Surface CO_2 Gradients Challenge Conventional CO_2 Emission Quantification in Lentic Water Bodies under Calm Conditions.

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Abstract. Lakes are hotspots of inland carbon cycling and are important sources of greenhouse gases (GHGs), such as carbon dioxide (CO_2). The significant role of CO_2 in the global carbon cycle makes quantifying its emission from various ecosystems, including lakes and reservoirs, important for developing strategies to mitigate climate change. The thin boundary layer (TBL) method is a common approach to calculate CO_2 fluxes from CO_2 measurements in both water and air, and wind speed. However, one assumption for the TBL method is a homogeneous CO_2 concentration between the measurement depth and the water surface, where gas exchange takes place. This assumption might not be true under calm conditions, when microstratification below the surface slows vertical exchange of gases. We used a floating outdoor laboratory to monitor CO_2 concentrations in 5 cm and 25 cm depth, CO_2 concentration in the air, wind speed, and water temperature profiles for one week in Bautzen Reservoir, Germany. While we found homogeneous CO_2 concentrations in the two depths during wind speeds above 3 m s^{-1}, there was a vertical gradient observed during wind still nights. The concentrations observed temporally ranged from undersaturation to supersaturation in 25 cm and 5 cm, respectively. Fluxes calculated from the measured concentrations therefore would change from negative to positive, depending on the measurement depth. Simultaneous Eddy Covariance measurements showed that even the measurements close to the surface underestimated the actual CO_2 concentration. Oxygen measurements support our hypothesis that plankton respiration~~al~~ at the water surface causes a periodic CO_2 concentration gradient from the surface to the underlying water. Until now, the depth of CO_2 measurements has not been questioned, as long as measurements were done in the upper mixed layer and close to the surface. Our results provide evidence that representative measurements of CO_2 in the water strongly depend on depth and time of measurements.

1 Introduction

Lakes are hotspots of inland carbon cycling and are important sources of greenhouse gases (GHGs), such as carbon dioxide (CO_2). The significant role of CO_2 in global warming and climate dynamics makes quantifying its emission from various ecosystems, including lakes and reservoirs, important for developing strategies to combat climate change. Proper measurement of CO_2 emissions from lakes is the basis for robust global CO_2 emission quantification (Raymond et al., 2013; Lauerwald et al., 2023).

Eddy Covariance (EC) is one of the most advanced methods to quantify GHG fluxes between water and atmosphere, as it provides direct measurements covering large footprint areas at high temporal resolution (Aubinet et al., 2012)~~The state-of-the-art method to quantify GHG fluxes between water and atmosphere is Eddy Covariance (EC) (Aubinet et al., 2012). Measurements by EC represent direct measurements covering large footprint areas at high temporal resolution~~. However, EC systems require a large homogeneous ~~lake~~ surface area to ensure accurate flux measurements. The limitation is increased by its costs and requirements for maintenance. Thus, there is a restricted number of EC sites on lakes worldwide (Golub et al., 2023). EC measures to total gas flux between water and atmosphere, including diffusion as well as ebullition. However, in contrast to methane, which is often emitted by gas bubbles, CO_2 fluxes at the water surface are nearly exclusively driven by diffusion. The by far most used approach to quantify diffusive aquatic GHG fluxes is the Thin Boundary Layer approach (TBL,

Lauerwald et al., 2023), also referred to as flux gradient method. The TBL approach, in contrast to EC, is an indirect measurement to quantify diffusive gas exchange across the water-atmosphere interface. The TBL flux is derived from the concentration gradient between the water ($c_{CO_2\ water}$) and the theoretical concentration at equilibrium with the air ($c_{CO_2\ equilibrium}$) multiplied by the gas exchange velocity (k, equation 1).

$$F_{CO_2} = k * (c_{CO_2\ water} - c_{CO_2\ equilibrium}), \quad (1)$$

This method is much simpler than EC, because it only requires concentration measurements in both the water and the atmosphere. The gas exchange velocity can be estimated from meteorological data, typically wind speed and eventually temperature or fetch, using empirical models (Cole and Caraco, 1998; McGillis et al., 2004; MacIntyre et al., 2010; Vachon and Prairie, 2013; MacIntyre et al., 2021). The temporal resolution can be comparable to the EC method when using submerged CO_2 probes. However, for both methods, uncertainties arise from situations involving very low wind speeds.

