Landscape-level terrestrial methane flux observed from a very tall tower

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\textbf{A B S T R A C T}

Simulating the magnitude and variability of terrestrial methane sources and sinks poses a challenge to ecosystem models because the biophysical and biogeochemical processes that lead to methane emissions from terrestrial and freshwater ecosystems are, by their nature, episodic and spatially disjunct. As a consequence, model predictions of regional methane emissions based on field campaigns from short eddy covariance towers or static chambers have large uncertainties, because measurements focused on a particular known source of methane emission will be biased compared to regional estimates with regards to magnitude, spatial scale, or frequency of these emissions. Given the relatively large importance of predicting future terrestrial methane fluxes for constraining future atmospheric methane growth rates, a clear need exists to reduce spatiotemporal uncertainties. In 2010, an Ameriflux tower (US-PFa) near Park Falls, WI, USA, was instrumented with closed-path methane flux measurements at 122 m above ground in a mixed wetland–upland landscape representative of the Great Lakes region. Two years of flux observations revealed an average annual methane (\text{CH}_4) efflux of 785 ± 75 mg C–CH\textsubscript{4} m\textsuperscript{−2} yr\textsuperscript{−1}, compared to a mean CO\textsubscript{2} sink of −80 g C–CO\textsubscript{2} m\textsuperscript{−2} yr\textsuperscript{−1}, a ratio of 1% in magnitude on a mole basis. Interannual variability in methane flux was 30% of the mean flux and driven by suppression of methane emissions during dry conditions in late summer 2012. Though relatively small, the magnitude of the methane source from the very tall tower measurements was mostly within the range previously measured using static chambers at nearby wetlands, but larger than a simple scaling of those fluxes to the tower footprint. Seasonal patterns in methane fluxes were similar to those simulated in the Dynamic Land Ecosystem Model (DLEM), but magnitude depends on model parameterization and input data, especially regarding wetland extent. The model was unable to simulate short-term (sub-weekly) variability. Temperature was found to be a stronger driver of regional CH\textsubscript{4} flux than moisture availability or net ecosystem production at the daily to monthly scale. Taken together, these results emphasize the multi-timescale dependence of drivers of regional methane flux and the importance of long, continuous time series for their characterization.

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1. Introduction

The contribution of microbial methane (\text{CH}_4) from wetlands remains a significant source of uncertainty in closing the global methane budget (Mikaloff Fletcher \textit{et al.}, 2004). In particular, wetland methane emissions may contribute as much as 25–40% of global \text{CH}_4 anthropogenic emissions and are the leading source of interannual variability in atmospheric \text{CH}_4 (Bousquet \textit{et al.}, 2006; Chen and Prinn, 2006; Crill \textit{et al.}, 1993). The recent increase in the growth rate of atmospheric \text{CH}_4 lends particular urgency to improving global simulations and inversions of the terrestrial methane source (Chen and Prinn, 2006; Collins \textit{et al.}, 2006). One
set of hypothesized mechanisms is the role of warming of high latitudes and wetting of the tropics (Dlugokencky et al., 2009). Because CH\(_4\) emissions are closely linked to changes in regional hydrology and temperature, and ongoing climate changes are likely to have a significant impact on regional water tables and wetland soil temperatures, there is a high likelihood that climate change will affect wetland CH\(_4\) emissions (Roulet et al., 1992; Sulman et al., 2009).

Model results provide motivation for long-term in situ observations of terrestrial CH\(_4\) sources and sinks. However, virtually all in situ measurements of surface to atmosphere CH\(_4\) flux have been conducted either at the plot scale, typically with chamber-based measurements (e.g., Jungkunst and Fiedler, 2007), or more recently at the ecosystem scale, particularly with eddy covariance flux towers (e.g., Hatala et al., 2012). In contrast, atmospheric tracer-transport inversions (e.g., Bergamaschi et al., 2010; Miller et al., 2013), global ecosystem models (e.g., Matthews and Fung, 1987; Tang et al., 2010; Tian et al., 2010), and global remote sensing based estimates of CH\(_4\) sources (e.g., Bloom et al., 2010) are provided at much larger spatial scales. Consequently, a scale mismatch arises for evaluation across methods. This scale mismatch is particularly difficult for CH\(_4\) because of fine-scale spatial heterogeneity of CH\(_4\) sources and sinks and sampling biases toward known CH\(_4\) sources (e.g. peatlands).

The primary objective of this study is to evaluate the first very tall tower continuous eddy covariance flux measurement of CH\(_4\) in a regional landscape. Further, we compared the magnitude and variability of these observations to plot-scale wetland and forest observations and model simulations. In late 2010, we instrumented a very tall tower in northern Wisconsin USA to observe CH\(_4\) fluxes at 122 m above the ground and CH\(_4\) concentration at 3 heights, sampling a spatially heterogeneous mix of upland forest and lowland wetland systems (Fig. 1). The site has been measuring CO\(_2\) and H\(_2\)O eddy fluxes at this height and two others since 1996.

Since the pioneering studies using tunable diode laser spectroscopy-based eddy covariance for CH\(_4\) fluxes (Fowler et al., 1995; Kim et al., 1998; Shurpali and Verma, 1998; Suyker et al., 1996), there have been growing a number of publications based on short-term CH\(_4\) flux observations (e.g., Friborg et al., 2003; Hargreaves et al., 2001; Nicoli et al., 2013). With the development of reliable, low-drift, closed and open path methane analyzers (McDermitt et al., 2011), it is now possible to maintain long time series of CH\(_4\) fluxes (e.g., Baldocchi et al., 2012; Hatala et al., 2012; Olson et al., 2013; Rinne et al., 2007; SMEETS et al., 2009; Wille et al., 2008). None of these measurements have been made at the landscape scale (25–100 km\(^2\)) from a very tall tower, and only a subset of these studies report simultaneously on CH\(_4\), CO\(_2\), and H\(_2\)O flux measurements.

The value of continuous observations at landscape scales is to directly observe to what extent episodic and spatially heterogeneous emissions influence the net annual budget of biospheric CH\(_4\) fluxes. Only continuous observations, for example, can regularly capture (or record) pulses of CH\(_4\) (e.g., after a rainstorm or during ebullition events) (Strack and Waddington, 2008) along with non-growing season fluxes, which may also be substantial (Pelletier et al., 2007; Yu et al., 2007).

We seek to understand the nature of regional or landscape-scale net ecosystem exchange of CH\(_4\) (NEE CH\(_4\)). In theory, we would expect that if wetland CH\(_4\) production (\(\text{R}_{\text{wetland}}\)) dominates forest CH\(_4\) consumption and wetland CH\(_4\) oxidation, then the landscape CH\(_4\) flux would be proportional to the wetland spatial extent and its mean flux as measured by chambers. Also, some ecosystem models simulate CH\(_4\) production based on assuming a constant ratio of either ecosystem respiration (\(\text{R}_{\text{eco}}\)) to CH\(_4\) or NEE CO\(_2\) to NEE CH\(_4\) at annual timescales (e.g., Potter, 1997). To investigate these claims, we ask:

- What is the magnitude of NEE CH\(_4\) in a mixed forest–wetland landscape and how does it compare to site-level chamber-based estimates?
- How predictive are environmental factors such as water table and temperature or other biogeochemical fluxes such as \(\text{R}_{\text{eco}}\) CH\(_4\) or NEE CO\(_2\) on daily to interannual variability of NEE CH\(_4\)?
- How well does a state–of–the–art ecosystem model simulate landscape NEE CH\(_4\)?

