed a more consistent diurnal trend than the wind speed above the canopy. With an increase in mid-canopy wind speed from 0 to 1 $m\,s^{-1}$, there was a consistent increase in uptake flux from −0.5 to −4.8 $nmol\,m^{-2}\,s^{-1}$ (Fig. 6). The mid-canopy wind speed is likely to be most representative of the impact of atmospheric dynamics on soil-level processes. Previous studies have shown that advection forced by pressure pumping related to atmospheric turbulence can increase gaseous flux through the snowpack (Massman et al., 1995) and landfill soils (McBain et al., 2005). The impact of this effect is most significant when the concentration gradients are weak (Massman et al., 1997). Typically this "pressure pumping effect" led to increased emissions in CO_2 and CH_4 resulting from ebullition or ventilation. In a study over the Siberian tundra, Sachs et al. (2008) observed that increased turbulence and wind speed in non-inundated areas could lead to a higher concentration gradient between the methane-enriched soil to the turbulent boundary layer, thus resulting in an increased diffusive flux. At Haliburton Forest, where uptake appears to dominate emission in the flux

BGD
9, 17743–17774, 2012

footprint of the tower, the concentration gradient is reversed. The soil layer would be relatively methane-depleted compared to the atmospheric boundary layer. Increased aeration in the coarse soils in Haliburton Forest may facilitate transport of CH_4-rich air from the overlying atmosphere to the methanotrophs and/or transport of CH_4-depleted air out of the soil into the atmosphere. Yonemura et al. (2000) observed wind-induced acceleration of gas transport in topsoil that could have played a role in gas uptake, where lower levels of uptake were observed during lower wind speed conditions.

The driving forces of monthly-scale variations in CH_4 fluxes were found to be different than for the diurnal trends. Wind speed remained relatively constant until October, when fall storms and a lack of a developed canopy likely contributed to an increase in wind speed (Table 1). However, CH_4 uptake decreased in October despite increased wind speed. This could have been the result of decreases in air and soil temperatures, and/or increased rain events that occurred in October, which could have reduced overall rates of microbial activity and limited rates of CH_4 and O_2 diffusion to aerobic methanotrophs. The strongest environmental correlation with CH_4 fluxes on monthly scales was soil moisture: as the soils progressively dried from June when the soil moisture was the highest, the uptake of CH_4 increase, reaching a peak in September.

4.3 Comparison of eddy covariance and chamber flux measurements

Early in the season, both the chamber and EC measurements indicate a small net flux, whereas in the months of August and September, both techniques indicate significant uptake, though the daytime EC flux values have a much higher magnitude. As chamber measurements were made in the daytime, this suggests a potential underestimation of CH_4 uptake rates due to the chamber blocking the wind, effectively shutting off the supply of CH_4 and O_2 to soil methanotrophs. While increased CH_4 uptake as a result of higher wind speed has not been widely reported in previous studies, it may be an important consideration for static chamber measurements that are conducted in enclosed containers with flow restrictions or a lack proper headspace mixing (Davidson et al., 2002; Christiansen et al., 2011). The better agreement between the magnitudes of

BGD
9, 17743–17774, 2012

the nighttime EC fluxes (lower wind speed) and the chamber measurements is suggestive of such an artefact in the chamber measurements, though certainly not conclusive evidence. A comparison between the tower-based EC measurements and the soil-level chamber measurements can also be used to investigate the presence of any significant above-ground CH_4 fluxes. Given that the canopy-level rates of CH_4 uptake are, if anything, higher than the soil-level rates of uptake, there is little evidence of an abiological plant source of CH_4 at Haliburton Forest.

