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Methane fluxes from a small boreal lake measured with the eddy covariance method

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Abstract

Fluxes of methane, CH_4, were measured with the eddy covariance (EC) method at a small boreal lake in Sweden. The mean CH_4 flux during the growing season of 2013 was 20.1 nmol m^{-2} s^{-1} and the median flux was 16 nmol m^{-2} s^{-1} (corresponding to 1.7 mmol m^{-2} d^{-1} and 1.4 mmol m^{-2} d^{-1}). Monthly mean values of CH_4 flux measured with the EC method were compared with fluxes measured with floating chambers (FC) and were in average 62% higher over the whole study period. The difference was greatest in April partly because EC, but not FC, accounted for fluxes due to ice melt and a subsequent lake mixing event. A footprint analysis revealed that the EC footprint included primarily the shallow side of the lake with a major inlet. This inlet harbors emergent macrophytes that can mediate high CH_4 fluxes. The difference between measured EC and FC fluxes can hence be explained by different footprint areas, where the EC system "sees" the part of the lake presumably releasing higher amounts of CH_4. EC also provides more frequent measurements than FC and hence more likely captures ebullition events. This study shows that small lakes have CH_4 fluxes that are highly variable in time and space. Based on our findings we suggest to measure CH_4 fluxes from lakes as continuously as possible and to aim for covering as much of the lakes surface as possible, independently of the selected measuring technique.

Methane, CH_4, is an important greenhouse gas with approximately 25 times higher global warming potential than carbon dioxide, CO_2, by mass and considering the effect of a single pulse emission over a 100 year period (Forster et al. 2007). Recent studies by Bastviken et al. (2011) and Ciais et al. 2013 highlighted the huge CH_4 emissions from lakes, estimated to correspond to 25% of the CO_2 equivalents sequestered by the terrestrial carbon sink reported by IPCC. However, these studies also recognized the large uncertainty of the measurements and the need for development of more representative measurement approaches.

CH_4 is mainly produced in the anoxic lake sediments and higher temperatures will normally result in higher CH_4 production (e.g., Duc et al. 2010; Marotta et al. 2014). From the sediment, CH_4 can be transported to the atmosphere along different pathways; diffusion, storage transport, ebullition (bubble flux), and transport through plants (e.g., Bastviken et al. 2004). The diffusive flux over the water–air interface is driven by the concentration difference between the water and the air and controlled by the transfer velocity. The transfer velocity describes the efficiency of the gas transfer and is controlled by, e.g., wind speed, and waterside convection (e.g., Rutgersson and Smedman 2010; Podgrajsek et al. 2014b).

The diffusive flux is substantially reduced by consumption of CH_4 in oxic sediments or waters by methane oxidizing bacteria (Bastviken 2009). Storage transport is a special case of diffusive transport: If a lake is stratified with anoxic bottom water, a large amount of CH_4 can be stored in the anoxic water layers (Michmerhuizen et al. 1996; Riera et al. 1999; Bastviken et al. 2004). With mixing of the lake, this CH_4 rich water is transported from the bottom to the surface, which will result in a large water–air gradient and a large diffusive flux. Other emission pathways, such as ebullition and transport through plants are direct and rapid, leaving less time for CH_4 oxidation before emission. Ebullition is controlled by CH_4 production rates, air pressure changes and bottom shear stress (Joyce and Jewell 2003), while transport

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through plants is influenced by the CH_4 production rates, the plant density and types, and the gas transport through the plants and subsequent exchange of the plant leaves (Kankaala et al. 2004). Juutinen et al. (2003) showed that for three lakes in Finland, the shallow littoral zone, with high CH_4 flux via emergent macrophytes and ebullition, accounted for 66–77% of the total CH_4 release. However, comparisons of fluxes by all above mentioned flux pathways are rare and may vary between lakes.

To estimate the total flux from a lake, all different pathways need to be estimated or measured. One common way to measure CH_4 fluxes from lakes is to use floating chambers (FCs) where the gas exchange across an area is assessed by monitoring the change in gas content inside the chamber headspace (the area of the chamber is generally < 1 m^2) (Bastviken et al. 2004; Bergström et al. 2007). This technique can be used with inexpensive field equipment, is conceptually simple, and has provided valuable information historically. However, extensive manual measurements in time and space are very labor demanding and often not practical. Therefore, other techniques may be beneficial for long-term assessment of fluxes across larger lake areas. The eddy covariance (EC) method (e.g., Aubinet et al. 2012) represents one such alternative technique that has become a standard approach for measuring greenhouse gas exchange in terrestrial environments (e.g., Baldocchi 2003). Fluxes measured with the EC method represent the fluxes of an upwind area called the footprint, which will vary in size and location depending on the measurement height, wind, atmospheric stability, and surface characteristics (e.g., Vesala et al. 2008). Depending on the height of the sensor and the weather and surface conditions, the footprint area can cover a few tens m^2 to several km^2.

