

Abstract

Greenhouse gas budgets quantified via land-surface eddy covariance (EC) flux sites differ significantly from those obtained via inverse modeling. A possible reason for the discrepancy between methods may be our gap in quantitative knowledge of methane (CH₄) fluxes. In this study we carried out EC flux measurements during two intensive campaigns in summer 2008 to quantify methane flux from a hydropower reservoir and link its temporal variability to environmental driving forces: water temperature and pressure changes (atmospheric and due to changes in lake level). Methane fluxes were extremely high and highly variable, but consistently showed gas efflux from the lake when the wind was approaching the EC sensors across the open water, as confirmed by floating chamber flux measurements. The average flux was $3.76 \pm 0.39 \mu\text{g C m}^{-2} \text{s}^{-1}$ (mean \pm SE) with a median of $1.42 \mu\text{g C m}^{-2} \text{s}^{-1}$, which is quite high even compared to tropical reservoirs. Fluxes increased exponentially with increasing temperatures, but were decreasing exponentially with increasing atmospheric and/or lake level pressure. A multiple regression using lake surface temperatures (0.1 m depth), temperature at depth (10 m deep in front of the dam), atmospheric pressure, and lake level was able to explain 35.4 % of the overall variance. This best fit included each variable averaged over a 9-h moving window, plus the respective short-term residuals thereof. We estimate that an annual average of 3 % of the particulate organic matter (POM) input via the river is sufficient to sustain these large CH₄ fluxes. To compensate the global warming potential associated with the CH₄ effluxes from this hydropower reservoir a 1.3 to 3.7 times larger terrestrial area with net carbon dioxide uptake is needed, which indicates the potential relevance of temperate reservoirs and lakes in local and regional greenhouse gas budgets.

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



1 Introduction

The global network of eddy covariance (EC) flux sites (Fluxnet; Baldocchi et al., 2001; Baldocchi, 2008) provides an excellent overview of the high diversity in terrestrial ecosystem functioning and how they influence the global greenhouse gas budget. Interestingly, the overall budget differs among estimates obtained via integration of land-surface EC flux sites and inverse modeling that use the atmospheric signal to deduce the carbon (C) uptake fluxes at the surface (Janssens et al., 2003; Schulze et al., 2009). Most of the focus on C fluxes in the Fluxnet community has been on carbon dioxide (CO₂), but a gap in knowledge of methane (CH₄) fluxes exists, which may be a reason for the discrepancy between methods in estimating global-scale greenhouse gas budgets. As ecosystem-scale CH₄ flux measurements are now becoming widely feasible with suitable fast-response sensors available on the market (e.g., Eugster and Plüss, 2010; McDermitt et al., 2010), it becomes realistic to quantify CH₄ fluxes for a wide range of ecosystems that have not been considered in the larger-scale European greenhouse gas budgets presented by Schulze et al. (2009), who focused on dominant land-use types, such as forests, croplands, and grasslands. Not included were lakes and reservoirs (Cole et al., 2007; Tranvik et al., 2009), which only cover a small fraction of the land surface area in the temperate zone of Europe, but could potentially be substantial local sources of methane (DelSontro et al., 2010).

Freshwater sediments are landscape-scale hot spots of methanogenesis, since they typically are anoxic below a few mm or cm depth, exhibit low concentrations of other electron acceptors used for anaerobic respiration (e.g. sulfate), and receive a continuous supply of particulate organic matter (POM) from both internal primary production and terrestrial sources (Bastviken, 2009). The question of linkage between organic carbon leaching from upland sites (Kindler et al., 2011) and the C inputs to riverine systems have received increasing attention in recent years as it has been shown that rivers and inland waters are not merely passive C conduits between the terrestrial biosphere and the worlds oceans (Siemens, 2003), but instead locations of active C

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



transformation and storage (Cole et al., 2007). Ultimately, inland waters, which cover just over 3 % of the continents, bury ≈ 50 % more C than the oceans and emit ≈ 1.4 Pg of C in gaseous form to the atmosphere per year (Tranvik et al., 2009). Methane, a much more potent greenhouse gas than CO_2 , is produced in the final stages of organic C degradation, and is particularly extensive in the anoxic sediments of lakes and reservoirs; thus, globally 0.1 Pg of CH_4 is released per year to the atmosphere, offsetting the terrestrial C sink by at least 25 % (Bastviken et al., 2011).

Reservoirs are of particular concern regarding CH_4 emissions as they tend towards higher trophic statuses and even more anaerobic conditions (St. Louis et al., 2000), especially the tropical ones, which emit most of their CH_4 via degassing of CH_4 -rich and oxygen-poor hypolimnetic waters at the turbines or further downstream after turbine passage (e.g., Guérin et al., 2006; Kemenes et al., 2007). Of the typical CH_4 emission pathways, most attention has focused on surface diffusion and much less on advection through plants or ebullition (bubbling), despite the latter emitting significantly more CH_4 (Bastviken et al., 2011). Ebullition remains underestimated, primarily due to its stochastic nature (Bastviken et al., 2011), which is a result of several environmental factors influencing its spatial and temporal variability.

While physical factors such as bottom shear stress (e.g., Joyce and Jewell, 2003) or pressure changes (e.g., Mattson and Lichens, 1990) may modify the timing of ebullition, it is factors like organic C input levels and temperature that most likely maintain the probability of ebullition occurring as they directly impact rates of methanogenesis (Bastviken et al., 2004). When CH_4 production rates exceed vertical diffusion through sediments, the consequent super saturation leads to bubble formation and growth, so long as the ambient CH_4 production maintains the gradient at the bubble perimeter (Algar and Boudreau, 2010). It has recently been shown that the highest ebullition rates in a small temperate reservoir occurred during the warm summer months (DelSontro et al., 2010), but in general not many small reservoirs, which far exceed the number of large ones (Downing et al., 2006), have been surveyed for ebullition. While global inland waters emit an order of magnitude less CH_4 than CO_2 , the greater global warming

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



potential of CH₄, along with the increasing number of manmade impoundments, make CH₄ emissions an important component of the global C cycle (Tranvik et al., 2009).

Hence, the aim of this article is to (1) critically validate earlier estimates of extreme CH₄ fluxes from a run-of-river hydropower reservoir on the Aare River in Switzerland with state-of-the-art EC flux measurements, (2) explore the importance of short-term variability of environmental conditions driving these CH₄ fluxes, and (3) relate the CH₄ fluxes from the reservoir to the net CO₂ uptake of the surrounding landscape to put this locally strong CH₄ source in the wider context of the regional-scale C budget.

We report the first direct EC flux measurements of CH₄ from a freshwater ecosystem, specifically a hydropower reservoir, from which the CH₄ fluxes from were large enough to be a potentially non-negligible C source. The processes discussed here are also quantitatively relevant for other similar systems in the temperate zone of Europe, which also receive substantial POM inputs from upstream (and hence upland) areas.

2 Material and methods

2.1 Site description

Lake Wohlen dam was completed in 1920, consequently creating the $\approx 2.5 \text{ km}^2$ reservoir that holds $\approx 22 \times 10^6 \text{ m}^3$ of water with a maximum depth of 18 m near the dam (mean depth $\approx 9 \text{ m}$). The Aare River, originating in the Central Alps and passing through several large lakes, directly feeds Lake Wohlen with an average flow of $122 \text{ m}^3 \text{ s}^{-1}$ (approximate range 4 to $400 \text{ m}^3 \text{ s}^{-1}$), which is equal to the discharge of this run-of-river reservoir, and amounts to a residence time no longer than a week and a fully oxic water column year round (Albrecht et al., 1998). Seasonal water temperature changes (from $\approx 5^\circ \text{C}$ in winter up to $\approx 20^\circ \text{C}$ in summer) have been shown to influence the variability in CH₄ emissions from the reservoir (DelSontro et al., 2010). Total organic carbon concentrations are typically $\approx 2.4 \text{ mg l}^{-1}$ at the inflow with $\approx 1.9 \text{ mg l}^{-1}$ of that existing as DOC. Lake Wohlen is characterized as meso- to eutrophic and receives relatively

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



large amounts of organic matter and moderately high phosphorus and nitrogen inputs (median concentrations of monthly measurements since 2001 were $17 \mu\text{g PI}^{-1}$ and 1.16 mg NI^{-1} , respectively; unpublished data from Water Laboratory of the Canton of Bern, Switzerland). Monitoring data on POM concentrations in the Aare river in Bern, right upstream Lake Wohlén, are available from 1994–1996 (Naduf, 2000), but not for 2008 during our measurements.

