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Key Points:

- In situ estimate of sediment gas flux from a small boreal lake
- Gas flux from lake sediment measured by EC was lower than estimated by sediment core incubations
- Low bottom water turbulence inhibits sediment-water gas exchange in small lakes

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Low sediment-water gas exchange in a small boreal lake

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Abstract Boreal lake sediments are carbon sources by producing CO₂. CO₂ flux from sediments is partly controlled by turbulence in the water column, which is not given the same attention as CO₂ production rates in current estimates of CO₂ fluxes from sediments. We quantified the in situ CO₂ flux across the sediment-water interface in a small (0.07 km²) lake in Sweden by measuring the in situ O₂ flux with the Eddy Correlation (EC) method and using the apparent respiratory quotient (CO₂ production:O₂ consumption) derived from sediment incubations. We demonstrate that median CO₂ flux estimated by EC was ~70% smaller than estimated by sediment incubations with artificial water mixing (1.0×10^{-2} and 3.6×10^{-2} μmol C m⁻² s⁻¹, respectively). Additionally, we show that inducing artificial mixing of supernatant water in the incubation experiment has a positive effect on observed fluxes, enhancing CO₂ flux by ~30% compared to not mixing supernatant water. We suggest that the difference between the methods is due to the strong artificial water mixing in sediment incubations compared to the turbulent mixing in this small lake. Additionally, low O₂ supply to sediment aerobic heterotrophic microbes during extended periods of low water currents can inhibit respiration and thus CO₂ production. These findings suggest that the sediment contribution to total lake CO₂ emission might currently be overestimated for small boreal lakes. Care should be taken when upscaling sediment CO₂ flux derived from incubation experiments to entire basins of small lakes, as incubation experiments are unlikely to accurately mimic in situ bottom water currents and gas exchange.

1. Introduction

Freshwater ecosystems play an important role in global biogeochemical cycles, such as the carbon (C) cycle [Tranvik et al., 2009]. Despite covering a small proportion (~3.7%) of the world's surface area [Verpoorter et al., 2014], inland waters are important links between the terrestrial landscape and marine systems as they process, transport, and retain large amounts of C [Aufdenkampe et al., 2011]. Lakes are hot spots for C cycling as they emit greenhouse gases, such as carbon dioxide (CO₂) and methane (CH₄), to the atmosphere [Bastviken et al., 2011; Raymond et al., 2013] and at the same time bury more C in their sediments than the world's oceans [Dean and Gorham, 1998]. Furthermore, with ~1000 times higher nutrient and carbon content than the water column, lake sediments act as sources of CO₂ and CH₄ through microbial respiration [Pace and Prairie, 2005] and may contribute considerably to the total CO₂ emission from lakes [Kortelainen et al., 2006]. The dual role as a source and sink of C in lake ecosystems makes lake sediments of particular interest to study, especially in the boreal zone that contains approximately one third of the world's soil C and has a high density of lakes [Gorham, 1991].

The flux of CO₂ and CH₄ from the sediment to the water column is a function of the production rate in the sediment and the rate of gas exchange between sediment and water. CO₂ production by microbial degradation of sediment organic C is strongly temperature controlled [Gudas et al., 2010] and can vary seasonally and spatially within the same lake as different parts of the sediment are exposed to different temperatures [Bergstrom et al., 2010; Den Heyer and Kalff, 1998]. Most of the bacterial respiration in lake sediments occurs in the uppermost oxygenated layer, where dissolved oxygen in the pore water decreases sharply and anoxia is reached at a few millimeters or centimeters sediment depth [Sobek et al., 2009]. As turbulent fluctuations of the bottom water currents are increasingly dampened close to the sediment-water interface (SWI), a thin diffusive boundary layer (DBL) is created where diffusion is the main transport mechanism for solutes such as oxygen (O₂) [Lorke et al., 2003]. Thus, efficiency of the transport of O₂ across the SWI is regulated by turbulence in the bottom water as it affects the thickness of the DBL [Bryant et al., 2010]. As diffusion is a slow process, the thickness of the DBL strongly controls the rate of gas exchange at the SWI [Lorke et al., 2003].

However, in assessments of lake sediment organic carbon (OC) mineralization, turbulence and thus DBL dynamics are often not given the same attention as CO₂ production rates [Bergstrom *et al.*, 2010; Gudas *et al.*, 2010]. The lack of representation of physical effects on gas exchange at the SWI makes studies on, e.g., the contribution of sediments to lake CO₂ emission inherently uncertain [Algesten *et al.*, 2005; Cardoso *et al.*, 2013].

Several methods have been used to estimate gas fluxes from sediments such as sediment core incubations and benthic chambers; both of which are based on enclosing sediment and measuring the difference in concentration over time in the overlying water to obtain flux [Glud, 2008]. One limitation of such methods is that they operate with reproducible but artificial turbulence in the overlying water by mixing with, e.g., a stirring magnet or a propeller, which might not reflect in situ bottom turbulence. Benthic landers that measure high-resolution microprofiles of O₂ are another option, where flux is calculated from the vertical concentration gradient without disconnecting the sediment from its natural turbulence condition. This method, however, only estimates flux from an area covered by the microsensor that is typically a few hundred micrometers in diameter, which is very little considering the large heterogeneity of sediments [Lewandowski *et al.*, 2002]. Similarly, pore water dialysis samplers (peepers) and diffusion equilibration samplers have been used to analyze vertical concentration gradients in sediment pore water, but these methods have limited spatial resolution [Harper *et al.*, 1998].

Eddy Correlation (EC) is a method that has mainly been used in atmospheric sciences to overcome the invasive nature of the enclosure methods used to estimate fluxes from ecosystems [Baldocchi, 2003]. In the last decade, subaqueous EC has been applied in different ecosystems to measure in situ gas fluxes from various types of aquatic sediment surfaces such as in studies from Berg and Huettel [2008], Berg *et al.* [2003, 2007], Glud *et al.* [2010], Kuwae *et al.* [2006], Long *et al.* [2013], Lorke *et al.* [2013], and many others. Most studies applying subaqueous EC have, however, been conducted in the ocean [Berg *et al.*, 2009, 2013] and only a few times in lakes [Brand *et al.*, 2008] and reservoir systems [Lorrai *et al.*, 2010; McGinnis *et al.*, 2008]. Boreal freshwater ecosystems have at present, to the best of our knowledge, not been studied at all by means of subaqueous EC. Furthermore, we are not aware of any studies of near-bottom turbulence in small boreal lakes. This shortcoming makes it difficult to judge the applicability of gas fluxes derived from, e.g., sediment core incubations when establishing C budgets of small boreal lakes, which are both very numerous [Verpoorter *et al.*, 2014] and important in terms of C burial and CO₂ emission [Kortelainen *et al.*, 2004; Raymond *et al.*, 2013].