The EC technique has recently been adopted for application on lakes. Most studies focused on CO_2 (e.g., Huotari et al. 2011; Podgrajsek et al. 2015) but in a few cases CH_4 fluxes have been studied (e.g., Eugster et al. 2011; Schubert et al. 2012; Podgrajsek et al. 2014a). In Podgrajsek et al. (2014a), it was shown that the FC and EC methods yield fluxes in the same magnitudes. However, there are still important differences between the two methods that need to be considered.

For EC measurements, homogenous surfaces are ideal. Thus, EC measurements located in the center of large lakes measuring open waters fluxes, are beneficial target areas for the EC technique. However, as CH_4 fluxes are higher from shallow, macrophyte covered areas (Bastviken et al. 2004; Bergström et al. 2007), and small lakes with such areas are prevalent globally (Verpoorter et al. 2014), the applicability of the EC technique for CH_4 flux measurements in small lake ecosystems and in near-shore areas is of high interest. Here, we report one of the first attempts to use EC CH_4 flux measurements under such conditions. We show, by footprint estimations and supplementary FC measurements, that the EC method may have potential to capture fluxes in small lake ecosystems. However, we also identify a number of issues that should be considered in similar future studies.

Methods

Site and instrumentation

Erssjön is a small lake located in southwest Sweden (58°22′N, 12°09′E, Fig. 1) in the Skogaryd research catchment (Klemedtsson et al. 2010). The lake has a surface area of 0.07 km^2 and a mean depth of 1.3 m. At the northeast and southwest parts of the lake, the water is approximately 1 m deep, and the maximum depth, 4.4 m, is located approximately in the center of the lake. Both the west and the east shores of the lake are surrounded by coniferous forest, to the northeast of the lake there is agriculture land (Fig. 2). The 6 m high meteorological tower positioned at the northeast shore of the lake (see tower position in Figs. 1, 2) had sensors mounted at three levels. Levels two and three, 4.7 and 6 m above ground, were equipped with propeller anemometers for wind speed and direction (Young, MI, U.S.A.) and radiation shielded and ventilated thermocouples for measurements of air temperature. At the first level, 2.4 m above ground, the EC instrumentation was mounted: a LI-7700 open gas analyzer for CH_4 measurements (LI-COR Inc., Lincoln, NE, U.S.A.) and a sonic anemometer (WindMaster, Gill Instruments, Lymington, UK) for measurements of the three dimensional wind components and virtual (sonic) temperature. The temperature in the water column was monitored every 2 h using temperature sensors (U22 Water Temp Pro v2 logger, Onset HOBO, Cap Cod, MA, U.S.A.). The sensors were deployed at every half meter depth at the deepest spot (i.e., approximately the center of the lake), to resolve the temporal development of water column mixing and stratification in Erssjön over the entire study period.

Eddy covariance measurements

The EC data, measured at 10 Hz, was both detrended and despiked over 30 min periods. The three wind vectors from the sonic measurements were rotated into the mean wind direction and tilt corrected, setting the mean vertical wind to zero. Time lag due to sensor separation between the sonic and LI-7000 was typically between 0.1 s and 0.2 s and was calculated according to Sahlée et al. (2008). The CH_4 density measurements from LI-7700 were corrected for temperature and humidity changes according to Webb et al. (1980) and additional corrections due to spectroscopic effects according to McDermitt et al. (2010). See Sahlée et al. (2014) for more detailed information on the performance of the EC instrumentation.

Data coverage

The measurements presented in this article are from the open water period in 2013 (April–December). The times