Measurements were carried out at the shore of Lake Wohlén at Jaggisbachau ($46^{\circ}57'52.17'' \text{ N}$, $7^{\circ}18'49.03'' \text{ E}$, 481 m a.s.l.), roughly 10 km northwest of Bern, Switzerland. The instruments were placed directly on the lake shore (cf. Eugster et al., 2003) in such a way that the flux footprint area during the prevailing west winds was entirely on the lake. Towards the prevailing wind direction (west) the fetch was still 1.2 km. At the sampling site clear evidence of ebullition was seen at the lake surface.

2.2 Eddy covariance flux measurements

The EC flux system was deployed on the shore of Lake Wohlén from 4 June to 30 June 2008 and again from 21 July to 12 August 2008. The system used in this study was described in full detail by Eugster and Plüss (2010). It consisted of a three-dimensional ultrasonic anemometer-thermometer (Gill, UK, model R2A; hereafter referred to as sonic anemometer) and an off-axis integrated-cavity output spectrometer (Los Gatos Research Inc., CA, USA, model 908-0001-0002; hereafter abbreviated as DLT-100) used for measuring CH_4 concentrations. An external vacuum pump (BOC Edwards XDS-35i, USA) was used for EC flux measurements, and main power (230 V AC) was drawn from the nearest building using a 130 m power cord with three leads of 4 mm^2 cross-section. Fully digital data acquisition at 20 Hz was achieved with an industry grade embedded box computer (Advantech ARK-3381, Taiwan). Both analyzers sent their data via RS-232 serial ports to the in-house data acquisition software running under the Linux operating system.

The sonic anemometer was installed at the lake border with a location that had undisturbed fetch over the lake towards the mean wind direction (west to north), and the

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



terrestrial surface with least disturbance in the east (large sand box for horse riding). At the location of measurements, the lake is 300 m wide at its narrowest spot (towards the north), whereas the longest fetch for EC at this site was 1.9 km for winds approaching from the northwest. The sensor height was 2.14 m and 2.13 m above the lake level at time of installation for the first and the second field campaign, respectively. A 6.7 m long Synflex-1300 tubing (Eaton Performance Plastics, OH, USA) with 10 mm outer diameter (8 mm inner diameter) was attached to the sonic anemometer 0.15 m below the center of the EC sensor head to draw air at the lake edge and send it to the DLT-100. A standard plastic funnel was used to protect the inlet against rain, and 1-mm mesh cloth was used to prevent mosquitoes from entering the hose. In contrast to Eugster and Plüss (2010), only a 5 μm filter was used in a combined water trap with a filter unit (SMC, Japan, model AF30-F03/0086095). This was sufficient during summer conditions to prevent mosquitoes from entering the instrument (note that the DLT-100 has an internal 2 μm Swagelok filter to protect the sampling cell from dust particles). An in-depth assessment of the flux equipment used in this study has been carried out (Tuzson et al., 2010), in which the system performed very well when measuring a predefined methane flux.

2.3 Flux data processing

Data processing was done with the in-house eth-flux software version 13.19 (Eugster and Senn, 1995; Mauder et al., 2008) and R for statistical analysis (R Development Core Team, 2010). Since no standard processing exists for CH_4 fluxes, however, the approach chosen for this application is described here.

As noted by Eugster and Plüss (2010), CH_4 fluxes are expected to be more variable than CO_2 fluxes over vegetation canopies as CH_4 fluxes are produced by episodic and stochastic processes rather than continuous processes, such as plant CO_2 uptake. In the case of Lake Wohlen, the dominant CH_4 emission pathway during summer is ebullition (DeSontro et al., 2010). The gas bubbles are produced in the bottom sediments of the lake and, while their release is not well understood, it is known to be

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



intermittent and vary in magnitude (e.g., Ramos et al., 2006). Hence, we tested various approaches to deal with the expected problem that bubbles may be released in intermittent plumes (i.e., extreme bursts of gas), and that perhaps the number of bubbles reaching the surface is not a random function of time. At the same time we tried to adhere to the accepted CarboEurope processing strategy for CO₂ as much as possible; that is, using block averages without detrending of the measured time series, and a two-step rotation to align the coordinates with the mean streamlines. The first rotation aligns the horizontal coordinates such that the mean wind speed \bar{u} is aligned with axis x and with zero mean in the lateral axis y . The second rotation step then corrects for the inclination angle between the mean streamlines and the horizontal plane spanned by the x - and y -axes of the sonic anemometer. Averages were computed for intervals of 5, 10, 30, and 60 min, but there was no clear indication that a specific averaging interval would necessarily lead to the highest accuracy in flux computations.

Moreover, the generally used tests of stationarity and integrated turbulence characteristics (Foken et al., 2004; Mauder et al., 2008) did not succeed in removing spurious data points (not shown). Since the EC instrumentation was mounted right at the lake border, our expectation was that whenever the wind blows along the lake shore with its shrubby vegetation, flux measurements should fail these tests. This was not the case, and hence we had to take a different approach that is detailed below to remove questionable flux data. It should be noted that a standard friction velocity (u_*) filtering approach (e.g., Gu et al., 2005) cannot be used over a lake surface. The higher heat capacity of water keeps the lake water warmer than its surroundings during the night, and hence near-neutral and unstable conditions were found over the lake at night 86 % of the time (between 22:00 and 05:00), but only 48 % of the time during daytime (between 10:00 and 17:00).

Cases with unrealistic CH₄ fluxes could be distinguished by inspecting the time lag between vertical wind speed and CH₄ concentration. There is an expected time lag that can be computed based on the length and inner diameter of the tube sending air to the DLT-100 and the pump rate (in our configuration 0.24–1.44 s; see Eugster and Plüss,

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2010). Hence, if the automatic cross-correlation procedure to find the lag stopped at the inner or outer boundary of the search window that we specified, then this was a clear indication that either (a) the physically correct lag was not clearly represented by the measurements (this could however also be indicative of a zero flux, which is the most difficult value to measure with EC), or (b) episodic events in the time series dominated the mixing of CH₄ in the atmosphere, and hence neither stationarity nor representativity for the upwind footprint area can be assumed. It is important to note that the established stationarity test in CarboEurope compares the mean of six 5-min averages with the 30-min flux, and deviations less than ±100 % are flagged as “good quality” (±30 % are flagged as “highest quality”; see Mauder and Foken, 2004). Hence, if one 5-min period in a 30-minute interval shows a flux that is 600 % higher (or lower) than during the other 5 intervals, then the stationarity test is still fulfilled and the data are considered “good quality” (a deviation of less than 180 % would be “highest quality”). For the measurement of CH₄ fluxes over a lake where ebullition is the responsible process and fluxes can range over several orders of magnitude (DelSontro et al., 2010; Ramos et al., 2006), the CarboEurope quality flags for CO₂ and momentum flux were not used. They were instead used only to remove the cases without a clear peak in the cross-correlation function that was inside the specified time window. With this data selection criterion we could still use the standard 30-min flux averages in our analyses.

Although we operated the CH₄ analyzer with a strong vacuum pump, the flushing of the sampling cell was not perfect (see Eugster and Plüss, 2010), and hence we applied a high-frequency damping loss correction according to Eugster and Senn (1995) to correct for the underestimation of EC fluxes. Using cases with well-developed cospectra (as in Fig. 1) we determined a damping constant $L \approx 0.14 \text{ s}^{-1}$, which was used for the Eugster and Senn (1995) correction.