5 Summary and conclusions

Methane fluxes inferred from tower measurements above the canopy and chamber measurements at the soil level indicate that the area of forest in the flux footprint is a net sink for CH_4. This was further supported by vertical gradients of CH_4 mixing ratios measured at three levels of the tower during two days of the measurement campaign. A diurnal trend was evident throughout the campaign with highest net uptake during the day and decreased uptake during the night. The diurnal cycle of CH_4 fluxes was most similar to the diurnal cycle in mid-canopy wind speed. The correlation of CH_4 uptake with wind speed can be interpreted to result from the increased substrate supply (CH_4 and O_2) to methanotrophs in the soil. At monthly timescales, soil moisture content appeared to be the major control on CH_4 fluxes. Chamber measurements indicate that high soil moisture could lead to hypoxic conditions and microbial methanogenesis at low elevation sites. Soil moisture may also influence the flux by limiting diffusion rates of substrate to soil methanotrophs. From measurements in August–October, daytime averaged EC fluxes were 4–7 times higher in net uptake than in chamber fluxes. However, when tower fluxes were averaged from the nighttime of the same day, the flux values were much closer. This suggests that static chambers may underestimate the magnitude of uptake within coarse soils if wind-driven ventilation is an important factor. It can also be concluded that plant-level CH_4 production, proposed by Keppler et al. (2006), does not detectably influence CH_4 dynamics in Haliburton Forest.

BGD
9, 17743–17774, 2012

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/9/17743/2012/ bgd-9-17743-2012-supplement.pdf.

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BGD

9, 17743–17774, 2012

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BGD

9, 17743–17774, 2012

Table 1. Monthly averages of eddy covariance CH_4 fluxes and ancillary measurements.

	CH_4 Flux ($nmol\,m^{-2}\,s^{-1}$)	Temperature (°C)		Soil Moisture (%)	Wind Speed ($m\,s^{-1}$)	
		Air	Soil		Above Canopy	Mid-Canopy
Jun	−1.28	12.3	13.3	65	2.35	0.43
Jul	−1.66	17.1	16.2	69	2.32	0.41
Aug	−3.39	14.1	15.8	59	2.29	0.41
Sep	−3.99	9.9	13.6	43	2.36	0.53
Oct	−2.95	3.8	10.1	67	2.92	1.07

BGD
9, 17743–17774, 2012

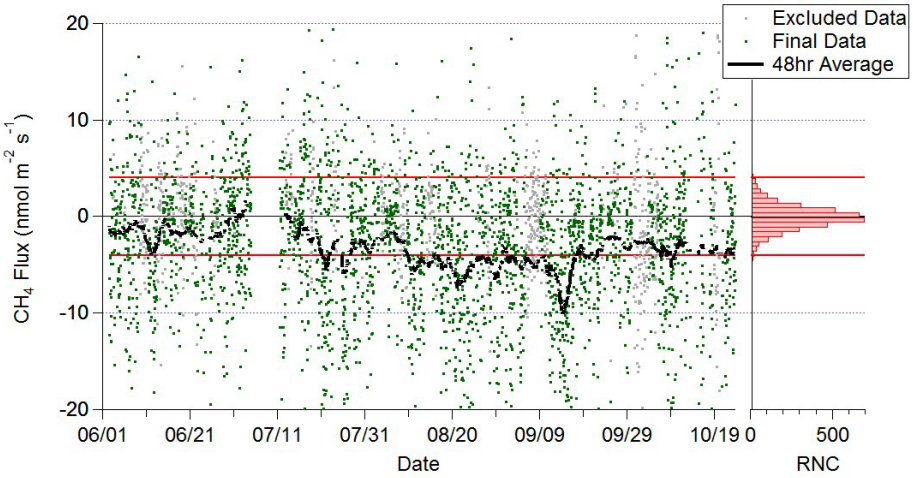

Fig. 1. CH_4 flux (green) with excluded data (grey) and limit of detection of individual flux measurements (dashed red) and distribution (right panel) from 2 June–24 October 2011.