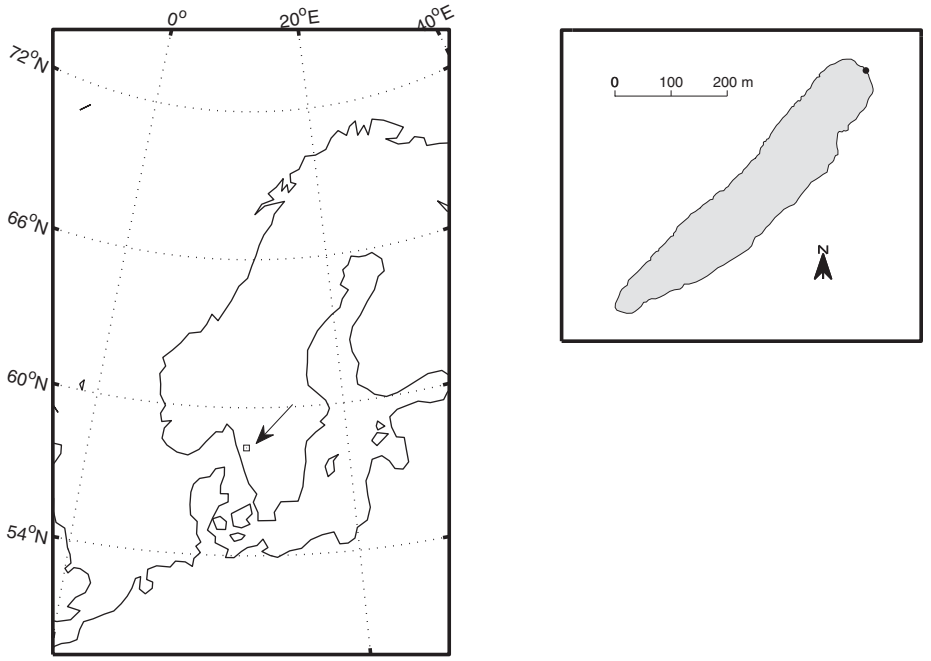

Fig. 1. Left; Map showing Scandinavia, where the arrow points to the position of Erssjön. Right; Lake Erssjön, black dot denotes the position of the EC tower.

Fig. 2. Photo of Erssjön taken from an airplane where the red circle highlights the position of the EC tower. Photo taken by: Jutta Holst.

presented are expressed in central European time. During part of the summer, the EC flux data are missing due to malfunction of the sonic. For quality control of the flux data and to select fluxes coming from only the lake, the data was further sorted by the following criteria: wind directions coming from the lake (between 200° and 260°), wind speed higher than 1 m s^{-1}, received signal strength indicator, from the LI-7700, higher than 10% and skewness and kurtosis in the range of −2 to 2 and 1 to 8, respectively (Vickers and Mahrt 1997). After this postprocessing, 1500 half-hourly CH_4 flux data points remained for further analysis.

Footprint

For interpretation of the EC data, the footprint is a crucial concept (see Schmid 2002; Vesala et al. 2008; Leclerc and Foken 2014). Under the assumption of stationarity, the footprint represents the area from where the measured fluxes originate. To estimate the footprint area for the study period, we used the footprint parameterization of Kljun et al. (2015), which is an updated version of Kljun et al. (2004), allowing derivation of two-dimensional footprint estimates. This footprint parameterization is based on the Lagrangian stochastic particle dispersion footprint model of Kljun et al. (2002), which is valid for a broad range of boundary layer conditions. The footprint parameterization was run with inputs derived from the EC measurements (Obukhov length, friction velocity, standard deviation of lateral velocity fluctuations). The boundary layer heights were estimated according to Kljun et al. (2015). For each half-hourly data point of the EC data, a footprint was calculated and then merged to one cumulated footprint for the whole study period.

Floating chambers

Repeated series of FC measurements with multiple FC for each occasion were made for comparison with EC data. The FCs consisted of inverted plastic buckets similar to the ones used by Cole et al. (2010) and Gålfalk et al. (2013). The FCs were covered with aluminum tape to reflect sunlight and minimize internal heating by direct sunlight. Styrofoam floats at the edges of the FCs kept them afloat; the FC walls reached approximately 3 cm into the water. The FCs covered an area of 0.08 m^2 and their volume was 7.5 L. Gas samples from the FC headspace were collected using an attached 25 cm long transparent PVC (Polyvinyl chloride) tubing

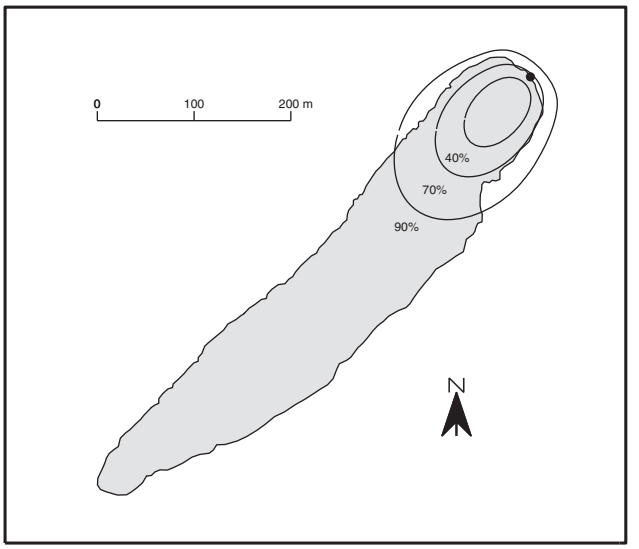

Fig. 3. Map of Erssjön, black dot denotes the position of the EC tower. Solid lines represent mean footprint areas cumulated over the study period, showing where 90%, 70%, and 40% of the fluxes in the study period originate from.