In this study we applied the EC method for the first time in a small boreal lake for the purpose of estimating in situ CO₂ fluxes from lake sediments. As current sensor technology only allows EC measurement of O₂ fluxes, we estimated CO₂ fluxes using the ratio between O₂ consumption and CO₂ production during in vitro sediment core incubations. We hypothesized that CO₂ flux estimated from in situ EC measurements is higher than CO₂ flux derived from sediment core incubation experiments, since enclosure techniques are unable to mimic natural turbulence conditions.

2. Methods

2.1. Site Description and General Instrumentation

This study was conducted in the small humic and mesotrophic Lake Erssjön situated in southwestern Sweden (58°23'N, 12°09'E; Figure 1), which has an area of 0.07 km² and a mean depth of 1.3 m (maximum 4.4 m). The lake is situated in the 750 ha Skogaryd Research Catchment (<http://www.fieldsites.se/en/>). Mean annual temperature and precipitation averaged over 30 years (1961–1990) were 6.9°C and 740 mm, respectively, measured at the closest weather station Vänersborg (www.smhi.se). Wind speed, wind direction, and air temperature over the lake were measured at a meteorological tower that was positioned at the lake shore adjacent to the deployment site as a part of another study [Podgrajsek *et al.*, 2015]. To obtain vertical temperature profiles of the lake, nine temperature sensors (U22 Water Temp Logger, Onset HOBO, Cape Cod, MA, USA) were deployed at the deepest spot of the lake in ~0.5 m intervals. From the temperature profiles, we calculated the maximum buoyancy frequency N_{\max} (s⁻¹) as

$$N_{\max} = [-(g/\rho_0)d\rho/dz]^{1/2} \quad (1)$$

where g is the gravitational acceleration, ρ_0 water density, and $d\rho/dz$ the maximum density gradient in the water column [Wuest and Lorke, 2003].

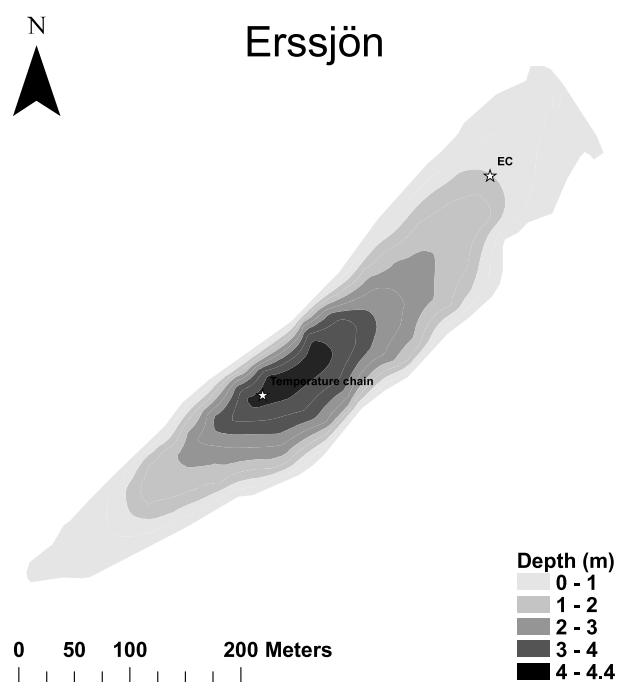


Figure 1. Bathymetric map over Lake Erssjön (58°23'N, 12°09'E). Eddy Correlation deployment and sediment core sampling were performed in the southwestern part of the lake at ~1–2 m, and the temperature chain was deployed at the deepest spot.

2.2. Eddy Correlation Measurements

The EC technique is a method for direct measurement of turbulent fluxes. Using Reynold's decomposition, the turbulent motions are represented as the deviations from a mean state (perturbations) and the turbulent fluxes as covariances of the perturbations. In the case of the vertical turbulent transport of an arbitrary scalar C , the vertical flux J is thus represented by the covariance between the vertical velocity (w) perturbation and the perturbation of the scalar:

$$\bar{J} = \overline{w'C'} \quad (2)$$

where the overbar represents the mean value and the prime denotes the perturbation. \bar{J} can be interpreted as the sediment flux when the solute transport equation fulfills steady state, horizontal homogeneity and when sources and sinks in the overlying water are negligible (see Brand *et al.* [2008] for a detailed discussion). In order to resolve all relevant time scales, fast-response measure-

ments of water velocity and the scalar of interest are necessary. The EC instrument used in this study (UNISENSE, Danmark) combines the 3-D water velocity measurement (horizontal u and v and vertical direction w) by an Acoustic Doppler Velocimeter (ADV, Vector, Nortek, USA) with a fast-response (0.3 s response time for a 90% response to concentration changes) Clark-type O_2 microelectrode for the flux calculation. More details on theory and instrumentation can be found in other papers that pioneered and evaluated the EC technique in the water such as Berg *et al.* [2003, 2009], Brand *et al.* [2008], and Lorrain *et al.* [2010].

2.2.1. Deployment and Data Analysis

We deployed the EC instrument at the northeastern side of the lake (Figure 1) at ~1.5 m water depth and measured at 8 Hz for three consecutive days in October 2013, with the instrument powered externally from the shore. The measurement volume was located at ~16 cm above the sediment. In September 2013, we also deployed only the ADV at the same site measuring at 64 Hz for ~8 days. Apart from the ADV and O_2 microelectrode, an O_2 optode (Aanderaa, 4330) logging at 0.1 Hz was used as a reference measurement to shift the mean O_2 signal from the microelectrode and to correct for sensor drift. Data analysis followed the procedures outlined in Lorrain *et al.* [2010] using 15 min window lengths and is summarized below.