2.4 Floating chamber flux measurements

Floating chamber campaigns for directly collecting surface CH₄ emissions were conducted in 2008 on 23, 24, 29, and 30 July and were part of the DelSontro et al. (2010)

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



whole-year sampling effort. Chambers consisted of a circular bucket (22 l, 26 cm high, 855 cm² surface area) that collected gas diffused from the water surface and released from emerging bubbles (if present) while the chamber was kept afloat by buoys and upright by weights. An air-tight tube (\approx 40 cm long) was attached to the top of each chamber via a brass hose fitting (0.4 mm inner diameter) screwed into the chamber and made air-tight with an o-ring. Chambers were unanchored and allowed to drift on the lake adjacent to the EC tower location. Transects were approximately 0.5 km long and lasted anywhere from 20 min to an hour and 45 min depending on wind speed. Gas was collected using a 60 ml syringe and a 3-way stopcock at the end of the tubing. Ten ml of gas was extracted and discarded to mix the gas inside the tubing and to flush the syringe. Then 20 ml of gas was collected and injected into 30 ml serum bottles pre-capped with a butyl-rubber stopper and aluminum cap. Bottles were also pre-filled with a saturated NaCl solution to prevent CH₄ dissolution and an open needle placed in the stopper allowed the displaced NaCl solution to exit the bottle while the collected gas was being injected. Samples were stored upside down until analysis on a gas chromatograph (Agilent 6890N) with a flame ionization detector.

2.5 Ancillary measurements

During the flux measurement campaigns, lake water temperatures (0.1 m depth) at the site of EC flux measurements were recorded as 5-min averages with a self-contained temperature mini-logger. Air temperature, relative humidity, cup anemometer wind speed and wind vane direction were recorded by an Aanderaa (Norway) weather station. Full-year measurements of Aare river discharge and temperature were obtained from the Schönau monitoring site upstream of our sampling area (daily resolution for discharge, hourly for temperature, obtained from the Swiss Federal Office for the Environment). The hydroelectric company BKW provided additional water temperatures at 10 m depth in front of the dam, together with lake level information (both at 15 min intervals), and high-precision air pressure information was taken from the nearest MeteoSwiss station Mühleberg, which was 2.5 km west of our flux measurement site.

3 Results

3.1 Performance of the system

The performance of the methane analyzer used here was already described by Eugster and Plüss (2010). The field data that were shown in this previous study were collected on a landfill site in Switzerland in the time period between the two campaigns that were carried out for this study at Lake Wohlen. The overall technical performance of the equipment was very similar between the two Lake Wohlen campaigns, showing well-defined spectra of wind speed components and CH₄ concentration fluctuations, but more variable cospectra of CH₄ fluxes depending on flux strength and stationarity of conditions. Figure 1 shows an example for ideal conditions when the wind direction was from the lake. A minor damping at the highest frequencies was still seen in the CH₄ spectra (Fig. 1b) with the configuration that we used, but the effect on CH₄ fluxes is rather small (Fig. 1d). The two idealized curves in Fig. 1d represent the damped (solid gray) and undamped (dashed gray) cospectrum as described by Eugster and Senn (1995). The damping constant was quantified at 0.14 s^{-1} , which requires a high-frequency damping loss correction that increases measured CH₄ effluxes by 16% on average (median is 9%). The CH₄ spectra clearly indicate a strong signal that is orders of magnitude larger than the white noise level of the DLT-100 instrument (Fig. 1b). The cospectrum shown in Fig. 1d shows an almost ideal period with continuous effluxes from the lake surface, whereas the vast majority of cases show a more variable and intermittent behavior of fluxes, even during periods where the vertical wind speed w and CH₄ spectra are rather smooth. As noted by Eugster and Plüss (2010), this was expected as we were measuring a phenomenon with episodic tendencies (i.e., bubble plumes released intermittently from the lake with less active or quiescent times of ebullition the rest of the time).

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2 Turbulent conditions at the measurement site

After having removed the conditions with instationary fluxes, the accepted fluxes primarily were measured at moderate ($<4 \text{ m s}^{-1}$) wind speeds when wind was coming from the lake, and during rather calm conditions ($<1 \text{ m s}^{-1}$) when winds were from the land surface (Fig. 2). The observed horizontal wind speed dependence on wind direction was expected as Lake Wohlen is located in a east-west running valley of the Aare River with the surrounding plateau $\approx 120 \text{ m}$ above lake level. The prevailing synoptic westerly winds could therefore approach our measurement station with minimal obstruction, whereas winds from other directions were always associated with very local thermo-topographical wind systems driven by differential heating between the cold lake surface and the warmer (day) or even colder (night) land surface during this time of year (see e.g., Whiteman, 2000, or Pielke and Avissar, 1990, for a general overview of such local secondary circulations).

Clear effects of obstructions to both sides of the flux tower system are apparent in the aerodynamic roughness seen by the sonic anemometer. The roughness length z_0 (m) can be computed from momentum flux $\overline{u'w'}$ ($\text{m}^2 \text{ s}^{-2}$) measured at height z above ground (m), horizontal wind speed \bar{u} (m s^{-1}), and Monin-Obukhov stability z/L (Monin and Obukhov, 1954) that are directly measured by the sonic anemometer,

$$z_0 = \frac{z}{\exp\left[\frac{\bar{u}k}{u_*} + \Psi(z/L)\right]}, \quad (1)$$

where u_* is the friction velocity derived from momentum flux measurements ($u_* = \sqrt{-\overline{u'w'}}$ for conditions where $-\overline{u'w'} < 0 \text{ m}^2 \text{ s}^{-2}$), and $\Psi(z/L)$ is the stability correction function parameterized by Paulson (1970) based on the concept of the universally valid diabatic wind profile (Monin and Obukhov, 1954). Overlines denote averaging over time (30 min in our study), and primes indicate the short-term deviation from such a mean. Since momentum flux measured with EC tends to require longer averaging

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



times than scalar fluxes (Wyngaard, 1990), we expect to see any effects of obstructions and inhomogeneous fetch most clearly in $-\overline{u'w'}$ or in an entity such as z_0 that is derived from $-\overline{u'w'}$. For the sector with wind speeds exceeding 1 m s^{-1} (around 220° – 310° in Fig. 2) where there is a fetch of several hundreds of meters over the water surface, median z_0 computed with Eq. (1) was 0.005 m. This is an appropriate order of magnitude as it is higher than that expected over large water bodies ($<0.001 \text{ m}$; Panofsky and Dutton, 1984), but lower than that tabulated for short-cut grass over flat ground ($\approx 0.007 \text{ m}$, Panofsky and Dutton, 1984).

Methane emissions from the lake (and from other potential sources in the valley) are strongly contained in the atmospheric boundary layer above the lake surface due to the relatively cold surface water (Fig. 3a; summer maximum $\approx 20^\circ\text{C}$), which limits convection during daytime, but enhances turbulent mixing during nighttime.

3.3 Lake methane effluxes

To analyze lake methane effluxes measured by EC we extracted the data where the 30-min vector-averaged wind direction was from the lake (between 200° and 10° , see also Fig. 2). During both deployments, CH_4 concentrations in ambient air at EC height were a minimum of 1.853 ppm, which is slightly above the background concentration (1.774 ppm; Forster et al., 2007). Figure 4 clearly shows very high concentrations in $>50\%$ of all cases when winds were from the NW, which was the direction towards the lake where ebullition was easily seen at the surface and chambers caught some of the highest effluxes. Using wind from the lake direction always resulted in positive CH_4 fluxes indicating an efflux from the lake to the atmosphere (Fig. 5). With the exception of a few measurements exceeding $80 \mu\text{g m}^{-2} \text{ s}^{-1}$ found in the near-shore sector of the lake (220° – 260°) during higher wind speeds, median fluxes were highest when winds were low ($<1 \text{ m s}^{-1}$, Fig. 2) and from the NW (310° – 340° ; Fig. 5). A detailed inspection of the flux footprint area contributing to the CH_4 fluxes observed during our two field campaigns shows that the shallow near-shore areas were best covered (Fig. 6). The flux footprint area as computed with the Kljun et al. (2004) model was much smaller

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



than we expected when we designed the field experiment. Figure 6 shows a composite of relative footprints for each 30-min period weighted by the respective CH₄ efflux. These calculations show that the most relevant surface areas that led to the strong effluxes were in the southwest where the high frequency of wind from this direction (west is the prevailing wind direction at the site) is combined with large effluxes, and an area in the northwest where infrequent winds were associated with the highest median fluxes that we measured (see Fig. 5).