(outer diameter 5 mm and inner diameter 3 mm) with a three-way luer-lock stopcock (Becton-Dickinson, U.S.A.). Each FC was attached to a float by a 1 m line and this float was anchored to the sediments using triplicate 50 mL centrifuge tubes filled with sand and water. The 1 m separation between the chamber and the float made sure influence from possible bubbles released when the plastic weights hit the sediment was avoided, and it enabled the light weight FC to readily follow up–down wave movements. The chambers were deployed biweekly from April to November for 24 h each time to capture diel variability in CH_4 emissions. The total fluxes in the chambers (including both diffusive fluxes and ebullition) were calculated as suggested by Bastviken et al. (2004). In this study, we include measurements from eight FCs distributed in the northeastern part of the lake from within the EC footprint.

Results

The eddy covariance footprint

The cumulative footprint area indicates that most of the fluxes, measured with the EC technique, represent fluxes originating from the shallow northeast part of the lake (Fig. 3). However, there is some influence of the surrounding land on the measured fluxes, i.e., some parts of the flux footprint spans over the land area (Fig. 3). CH_4 fluxes from forest soils are often negligible or negative (e.g., Klemedtsson and Klemedtsson 1997; Wang et al. 2013). Still, to make sure that the soils around Erssjön do not contribute significantly to the measured EC flux, measurements of CH_4 fluxes in the forest surrounding Erssjön were made in the summer of 2014 using distributed soil FC measurements around the lake. The CH_4 fluxes in the forest were always small and spanned from approximately -0.3 nmol m^{-2} s^{-1} to -0.1 nmol m^{-2} s^{-1}. This means that these fluxes are typically in the order of 100 times smaller than fluxes from the lake. We can, thus, assume that even if a minor part of the footprint covered the surrounding forested area, the majority of the fluxes originate from the lake. The lake shores, with dense stands of emergent macrophytes (primarily *Carex rostrata* and *Phragmites australis*), were partially located within the footprint, meaning that fluxes from this vegetation were most likely contributing to the measured EC fluxes.

Methane fluxes measured with the eddy covariance method

The mean CH_4 fluxes for the entire data set (April–December) was 20.1 nmol m^{-2} s^{-1} with maximum values of around 130 nmol m^{-2} s^{-1} and a median flux of 16 nmol m^{-2} s^{-1}.

From 06 April to 01 June (Fig. 4a), most of the daily mean EC fluxes ranged between 8 nmol m^{-2} s^{-1} and 20 nmol m^{-2} s^{-1} with a few values exceeding this range (up to 45 nmol m^{-2} s^{-1}). The mean value for this period was 14.5 nmol m^{-2} s^{-1}. The fluxes after the data gap in summer (Fig. 4b) had a mean value of 23 nmol m^{-2} s^{-1} and maximum daily mean values in August and September, reaching up to approximately 55 nmol m^{-2} s^{-1}, while fluxes declined to low values in December. The temporal pattern of the fluxes seems to follow a marked seasonality with higher fluxes at or slightly after the peak of the summer season.

As mentioned above, CH_4 is produced in the sediment and is transported to the atmosphere by mainly three pathways. These pathways and production of CH_4 are controlled by different environmental variables. For example, wind speed (u), air temperature (T), incoming solar radiation (RIS; affecting water temperature and macrophyte activity), and atmospheric surface pressure (p), can affect production of CH_4 or the fluxes of CH_4. CH_4 fluxes as a function of these four variables measured at the site are shown in Fig. 5. The linear fit to the data shows no strong effects of wind speed and pressure increases (Fig. 5a,b). The low R^2 (> 0.03) in these cases makes the explanatory power negligible in spite of significant relationships due to large amounts of data. Conversely, there seems to be a weak increase in CH_4 fluxes with increasing temperatures (Fig. 5c). For incoming solar radiation (Fig. 5d) the linear fit has again a very low explanatory power (low R^2) and hence CH_4 fluxes do not seem to depend on the amount of incoming solar radiation.

Methane fluxes measured with the floating chambers in comparison with fluxes measured by eddy covariance

The mean FC flux for 2013 was 14 nmol m^{-2} s^{-1} with maximum monthly mean values of approximately 35.6 nmol m^{-2} s^{-1}. The EC method measured higher fluxes than the FC method during all months (Fig. 6). The largest difference, 168%, was found in April and the mean difference for all months was 74% (Table 1). The flux variability for FC and