When there is no wind, EC systems cannot measure at all, because the method relies on air movements (Aubinet et al., 2012; Podgrajsek et al., 2014). But also, the TBL approach encounters difficulties under wind still conditions. While wind is the primary driver of gas transfer velocity and surface turbulence in large water bodies, its influence decreases under calm conditions, where factors such as the mixed layer depth (Rutgersson et al., 2008), surface cooling-induced convection (MacIntyre et al., 2010; Heiskanen et al., 2014; Andersson et al., 2017; MacIntyre et al., 2010) and the lake's morphometry (Schilder et al., 2013) gain prominence in controlling gas exchange velocities. For example, Podgrajsek et al. (2015) showed that high CO_2 fluxes in calm conditions can be explained by convective cooling. Further, most parametrizations of *k* rely heavily on wind speed and assume a zero intercept, leading to significant uncertainties under calm conditions. While factors like turbulence and convection are often considered in the context of their influence on *k* only, they also play a critical role on near-surface CO_2 gradients. This may lead to systematic underestimation of k under wind still conditions. Although studies to improve *k* models exist (Cole et al., 2010; Crusius and Wanninkhof, 2003; MacIntyre et al., 2010; Vachon and Prairie, 2013), to our knowledge, the effect of uncertainty in $c_{CO_2 Water}$ measurements on the TBL approach received little attention.

In calm conditions, the absence of wind significantly reduces turbulence, leading to surface microstratification. This microstratification creates distinct layers within the mixed layer, each with varying temperatures and solute concentrations, including CO_2 (Åberg et al., 2010). Specifically, microstratification may result in dissolved gas gradients near the water's surface, suggesting that the actual layer engaging in atmospheric exchange becomes very thin. This challenges the assumption of the TBL approach that water just below the surface accurately represents the layer of gas exchange.

The critical layer of diffusive gas exchange is the surface microlayer (SML). The SML is the interface between the water and the atmosphere and is characterized by high biological activity and physical processes that affect the interaction with the atmosphere (Gladyshev, 2002; Wurl et al., 2011). The SML is known for its enriched concentrations of phytoplankton, organic and inorganic solutes, and particles (Hardy, 1982). These components define the SML as a distinct layer situated between the atmosphere and the underlying water. In the ocean, surfactants in the SML were found to reduce the diffusive gas exchange with the atmosphere by 32 % (Pereira et al., 2018). Mustaffa et al. (2020) found a reduction of fluxes of 62 % in the presence

of natural slicks. Under calm conditions, the SML thickens, becoming even more crucial for the diffusive exchange of gases (Rahlff et al., 2017). However, despite its importance, there remains a gap in our understanding of the interactions between the atmosphere, the SML, and the water beneath, particularly in freshwater environments.

Dynamics in gas exchange between the epilimnion, the surface layer and the atmosphere could lead to systematic uncertainty in the quantification of the surface gas concentration, as samples might be collected from depths that do not accurately reflect the conditions of the water-atmosphere interface. In a recent study, Rudberg et al. (2024) explored how spatial and temporal differences affect the influence of k and c_{CO2} on the variability of F_{CO2}. By deploying a floating chamber over several hours, the gas concentration in the chamber equilibrated with the gas concentration in the surface water and enabled the quantification of the surface CO_2 concentration. With this approach they demonstrated that over periods of 1-9 days, c_{CO2} contributed more to F_{CO2} variability than k. This finding emphasizes the need for precise c_{CO2} measurements when estimating fluxes using models. Similar research has been conducted in the ocean. Although CO_2 samples for gradient-based flux models are usually collected a few meters below the water surface, Calleja et al. (2013) found significant gradients between depths of 5-8 meters. Hari et al. (2008) found different CO_2 concentrations in 0.1 m and 0.5 m depth while investigating a new method for CO_2 measurements. However, while these differences are attributed to varied biological activities in the different depths, there is a lack of knowledge about the formation and characteristics of such gradients, especially with regard to the SML and diffusive gas exchange with the atmosphere.

To better understand the importance of vertical CO_2 gradients at the water-atmosphere interface, we conducted an extensive field experiment in a eutrophic reservoir. Our study is based on two key hypotheses:

1. Temporal gradients of CO_2 concentrations are present close to the water surface, and
2. These gradients are influenced by meteorological factors, such as wind.

To assess the effect of an eventual surface CO_2 gradient on CO_2 fluxes we compared fluxes calculated by the TBL approach with fluxes measured by EC during the same period.