3.4 Comparison with chamber fluxes

Since we expected a larger footprint area with the EC system than the posteriori computations actually showed for the subset of data with wind from the lake (Fig. 6), the drifting chambers were deployed just outside the footprint of the EC flux measurements. Still, if we assume that our EC flux measurements should be representative for the lake, then a general agreement with the chamber flux measurements should be found. In fact, the flux data obtained from 29 chamber deployments show the same order of magnitude and variability of fluxes (boxplot in Fig. 5) as measured by the EC flux system. The median CH₄ efflux from the lake measured by EC (which includes the necessary high-frequency damping loss corrections) was 1.42 μg C m⁻² s⁻¹ (interquartile range 0.66–2.77 μg C m⁻² s⁻¹; mean ± SE 3.76 ± 0.39 μg C m⁻² s⁻¹; N = 513 half-hour averages), whereas the chamber flux measurements obtained a median flux of 7.43 μg C m⁻² s⁻¹ (interquartile range 1.53–11.11 μg C m⁻² s⁻¹; mean ± SE 7.43 ± 1.33 μg CH₄ m⁻² s⁻¹; N = 29 chamber deployments). This flux is extremely high for a temperate hydropower reservoir, but agrees well with the values expected for summer conditions based on data obtained by DelSontro et al. (2010) using a multi-temporal discrete water sampling and mass balance approach from June 2007 to June 2008.

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.5 Methane fluxes from contrasting surfaces

Eddy covariance flux measurements may be very accurate point measurements, but may not be representative (Wyngaard, 1990) for a larger upwind surface area (the flux footprint area) if a handful of simplifying assumptions cannot be made. To be able to relate a high-quality EC flux to the larger surface area, the common assumptions to be made are (1) that turbulent conditions are stationary such that the time-for-space substitution (Taylor's frozen turbulence field hypothesis; Taylor, 1938) is valid; (2) that CH₄ sources and sinks are randomly distributed in space (homogeneity of surface); and (3) that source or sink strengths must be spatially representative (see Schmid, 2002 for an overview of footprint concepts and assumptions). With our placement of instruments these conditions are met in the undisturbed sector facing the prevailing wind (the lake sector, which allows us to measure fluxes from the water body), and possibly in the SE wind sector, where the sand box is found. Other directions are heavily disturbed and are hence only shown for reference. Figure 5 shows the fluxes measured from all directions without eliminating conditions where the above assumptions are not met. This is of particular interest to test a common but largely untested hypothesis that EC flux measurements are useless if the above assumptions are not perfectly met. And as a second objective, it allows us to test whether the CH₄ flux to or from the sand box in the SW is small. In such well-aerated sandy soils either a small CH₄ sink (e.g., Hütsch et al., 1994; Castaldi et al., 2007) or a small source should be expected (e.g., Radl et al., 2007). Using Radl et al.'s fluxes from moderately impacted pastures in spring a flux in the range 0.03 to 0.14 μg C m⁻² s⁻¹ would be expected from the sand box.

Our results show similarly small fluxes for the wind sectors between 135 and 160° (from the sandbox, 0.07 ± 0.11 μg C m⁻² s⁻¹, mean ± SE), which agrees well with our expectation. This indicates a rather good performance of the system, although it should be noted that the alignment between these relatively small minimum fluxes and the center of the sand box is not perfect. Still, from this comparison we expect our EC system to be suitable also for efflux measurements from the lake sector. In strong

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



contrast to the sand box fluxes, there were no cases with CH₄ uptake over the lake (220°–10°), whereas the obstructed lake border and terrestrial hinterland surfaces did show downward CH₄ fluxes, namely in the sector 160° to 200°.

3.6 Environmental drivers

5 The hydropower generation of the lake follows a typical diurnal pattern with highest lake level in the morning and lowest in the evening (before 22:00), which most likely caused the diurnal variability observed in CH₄ effluxes (data not shown). The regression against lake level measurements is able to explain 23.1 % of the variation in CH₄ efflux from the lake (Table 1, adj. $R^2 = 0.231$, $p < 0.000001$, based on log-transformed
10 30-min flux averages), despite the relative change of lake level with respect to a 2-day retrospective moving average only being ± 0.1 m (or 10 hPa and not much stronger than atmospheric pressure variability due to changing weather patterns). Also every 3 to 4 weeks in summer, the hydropower company lowers the lake level artificially by an extra 0.15 m (Fig. 3b), which superimposes a longer-term variability that we were not
15 able to resolve with two field campaigns of a few weeks each, but most likely affects the seasonal efflux as shown by Ostrovsky et al. (2008).

A strong diurnal cycle is also found in the near-surface water temperatures that we measured at the field site (mean diurnal range was 2.91, 3.04, 3.05, and 2.04 K in June, July, August, and September, respectively), but synchronous measurements of
20 temperatures and CH₄ fluxes only showed a weak correlation (Fig. 7, $R^2 \leq 0.143$). Hence we wanted to know whether (1) time lag effects or (2) time integration effects might be essential for the explanation of CH₄ fluxes from this dynamic aquatic system. To address these two components we used (1) lagged cross-correlation analysis and
25 (2) smoothing of the variables under consideration.

The time lag analysis directly showed the time delay between the temperature measurements taken at the hydropower dam at 10 m depth and lake surface temperature measured at the flux site, which was 4.5 h. Since no other relevant time lag effects could be found we shifted this time series by -4.5 h. This allowed us to conclude that

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



time lag effects in our system are associated purely with the time it takes the water in the flux footprint of our measurements to reach the dam. In contrast, the second component (i.e., time integration) revealed more significant results. Since CH₄ fluxes are not normally distributed (see e.g., Ramos et al., 2006; Eugster and Plüss, 2010), for these analyses we used the log-transformed CH₄ fluxes.

The smoothing was done under the theoretical consideration that CH₄ production and transport in the lake may not respond to the environmental variables at the 30-min timescale of our averaging intervals, but to longer integration periods of up to 5 days, well beyond the peak of best multivariate correlations (Fig. 7). Hence we generated new averaged (smoothed) time series of atmospheric pressure (P), lake surface temperature measured in the footprint of the flux site (T_{lake}), and 10-m deep water temperature measured at the dam (T_{dam}). We used a retrospective moving average filter with equal filter weights to produce these new time series. The computations were carried out for integration periods (i.e., filter lengths) of 0 to 5 days in 1-h time steps. For each time step (except for lag 0) both the smoothed values and the residuals were used in the regression analysis. This was considered meaningful because, for example, a change in pressure might increase or decrease the bubble flux in the water column, but only during a certain time period until a new equilibrium is established. In this setting, a good correlation with a smoothed variable would indicate a buffered system with slow adaptation to changing conditions. Contrastingly, a better correlation with the residuals than with the smoothed variable implies a rapid adaptation of the relevant mechanisms influencing CH₄ efflux in response to environmental conditions changing on relatively short timescales.