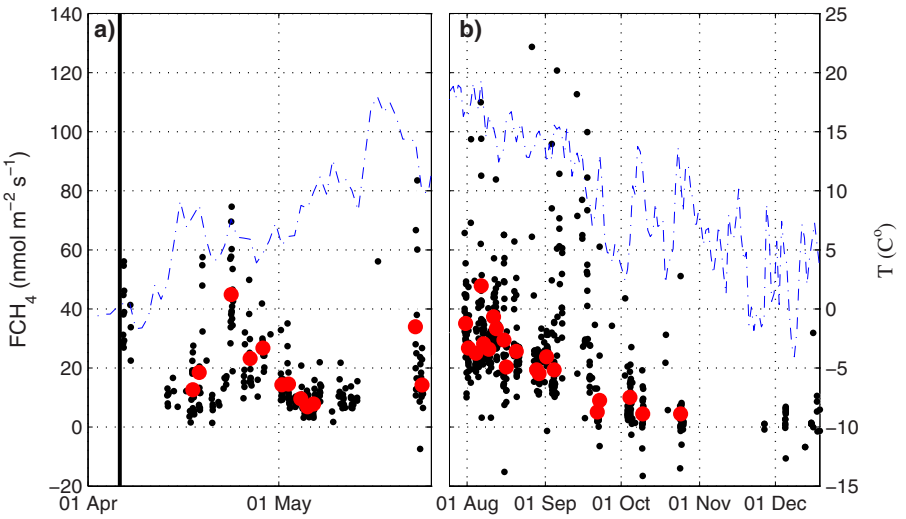

Fig. 4. Half-hourly mean values of CH_4 flux ($n = 1500$) in grey and daily mean values CH_4 flux ($n = 56$) in red. Blue dash dotted line represents daily mean air temperatures (right *y*-axis). Daily mean values are only shown for days with more than 10 half-hourly values available. The solid black vertical line denotes the date of ice melt (06 April).

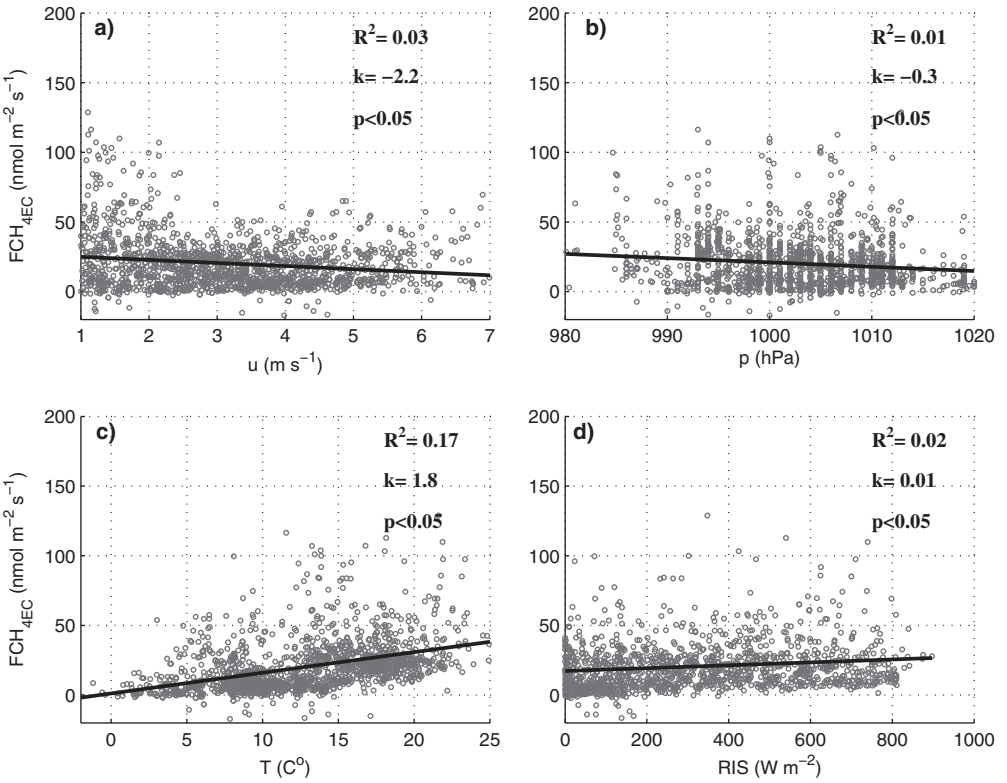

Fig. 5. Half-hourly mean values of CH_4 flux plotted as function oft (a) wind speed, *u*, (b) atmospheric surface pressure, *p*, (c) air temperature, *T*, and (d) solar incoming radiation, RIS. The black lines represent linear fits to the data. The values in the upper right corners show the R^2, the slope of the fit, *k*, and the *p* value of the slope of the fit.

EC measurements were of similar magnitudes and the monthly mean FC fluxes confirm the seasonal flux pattern indicated by the EC measurements (Table 1; Fig. 6).

A closer look at two high flux events in April

Two days; 06 April and 23 April, had substantially higher daily mean fluxes than the rest of the days in April (Fig. 4a).