We excluded data with $\bar{u} < 1 \text{ cm s}^{-1}$ (low turbulence) and series that showed electrical noise and jumps in the O_2 microelectrode. Despiking was performed on all signals according to Goring and Nikora [2002]. To correct for tilt in the ADV, a double coordinate rotation was applied by setting $\bar{v} = 0$ and $\bar{w} = 0$ [Lee *et al.*, 2004]. To correct for sensor placement of the O_2 microelectrode in relation to the ADV measuring volume and lag time due to the internal sensor electronics, we applied time-lag correction by stepwise shifting the ADV data against the O_2 microelectrode data to find the maximum point of correlation, which was ~2 s. To calculate fluctuations, we used 5 min running mean detrending on the 15 min window lengths. Spectral analysis was performed using the Welch's method [Welch, 1967]. From the ADV measurement we also calculated the Reynolds number:

$$Re = \frac{L\bar{u}}{\nu} \quad (3)$$

where L is the characteristic length scale (the distance of the ADV measurement volume to the sediment following Brand *et al.* [2008], corresponding to ~16 cm in our case) and ν the kinematic viscosity of water. By using a fixed L , our calculations are most likely conservative as in reality L changes over time and is most

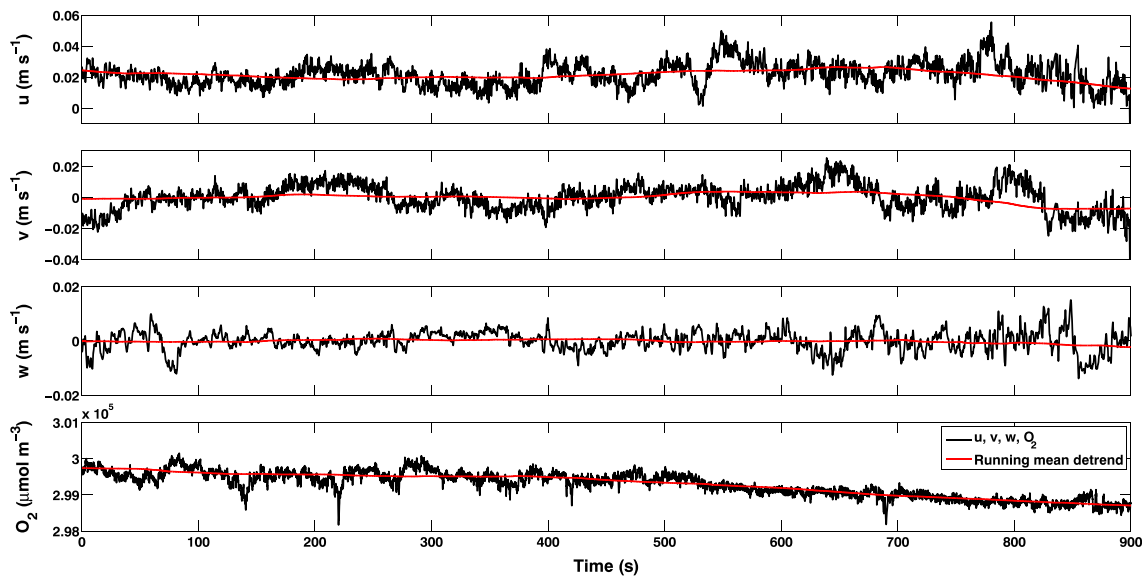


Figure 2. Time series for a 15 min segment for (first to third panels) the three velocity components u , v , and w and (fourth panel) O_2 concentration. Red lines represent the running mean for calculating turbulent fluctuations. The time series for O_2 concentration is showing raw data with no optode correction for sensor drift.

likely thicker as the lake is generally mixed throughout the 1.5 m water column at the measurement site (see further results in section 3.2). All data analysis was performed using MATLAB scripts.

For some time periods in the velocity signal, we saw vortex shedding from the O_2 microelectrode at ~ 1 Hz (e.g. time series in Figure 2, spectra Figure 3) thus, we applied a low-pass filter with 1.1–1.6 Hz as cutoff frequency. No flux is lost due to this procedure as the signal is weaker in the w spectra and it does not affect the flux calculations, since flux-contributing eddies start at ~ 0.8 – 0.9 Hz (Figure 4). While some of the disturbance is still visible in the u and v spectra, the filtering proves sufficient for the flux determination since the disturbance was above the cutoff frequency of the filter and therefore negligible in this case. For additional quality control, all individual spectra of u , v , w , and O_2 and cospectra as well as ogives for the flux $\overline{w'O_2'}$ were visually inspected.

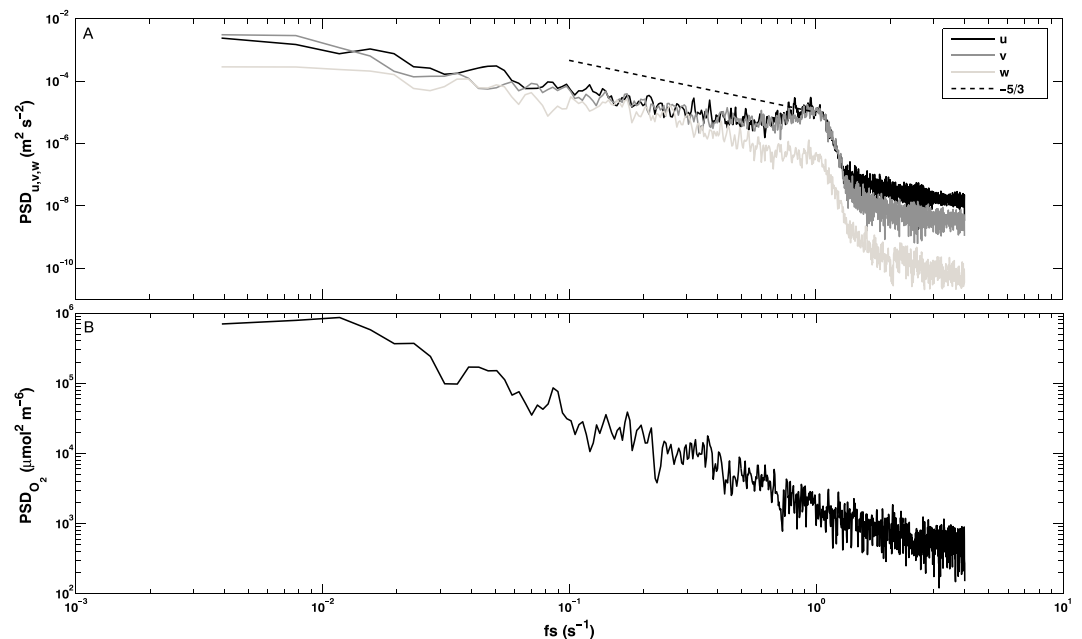


Figure 3. Spectral analysis for the same time period as in Figure 2, (a) power spectral density of low-pass filtered velocity u , v , and w with a $-5/3$ line showing the slope of the inertial subrange according to Kolmogorov's law and (b) for O_2 .