2. Methods

2.1. Site description

Methane flux and profile measurements were made at the WLEF very tall tower US-Pfa Fluxnet site (Davis et al., 2003) in Wisconsin, USA (45.945° N, 90.273° W). The surrounding landscape (Fig. 1) is a representative mix of forested and open wetlands (28% in entire region (~50 km radius), 18% within 5 km radius of tower) with the remainder primarily composed of mixed deciduous and evergreen forests with most stands ranging from 30 to 70 years old. Most of the landscape is within the Chequamegon-Nicolet National Forest and forests that are actively managed for multiple purposes, including recreation, wildlife habitat, and timber production. Wetlands in the region include both open fens and forested bogs and a smaller proportion of open-water bodies. Upland stands are generally characterized by mixed northern hardwood species (Acer saccharum, Tilia americana, Fraxinus pennsylvanica, Betula
Table 1: Very tall tower site and instrument characteristics.

<table>
<thead>
<tr>
<th>Coordinates</th>
<th>45.945° N, 90.273° W</th>
</tr>
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</table>

<table>
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<tr>
<th>Land cover (general region)</th>
<th>28% wetland, 67% upland mixed forest, 5% grass or other</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annual mean temperature (1995–2013)</td>
<td>5.7°C</td>
</tr>
<tr>
<td>Annual total precipitation (1995–2013)</td>
<td>586 mm</td>
</tr>
<tr>
<td>Summer mean temperature (JJA, 1995–2013)</td>
<td>18.4°C</td>
</tr>
<tr>
<td>Summer total precipitation (JJA, 1995–2013)</td>
<td>243 mm</td>
</tr>
<tr>
<td>Measurement height Instruments</td>
<td>122 m above ground</td>
</tr>
<tr>
<td>Flux gas analyzer (CH4)</td>
<td>Picarro, Inc. 1301-f</td>
</tr>
<tr>
<td>Flux gas analyzer (CO2/H2O)</td>
<td>Licor, Inc. LI-6262</td>
</tr>
<tr>
<td>Storage profile (CH4)</td>
<td>Los Gatos, Inc. LGR Fast Methane Analyzer</td>
</tr>
<tr>
<td>Storage profile (CO2)</td>
<td>Licor, Inc. LI-7000</td>
</tr>
<tr>
<td>Sonic anemometer</td>
<td>ATI, Inc. Type K</td>
</tr>
<tr>
<td>u* cutoff</td>
<td>0.2 m s⁻¹</td>
</tr>
</tbody>
</table>

*papyrifera*); early to mid-successional aspen–fir (*Populus tremuloides, Populus grandidentata, Abies balsamea*); and pine–spruce (*Pinus resinosa, Pinus banksiana, Picea glauca*). Lowlands are generally characterized by wetland shrub and sedge species in fens and along stream banks (*Alnus rugosa, Salix spp., Carex spp.*); deciduous hardwood species in retired and seasonal drainage ways (*Fraxinus nigra, Ulmus rubra, Acer rubrum*); ericaceous shrubs and moss in open bogs (*Chamaedaphne calyculata, Ledum groenlandicum, Sphagnum spp.*); and wetland conifers in drier peatlands and bog edges (*Thuja occidentalis, Larix laricina, Picea mariana, A. balsamea*).

The site has an interior continental climate with cold winters and warm summers (Table 1). Precipitation is greatest in the spring and fall, though there is regular and abundant winter snowfall. Over the two decades of flux tower CO₂ measurements, the site has varied from being a small source of CO₂ to a modest sink for CO₂ (Desai, 2014). Previous studies (Desai et al., 2008a) have indicated that the mean tower footprint samples a landscape that is representative of much of the Upper Midwest U.S. forested region, and the proportions of wetland and forest sampled are representative of the average wetland/forest coverage in the entire National Forest.

2.2. Very tall tower measurements

Flux measurements of CO₂, H₂O, heat, and momentum and associated tower profile meteorology and surface micrometeorology have been made continuously at the site since the middle of 1996 (Davis et al., 2003). Flux measurements have been made at three heights above ground, 30 m, 122 m, and 396 m. CO₂ and H₂O flux measurements at each level were made with Licor, Inc. LI-6262 infrared gas analyzers and ATI Type K sonic anemometers (Table 1). Each level has a gas analyzer in a trailer at the tower base with large vacuum pumps drawing air to them. For the upper two levels, an additional gas analyzer was placed on tower at their respective heights to minimize data loss and account for flux loss for long tube lengths. Generally, fluxes between the on tower sensors and the long tube length sensors compared favorably, especially after high frequency spectral loss corrections were applied (Berger et al., 2001). All flux instruments were sampled initially at 5 Hz, but switched to 10 Hz in 2006. In addition to the flux measurements, each level has measurements of temperature and humidity (Vaisala, Inc. HMP45C). Measurements of incoming above-canopy photosynthetically active radiation (PAR) were made in the clearing at the base of the very tall tower. Precipitation and soil moisture were made at a nearby stand-scale flux tower (US-WCr) and compared and gap-filled with other micrometeorological stations within 30 km of the tower.

In the middle of 2010, we installed a cavity ring-down spectrometer (Picarro Inc., model 1301-f) for measurement of continuous CO₂ and CH₄ concentration. This instrument is one of several new instruments with high sensitivity for continuous high-frequency CH₄ measurements that have arisen since the development of low-cost quantum cascade and infrared lasers (Kroon et al., 2007), with limited sensor calibration drift (Hendriks et al., 2008). The instrument was housed inside a temperature-controlled trailer and sub-sampled air diverted from the 122 m level LI-6262 analyzer. A second pump was applied to draw air into the Picarro cavity. The Picarro analyzer maintains a constant pressure and temperature in the cavity and directly reports mole fraction of the gas species. We did not attempt to sync the LI-6262 water vapor signal to estimate 10 Hz CH₄ dry air mixing ratio, but rather applied a Webb–Pearson–Leuning (WPL, Webb et al., 1980) correction as discussed below.

Storage flux was derived from profile measurements of CO₂ and CH₄ made on the tower. CO₂ profile measurements were made with a Licor, Inc. LI-7000 analyzer maintained by the National Oceanographic and Atmospheric Administration (NOAA) Earth Systems Research Lab (ESRL) (Andrews et al., 2014). These measurements have been made since 1995 with a Licor 6251, which was replaced by the LI-7000 in May 2009. A separate set of opens at the same heights as the flux tower levels provided air to the analyzer, which performed 5-minute sequential sampling of each level. These air samples were dried, flow controlled, and calibrated with zero and span gases multiple times per day. In spring 2010, we installed a Los Gatos, Inc. LGR Fast Methane Analyzer, drawing dried and conditioned air from the NOAA ESRL system and added standards with known CH₄ concentration for calibration. Both profile measurements used in this study were acquired from calibrated and interpolated time series of CO₂ and CH₄ concentrations from the three flux heights.

Flux and meteorology measurements were acquired with Campbell Scientific, Inc. data loggers, except for the Picarro, which has its own internal storage system. To maintain time alignment, all loggers and computers were synced to NIST UTC internet time on an hourly basis. Flux data processing for CO₂ and H₂O fluxes was virtually unchanged from Berger et al. (2001). The observed CO₂ concentrations were calibrated against the NOAA ESRL tower CO₂ observations within a 24-hour window, and similarly water vapor was calibrated to water vapor mixing ratio obtained from on tower Vaisala HMP45C sensors and surface barometric pressure measurements. Picarro CO₂ and CH₄ observations had very small drift and have not shown any need for calibration beyond factory calibration. A WPL correction for dilution by water vapor is needed to obtain the dry air mole fraction of CO₂ and CH₄ using the approach of Hiller et al. (2012). We opted not to apply the direct correction method of Baldocchi et al. (2012) and Detto et al. (2011) because lining up H₂O observations from the LI-6262 to the Picarro at 10 Hz was not easily possible, except for limited periods, where we did compare the two approaches.