Photographs of the ice conditions on the lake on 05–07 April (Fig. 7), show that on 06 April, a large part of the lake became free from ice. Thus, the flux peak of approximately 20 nmol m^{-2} s^{-1} (daily average) on 06 April seems associated with ice out.

Between 19th April and 20th April, the wind speed dropped from 7 m s^{-1} to 4 m s^{-1} (Fig. 8b) and the water became thermally stratified for three days (Fig. 8c). The temperature difference between bottom water (3.5 m below the surface) and surface water (1 m below the surface) was approximately 2.5°C during stratification (Fig. 8c). While the water was stratified, CH_4 fluxes were low (Fig. 8a). On 23 April, wind speed increased and the water mixed all the way to the bottom, i.e., the lake became well mixed (Fig. 8c). The mixing of the lake coincides with a peak flux of CH_4 observed on 23 April (Fig. 8a).

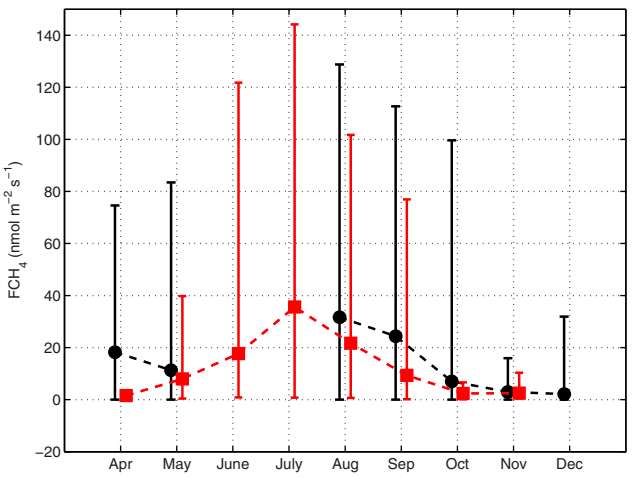

Fig. 6. Monthly mean values of CH_4 fluxes. Black dots represent the EC measurements, red squares the FC measurements. The error bars mark the maximum and minimum EC and FC measurements during each month. For the EC values only months with at least 30 half-hourly mean values are included.

Discussion

Both the study period, mean and the median CH_4 fluxes observed at Erssjön were approximately twofold higher than the average total open water fluxes from lakes at similar latitudes as Erssjön (Bastviken et al. 2011). The fluxes in this study are also higher than the median flux value from EC measurements at a much larger Swedish lake (Podgrajsek et al. 2014b). The higher fluxes at Erssjön are likely due to the fact that the measured fluxes originate from the shallow littoral zone, a strong source region for atmospheric CH_4 (Bastviken et al. 2004; Bergström et al. 2007). Additional FC measurements at Erssjön also showed that near-shore fluxes were significantly higher than fluxes from central lake parts (Natchimuthu et al. 2015).

The FC fluxes here were more similar to the average boreal total open water value presented in Bastviken et al. (2011). It is not surprising that FC fluxes over open water, in our case measuring only during 24 h periods every other week, are lower than the fluxes measured with EC. The EC fluxes will, in contrast to the FC fluxes, more likely capture ebullition events, because of their greater temporal coverage, and in this case also included fluxes from emergent macrophytes, known to produce larger fluxes than observed over open water. Nevertheless, the fact that these two independent methods yield rather similar fluxes and similar monthly pattern (Fig. 6), in spite of the large differences in temporal and spatial coverage, indicates that they both produce realistic and reliable results within the general constraints of each respective method.

CH_4 can accumulate under ice both as dissolved CH_4 in the water (e.g., Michmerhuizen et al. 1996) as well as CH_4 rich air bubbles trapped under the ice and in the ice (e.g.,

Table 1. Monthly mean values of the EC and FC fluxes and the difference between monthly mean values measured with the two methods.

	EC (nmol m^{-2} s^{-1})	Number of half-hourly EC data	FC (nmol m^{-2} s^{-1})	Number of FC deployments (deployment time 24 h)	Percentage difference between EC and FC mean values (%)
Apr	18.3	241	1.6	16	168
May	11.3	285	8	14	34
Jun			17.7	16	
Jul			35.6	16	
Aug	31.6	488	21.7	16	37
Sep	24.4	237	9.3	22	90
Oct	7	144	2.4	6	98
Nov	3	33	2.5	8	17
Dec	2.2	69			
Mean	20.1		12.4		62