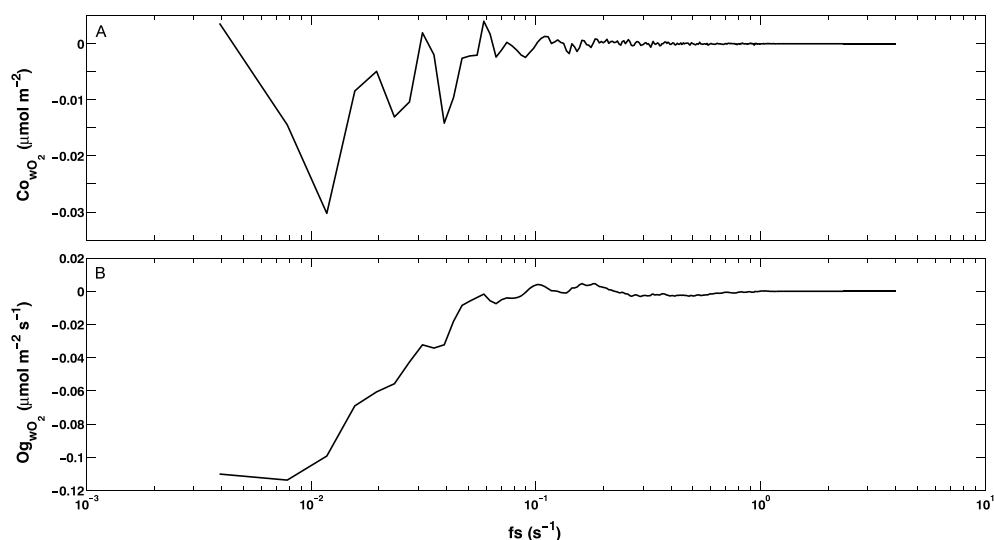


Figure 4. (a) Cospectral density for O₂ flux and (b) cumulative cospectral density (ogive) for O₂ flux, for the same time period as Figure 2.

2.3. Experimental Determination of CO₂ and O₂ Flux From Sediments

Six sediment samples were collected after the EC deployment nearby the measurement site with a gravity corer (UWITEC, Austria) from Lake Erssjön. Following the same protocol as Gudas *et al.* [2010], approximately 5 cm of top sediment with overlying water was transferred without any visible disturbance of sediment to polycarbonate cores (diameter 53 mm) used for the incubation experiment. The cores were kept open at in situ temperature (8°C) with the supernatant water mixing for 7 days to establish steady state between sediment respiration and gas flux to the water. Mixing the supernatant water was achieved by placing a floating stirring device (a buoyant 2 mL Eppendorf vial containing small magnets and construction foam) inside the cores and agitating by an axle with four magnetic arms rotating in the water bath outside the cores at ~40 rpm. This assured continuous mixing of the water column inside the incubation tubes, without resuspending the sediment. The samples were thereafter enclosed and incubated for ~3 days in the dark at constant temperature (8°C, corresponding to in situ temperature) in water chambers, and the water overlying the sediment was kept mixing at a constant rate using the magnetic stirring device. Dissolved O₂ concentration in the water overlying the sediment was monitored continuously during the incubation period with oxygen sensor spots (PreSens, model PSt3) and was approximately at in situ (~8.7 mg L⁻¹) concentration at incubation start and ~6 mg L⁻¹ at the end of incubation. In order to get a measure of biological CO₂ production that is independent of pH-related shifts in the carbonate equilibrium, we measured CO₂ production as the change of dissolved inorganic carbon concentration at the start and end of the incubation, using a Total Carbon Analyzer (Sievers 900). CO₂ flux reported in this study therefore refers to the flux of both gaseous CO₂ and its reaction production with water (HCO₃⁻ and CO₃²⁻). CO₂ flux was calculated in μmol m⁻² s⁻¹.

Additionally, we tested the effect of artificially induced mixing of the overlying water on measured gas exchange at the SWI. We incubated another set of sediment cores (sampled in late summer 2013 as a part of another study [Chmiel, 2015]) at three different mixing conditions of the overlying water by means of magnetic stirring: no stirring, intermediate stirring speed (stirring 1, ~20 rpm), and high stirring speed (stirring 2, ~40 rpm) (experiment performed twice with three replicates each). The high stirring speed was the highest rotation speed of the magnetic stirring device without any visible resuspension of sediment particles. In this experiment, solely O₂ concentration above the sediment was continuously monitored during the course of the experiment (~3 days).

2.4. In Situ CO₂ Flux Estimation

To estimate in situ CO₂ flux from O₂ flux measured by the EC, we used the apparent respiratory quotient (RQ) calculated from the sediment core incubation, which is the molar ratio of CO₂ consumed and O₂ produced [Granéli, 1979]:

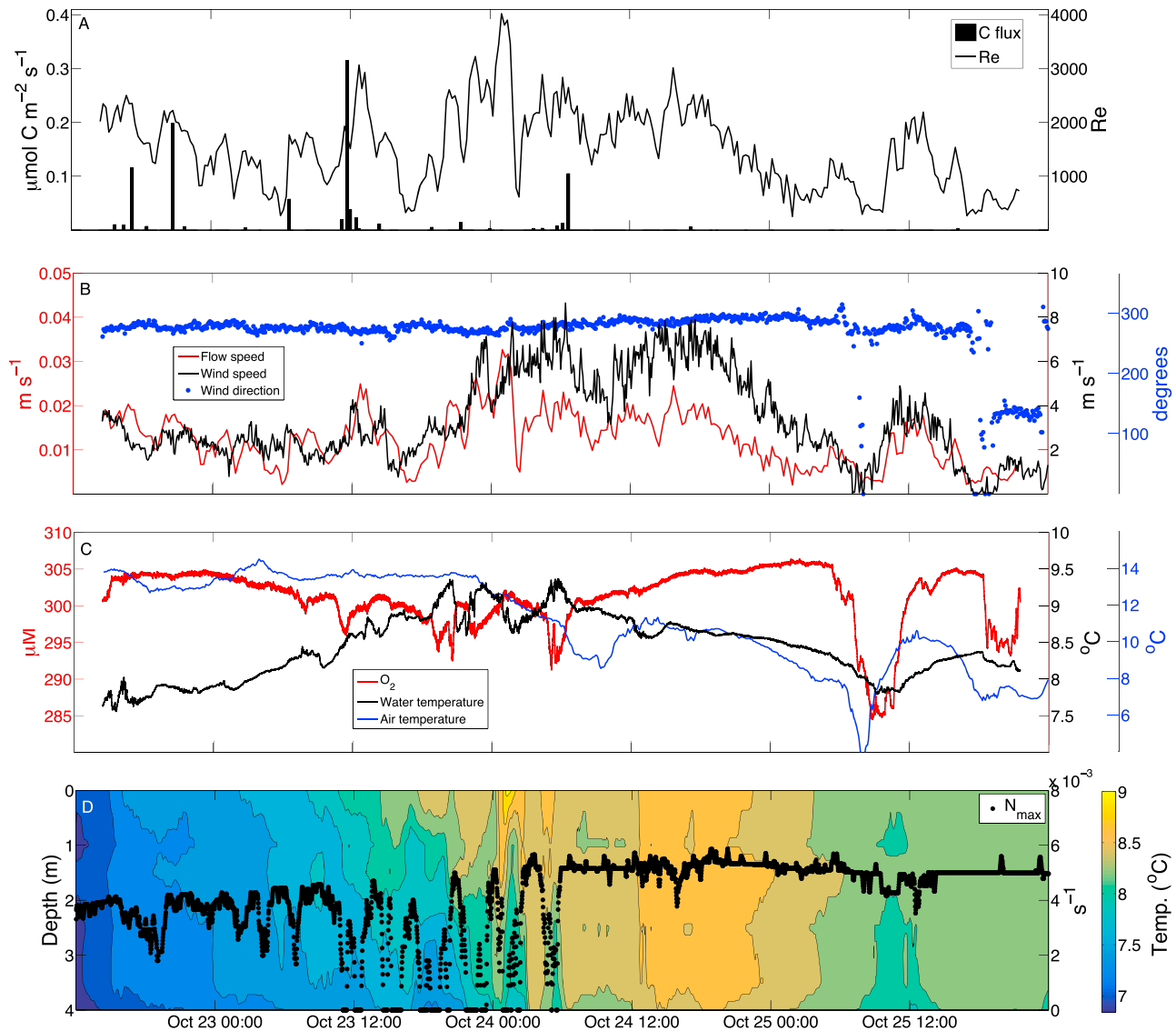


Figure 5. In Fig5C, the legend is missing the black line for water temperature on my version. Time series of (a) C flux ($\mu\text{mol m}^{-2} \text{s}^{-1}$) from Lake Erssjön sediment (where C flux = $\text{EC}_{\text{O}_2 \text{ flux}} \times \text{apparent RQ}$; see section 2.4) and Reynolds number; (b) horizontal flow speed (u), wind speed, and direction; (c) water temperature and O_2 measured by the low-frequency optode and air temperature; and (d) 0.2°C isotherms over depth (left y axis) and corresponding maximum buoyancy frequency N_{max} (right y axis) calculated from the temperature profile measured at the deepest spot of the lake for the measurement period of ~3 days.

$$\text{RQ} = \frac{\text{CO}_2 \text{ prod}}{\text{O}_2 \text{ cons.}} \quad (4)$$

Assuming that the apparent RQ observed in sediment core incubations also is valid for in situ gas fluxes, turbulent O_2 fluxes derived from the EC measurements were multiplied with the apparent RQ to calculate turbulent CO_2 flux across the SWI. Applying EC measurements with discrete sediment sampling has similarly been done by Yamamoto *et al.* [2015] to estimate alkalinity flux from coral reefs.

3. Results

3.1. O_2 Fluxes From Eddy Correlation Measurements

From the total data set of 3 days of EC measurements in Lake Erssjön, <5% were used for flux calculation. Most of the data excluded were periods of low horizontal mean flow ($\bar{u} < 1 \text{ cm s}^{-1}$) and jumps in the O_2 sensor. Several periods in the data set were excluded after visual inspection of the spectra and cospectra

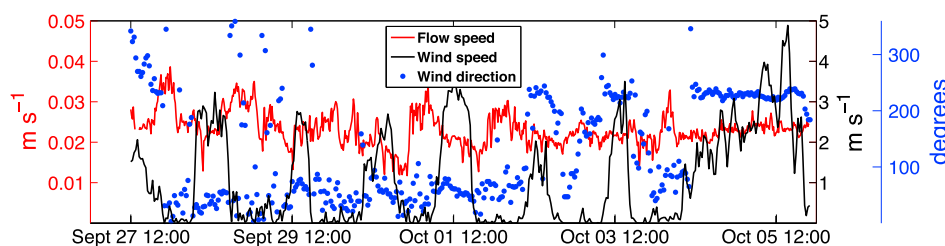


Figure 6. Horizontal flow speed (u) and wind speed and direction during a ~8 day measurement period (26 September to 5 October 2013) when only the ADV was deployed.

as well as ogives, removing cases that were apparently incongruent with theory. An accepted time series of u , v , w , and O_2 , and corresponding spectral analysis of w and O_2 , is shown in Figures 2 and 3 (displaying the expected $-5/3$ slope in all three velocity components). For the majority of the data set, individual 15 min periods displayed both positive and negative flux contribution in the low-frequency range of the wO_2 cospectra, the source most likely being mesoscale motions not scaling with local gradients. For these time periods we therefore set a cutoff frequency at ~ 0.1 Hz, thereby excluding flux contributions from lower frequencies.

Median O_2 uptake from Lake Erssjön sediment at the EC deployment site was $9.2 \times 10^{-3} \mu\text{mol } O_2 \text{ m}^{-2} \text{ s}^{-1}$ with a range of 2.2×10^{-3} – $2.9 \times 10^{-1} \mu\text{mol } O_2 \text{ m}^{-2} \text{ s}^{-1}$ where most fluxes were distributed on the low end (Figure 7 for C flux). We excluded a few cases with positive fluxes that were in the smallest range compared to the negative fluxes. We deem these as artifact as it is very unlikely for photosynthesis to occur to a greater extent than the heterotrophic respiration in the organic matter-rich sediment of our highly colored study lake, where at 1.3 m depth there is less than 1% of incident photosynthetically active radiation (PAR) (measured as part of another study [Groeneveld *et al.*, 2015]).

3.2. Experimental and Estimated In Situ CO_2 Fluxes

Experimentally determined fluxes during sediment core incubations were on average $3.9 \times 10^{-2} \mu\text{mol } O_2 \text{ m}^{-2} \text{ s}^{-1}$ (range 2.5×10^{-2} – $5.5 \times 10^{-2} \mu\text{mol } O_2 \text{ m}^{-2} \text{ s}^{-1}$) for O_2 and $3.8 \times 10^{-2} \mu\text{mol } C \text{ m}^{-2} \text{ s}^{-1}$ (range 3.4×10^{-2} – $4.3 \times 10^{-2} \mu\text{mol } C \text{ m}^{-2} \text{ s}^{-1}$) for CO_2 . This resulted in a mean apparent RQ of 1.1 (range 0.63–1.4) for Lake Erssjön sediment. Multiplying in situ O_2 fluxes measured by EC with the apparent RQ derived from sediment core incubations, in situ CO_2 fluxes were calculated (Figure 5a). Accordingly, median in situ CO_2 production (i.e., positive fluxes) was calculated to $1.0 \times 10^{-2} \mu\text{mol } C \text{ m}^{-2} \text{ s}^{-1}$ (range 2.4×10^{-3} – 3.2×10^{-1}).