BGD
9, 17743–17774, 2012

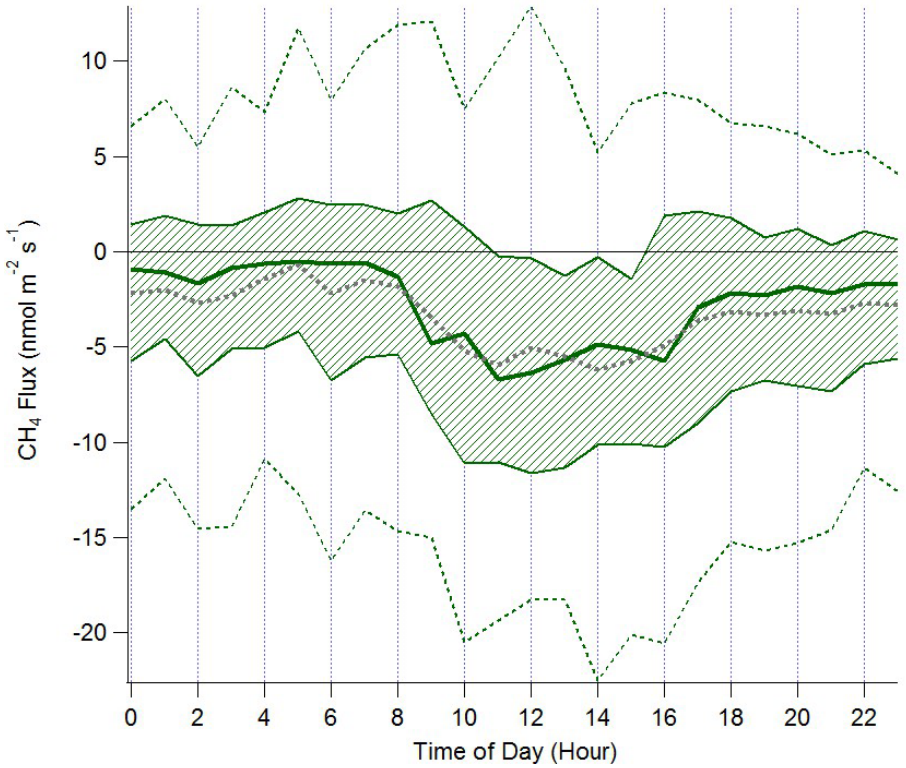

Fig. 2. Median diurnal cycle of CH_4 fluxes (bold) and average flux (dashed gray), with interquartile range (shaded area), and the 5th and 95th percentile range (indicated by dashed line).

BGD
9, 17743–17774, 2012

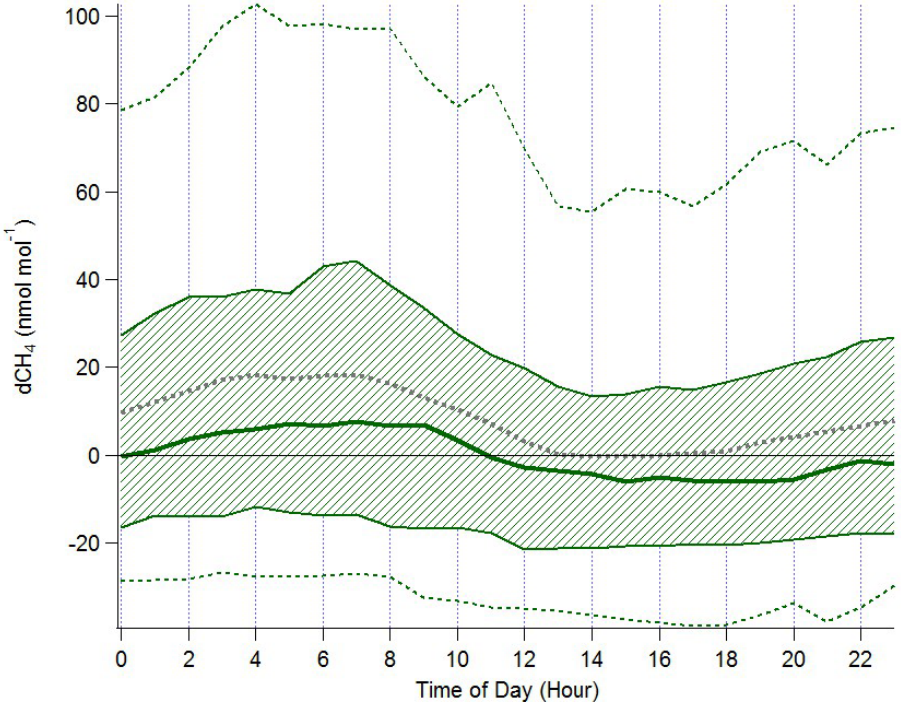

Fig. 3. Mean (grey dashed line) and median (bold green) diurnal cycle of change in CH_4 mixing ratio, with interquartile range (shaded area), and the 5th and 95th percentile range (indicated by green dashed lines) normalized to the median value.