Sonic anemometer data were rotated to long-term (12-month) planar fits. Air sampling lags were identified with maximal lagged covariance, and high-frequency empirical spectral corrections were applied (Berger et al., 2001). Given the larger eddies present at 122 m than lower heights, we have previously showed that an hour-long averaging time is more appropriate (Berger et al., 2001).

One particular issue with our setup was drifting clocks between the Picarro and the datalogger that stores the sonic data, even with regular time syncing. Further, the Picarro’s raw data are not stored at regular time intervals owing to data processing and laser control sequence. We used a nearest neighbor approach for each time stamp, especially following the method of Eugster and Plüss (2010) to line up time stamps to the sonic anemometer, with replication if needed. Lag corrections were applied after this.
drift owing to malfunctioning computer clocks was obvious in the long-term time series of lag times, requiring manual adjustment of the window of acceptable lag times.

Additional quality control was applied, including range checks, spike detection, and low turbulence filtering. We applied a 0.2 m s⁻¹ filter for low turbulence at night. For CO₂ and H₂O fluxes, where multiple heights and sensors were available, a preferred intake height algorithm (Davis et al. 2003) was applied to combine the independent flux observations, preferring higher levels in daytime and the lowest level at night during periods of negative heat flux, indicating decoupling of higher intake heights from the surface layer, as described in Davis et al. (2003).

While systematic biases are possible from assumptions made in data filtering, calibration, and flux algorithms, there is also the issue of random flux uncertainty. Given the sporadic nature of CH₄ emissions against a low background flux at most sites, turbulent flux uncertainty can be large relative to flux magnitude (Kroon et al., 2010). To estimate flux uncertainty for CH₄, we applied the method of Salesky et al. (2012). Flux uncertainty was derived from successive computation of eddy fluxes with longer averaging times, estimating the standard deviation of these sub-hour fluxes and extrapolating them to the hour to estimate flux uncertainty. Computationally, this calculation of fluxes at all averaging times up to one hour was done in Fourier spectrum to speed computation time. The method has been shown by Salesky et al. (2012) to be reliable and comparable to other methods based on random flux shuffling (Billesbach, 2011). For daily and cumulative errors, hourly errors were summed by squares after accounting for temporal autocorrelation up to a 24 h lag.

For calculation of seasonal and annual fluxes, we also gap-filled the flux measurements of CO₂ and CH₄ and inferred gross primary production (GPP) and ecosystem respiration (ER). CO₂ fluxes were gap-filled and partitioned by using the method described in Desai et al. (2005), based on a moving-window regression of quality controlled nighttime net ecosystem exchange of CO₂ (NEE CO₂) and a fit of daytime observations to incoming photosynthetically active radiation (PAR). This method has compared favorably to other methods in common usage (Desai et al., 2008b).

There is currently no generally-accepted method for gap-filling for CH₄ fluxes. Our initial attempts at similar regression approaches as for NEE CO₂ at the hourly scale did not find strong relationships, similar to what has been reported by others (e.g., Dengel et al., 2013). Short gaps (<4 h) at the hourly scale were filled with linear interpolation. However, at the daily scale, a stronger relationship with temperature allowed us to apply a second order polynomial fit between CH₄ daily flux and air temperature, accounting for random flux uncertainty as described above. While soil temperature would be possible for a short tower, there is no single estimate of regional soil temperature, and thus air temperature is the best metric of regional average ecosystem temperature. Further, we modeled random flux uncertainty as a linear function of mean flux to extrapolate random uncertainty of the gap-filled daily fluxes, to which we summed with the one-sigma uncertainty of the regression to estimate total random uncertainty. We also separately estimated gap-filling uncertainty by repeated calculation of annual sums of NEE CH₄ with differing regression coefficients based on their uncertainty.

Finally, flux footprints were estimated for each hour to estimate source contributions and potential footprint bias. We applied the empirical CBL model of Wang et al. (2006), which relies on similarity theory to derive mean Gaussian surface influence functions as a function of boundary layer characteristics such as convective velocity scale (w*), boundary layer depth (h), roughness height (z₀), and Monin–Obukhov length (z/L). These were used to confirm representative sampling of land cover in the tower climatological footprint as shown in Fig. 1.

2.3. Plot-level observations

For comparison of regional fluxes from the tower to in situ CH₄ fluxes, we analyzed static chamber flux measurements made in four wetlands and three upland forests near the very tall tower (within 20 km, though not necessarily within the flux footprint). Static chamber measurements were made in the growing season (May–Sep) of 2005 and 2006 based on syringe sampling from closed, vented PVC chambers (25 cm diameter, 10 cm height). Chamber headspace samples (15 mL) were collected four times during a 30-minute period, with each sample transferred to an air-tight vial for transport to the laboratory. Vials were analyzed for CH₄ concentration by gas chromatography using a flame ionization detector (Hewlett Packard, 5890A) with calibrated standards (Scott Specialty, Inc.). Fluxes were calculated based on the increase in headspace concentration over time (Weishampel and Kolka, 2008). At each site, 3 plots containing 4 subplots each with 3 fixed, static chamber collars were sampled approximately monthly across the growing season (days of year 100 to 278). Mean soil temperature and volumetric soil water content were also measured in the plots at each flux sampling time point.

Wetland sites included an open, sphagnum-dominated bog (South Fork, SF; 45°55.37′N 90°07.92′W), a sedge-dominated riparian fen (Wilson Flowage, WF; 45°48.99′N, 90°10.29′W), an alder-dominated riparian wetland (Lost Creek, LC; 46°04.96′N, 89°58.72′W), and a cedart swampland (CS; 45°56.53′N 90°16.21′W). Forest sites included one mature deciduous forest, Willow Creek (WC; 45°48.47′N, 90°4.72′W), and two recent clear-cut (<10 years at time of sampling) deciduous forests, Riley Creek (RC; 45°54.53′N, 90°07.27′W) young aspen and Thunder Creek (TC; 45°40.23′N 90°03.25′W). In this study, we were primarily interested in the mean and range of the wetland emissions and forest soil methane consumption over the entire growing season.

In addition, for comparison purposes, we also upscaled the chamber measurements using flux footprint-weighted estimates of wetland and forest cover multiplied, respectively, by mean and standard deviation of wetland and forest chamber fluxes over all collars, all sites, and all growing season sampling dates (assuming 179 day growing season), assuming no methane exchange in winter or for other land cover types. Intra and inter site variability across collars was propagated via Monte Carlo sampling to estimate sensitivity of upscaling.

2.4. Numerical modeling

The Dynamic Land Ecosystem Model (DLEM) is a comprehensive terrestrial ecosystem model that couples carbon, nutrient and water cycles in terrestrial ecosystems for estimating the hydrological and biogeochemical fluxes and pool sizes at multiple scales from site to region/globe and with time step ranging from day to year. Through carbon–nutrient–water coupling, DLEM is capable of simultaneously depicting the biosphere–atmosphere exchange of CO₂, CH₄ and N₂O under multiple natural and anthropogenic disturbances (Tian et al., 2010). The model can simulate regional hydrology including evapotranspiration, runoff and soil moisture (Liu et al., 2013). Here, we ran the model in two modes over the study period: a cut-out of a previously continental-scale regionally parameterized model and a single site-level model. The regional model was cut-out from a spatial resolution of 5 by 5 arc-minutes (around 9.2 × 9.2 km grid at the equator), using default land cover for the grid cell. The site model was run with local estimates of wetland and forest cover. There is large difference in the percent area of three major plant functional types between regional data and site data (Table 2). The site model experiment was run with gap-filled tower observed meteorology, whereas the regional model was run with large-scale gridded meteorology (Climate Research Unit
National Center for Environmental Prediction—CRUNCEP). We ran the model in site and regional modes to assess biases in modeling of regional CH₄ flux.