Figure 7 shows the result of this analysis as a function of retrospective time integration (smoothing). The highest explained variance – which indicates an optimum integration time over 9 h – reached a modest adj. $R^2 = 0.3542$ ($p < 0.000001$; Table 2 and arrow in Fig. 7). While (smoothed) T_{lake} increases methane efflux (Table 2; Fig. 8c), the short-term deviation (residuals) of the lake water level tends to decrease the flux (Fig. 8a), similar to the short-term atmospheric pressure variations (Fig. 8b). Each of

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the temperature variables (Fig. 8c, d and e) suggests an increasing flux with increasing temperature. In combination, however, T_{lake} has the strongest explanatory power in the analysis (Table 2), whereas T_{dam} corrects for the exaggerated diurnal temperature range of T_{lake} (negative regression slope in Table 2). This means that the best place for the temperature measurements to explain CH_4 fluxes would have been at a depth between the T_{lake} (surface) and T_{dam} (-10 m). Overall, our linear model explained $\approx 35\%$ of the variation seen in CH_4 emissions from Lake Wohlen (adj. $R^2 = 0.3542$, $p < 0.000001$, Table 2). This suggests that although short-term variability responds to temperature and pressure effects other unmeasured components are also essential. We suspect that this may be the substrate supply for methanogenesis in the sediments (i.e., POM inputs from the river). We were however unable to find a strong relationship between POM import and CH_4 emission on the short timescales studied here, since it takes some time (one year or longer) for deposited POM to reach the deep sediment layers responsible for ebullition (data not shown).

4 Discussion

Eddy covariance flux measurements showed extremely high CH_4 emissions from Lake Wohlen, which confirms a previous study's results using a system analysis mass balance approach, as well as floating chambers, to assess the fluxes (DelSontro et al., 2010). These extreme fluxes were mainly driven by water temperature, but are strongly reduced whenever pressure exerted by lake level and air pressure increased. All temperature variables show increasing CH_4 flux with increasing temperature, as would be expected from a biologically-sourced CH_4 flux that depends on the metabolic activity of methanogens decomposing organic matter under anoxic conditions (Fig. 8c and d) (Takita and Sakamoto, 1993; Conrad, 1989). However, on an annual timescale with periodic flux measurements using drifting chambers (DelSontro et al., 2010) a much clearer water temperature dependency of fluxes was found for temperatures exceeding 10°C . Even if CH_4 emission fluxes measured by eddy covariance generally agreed with

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



chamber-derived fluxes, it was not possible to relate individual chamber flux values to EC fluxes from the same periods. In a few cases the agreement was quite good, but in general the lack of overlap between the chamber transects and the EC flux footprint, as well as the difference in temporal resolution of the sampling methods, makes for a difficult direct comparison.

With the high temporal resolution of EC flux measurements, the short-term process of pressure changes due to changes in reservoir level and/or changes in atmospheric pressure became an important confounding factor of CH₄ emission. In the following, we first address the question of whether biological (temperature-driven) or physical (pressure-driven) processes – or both together – are crucial for understanding CH₄ fluxes from this hydropower reservoir. Next we discuss what the C sources are and whether they are sufficient to sustain the extreme CH₄ emissions measured. Finally, the CH₄ fluxes will be put in relation to net C uptake of the surrounding terrestrial ecosystems to estimate the potential relevance of aquatic ecosystem fluxes to the local and regional greenhouse gas budgets.

4.1 Temperature versus lake level response

Earlier surveys (DelSontro et al., 2010) did not use the temperature measured at the locality of flux measurements, but the upstream river temperature from the routine long-term measurements by the local authorities at Schönau, Bern (Naduf, 2000). To rule out the possibility that such a methodical difference could be responsible for the important differences in correlation between CH₄ flux and temperature, we also carried out our analysis with these temperature readings (Fig. 8e) instead of those measured on site (Fig. 8c). There is however no indication that this is an issue as our EC-measured CH₄ fluxes show a similar response to both temperatures and in both cases the order of magnitude corresponds with that reported by DelSontro et al. (2010) (DS2010 lines in Fig. 8c and e).

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



In addition, the high resolution flux sampling provided by EC allowed the introduction of short-term “noise” from processes acting on shorter timescales. The physical processes related to the short-term deviations from the smoothed lake water level (Fig. 8a) and atmospheric pressure (Fig. 8b) tend to decrease fluxes when pressure increases, or enhance the efflux when pressure decreases, but only until a new equilibrium is reached. This, however, does not change the fact that biological activity (i.e., decomposition of organic matter in the sediments) is responsible for the CH₄ fluxes observed over longer time periods. As well, our flux footprint (Fig. 6) only covers the shallowest areas of the lake (depth <3 m) and it is known from other studies that episodic bursts of CH₄ are characteristic of the shallow littoral zone (e.g., Hofmann et al., 2010).

Also, we found an order of magnitude larger fluxes during the few cases where wind was approaching from the Northwest (Fig. 5), which corresponds to the only cases where our flux footprint reaches beyond the shallow littoral zone (Fig. 6). These larger fluxes from the direction of the old river channel also agree best with the floating chambers, which actually bypassed the shallow littoral zone and drifted along the old river channel only. Regardless, it may be that in Lake Wohlen the methane production in the sediments of the deeper parts of the lake dominates the overall CH₄ emissions. Due to the lack of stratification of the lake, mean temperatures at depth are not expected to dramatically differ from the temperatures that we measured for the surface waters when averaged over days or longer.

4.2 What are the C sources and are they sufficient to sustain CH₄ emissions?

Since Lake Wohlen is oxic in summer without a clear stratification (see Fig. 3a), it is unlikely that substantial CH₄ production occurs in the water column itself; hence production must be constrained mostly to the anoxic sediments underlying this oxic and well-mixed lake (Kiene, 1991; McGinnis et al., 2008).

Three studies in 2008 investigated the water quality of the Aare river, including Lake Wohlen, using three different indicators: (1) bioindication of algae (von Känel, 2008), (2) silicious algae (AquaPlus, 2008), and (3) macroinvertebrates (Mürle et al., 2009).

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



All three assessments found very high water quality (highest mark) for most biological and chemical aspects investigated. Good quality (second highest mark) was found for DOC, nitrite and total phosphorous. However, these are only qualitative measurements, whereas quantitative estimates only were made more than a decade ago. If we consider these monitoring data from 1994–1996 to be representative for 2008, an average POM import by the Aare river of 139 g C s^{-1} to Lake Wohlen can be expected. When put in relation to the 2.5 km^2 lake surface, and assuming that all imported POM settles to the sediments, this corresponds to a POM sedimentation of roughly $56 \mu\text{g C m}^{-2} \text{ s}^{-1}$. These calculations indicate that only a small fraction of the river POM import, on the order of 3%, is needed to account for the observed extreme CH_4 emission ($3.76 \mu\text{g C m}^{-2} \text{ s}^{-1}$) from Lake Wohlen.

5 Conclusions

So far CH_4 effluxes from inland waters have been largely ignored by the terrestrial ecosystem flux community, and hence it is of interest to make a rough estimate of how the CH_4 fluxes from Lake Wohlen relate to typical C uptake rates of the surrounding landscape. The compilation of multi-year net ecosystem exchange (NEE) of grasslands, croplands and forest by Kindler et al. (2011) resulted in an average NEE of the European sites under investigation to be $296 \pm 61 \text{ g C m}^{-2} \text{ yr}^{-1}$ (in CO_2 equivalents this is $34 \pm 7 \mu\text{g CO}_2 \text{ -eq m}^{-2} \text{ s}^{-1}$). Our measured summer CH_4 effluxes from Lake Wohlen (average, $3.76 \pm 0.39 \mu\text{g C m}^{-2} \text{ s}^{-1}$) expressed as CO_2 equivalents (factor 25 for a 100-yr time horizon, Solomon et al., 2007) to quantify their global warming potential yields $125 \pm 13 \mu\text{g CO}_2 \text{ -eq m}^{-2} \text{ s}^{-1}$, whereas DelSontro et al. (2010) found $\approx 45 \mu\text{g CO}_2 \text{ -eq m}^{-2} \text{ s}^{-1}$ for the annual average. Hence, for each square meter of Lake Wohlen, at least 3.7 m^2 terrestrial surface area with the sufficiently large net C uptake estimated by Kindler et al. (2011) is required to neutralize the greenhouse forcing exerted by the summer CH_4 effluxes from the reservoir, but less ($\approx 1.3 \text{ m}^2 \text{ m}^{-2}$) on the

BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



annual average. Therefore, temperate reservoirs can be a relevant component in local and regional greenhouse gas budgets, even if the total lake surface appears small at larger scales.