BGD
9, 17743–17774, 2012

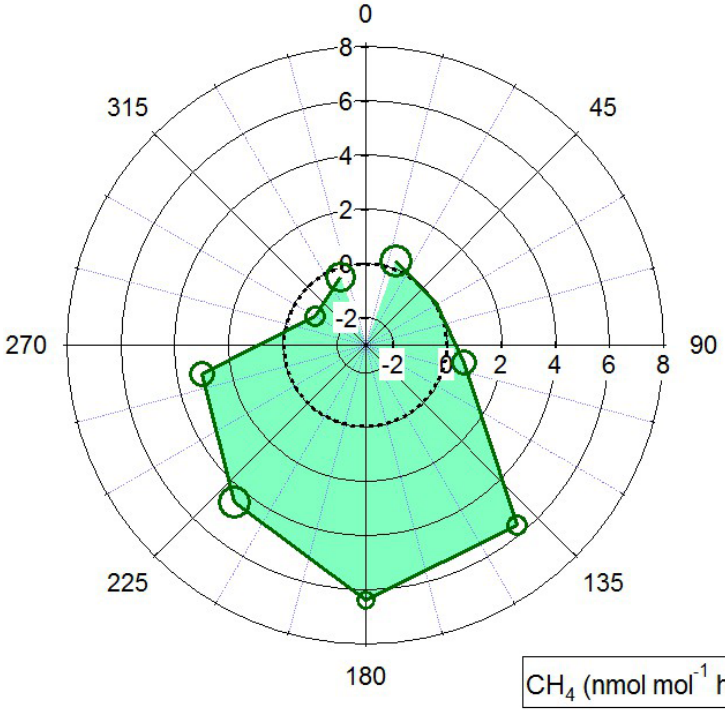

Fig. 4. Polar plot of bin-averaged slope of the change in CH_4 mixing ratio between 00:00–05:00 from each day during the measurement period with each the size of the circular markers indicating the number of points within the bin (8–19 points).

BGD
9, 17743–17774, 2012

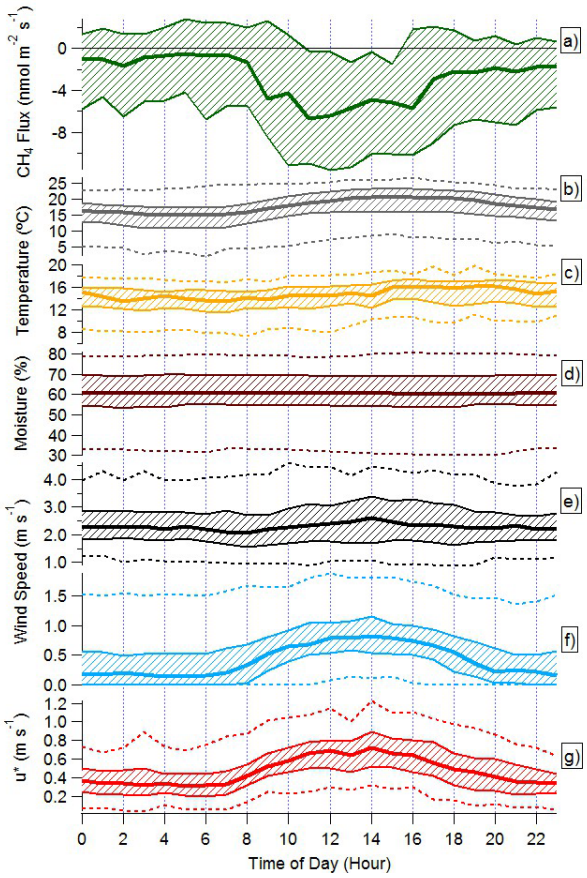

Fig. 5. Median (bold) diurnal cycle of CH_4 fluxes with average flux (dashed gray) **(a)**, ambient temperature **(b)**, soil surface temperature **(c)**, soil moisture **(d)**, wind speed above canopy **(e)**, mid-canopy **(f)**, and friction velocity, u^* **(g)**; with interquartile range (shaded area), and the 5th and 95th percentile range (indicated by dashed line).