3. Results

3.1. Fidelity of very tall tower flux

Methane eddy covariance flux measurements in 2011 and 2012 were successfully made over 68% of the time (Table 3). An additional 13% of all available hours were filtered for low turbulence conditions (\(u^* < 0.2 \text{ m s}^{-1}\)). Spectral loss from long tube lengths and lag times were nearly identical for NEE CO₂ and NEE CH₄ and similar to earlier results published in Berger et al. (2001).

Flux observations sampled a footprint (Fig. 1) with an average fetch in any one direction of 1–4 km and sampled all wind sectors. The relatively self-similar pattern of wetlands and forests in the fetch allowed for a “homogenous” sampling of diverse upland and lowland ecosystems around the tower. However, given the lower amount of wetland in the immediate vicinity of the tower compared to the larger region, the 2011 footprint climatology showed an average wetland sampling of 17%, with forests at 70%, and other covers (grass, water, roads, shrubs) at 13%. Daytime and nighttime footprints were similar, except for slightly enhanced contribution of the ~100 m diameter grassy clearing surrounding the tower during the daytime.

Flux observations of methane had turbulent behavior quite similar to CO₂. WPL correction for water vapor dilution was found to be modestly important for NEE CH₄ from closed path analyzers (Fig. 2). WPL corrected NEE CH₄ was on average 1.2% larger than uncorrected. We also tested whether a WPL correction was similar to the direct dry air mixing ratio flux calculation. Over a one month period, H₂O mixing ratio observations were synced in time and used to directly compute dry mole fraction CH₄ at 10 Hz. Our results showed strong correlation and low bias, but on average, the direct dry-air NEE CH₄ were 1.6% larger than WPL-corrected flux, or overall nearly 3% larger than uncorrected NEE CH₄ (Fig. 2).

Because methane fluxes at the site were small, random turbulent uncertainty could be a significant component. Our application of the Salesky et al. (2012) method revealed a baseline uncertainty (level of detection) of NEE CH₄ to be 0.13 nmol CH₄ m⁻² s⁻¹ at the hourly scale and 0.42 mg C−CH₄ m⁻² day⁻¹ at the daily scale. Over the two year study period, 2.2% of hours had an NEE CH₄ magnitude below that amount, though 15.2% of daily NEE CH₄ was below the daily threshold, primarily during the winter. Average uncertainty was 20% for hourly fluxes and 12% for daily fluxes (Fig. 3). However,

Table 2

<table>
<thead>
<tr>
<th>Plant functional type</th>
<th>DLEM regional (%)</th>
<th>DLEM site (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wetland</td>
<td>44</td>
<td>28</td>
</tr>
<tr>
<td>Forest</td>
<td>43</td>
<td>67</td>
</tr>
<tr>
<td>Grass and other</td>
<td>13</td>
<td>5</td>
</tr>
</tbody>
</table>

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Table 3

<table>
<thead>
<tr>
<th>Observed annual fluxes and meteorology during study period.</th>
<th>2011</th>
<th>2012</th>
</tr>
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<tbody>
<tr>
<td>Annual mean temperature (°C)</td>
<td>5.7</td>
<td>7.0</td>
</tr>
<tr>
<td>Annual precipitation (mm)</td>
<td>458</td>
<td>568</td>
</tr>
<tr>
<td>Summer ([JJA]) temperature (°C)</td>
<td>19.1</td>
<td>19.4</td>
</tr>
<tr>
<td>NEE CO₂ (g C−CO₂ m⁻² yr⁻¹)</td>
<td>58.0</td>
<td>101.4</td>
</tr>
<tr>
<td>GPP (g C−CO₂ m⁻² yr⁻¹)</td>
<td>858.1</td>
<td>1160.7</td>
</tr>
<tr>
<td>Sasa (g C−CO₂ m⁻² yr⁻¹)</td>
<td>799.7</td>
<td>1059.3</td>
</tr>
<tr>
<td>NEE CH₄ (mg C−CH₄ m⁻¹ yr⁻¹)</td>
<td>911 ± 84</td>
<td>659 ± 64</td>
</tr>
<tr>
<td>Ratio NEE CH₄:NEE CO₂ (%)</td>
<td>−1.57</td>
<td>−0.65</td>
</tr>
<tr>
<td>Ratio NEE CH₄:Sasa (%)</td>
<td>0.0011</td>
<td>0.00062</td>
</tr>
<tr>
<td>Missing NEE CH₄ (%)</td>
<td>29</td>
<td>36</td>
</tr>
<tr>
<td>Screened NEE CH₄ (%)</td>
<td>12</td>
<td>13</td>
</tr>
</tbody>
</table>

Fig. 2. Comparison of in-line water vapor correction and post-WPL correction for water vapor dilution applied to CH₄ efflux fluxes. (a) Comparison of “wet” mole fraction CH₄ flux to “wet” mole fraction CH₄ flux with WPL applied, showing the effect of water vapor dilution is to underestimate fluxes by ~1%. (b) A direct dry mole fraction estimated flux shows high correlation and low bias with WPL-corrected CH₄ flux, but the direct computed fluxes are on average 1.6% larger.

Fig. 3. Estimate of flux random turbulent uncertainty (y-axis) versus absolute magnitude of NEE CH₄ for (a) hourly and (b) daily scale. The blue line shows bin-averaged NEE CH₄ for intervals of (a) 10 nmol CH₄ m⁻² s⁻¹ or (b) 4 mg C−CH₄ m⁻² day⁻¹, while the red line shows the result of linear regression. In general, uncertainty scales linearly with flux. The intercept is an estimate of minimal detectable flux.
at the hourly or daily scale, uncertainty only weakly scales with flux magnitude. These uncertainty estimates were propagated in estimates of total annual flux, as discussed below.

For very tall tower measurements, the contribution of below sensor height storage flux can be significant for all fluxes with strong surface sources or sinks, especially at night (Fig. S1). Storage flux magnitude contributed a median of 48% of the total NEE CH₄ magnitude around noon, but 75% of the nighttime NEE CH₄ at the hourly scale. Storage flux declines to zero as averaging timescale increases. Nonetheless, this flux cannot be neglected for hourly to daily NEE CH₄ observations from very tall towers, especially at night. For NEE CO₂ and NEE CH₄, storage flux is on the same order as eddy flux at night, though the largest magnitude contribution of storage flux occurs shortly after sunrise, when flushing of accumulated nighttime CO₂ or CH₄ near the surface leads to a strong negative storage flux, which quickly declines to zero by solar noon. However, for CH₄, this peak occurs roughly 1–2 h later than for CO₂, and the decline to zero is more gradual and also shifted by a similar amount. Further, in the morning during the growing season, flux and storage terms for NEE CO₂ are the same sign (negative), while for NEE CH₄, they are opposite signs (positive for eddy flux, negative for storage), leading to a possibly greater source of error for diurnal fluxes of NEE CH₄, especially if storage and eddy fluxes have differing source area contribution. For daily NEE, this effect is negligible as average daily storage flux for CH₄ is <4% of daily NEE CH₄.

3.2. Comparison to plot-level chamber observation

Plot level chamber methane fluxes (Fig. 4a) reveal significant within and across site differences in collar-averaged daytime CH₄ fluxes across the four wetland (193 measurements) and three upland forest study sites (152 measurements) in the region. Tower observed daytime growing-season NEE CH₄ have efflux rates that bracket the static chamber observations, with most tower observations occurring in-between the largest and smallest wetland flux observations. Tower maximum efflux rates do not generally exceed those observed at the high CH₄ emission sedge site, where plant-mediated pathways and high proportion of labile carbon likely facilitated CH₄ flux. Chamber CH₄ exchange from wetland or upland forest sites had significantly different distributions than tower NEE CH₄ (Wilcoxon Rank-Sum U-Test p < 0.001). The average daily efflux of CH₄ from all sampled wetlands was 5.08 ± 15.3 nmol CH₄ m⁻² s⁻¹ and average forest soil uptake was −1.8 ± 1.1 nmol CH₄ m⁻² s⁻¹. Tower mean NEE CH₄ averaged over the period corresponding to the earliest and latest sample dates (days of year 100–278) was 3.9 ± 11.2 nmol CH₄ m⁻² s⁻¹. Large negative values of NEE CH₄ observed by the tower were much larger than any observed at chamber sites. The highest magnitude of chamber CH₄ emissions was observed from the groundwater fed sedge dominated wetland (WF), which promoted plant-mediated transport and was wetter than the other sites.