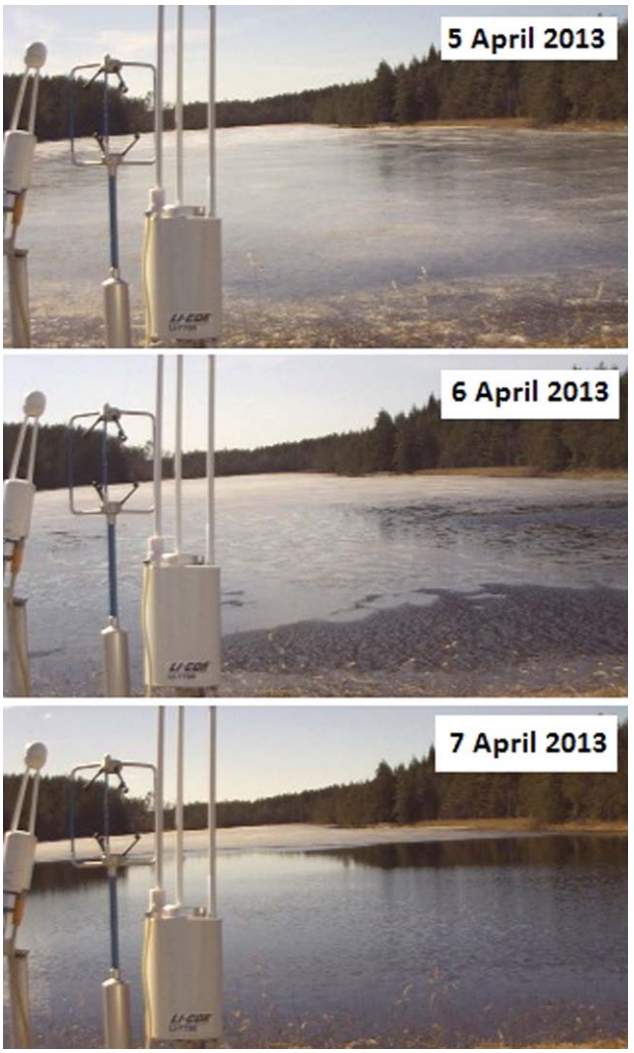

Fig. 7. Photographs taken at 12:00 on 05–07 April showing the EC instrumentation and the lake surface during ice melt.

Wik et al. 2011). Several previous studies have highlighted ice melting as one of the most important periods for gas evasion from lakes (Phelps et al. 1998; Huttunen et al. 2003; Boereboom et al. 2012). According to Karlsson et al. (2013), CH_4 fluxes during ice melt can account for 3–84% of the annual CH_4 fluxes. However, under-ice measurements of dissolved CH_4 during winter 2012/2013 at Erssjön did not show any accumulation of CH_4, and O_2 was present throughout the water column. But high CH_4 fluxes after ice melt, without any significant accumulation of CH_4 under ice, have been observed previously (Miettinen et al. 2015). One possible explanation is that CH_4 can be transported from the catchment and the frozen littoral zone if there is simultaneous soil thaw and increase the hydraulic connectivity around the lake. In accordance to Miettinen et al. (2015), we hypothesize that the high CH_4 flux peak on 06 April is a result of horizontal transport of CH_4 from the catchment and the littoral zone. It should be noted that this flux peak was relatively small in comparison and was of only a minor contribution to the total CH_4 release during the total open water period.

The flux pattern between 06 April and 23 April (Fig. 8) can be explained as follows: During stratified periods, dissolved CH_4 released from hypolimnetic sediments will get trapped below the stratification and reduce the amount of CH_4 emitted to the atmosphere (Bastviken et al. 2008). During periods of whole lake mixing on the other hand (23 April), CH_4 released from the sediments, across the whole lake, will be transported rapidly through the water column and emitted to the atmosphere (storage transport). The mixing of the whole water column may also trigger ebullition events (Joyce and Jewell 2003), which together with the storage transport can result in periods with high CH_4 fluxes.

CH_4 fluxes show an increase from middle of April until end of April before the fluxes start to decrease again (Fig. 4). Because the air temperature increases throughout this period, i.e., also when fluxes decreased, this flux pattern cannot be explained by temperature changes. We instead argue that this flux pattern is explained by the discharge from the catchment, as the discharge increases towards the end of April before it starts to decrease.

For CO_2, which is emitted across the lake surface largely by diffusive fluxes, it is well recognized that the diffusive fluxes will increase with increased wind speed (e.g., Cole and Caraco 1998; Wanninkhof et al. 2009). Yet, for the CH_4 fluxes measured in this study there is no clear relation to the wind speed pattern and the tendency is the opposite (with a very low R^2); lower wind speeds corresponded to higher fluxes while high wind speeds corresponded to lower fluxes (Fig. 5a). As discussed in the introduction, the EC method will not only measure the diffusive fluxes of CH_4 but also ebullition and fluxes through plants, with the two latter flux pathways often dominating in small lakes. The lack of a strong correlation between total CH_4 fluxes and wind speed could be an indication that ebullition and flux through plants dominates over the wind driven diffusive fluxes at this site.