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Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



P., Eugster, W., Gleixner, G., Grünwald, T., Heim, A., Ibrom, A., Jones, S. K., Jones, M., Klumpp, K., Kutsch, W., Steenberg Larsen, K., Lehuger, S., Loubet, B., McKenzie, R., Moors, E., Osborne, B., Pilegaard, K., Rebmann, C., Saunders, M., Schmidt, M. W. I., Schrupp, M., Seyfferth, J., Skiba, U., Soussana, J.-F., Sutton, M. A., Tefs, C., Vowinckel, B., Zeeman, M. J., and Kaupenjohann, M.: Dissolved carbon leaching from soil is a crucial component of the net ecosystem carbon balance, *Glob. Change Biol.*, 17, 1167–1185, doi:10.1111/j.1365-2486.2010.02282.x, 2011. 5021

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BGD

8, 5019–5055, 2011

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Methane emissions
from hydropower
reservoir**

W. Eugster et al.

Table 1. Linear regression between the log-transformed CH₄ effluxes from Lake Wohlen and lake level changes as a potential driving variable for fluxes.

Variable	Estimate	Std. Error	<i>t</i> value	Pr (> <i>t</i>)
Residual lake level (m)	−17.42	1.40	−12.42	<0.000001
Intercept	−10.769	0.050	−15.50	<0.000001

Residual standard error: 1.1 on 511 degrees of freedom
 Multiple *R*-squared: 0.232, Adjusted *R*-squared: 0.2305
F-statistic: 154.4 on 1 and 511 DF, *p*-value: <0.000001

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions from hydropower reservoir

W. Eugster et al.

Table 2. Multiple linear regression between the log-transformed CH₄ effluxes from Lake Wohlen and potential driving variables with 9-h retrospective boxcar smoothing that led to highest overall explanation of variance (adj. $R^2 = 0.35$). Lines in italics are not significant ($p > 0.05$). T_{lake} and T_{dam} denote lake surface (−0.1 m) and dam water (−10 m) temperatures, respectively.

Variable	Estimate	Std. Error	<i>t</i> value	Pr (> <i>t</i>)
Residual lake level (m)	−19.07	1.98	−9.654	<0.000001
T_{lake} (9-h mean lake surface temperature, °C)	0.4828	0.0671	7.200	<0.00001
Residual pressure (hPa)	−0.3695	0.05924	−6.237	<0.00001
T_{dam} (9-h mean dam water temperature, °C)	−0.3088	0.0697	−4.431	<0.000012
T_{lake} residual (°C)	−0.2572	120.0715	−3.597	<0.00035
<i>Intercept</i>	<i>1205.9</i>	<i>719.0</i>	<i>1.677</i>	<i><0.094</i>
<i>9-h mean lake level (m)</i>	<i>−2.4870</i>	<i>1.497</i>	<i>−1.661</i>	<i><0.097</i>
<i>9-h mean pressure (hPa)</i>	<i>−0.0143</i>	<i>0.0145</i>	<i>−0.983</i>	<i><0.33</i>
T_{dam} residual (°C)	−0.1124	0.1313	−0.856	<0.39

Residual standard error: 1.017 on 504 degrees of freedom
 Multiple R -squared: 0.3643, Adjusted R -squared: 0.3542
 F -statistic: 36.1 on 8 and 504 DF, p -value: <0.000001

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions
from hydropower
reservoir

W. Eugster et al.

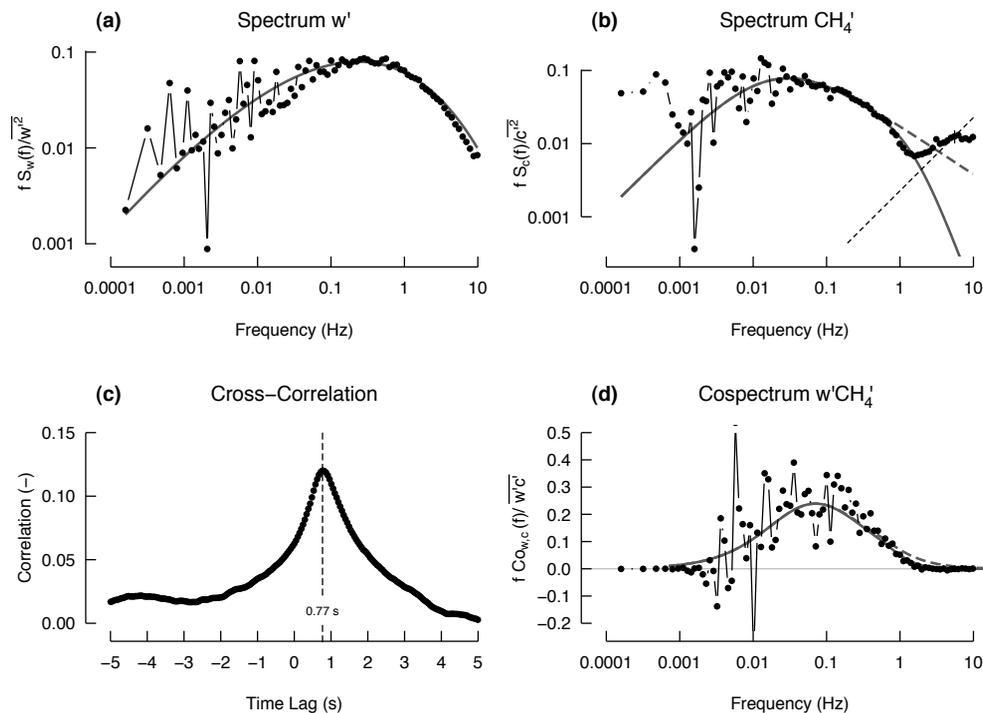


Fig. 1. Example spectra of **(a)** vertical wind speed w and **(b)** CH_4 concentration c , **(c)** time lag between w and c , and **(d)** cospectrum of CH_4 flux over Lake Wohlensee. The data used were collected during 1.75 h (217 records) between 18:00 and 19:45 on 21 July 2008 with a mean horizontal wind speed of 1.5 m s^{-1} and wind direction 284° . Spectra and cospectrum were bandwidth averaged using 100 bands of equal spacing on the log frequency axis. Idealized curves are shown in gray. Dashed gray lines in **(b)** and **(d)** are idealized curves for an ideal system without high-frequency damping losses, and black dashed line in **(b)** shows white noise level of CH_4 analyzer.

Methane emissions from hydropower reservoir

W. Eugster et al.

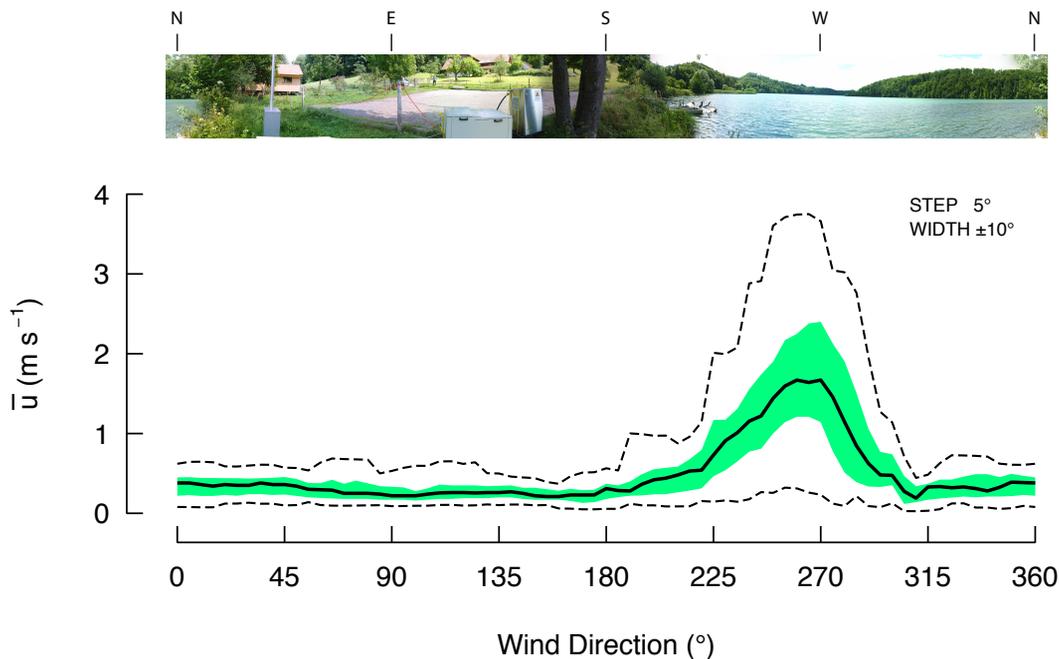


Fig. 2. Horizontal wind speed as a function of wind direction. For reference, a panorama image taken at the position of the sonic anemometer sensor head is shown in the top section. Data were aggregated for overlapping wind direction sectors of 10° with 50% overlap. Median (bold line), interquartile range (shaded area; 50% of all values), and maximum and minimum values (dashed lines) are shown. The predominant wind direction from the west is also reflected by highest wind speeds.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