While upscaling is of limited value given the amount of chamber data available, it can provide some estimate of whether the chamber fluxes are representative of the landscape flux. Mean chamber-based upscaled NEE CH₄ was 145 ± 436 mg C–CH₄ m⁻² s⁻¹ from wetlands and −214 ± 131 mg C–CH₄ m⁻² s⁻¹ from forests. This amounts to a total upscaled NEE CH₄ of −64 ± 567 mg C–CH₄ m⁻² s⁻¹, as the forest CH₄ sink essentially cancels out wetland emissions. Tower observations show a net source of CH₄ of 785 ± 75 mg C–CH₄ m⁻² s⁻¹ observed by eddy covariance. Wetland chamber emissions alone are less than 20% of the tower observed source. Caution is required as the chambers were sampled in different years (2005–2006) from the tower (2011–2012). Summer mean temperatures for chamber observations in 2005–2006 were 0.25 °C warmer and 2% wetter on average compared to tower observations in 2011–2012. These findings highlight the need to better delineate wetland type and area, peat depth, edge effects, and decomposability for accurate upscaling.

3.3. Seasonal and interannual patterns of carbon fluxes

Patterns of daily CH₄ (Fig. 5a), CO₂ (Fig. 5b) fluxes and inferred GPP (Fig. 5c) and Rₑ (Fig. 5d) at the site showed seasonal patterns typical of temperature-limited temperate mixed forest regions. NEE of CO₂ and CH₄ were generally negatively correlated at a monthly scale (Table 4). Peak uptake of NEE CO₂ was in early to midsummer, while NEE CH₄ showed higher daily variability and lacked a distinct early-mid summer peak. Patterns of NEE for CO₂ and CH₄ were similar in both years, but 2012 featured both an earlier growing season start and a pronounced drought in the mid-summer (Jul–Sep) (Fig. 6c). While drier in the growing season, the earlier green-up led to higher GPP in 2012 for most of the growing season (Fig. 5c), and higher Rₑ from mid-summer onward. The period of high ecosystem respiration was not directly related to any reduction of CH₄ emissions, a feature only apparent at the annual scale. Both years had growing seasons (May–Sept) that 10–28% drier and 0.4–0.8 °C warmer than the long-term (1995–2013) average.

NEE CH₄ exhibited periods in both the winter and growing season of high emissions relative to the average for the time period (Fig. 5a). These “bursts” were primarily generated in the turbulent flux term, were more common and prominent for CH₄ than CO₂, were skewed in the positive direction, and were not coincident with excursions in NEE CO₂, nor were they consistently co-occurring with large pressure or turbulence changes or any known fossil-fuel CH₄ sources. These high emission days in summer also exhibited relatively high turbulent flux uncertainty and were more pronounced in 2011 than 2012. NEE CH₄ hourly bursts that exceed two standard deviations from a background seven-day average over the measurement period occurred only 6% of the time, but
**Fig. 5.** Time series of daily (a) NEE of CH$_4$, (b) NEE of CO$_2$, (c) GPP and (d) $R_{eco}$ for a two-year period at the tower site. Red crosses are gap-filled, and gray bars show turbulent flux uncertainty. Blue line shows a 10 day smoothed average. CH$_4$ fluxes show a decline from 2011 to 2012 in contrast to increases seen in GPP and $R_{eco}$ and no change in NEE.

**Fig. 6.** Similar to Fig. 5 but for meteorological forcing of gap-filled (a) daily mean temperature, (b) daily cumulative photosynthetically active radiation, (c) cumulative precipitation, and (d) near surface soil moisture from an upland, mixed forest in the flux tower footprint. Both years had similar temperature and cloudiness, but differing patterns of growing season precipitation leading to lower soil moisture in 2012.

**Table 4**

Pearson linear correlation coefficient (r) between NEE CH$_4$ and other observations at hourly to monthly averaging scales. Only significant correlations ($p < 0.1$) are shown after correcting for time series auto-correlation. No significance denoted with ns. NEE CH$_4$ is not strongly correlated to soil moisture, but instead most positively correlated to temperature and GPP and $R_{eco}$ at these time scales.

<table>
<thead>
<tr>
<th>Averaging time</th>
<th>Temperature</th>
<th>Photosynthetically active rad-iation (PAR)</th>
<th>Volumetric surface soil moisture</th>
<th>Net ecosystem exchange CO$_2$ (NEE CO$_2$)</th>
<th>Gross primary production (GPP)</th>
<th>Ecosystem respiration ($R_{eco}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hour</td>
<td>ns.</td>
<td>ns.</td>
<td>ns.</td>
<td>0.09</td>
<td>ns.</td>
<td>ns.</td>
</tr>
<tr>
<td>Day</td>
<td>0.49</td>
<td>0.43</td>
<td>ns.</td>
<td>ns.</td>
<td>0.49</td>
<td>0.53</td>
</tr>
<tr>
<td>Week</td>
<td>0.71</td>
<td>0.66</td>
<td>ns.</td>
<td>-0.49</td>
<td>0.72</td>
<td>0.74</td>
</tr>
<tr>
<td>Month</td>
<td>ns.</td>
<td>ns.</td>
<td>ns.</td>
<td>-0.68</td>
<td>0.80</td>
<td>0.79</td>
</tr>
</tbody>
</table>
they contributed nearly a quarter of the absolute flux, which adds a further challenge to gap-filling, which in our current version cannot capture these events. Unfortunately, during the anomalously warm early spring of 2012, CH4 flux observations were not available. Spectral analyses of the modes of variability for gap-filled NEE CO2 and NEE CH4 from 2011–2012 show that the contribution of timescale to NEE CH4 is relatively similar to NEE CO2, though NEE CH4 scale has reduced contribution of variation from the monthly (20–30 day) scale and greater contribution at the seasonal (>100 day) scale (Fig. 7b).

Overall, annual NEE CH4 from the region is relatively small in magnitude, on average 1.1% of the NEE CO2 by mole or mass fraction (Table 3). Cumulative NEE CH4 (Fig. 7a) in the two years averaged 785 ± 75 mg C m⁻² yr⁻¹ while NEE of CO2 was -80 g C m⁻² yr⁻¹. CH4 fluxes were lower in 2012, though just outside the uncertainty bounds arising from both gap-filling and flux random uncertainty. In 2012, CH4 fluxes appear to be suppressed in the early to mid-growing season in slightly warmer, but wetter conditions compared to the previous year, though the presence of gaps in part of this period complicates the analysis. The remaining part of the growing season has a similar pattern of net emissions as the prior year (Fig. 5).

The shifts in RCO and CH4 NEE in 2012 were likely related to the 1.3 °C higher annual air temperature in 2012 and lack of precipitation in late July through August in 2012 (Table 3). Warmer air temperatures in 2012 led to a very early growing season, and a quasi-stationary ridge of high pressure promoted longer periods of dry, warm conditions in summer 2012. While the reduction in precipitation is not particularly large, there was a significant change in timing of precipitation (Fig. 6c), depressing 2012 soil moisture through the late summer and fall (Fig. 6d).