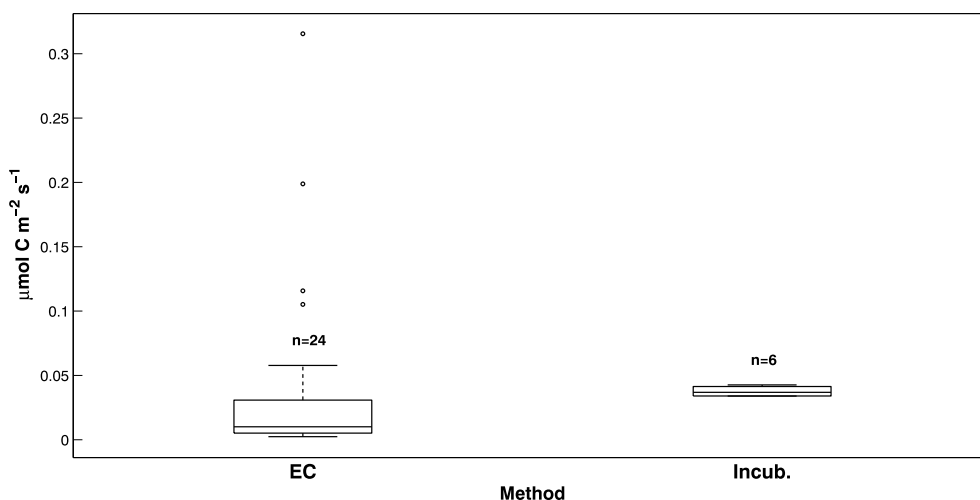


Figure 7. Boxplot comparing CO_2 flux obtained by EC measurements and experimental core incubations; n denotes sample size. Box shows median value and interquartile range. Samples are considered outliers above or below two standard deviations (2σ) from the mean. Whiskers show the adjacent value that is the most extreme data point not considered an outlier.

Increases in mean horizontal flow velocity \bar{u} (Figure 5b) seemed to coincide with increases in CO_2 flux (e.g., 23 October 12:00), but the many data gaps caused by the quality control prevented statistical analysis of any correlations between EC-derived gas fluxes and other variables. There is further no clear connection between the CO_2 flux and the Reynolds number (Figure 5a), a change in temperature, or O_2 at the deployment site (Figure 5c); yet again, the many data gaps in CO_2 flux make the regulation of CO_2 flux difficult to assess. However, during this period, \bar{u} was significantly and positively correlated to wind speed over the lake (Spearman's correlation coefficient = 0.75, $p < 0.01$) and water column stratification was weak with complete water column mixing at highest wind speed (Figure 5d). Moreover, during the time period when only the ADV was deployed (Figure 6, 27 September to 5 October), a positive relationship between wind speed and \bar{u} with a 10 h time lag was observed (Spearman's correlation coefficient = 0.25, $p < 0.01$, time series cross correlation analysis).

The maximum buoyancy frequency N_{max} (Figure 5d) was always lower than $5.9 \times 10^{-3} \text{ s}^{-1}$, and thereby well below the frequencies where we see the turbulent EC fluxes (Figure 4b, starting from $\sim 1.0 \times 10^{-2}$ to $1.0 \times 10^{-1} \text{ s}^{-1}$), indicating a spectral gap between turbulent length scales and potential internal waves. At some time points during the EC measurement period (Figure 5), we see indications of potential seiche-like movements in the water column (e.g., 24 October around 00:00, Figure 5d), where there is a downwelling of warm water followed shortly by an upwelling of colder water and then another downwelling followed by an upwelling, within a ~ 6 h period. Correspondingly, at the EC measurement site, colder temperatures are accompanied by an increase in O_2 concentration and subsequent warming of the water with lower O_2 concentration (Figure 5c). However, during this period of potential seiching, no EC fluxes could be calculated. For the following period (approximately 24 October 12:00 and onward), a concomitant cooling and later warming of both air and water at the EC measurement site could be observed.

3.3. Comparison of Gas Fluxes Derived From Eddy Correlation and Sediment Core Incubations

Median CO_2 fluxes derived from sediment core incubations were ~ 2 times higher than those derived from EC measurements (Figure 7, Kruskal-Wallis test, $p = 0.01$, $n = 25$). Only the highest EC-based CO_2 fluxes were of similar magnitude as the CO_2 fluxes derived from sediment core incubations. Note that in this incubation experiment, the water overlying the sediment was continuously mixed.

In another experiment, we tested if the difference between EC-derived and incubation-derived CO_2 flux (Figure 7) may be related to different turbulence regimes by manipulating artificial turbulence in sediment core incubation experiments. This experiment showed that the average O_2 consumption rate was about 20% higher if the overlying water was artificially mixed by a magnetic stirring device than if the overlying water was not mixed (Figure 8). Different magnetic stirring speeds (i.e., intermediate or high stirring speed) had no effect on observed O_2 uptake (Kruskal-Wallis test, $p = 0.8$, $n = 18$), but not stirring the overlying water in sediment incubation experiments resulted in significantly lower O_2 uptake than if the overlying water was stirred (Kruskal-Wallis test, $p = 0.04$, $n = 18$). The lowest observed O_2 uptake at nonstirring condition was lower by a factor of ~ 2 compared to the highest observed O_2 uptake at stirring condition. Experimental gas flux at no stirring of overlying water (Figure 8) is overlapping with EC-derived gas flux (Figures 5 and 7), but since the sediment cores for this experiment were sampled at another point of time and not at the EC measurement site, fluxes may not be directly comparable.

4. Discussion

4.1. Low In Situ Sediment-Water Gas Exchange

In this study we demonstrate that sediment-water gas exchange in a small boreal lake was on average smaller when measured in situ by EC than when derived from sediment core incubation experiments. Based on the first study to apply the EC method for estimating CO_2 fluxes from lake sediments, this finding suggests that bottom water turbulence strongly affects the contribution of sediment OC mineralization to lake CO_2 emissions, particularly in the many small boreal lakes that are typically wind sheltered by surrounding forest and exhibit low turbulence at the bottom due to near-surface thermoclines [Xenopoulos and Schindler, 2001].