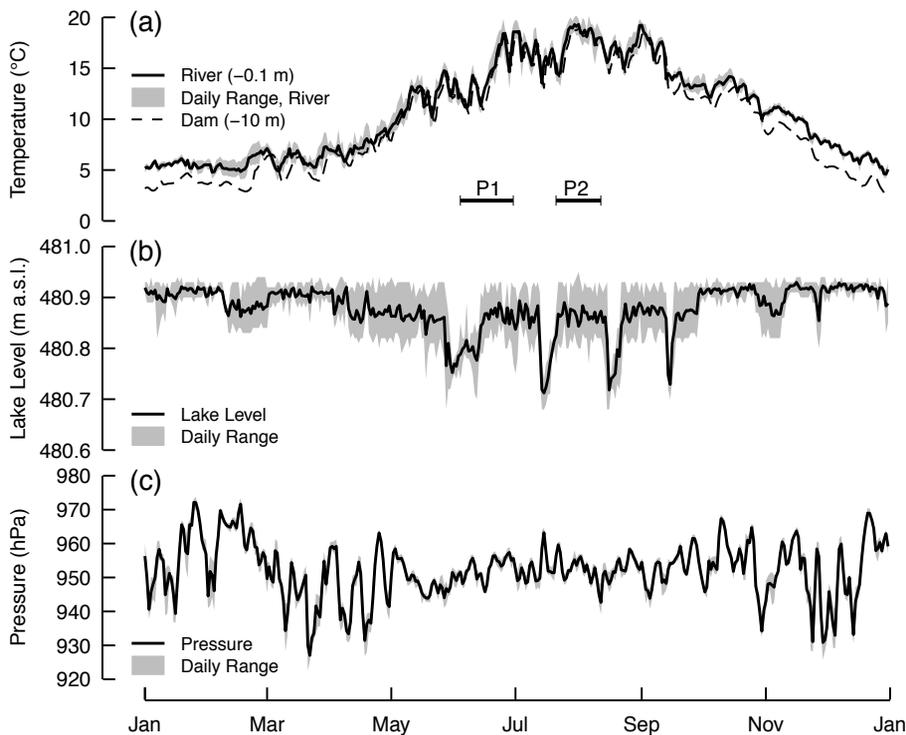


Fig. 3. Driving variables of CH_4 efflux from lake Wohlensee at daily resolution, **(a)** temperature of the river waters (0.1 m; bold line with gray band showing daily range of values) and the dam (10 m, dashed line); **(b)** lake level measured at the dam (bold line with gray band showing daily range of values); and **(c)** atmospheric pressure (bold line with gray band showing daily range of values). Data courtesy of Swiss Federal Office for the Environment (**a**, river temperature), Bernische Kraftwerke BKW (**a**, dam temperature, and **b**), and MeteoSwiss (**c**). P1 and P2 indicate the period when eddy covariance flux measurements were carried out.

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Methane emissions from hydropower reservoir

W. Eugster et al.

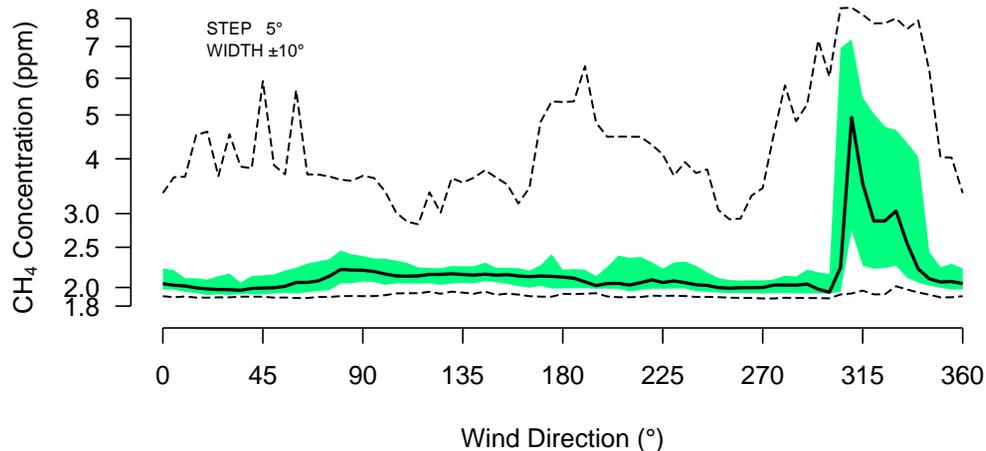


Fig. 4. Ambient CH_4 concentrations as a function of wind direction. The effect of ebullition from the water is clearly seen when winds are from the NW (315°), and these high concentrations also influence the maximum values observed when wind arrives from other directions. Data were aggregated for overlapping wind direction sectors of 10° with 50% overlap. Median (bold line), interquartile range (shaded area; 50% of all values), and maximum and minimum values (dashed lines) are shown.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Methane emissions from hydropower reservoir

W. Eugster et al.

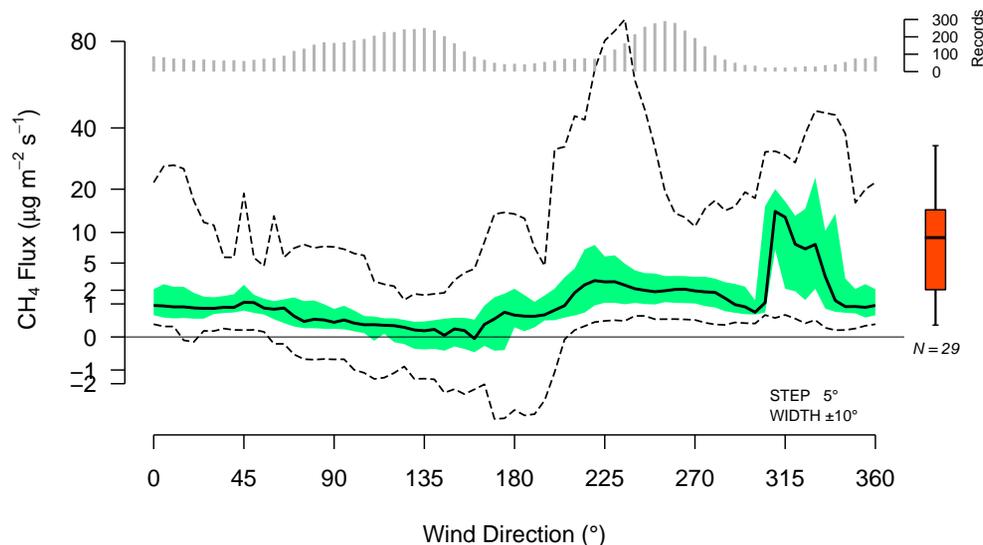


Fig. 5. Methane fluxes as a function of wind direction. Data were aggregated for overlapping wind direction sectors of 10° with 50% overlap. Median (bold line), interquartile range (shaded area; 50% of all values), and maximum and minimum values (dashed lines) are shown. The top inset shows the number of records available for each wind direction sector, and the box and whisker plot at right shows the range of CH_4 fluxes obtained by floating chambers. Note that CH_4 fluxes were always positive when wind was approaching over the lake surface (200° to 10°).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