Interannual variability of CH4 flux between the two years is 32% of the mean flux, slightly larger than variability in GPP (29%) and RCO (28%), but different than for NEE CO2 fluxes over this time (34%), as the longer growing season (increased GPP) in 2012 more than offset the warmer, drier conditions in the same year (increased RCO). The consequence of the longer growing season and warmer conditions was that GPP increased by 35%, while RCO increased by 32% between 2011 and 2012, whereas annual CH4 fluxes declined by 28%. Interannual variability in prior years for CO2 NEE has been larger. The range of annual CO2 fluxes measured from 1996–2012 exceeded 296 g C m⁻² yr⁻¹ (Desai, 2014), as the site has shifted from being a net source of CO2 to a net sink in some years.

3.4. Growing season diurnal patterns

Diel patterns for NEE CH4 are particularly unique showing an early to mid-morning negative peak in CH4 fluxes in contrast to a late morning peak for NEE CO2, and near noon peak for GPP, and afternoon peak for RCO (Fig. 8), as the latter two follow patterns of PAR and air temperature. NEE CH4 reaches a minimum between 8 and 10 local time (LST), but the minima shifts earlier in 2012, and variability in diurnal pattern is large. While the relative change in hourly NEE was small between 2011 and 2012, there are distinguishable changes in RCO and GPP which were large and compensating. For CH4, a decrease in NEE CH4 from 2011 to 2012 is seen in the average for all hours, but variability in this mean is large. There is, however, a decrease in variability around the mean in 2012 compared to 2011, perhaps reflecting the changes in areal coverage of inundated areas contributing episodic methane emissions, given lower soil moisture as a result of decreased late summer precipitation in 2012.
3.5. Environmental controls on regional methane flux

Variations in CO₂ NEE are typically well described by variations in PAR and temperature at the hourly scale (Desai, 2014), but these correlations were only apparent for CH₄ when averaged at daily to weekly time scales (Table 4). Correlation of NEE CH₄ to NEE CO₂ is significant and negative, but weaker in effect size than for PAR and T. Further, at monthly timescales, the correlation for NEE CH₄ is greatest for Reco and GPP. Interestingly, this relationship with Rco is positive, implying that greater Rco is associated with greater emissions of CH₄ in the region. However, this relationship does not hold at the interannual scale, where increased Rco in 2012 is accompanied by decreased NEE CH₄ (Table 3).

It is likely that the positive correlation of Rco and NEE CH₄ at the shorter time scales primarily reflects the exponential nature of these processes with respect to temperature (Fig. 9). Scatterplots of NEE CH₄ versus temperature and GPP are only weakly correlated at the hourly scale, partly owing to the high uncertainty of NEE CH₄. For daily average NEE CH₄, a linear relationship to GPP and exponential relationship to temperature are more apparent. For the exponential relationship to temperature, daily NEE CH₄ is relatively insensitive for air temperature of 0–15 °C, followed by a large increase in emissions with higher temperature (Fig. 9d). Regionally, it appears at short timescales that CH₄ production and its relationship to temperature dominate any increase in longer-timescale changes in CH₄ oxidation that would occur with the lower soil moisture that co-occurs with high temperature.

3.6. Comparison to ecosystem models

The DLEM model output of daily NEE CH₄ for the region (only available in 2011) and site (2011–2012) reveals similar seasonal patterns to the very tall tower observations, but several discrepancies exist (Fig. 10). First, the regional model, run with an estimate of land-cover based on a continental gridded map, generated CH₄ emissions significantly larger than observed NEE CH₄, likely owing to the larger estimation of wetland area fraction in the regional model (Table 2). It also resulted in CH₄ emissions earlier in the spring and later in the autumn compared to observations. The site level run, using local estimates of wetland extent and local meteorology, had seasonal magnitudes much more in line with the tower. The site model still overestimated CH₄ emissions in the autumn. Further, the site model showed very little interannual variability, while the observations clearly showed a mid to late summer suppression of CH₄ emissions in 2012, likely in response to the lack of precipitation in this time period. Finally, both models tended to have relatively modest sub-weekly variability in CH₄ emissions, while observations showed much larger day-to-day and monthly variation.

4. Discussion

4.1. Uncertainty of regional CH₄ flux

Our analysis confirms that current generation closed path methane analyzers can reliably measure CH₄ fluxes, even in regions of small flux magnitude, as long as high-frequency spectral corrections were applied, confirming recent cross-comparison studies (e.g., Iwata et al., 2014). WPL water vapor dilution corrections were more important for CH₄ than CO₂ given the two orders of magnitude smaller concentration of CH₄ than CO₂ in air. Still, even with long tube lengths, CH₄ fluxes could be measured reasonably to ~20% accuracy at the hourly scale, similar to results shown in recently published papers on methane eddy covariance (Detto et al., 2011; Smeets et al., 2009). Both large positive and negative short-term CH₄ pulses appear to be real, but could arise from either ecosystem processes or vertical flux transport.

A bigger challenge in quantifying net CH₄ ecosystem exchange appears to be finding an adequate gap-filling strategy, as relationships of CH₄ flux at the hourly scale to meteorological drivers have far greater variability than for CO₂. New approaches using artificial neural networks have shown promise (Dengel et al., 2013; Hatala...
over hotspots, published by CH for Our 2012 study compared dynamic 785 wetland regions. CH showed that 4 seasonal variations and magnitudes of chamber fluxes agree well to stand-level eddy covariance observations of NEE CH4 (Yu et al., 2013), upsampling these plot and stand level observations to the region is not straightforward, as high spatial heterogeneity complicates sampling strategies. Further, since production, consumption, and oxidation responses of CH4 to climate are non-linear, extrapolating flux sensitivity from spatial variations across sites does not necessarily lead to the same conclusions about CH4 drivers as temporal variation within sites (Sabrekov et al., 2014).

Finally, estimates of scaled fluxes are highly sensitive to estimates of wetland and forest extent in the case of chambers and for temporal variation of these within the flux footprint for towers. Our chamber estimates argue that the small forest CH4 sink overwhelms wetland CH4 emission mainly because forests have a much larger spatial extent. Additionally, drier conditions in 2005–2006 compared to 2011–2012 may have decreased wetland CH4 production. It could also be the case that the higher CH4 estimate from the flux tower suggests that chambers did not adequately sample high sources of wetland CH4 emission or over-estimated the forest CH4 sink. For example, upland–wetland edges could be particularly dynamic sources of CH4 production, but are rarely sampled.

The purpose of our upsampling was not to build a defensible NEE CH4 from chambers, but to estimate how well plot-scale measurements can sample landscape CH4 flux. Our approach was necessarily simplistic due to constraints of sampling design. Other upsampling based on vegetation maps (e.g. Reeburgh et al., 1998) point to the importance of capturing landscape CH4 hotspots, such as wetlands. Within site and across site variation in CH4 exchange among fens and bogs is large (Baldocchi et al., 2012), and attempts to find optimal and efficient sampling designs for upsampling are not at hand.

Our results call into question the reliability of extrapolation of CH4 plot scale flux studies for estimating global natural CH4 emissions, which is urgently needed given that recent studies have suggested, but not conclusively shown, increases in global wetland CH4 emissions in the past decade (Sphani et al., 2011).

4.2. Magnitude of regional CH4 flux

Average annual CH4 efflux was a relatively small 785 ± 75 mg C–CH4 m⁻² yr⁻¹, compared to a mean CO2 sink of −80 C–CO₂ m⁻² yr⁻¹. The two years showed a 30% shift in CH4 flux from one year to the next that was detectable outside the bounds of our uncertainty analysis. Regional CH4 fluxes by eddy covariance also bracketed those observed by chamber fluxes in prior years in wetlands within the tower landscape.