Our estimates of median CO_2 flux from the EC method are about a factor of 2 smaller than median CO_2 flux estimated by the incubation experiments (Figure 7). Our core incubations return very similar rates as previously reported rates from core incubations of similar lakes [Algesten et al., 2005; Bergstrom et al., 2010;

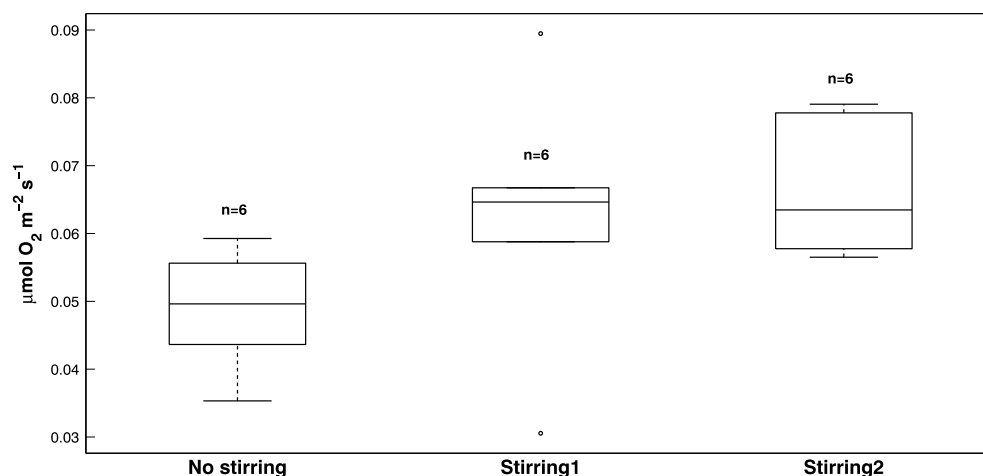


Figure 8. Boxplot of sediment O_2 consumption rate obtained from a core incubation experiment, with no stirring versus two different magnetic stirring levels; n denotes sample size.

Gudas et al., 2010]. This finding has implications for how sediment CO_2 fluxes have previously been studied and used in upscaling approaches and suggests that the sediment contribution to total lake CO_2 emission may currently be overestimated for small boreal lakes. To understand why this is the case, we must discuss how the two methods differ in what processes are measured.

The sediment cores are taken from a small area, then enclosed, subjected to artificial turbulence and incubated in the dark. The sediment core incubations therefore only estimate CO_2 flux from respiration, while the EC method measures the net CO_2 flux (photosynthesis – respiration). However, since our study lake is highly colored with $<1\%$ incident PAR at 1.3 m [Groeneweld et al., 2015], our EC measurement was at a depth where photosynthesis by benthic algae living on the sediment is most probably very limited. Therefore, it is unlikely that benthic photosynthetic activity, which can be important in clear-water lakes [Ask et al., 2009], contributes to the difference in CO_2 flux between EC measurements and core incubations.

Inducing artificial turbulence in the sediment cores is done to achieve a steady state between respiration in the sediment and flux to the water and to prevent measuring only diffusive flux from the sediment. However, core incubation experiments estimate rather a potential CO_2 flux, as opposed to the actual instantaneous CO_2 flux that is measured by the EC. The artificial turbulence induced in the experimental setup is intended to mimic natural conditions, i.e., nonstagnant bottom water, but apparently seems to create turbulence conditions that were rather high for the small study lake. Accordingly, the highest EC fluxes, which coincide with high \bar{u} and high wind speed (Figure 5), were of similar magnitude as the CO_2 fluxes derived from sediment core incubations under artificially induced mixing (Figure 7).

The effect of turbulence on gas fluxes across the SWI has been demonstrated by several studies of sediment O_2 uptake [Bryant et al., 2010; Lorke et al., 2003]. Studying micropores of O_2 during a full seiche cycle, Bryant et al. [2010] showed the effect of turbulence on the thickness of DBL and in turn the uptake of O_2 by the sediment, where the O_2 uptake increases with increasing turbulence as the thickness of the DBL decreases. During low turbulence, CO_2 and other reduced substances produced in respiration processes accumulate in sediment pore water rather than being released to bottom water via diffusion from the top layers of the sediment during these periods. Accumulated CO_2 can then subsequently be released periodically with higher turbulence [Brand et al., 2008; Bryant et al., 2010]. This effect of intermittent turbulent transport on solute fluxes was first investigated with EC measurements in seiche-driven lakes by Brand et al. [2008]. Accumulation and release of reduced substances may help to explain why we measured a smaller difference in gas flux in the incubation experiment with stirring and no stirring of the water (Figure 8), as compared to the variability in EC-derived gas fluxes. By stirring the sediment cores during preincubation prior to the actual incubation experiment, we aimed to achieve steady state between CO_2 production and CO_2 flux, and consequently, any accumulated CO_2 was released prior to the experiment. Hence, varying mixing regime in the overlying water had a comparatively smaller effect on observed gas flux since there was little accumulated

CO₂ to be liberated from the sediment by high turbulence. Conversely, while in situ, extended periods of low turbulence are more likely to have resulted in the accumulation of CO₂ and hence high flux at instances when turbulence was high enough to allow for the release of the accumulated CO₂ (Figure 5). Following this argumentation, one may conclude that if integrated over longer periods of time, in situ gas exchange will become more similar to gas exchange derived from core incubations, since events of higher turbulence and gas flux may compensate for periods of very low gas flux. However, during periods of low gas exchange, the supply of O₂ to aerobic respiration is very limited which may result in reduced overall CO₂ production. The magnitude of overall OC mineralization is strongly limited by O₂ supply, particularly in sediments dominated by terrestrial OC such as in our study Lake Erssjön (sediment C:N ~ 14: *Chmiel* [2015]) [*Rasmussen and Jorgensen*, 1992; *Sobek et al.*, 2009]. Hence, it is likely that extended periods of low gas exchange at the SWI limit the overall extent of sediment OC mineralization and CO₂ production by reducing O₂ supply to aerobic respiration. This may contribute to the lower observed EC gas exchange fluxes relative to core incubation-derived gas fluxes. In addition, the potential initially high gas fluxes at the onset of turbulence may be missed by the EC, since these data are highly instationary and do not fulfill the EC data quality criteria.

4.2. In Situ Measurement of Sediment-Water Gas Exchange in a Small Boreal Lake

While our study was conducted in a lake with low water velocity and therefore low flux (median flux $1.0 \times 10^{-2} \mu\text{mol O}_2 \text{ m}^{-2} \text{ s}^{-1}$ at a \bar{u} of 1.3 cm s^{-1}), most studies applying the EC method have been conducted at high current velocities \bar{u} . For example, *McGinnis et al.* [2011] presented an average flux of $0.46 \mu\text{mol O}_2 \text{ m}^{-2} \text{ s}^{-1}$ for a run-of-the-river reservoir with $\bar{u} > 16 \text{ cm s}^{-1}$. Even fewer studies have been conducted in lakes with low \bar{u} . *Brand et al.* [2008] observed turbulent fluxes at \bar{u} above 2 cm s^{-1} and Re above ~ 1700 with a corresponding minimum flux of $4 \times 10^{-2} \mu\text{mol O}_2 \text{ m}^{-2} \text{ s}^{-1}$. This is similar to our measurements, as we observed turbulent fluxes above \bar{u} of 1.0 cm s^{-1} and $Re \sim 1300$. However, the Re is not directly comparable since they used an L (characteristic length scale, measurement height of the ADV to the sediment) of 10 cm while we used 16 cm (equation (3)). Considering the low \bar{u} in our study lake, the measured O₂ fluxes reported here compare well to earlier results that under low turbulence levels, the EC fluxes do not reflect the full sediment uptake dynamics [*Brand et al.*, 2008]. It is important to point out that the EC method measures turbulent gas exchange; in apparently low turbulence systems such as our study lake, diffusive gas exchange may be one of the main pathways of solute exchange at the SWI.