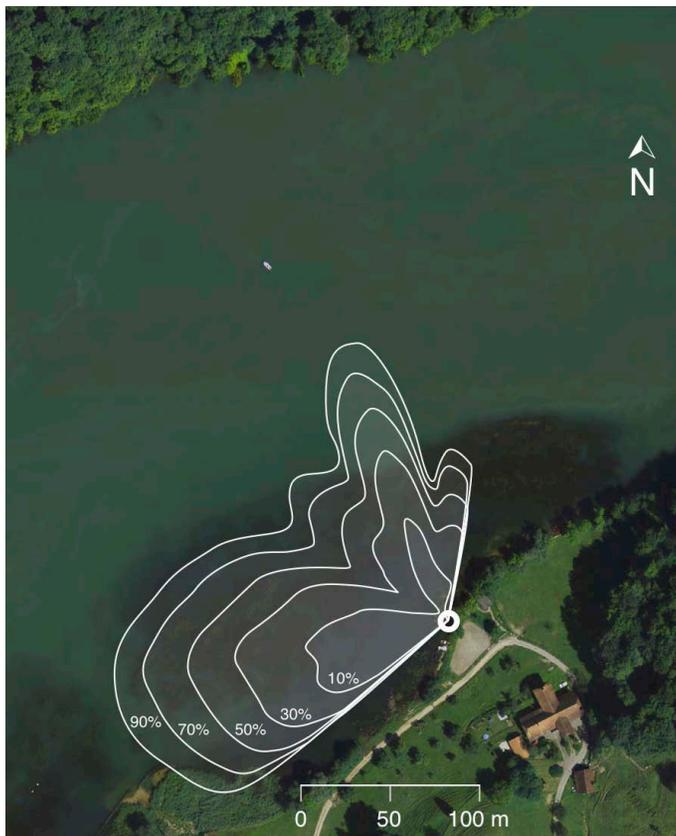


Fig. 6. Flux footprint for CH₄ efflux from Lake Wohlen. Isolines show percentage of contribution to flux measurements of both periods. The white circle shows the position of the flux tower on the lake shore (background image ©2010 swisstopo, reproduced with the authorization of swisstopo JD100042/JA100020).

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



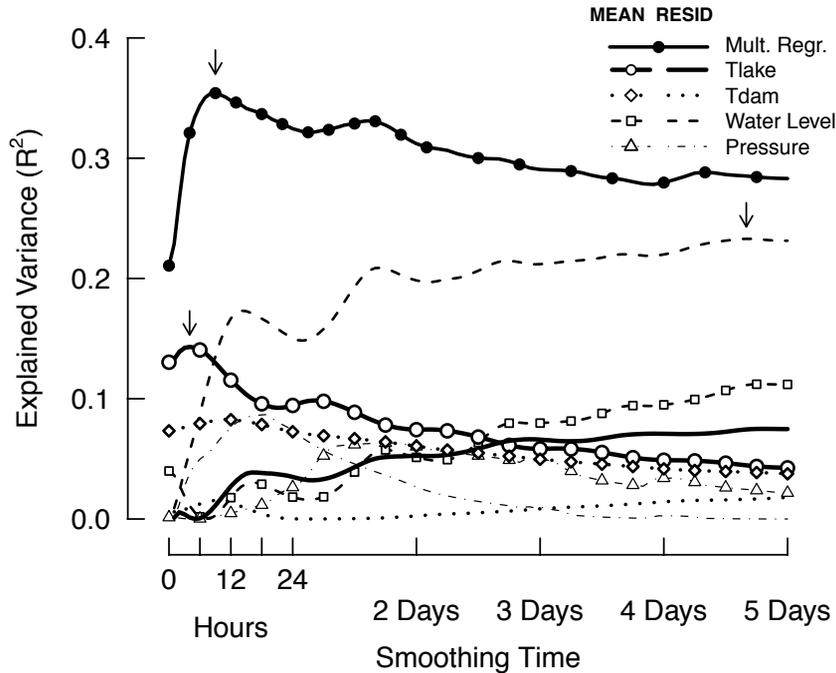


Fig. 7. Correlation analysis with smoothed driving variables (lake surface water temperature T_{lake} , dam 10-m temperature T_{dam} , water level height and atmospheric pressure) and the multiple regression shown in Table 2. For each driving variable, the evolution of R^2 (adj. R^2 for the multiple regression) with increasing smoothing time (running means: open symbols; residuals from running means: lines without symbols) is shown. Although lake surface water temperature shows the greatest explanatory power for short averaging times, none of the single drivers reach the level that the linear combination used in the multiple regression approach achieved. Arrows show the optimum smoothing time for the three most important curves.

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



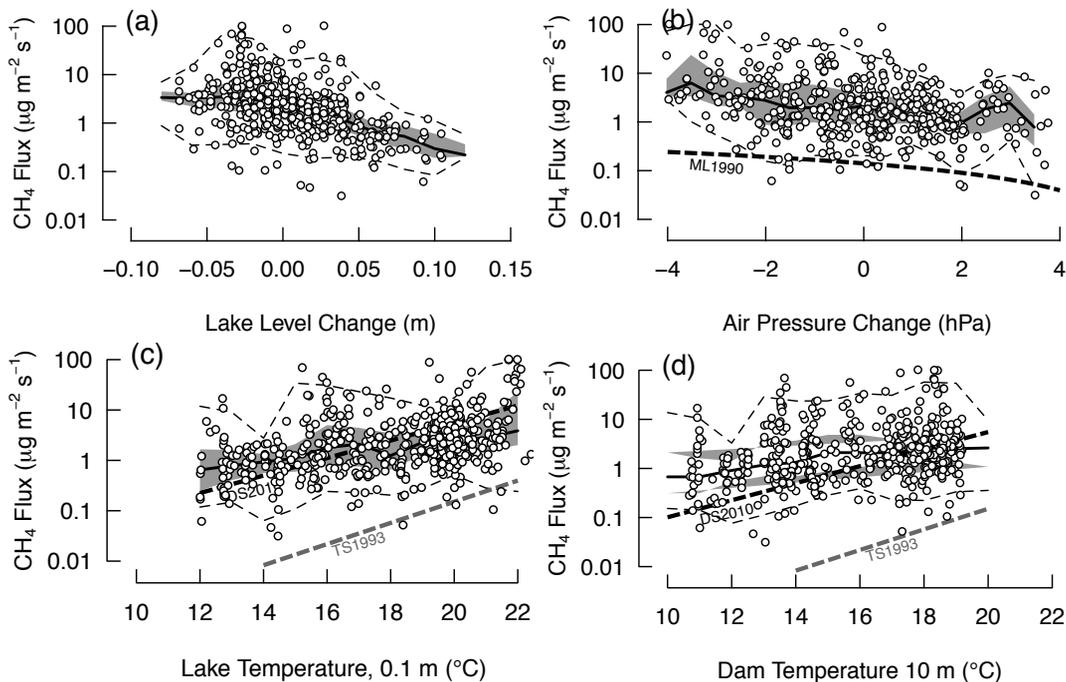


Fig. 8. Dependence of lake CH_4 effluxes from **(a)** lake level changes, **(b)** air pressure changes, **(c)** near-surface lake temperature, **(d)** 10-m water temperatures, and **(e)** upstream near-surface water temperatures. Individual 30-min flux averages (open circles) are plotted on top of bin-averaged median (bold line), interquartile range (gray band), and 95% confidence interval (broken lines). Bin sizes are: 0.02 m for lake level changes, 0.5 hPa for air pressure changes, and 1.0 K for temperatures. As a reference for published pressure response the Mattson and Lichens (1990) curve is shown in panel **(b)**, and the temperature responses reported by Takita and Sakamoto (1993) and DelSontro et al. (2010) are shown in panels **(c–e)** with thick broken lines.

Methane emissions from hydropower reservoir

W. Eugster et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