BGD
9, 17743–17774, 2012

Methane fluxes measured by eddy covariance and static chamber techniques

J. M. Wang et al.

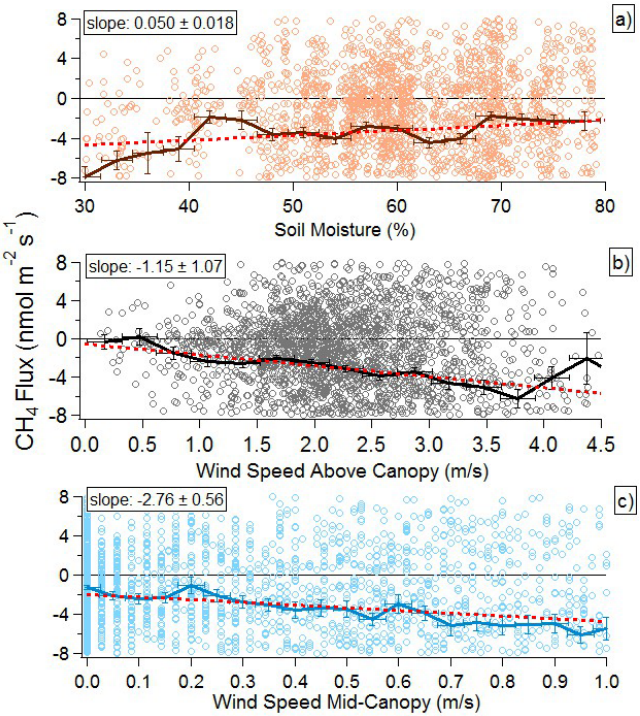

Fig. 6. Scatter plot of CH_4 fluxes with soil moisture **(a)**, and wind speed above canopy **(b)** and mid-canopy **(c)** with a line of best fit (red dashed line), error bars indicating standard deviation of the mean (vertical), and size of the bin (horizontal). Dashed red line is a linear regression through the data, with the slope $\pm 2\sigma$ reported for each panel.

BGD
9, 17743–17774, 2012

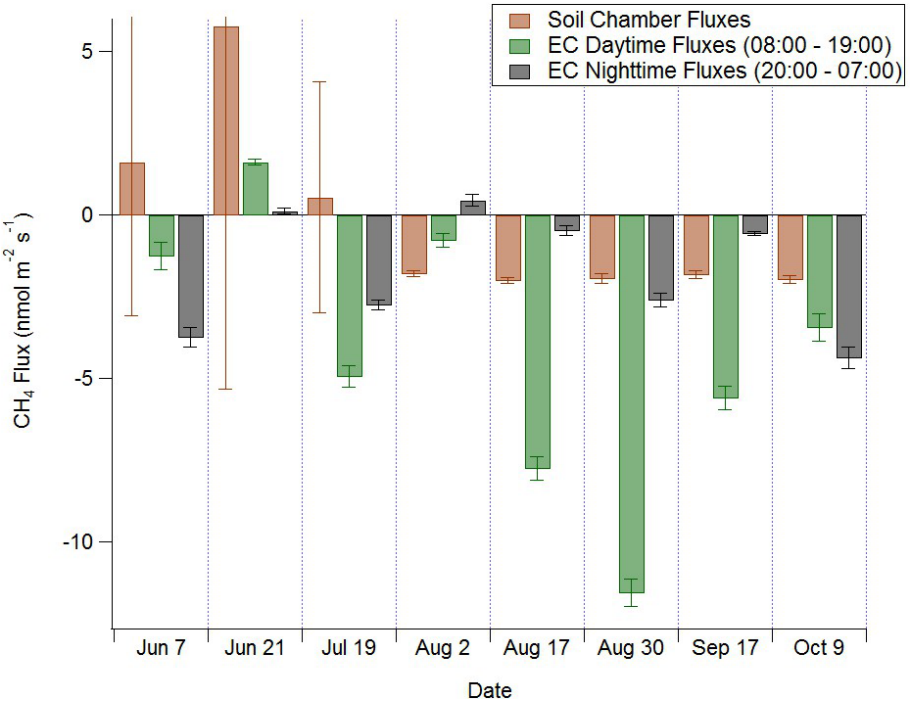

Fig. 7. Bar graph of fluxes calculated from chamber measurements (brown), and FGGA measurements during the daytime (green) between 08:00–19:00 and nighttime (black) between 22:00–07:00 with error bars indicating standard deviation of the mean within the averaging period for the EC fluxes, or across the sites for the chamber fluxes.

BGD
9, 17743–17774, 2012