Our results are similar qualitatively to the early CH4 emission work of Shurpali and Verma (1998), which showed modest CH4 emissions and lack of strong short-term coupling between CH4 fluxes and GPP in a Minnesota bog. Overall, our regional observations are about an order of magnitude larger than recently published eddy covariance forest CH4 flux estimates (Shoemaker et al., 2014) and 1–2 orders of magnitude smaller than a range of CH4 eddy flux studies in a variety of wetlands, including deltas (Baldocchi et al., 2012), rice paddies (Hatala et al., 2012), grazing fields (Herbst et al., 2011), boreal fens (Long et al., 2009; Rinne et al., 2007), peatlands (Pelletier et al., 2007), marshes (Chu et al., 2014), and tundra (Sachs et al., 2008; Tagesson et al., 2012; Wille et al., 2008).

Areas of significant CH4 emission do occur in the region. For example, recent eddy covariance estimates of NEE CH4 in a Minnesota fen from 2009–2011 show emissions of 11.8–24.9 g C–CH4 m⁻² yr⁻¹, a value that amounted to 23–39% of the NEE CO₂ sink (Olson et al., 2013). Similarly, Pyper et al. (2013) finds a northern Michigan poor fen with May–Sept emission of 13 g C–CH4 m⁻² yr⁻¹ and Chu et al. (2014) show freshwater marsh emissions of 49.7 g C–CH₄ m⁻² yr⁻¹ and cropland emissions of 2.3 g C–CH₄ m⁻² yr⁻¹ in northern Ohio.

Another independent approach to regional NEE CH4 is the very tall tower modified Bowen ratio technique based on assuming similarity in the flux–gradient relationship in profiles of CO2 and CH4 concentration (Werner et al., 2003). This method, when applied to the tall tower site, showed average emissions of 2.7 g C–CH4 m⁻² yr⁻¹ in 1998, which is more than three times the estimate here (Fig. 4b), and with a longer NEE CH4 emission season (Mar–Oct). However, those results were from 1998, a year that was much warmer (average annual temperature of 7.8 °C) than the 2011–2012 average (5.4 °C). Further, the similarity approach has known biases during periods of weak vertical gradients of CH4 or CO2 and assumption of directly scaling of NEE CH4 with NEE CO₂, whose correlation is weak at the hourly and daily scale in our study (Table 4). The authors concluded that this region emits 40% less CH4 than other regions at the same latitude.

Another regional carbon cycling upsampling study in the nearby Northern Highland Forest, based on the literature, found a range of 1 to 20 g C–CH₄ m⁻² yr⁻¹ for CH4 emission, roughly 1–2% of the estimated net carbon uptake in the region, but nearly 10% of that for wetlands and 10% of that for lake evasion (Buffam et al., 2011). This estimated range of CH4 flux was also found to be similar to the amount of carbon lost from the terrestrial landscape as SOC runoff. While Buffam et al. (2011) noted large uncertainty on the CH4 emission term, our regional observation results are consistent with a value closer to the lower end of the range used.

4.3. Drivers of CH4 regional net exchange

We were able to discern shifts in annual CH4 flux arising from shifts in growing season length, air temperature, and late summer drought. The late summer 2012 drought was primarily a consequence of shifts in precipitation timing (earlier) instead of total precipitation magnitude. The early start of the growing season, which likely increased transpiration demand, along with the lack of rain in late summer of 2012 conceivably suppressed CH4 production from wetlands in the tower footprint, while simultaneously increasing upland forest soil CH4 uptake, though no single driver can adequately explain hourly to daily NEE CH₄.

Our results are generally consistent with the numerous site-level studies that have attempted to correlate CH4 observations to environmental parameters such as water table depth, temperature, vegetation type, CO2 fixation and respiration rates, atmospheric O3, and/or microbe/organic matter quality. A review paper by Jungkunst and Fiedler (2007) noted that most studies point to water table and soil temperature as strong controlling factors, and they further note that latitudinal trends suggest that anaerobic and aerobic decomposition are both important in boreal regions.

While the modified Bowen ratio study of Werner et al. (2003) showed precipitation explained a greater fraction of variance in regional NEE CH4 than temperature in 1997–1998, our results support temperature as the primary driver at the monthly to seasonal timescale and precipitation, which may drive the availability of
substrate suitable for anaerobic decomposition as the most likely explanation for variation at the interannual scale. Enzyme kinetics of CH₄ production, primarily controlled by temperature, seem to drive most of the daily to seasonal scale variability, with an exponential dependence consistent with a recent report by Yvon-Durocher et al. (2014). Other studies have further confirmed the strong role of temperature for short-term CH₄ dynamics (Bloida et al., 2007; Tagesson et al., 2012; Rinne et al., 2007).

Hydrology and long-term moisture status appear to be the key controls for seasonal to annual variability of NEE CH₄, Reco and GPP, consistent with a recent water-table manipulation study by Ballantyne et al. (2013). Thus, long-term changes in water table are expected to have a strong impact on wetland CH₄ and CO₂ emission ratios (Davidson and Janssens, 2006). Results at other sites concur that peatlands and tundra systems are particularly sensitive to water availability within the active layer (e.g., Hendriks et al., 2007; van Huissteden et al., 2005), and peatland drainage or restoration by flooding strongly influences CH₄ production (Merbold et al., 2009; Turetsky et al., 2008; Waddington and Day, 2007). Long-term declines in water table may lead to soil subsidence, community change, and invasion of upland species (Strack and Waddington, 2007; Sulman et al., 2013), significantly altering CH₄ production and oxidation.

Our results do not support net ecosystem photosynthesis (NEE, NPP, or GPP) as the primary controller on CH₄ net flux at the regional scale. The concept of a fixed ratio of GPP, NPP or NEE to CH₄ production or NEE that has been argued based on field measurement synthesis and process-based models (Potter, 1997; Walter and Heimann, 2000; Whiting and Chanton, 1993) is not apparent in the short term. The ratios of NEE CH₄ to NEE CO₂ observed here (~1%) at the annual timescale fall within values measured in short term experiments (~1–3%: King and Reeburgh, 2002; King et al., 2002; Megonigal et al., 1999). Whiting and Chanton (1993) call net ecosystem production (equivalent to NEE CO₂) the “master variable” in controlling NEE CH₄, suggesting that a fixed 3% of NEE CO₂ is emitted as NEE CH₄. Clearly, even if this holds to be the case in general, variation around the value can be large and is timescale-dependent.

King et al. (2002) report on input of new substrate from GPP as a source of CH₄ emission, arguing that increased productivity provides greater labile substrate and increased transport. In contrast, greenhouse studies have shown that CH₄ emissions related to plant type tended to decrease with increasing plant biomass (Kao-Kniffin et al., 2010). While GPP does correlate with NEE CH₄ at our site, much of the correlation appears to be a co-varying effect of temperature on both processes at the seasonal scale. Short-term variations in GPP or NEE CO₂ do not correlate highly with NEE CH₄, as the primary role of production is not to directly promote methanogenesis, but provide substrate, while redox conditions provide conditions favorable for CH₄ production. However, plants can serve as a conduit of CH₄, and thus GPP may be a proxy for plant-mediated transport (King et al., 1998; Matthes et al., 2014). However, these results are difficult to interpret regionally, as the primary GPP signal is coming from forests in the flux footprint. Perhaps higher forest GPP implies greater export of carbon to the watershed, providing greater substrate for methanogenesis, which would require monitoring of aquatic and dissolved carbon.

Our results also showed a relatively high amount of short-term scale variation in NEE CH₄, greater seasonal variation than for CO₂, and an unusual diurnal pattern to CH₄ flux, with minimum fluxes in early to mid-morning. Several studies have argued that atmospheric pressure changes (Sachs et al., 2008) or shear turbulence (Wille et al., 2008) could drive episodic CH₄ emissions, and perhaps a venting effect (for the diurnal cycle) and synoptic pressure changes (for the weekly–monthly variation) are leading to the variation we observed. For example, storage fluxes of CH₄ act in the opposite direction (negative) to turbulent flux (positive) during the day. It is the strong negative storage fluxes associated with atmospheric venting that drive the minima.