To avoid measuring fluxes which do not originate from the lake surface, the EC instrumentations were mounted at only 2.4 m above ground. This low measuring height resulted in footprints covering only approximately one third of the lake surface (Fig. 3). It is important to acknowledge that CH_4 fluxes from lakes can be highly heterogeneous from seemingly homogenous water surfaces, ranging from high fluxes in the emergent plant belt and shallow waters with frequent ebullition, to lower fluxes in central parts of the lakes where fluxes are dominated by diffusion. As the actual half-hourly footprints vary in time, the measured fluxes may include more or less plant flux, shallow water ebullition, or open water diffusive fluxes, which could explain some of the variability observed in the EC measurements.

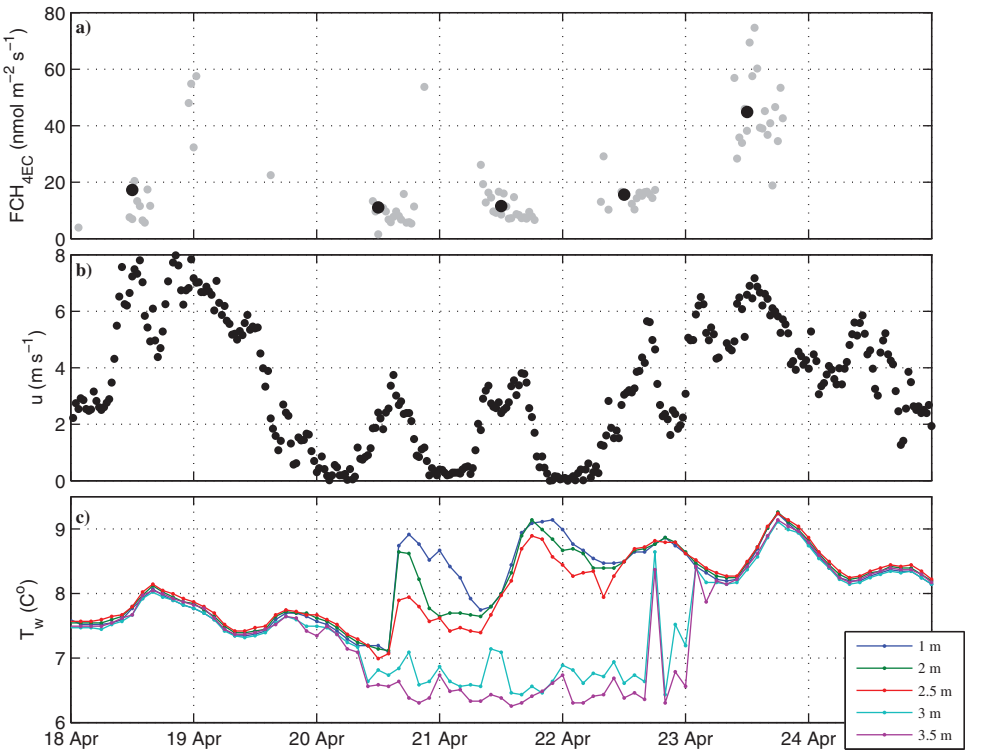

Fig. 8. Time series of the period 18th–25th April (a) Half-hourly mean values of CH_4 flux in grey and daily mean values CH_4 flux in black, (b) wind speed, *u*, and (c) water temperature, T_w, at different levels, denoted by the colors in the legend.

Also, the estimations of the total CH_4 flux from Erssjön, measured with the EC method, could be biased because only one sector of the lake is represented in the measurements.

As Erssjön is a small lake, the EC method will also lack measurements from many days and especially nights, mainly due to wind directions from other sectors than from the lake or too low turbulence. Missing out on the night-time measurements might severely bias long term flux estimates (Podgrajsek et al. 2014a). This is one of the main challenges associated with EC measurements in small and often wind sheltered lake systems. Still, this study shows that EC measurements from small lakes can reveal interesting flux patterns and their possible controlling parameters. The possibility of quantifying fluxes during time periods when the lake is inaccessible by boat, such as during ice out is valuable, as is the capability to identify how changes in stratification or mixing of the water column affects the fluxes (Fig. 8).

Conclusions

We measured the CH_4 fluxes from a small lake in western Sweden during 2013 using both the EC and FC methods. This study supports the idea that small lakes have CH_4 fluxes that are highly variable in time and space and may significantly contribute to the global CH_4 budget. Additionally, the EC fluxes were generally found to be larger compared to the FC fluxes, largest difference in April due to the ability of the EC method to capture the high flux events during lake mixing after ice melt. Based on the findings of this study, we suggest to measure CH_4 fluxes from lakes as continuously as possible and to aim for covering as much of the lake's surface as possible, independently of the selected measuring technique.

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