Advection of water masses with different O₂ concentrations past the EC sensors has the potential to result in the calculation of fluxes that are not related to gas exchange at the SWI of the measurement site. During the time period of the EC measurements, we observed time points with potentially seiche-like water movements in the lake (Figure 5d, e.g., starting 24 October 00:00). These downwelling and upwelling water movements were observed at the deepest spot in the middle of the lake and might cause the converse water motions at the EC measurement site. This may be indicative of near-bottom currents driven by internal waves. With internal waves, an upwelling of colder water can cause advection of comparatively O₂-poor water close to the sediment bottom and may thereby enhance the calculated EC fluxes. However, at this time point, very few EC fluxes were calculated. Additionally, estimated EC fluxes were relatively low. Similarly, around 25 October 12:00 there are indications of advection where the water is getting colder and O₂ is dropping (Figure 5c), even if there is also a decrease in air temperature that might correspond to this change. Also during this time, \bar{u} was too low for any turbulent fluxes to be observed by the EC. At the time points when we observed the highest fluxes (around 23 October 12:00), there were only small changes in O₂ concentration and no cooling of water at the EC deployment site (Figure 5c) which suggests that advection due to upwelling is absent during this time. However, since we do not have any measurement of horizontal or vertical O₂ gradients, we cannot rule out the possibility that advection may have affected and enhanced the fluxes presented here. If so, the contribution of gas exchange at the SWI to the observed fluxes would be even smaller, reinforcing our conclusion that gas exchange at the SWI is low in the studied lake.

The EC method has further been evaluated and compared to other methods for estimating O₂ flux from sediments (mostly marine) such as chambers [*Berg et al.*, 2013], O₂ microprofiles in combination with

chambers [Berg *et al.*, 2009], and microprofiles and core incubations [Glud *et al.*, 2010]. These studies have shown both good agreement among the methods and a higher estimation of EC flux compared to other methods. These studies have, however, been conducted in very different systems with much higher bottom current speeds than in our study, corresponding to higher levels of turbulence. While there are challenges in measuring EC fluxes in small lakes with low \bar{u} , our EC fluxes are similar to previously reported EC fluxes in lakes with low \bar{u} , and we observed a similar pattern of intermittency in flux during a strong wind event, as Brand *et al.* [2008] observed during seiche events. Also, our EC fluxes are on the same order of magnitude as fluxes derived from sediment core incubations.

4.3. Applicability to Other Aquatic Systems

During our EC deployment we had periods of relatively high wind speeds, ranging from 0 to 8.6 m s^{-1} with a mean 3.3 m s^{-1} , which correlated with horizontal water velocity \bar{u} (Figure 5b). During the year 2013, less than 1% of the wind speed was above 7 m s^{-1} . Even though wind speed during our EC campaign was reaching one of the highest levels measured during the entire year (8.6 m s^{-1}), corresponding \bar{u} reached only up to 3.3 cm s^{-1} during the EC campaign (Figure 5b). Apart from the measurement series of EC fluxes (Figure 5), we also present a measurement series of near-bottom water velocity measurements by ADV at the same measurement site at a different time period (27 September to 5 October; Figure 6). During this time period, we observed a 10 h time lag between wind and water velocity (Figure 6, e.g., wind peaks 27 September to 3 October ~12:00 with ~10 h lag in flow peaks), a more regular pattern than during the time period of EC fluxes and is indicative of seiche-driven water motions. Seiche-driven bottom water turbulence has previously been found to be important for sediment-water solute exchange [Brand *et al.*, 2008; Lorke *et al.*, 2003]. However, in Lake Erssjön, not even during a longer period of a seiche event did the bottom water velocity exceed 3.9 cm s^{-1} (Figure 6). Hence, turbulent gas fluxes in Lake Erssjön during seiche events can be expected to be similar to those observed during strong wind events (when peak \bar{u} was 3.3 m s^{-1} ; Figure 5b). This strongly suggests that in our study lake, strong wind events with neither short-term seiche (Figure 5) nor regular seiche (Figure 6) induce strong bottom water velocity, and thus, sediment-water fluxes are probably low.

Most small boreal lakes have similarly organic-rich sediments (OC: 12–39%) [Algesten *et al.*, 2005; Pajunen, 2000] as Lake Erssjön (OC: 22%), and most of them are wind sheltered by the surrounding forest. It is therefore likely that the in situ sediment gas flux conditions observed in Lake Erssjön are applicable to other small boreal lakes that are mostly situated in fairly wind-sheltered areas in the forest. However, since there are no systematic turbulence measurements in small boreal lakes in the literature, the findings of low gas exchange at the SWI in Lake Erssjön cannot be generalized. For future studies, we recommend to aim at investigating turbulence when measuring gas exchange at the SWI. We also recommend that care should be taken when upscaling fluxes estimated by sediment core incubation experiments to small boreal lakes, as they may overestimate in situ fluxes because in situ bottom water mixing is probably lower than artificially controlled mixing in sediment core incubations.

5. Conclusions

This is the first study that estimates in situ CO_2 flux from lake sediments by the combining the EC method with sediment core incubation experiments, and we conclude that EC-derived fluxes were smaller than if estimated by sediment core incubation experiments. We attribute the difference between methods to (a) artificial water mixing in sediment core incubation experiments being stronger than in situ bottom water current velocities and (b) low O_2 supply to aerobic heterotrophic microbes during extended periods of low current velocities inhibiting overall OC mineralization and CO_2 production. Applying the EC method in small lakes is challenging and may result in a relatively small set of useable data due to the high intermittency of turbulence-driven mixing in these systems. Most of the lakes in the boreal region are small and fairly wind sheltered, and we propose that our results found in Lake Erssjön are probably applicable to similar type of lakes. Care should be taken when upscaling sediment CO_2 flux derived from sediment core incubation experiments to entire basins of small lakes, as considerable overestimates may result.

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