Mastepanov et al. (2008) observed CH₄ bursts before soil freezing in a tundra ecosystem. While our results also show a variety of emission spikes in winter and summer, we have yet to find any particularly strong correlation to barometric pressure, changes in atmospheric pressure, friction velocity magnitude (both above and below the filtering threshold), or other measures of processes that could lead to “pumping” of CH₄ from the soil and snow surface. Initial experimental tests involving melting snow and changing suction pressure with a static chamber did not reveal any significant variation in CH₄ fluxes. Fossil fuel combustion could be a source for CH₄, but the timing of the bursts were not consistent with possible generator or traffic sources, which are quite limited in the flux footprint.

Despite the predominance of upland forest in the flux footprint, the site still is a net emitter of CH₄ in both years. Upland plants have not been shown to emit significant quantities of CH₄ in the field (Kirschbaum and Walcroft, 2008). Generally, upland soils promote methanotrophs and thus dry soils tend to consume CH₄ (Ullah and Moore, 2011). This rate is controlled primarily by diffusion processes in the soil (Ridgwell et al., 1999). A recent synthesis of micrometeorological CH₄ emission estimates in forests generally shows net CH₄ sources with an interquartile range of 1.33–5.45 nmol CH₄ m⁻² s⁻¹ (Nicolini et al., 2013). Another review of 120 papers on soil CH₄ consumption found no universal predictive ability of soil consumption by environmental drivers, but showed that coarser soils had the largest CH₄ uptake in temperate forests, with a mean uptake in temperate forests of 428 ± 2360 mg C m⁻² yr⁻¹ (Dutart and Verchot, 2007). This reported uptake is larger than the average observed in our plot-level chamber measurements in upland forests.

Our study site did include a few lakes in the landscape, and recent studies have argued that lakes and rivers may be large sources of CH₄ (Bastviken et al., 2011; Buffam et al., 2011; Grossart et al., 2011; Juutinen et al., 2009). Some evidence from chambers also suggests particularly large CH₄ flux variability at wetland–upland edges (unpublished data). Finally, winter emissions have generally been undersampled in most studies (Merbold et al., 2013), given logistical difficulty in measurement and assumption of small CH₄ fluxes. Our results also support limited CH₄ fluxes during periods of frozen soil and inactive vegetation. However, fluxes outside the growing season (May–Sept) still contributed 17% of the net annual flux, averaged over the two years, and thus cannot be neglected.

4.4. Recommendations for simulations

Demand for quantification of regional CH₄ balances is increasing (Luyssaert et al., 2012), and models are ultimately required to move from diagnosis to prediction. While several wetland and CH₄ models exist (Cao et al., 1996; Melton et al., 2013; Petrescu et al., 2008; Potter, 1997; Sonnentag et al., 2008; Walter et al., 2001; Zhang et al., 2002; Zhuang et al., 2004), many only weakly constrain hydrology, and only a few also include upland CH₄ biogeochemistry. Walter et al. (2001) review the most common approach, based on temperature, net primary production, substrate availability, and water table depth and show the importance of hydrologic drivers for latitudinal variation in CH₄ efflux.

Our analysis of the commonly used DLEM model results revealed a general agreement between model and very tall tower observations on seasonal pattern, but lack of correspondence at shorter or longer timescales. Further, the regional model significantly overestimated CH₄ emissions primarily due to differences in wetland extent in the regional (based on a cut-out of a continental model.
of greenhouse gas fluxes) versus site simulation (based on local meteorology and land cover), a common source of uncertainty for regional to global modeling of NEE CH₄ (Melton et al., 2013). Most models tend to show a strong sensitivity to water table (Petrescu et al., 2007), wetland extent (Ringleval et al., 2010), and vegetation decomposition rate (van Huissteden et al., 2005). Over North America, DLEM shows enhanced CH₄ emissions from increased climate variability, nitrogen deposition, and atmospheric CO₂, with climate variability dominating interannual variability (Tian et al., 2010; Xu et al., 2010). Simple models that rely on a fixed CO₂ uptake to CH₄ emission ratio for a base amount and exponential temperature functions to capture seasonal or short-term variability (Potter et al., 2006) are likely to neglect the importance of variations in water table which can cause a site to shift between CH₄ source and CH₄ sink. Similar to the results here, other models have generally been unsuccessful at capturing short-term variability in CH₄ emissions (Petrescu et al., 2007; Zhang et al., 2012).

Wetland extent and methane emission datasets both lead to wide variation in modeled (Melton et al., 2013) and extrapolated (Petrescu et al., 2010) estimates. Further, scaling methane emissions as a function of GPP or NEE, as some models do, is not universal. While some sites show as much as 20% of CO₂ uptake returned as methane emissions on a per mole basis (Rinne et al., 2007), the regional evaluation here showed only a fraction of a percent.

5. Conclusion

Our results confirmed the suitability of very tall towers for observation of regional CH₄ fluxes. While mixed forest dominates the landscape and the net CO₂ exchange budget, wetlands dominate the CH₄ emission budget. However, uncertainty on our very tall tower flux measurement, owing to random uncertainty, lack of well-established gap filling protocols, and flux footprint variability all need better quantification in future studies to better constrain the components of the regional CH₄ budget.

The net fluxes over two years showed modest CH₄ emissions in the region, representing less than 1% of NEE CO₂ in a productive mixed forest—wetland landscape. While individual fens or bogs can have large emission rates, as seen in some of our chamber flux observations, the region as a whole may be a minor contributor. We found that the landscape-scale CH₄ fluxes positively correlate with temperature at diurnal to monthly timescales, similar to ecosystem respiration. However, from one year to the next, ecosystem respiration and net CH₄ flux responded in opposite directions, reflecting the shifts in aerobic to anaerobic respiration that occur in wetlands with changes in moisture availability, the availability of organic substrates for decomposition, and the presence of living plants (e.g., sedge species) that can facilitate the exchange of gases between soil subenvironments and the atmosphere.

Simple models that scale CH₄ emissions with Rₑₑₑₑ or NEE of CO₂ are thus both spatial- and temporal-scale dependent. Interestingly, our results also showed higher CH₄ fluxes from the tower than simple upscaling based on chambers but lower than flux tower studies in nearby fens, confirming the relatively high spatial variability of CH₄ fluxes in the landscape. These results are contrary to a general assumption that chambers and plot-level studies always overestimate CH₄ emissions due to their typical placement in ecosystems with high CH₄ emission.

The regional flux time series was able to reveal limitations in modeling of short-term and interannual variability in CH₄ emissions by a dynamic ecosystem model. While temperature and moisture appear to be the strongest controls of CH₄ flux in the region, they have a clear timescale dependence. Our results suggest that models built on (1) temperature for short-term methane emission rate, (2) water table or moisture availability for long-term base emissions amount (or interannual variability), and (3) an estimate of wetland extent are most likely to successfully simulate regional methane fluxes. However, similar to other studies, we find models are unable to simulate short-term (sub-daily) variation in CH₄ emissions (Melton et al., 2013). Future work on decomposing the regional fluxes by land cover will further aid in developing appropriate metrics for evaluation of regional-scale simulations of CH₄ cycling.

While wetlands and other natural sources of CH₄ are only 15–30% of the global CH₄ budget, they are the largest source of variability and a major source of uncertainty for atmospheric chemistry, air quality, and climate models (Arness et al., 2010). The vast majority of observational studies of CH₄ emissions are made at the scale of a plot or individual ecosystem. Regional scale datasets, like the one conducted here, can provide estimates of CH₄ flux at a scale relevant to model evaluation.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jagrfomet.2014.10.017.

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