2 Materials and methods

2.1 Study site

Our study site Bautzen reservoir in Germany (51.218 °N, 14.466 °E)is a dimictic reservoir, High wind exposure, a relatively large surface area (533 ha) as well as a circular shape, and the shallow depth (mean 7.4 m, maximum 13.5 m) result in a weak stratification during summer in some years (Benndorf, 1995; Spank et al., 2023). The eutrophic to hypereutrophic reservoir (Kerimoglu and Rinke, 2013) serves for flow regulation of river Spree – the main river of Berlin. A preceding study showed CO_2 uptake of −9.8 g C m^{-2} and −71.0 g C m^{-2} during the ice-free seasons of 2018 and 2019, and CH_4 fluxes of 24.0 g C m^{-2} and 23.2 g C m^{-2}, respectively (Spank et al., 2023).

2.2 Meteorological field observatory

A floating outdoor laboratory was operated to continuously observe the mass- and energy exchange between the water surface and the atmosphere. A detailed description of the floating outdoor laboratory and its instrumentation can be found in Spank et al. (2020, 2023, 2024). The floating outdoor laboratory provided reference data of the carbon dioxide flux (F_{CO2}) between the water surface and the atmosphere as well as data of wind speed (U), air temperature (T_a), relative air humidity (RH), air pressure (p_a), solar radiation (R_g) and water temperature (T_w) in a temporal resolution of 30 minutes. In addition, the floating outdoor laboratory served as a carrier for the devices used during the measurement campaigns. The measurement height of meteorological sensors was 2 m above the water surface in accordance with international standards. W~~The w~~ater temperature ~~thermistor chain measured~~ ~~(~~T_w) was ~~-~~measured at depths of 0.25, 0.50, 0.75, 1.0, 1.5, 2.0, 2.5, 3.0, 4.0, 5.0, 6.0, 8.0, 10.0, 12.5, and 15.0 m. The eddy covariance measuring system, which provides F_{CO2}, was instrumented according to standards and guidelines given by Lee et al. (2004), Foken and Mauder (2024), Aubinet et al. (2012) and Burba (2013). ~~The~~ EC post processing was based on the methodologies of Carbo Europe (Aubinet et al., 1999) and ICOS (Sabbatini et al., 2018) ~~and had been performed~~ utilizing the software EddyPro 7.0.8 (LI-COR Biosciences 2023). T~~However, t~~he special site conditions and the platform movement were taken into account and addressed by a ~~special~~ correction of the sensor misalignment beforehand (Spank et al., 2020, 2023). The EC data are representative for the pelagic zone of Bautzen reservoir. In particular, ~~carefully~~ a ~~performed~~ footprint analyses proved that effects and disturbances from surrounding terrestrial sites can ~~be~~ almost completely be ruled out (Spank et al., 2023)~~(Spank et al. 2023). The EC post processing was based on the methodologies of Carbo Europe (Aubinet et al., 1999) and ICOS (Sabbatini et al., 2018) and had been performed utilizing the software EddyPro 7.0.8 (LI-COR Biosciences 2023). The special measurement condition on a floating platform required an upstream correction of the sensor misalignment (Spank et al., 2020, 2023)~~. The correctness and quality of data were ~~carefully~~ checked using the methodology of ~~(~~Foken and Wichura~~,~~ ~~(~~1996)~~Foken and Wichura (1996)~~. Quality features, i.e., stationarity and turbulence characteristics, were computed for the individual 30-minute data records and classified according to the nine-step classification scheme of ~~(~~Mauder ~~-~~et ~~-~~al.~~,~~ ~~(~~2006)~~Mauder et al. (2006)~~, where 1 is best and 9 is worst. Only 30-~~minute~~ data records were considered that fulfilled criteria for quality levels 1-6. In addition, the random sampling error was calculated according to ~~(~~Finkelstein and Sims~~,~~ ~~(~~2001)~~Finkelstein and Sims (2001)~~ and used to capture the non-systematic uncertainty of F_{CO2} and F_{CH4}. Further information on the uncertainty analysis and the analysis of systematic measurement uncertainties in the EC data can be found in~~ –~~ ~~(~~Spank et al.~~,~~ ~~(~~2024)~~Spank et al. (2024)~~. ~~Although the~~While this study ~~here addresses~~focusses on low wind situations ~~in~~where ~~which~~ EC measurements ~~could~~can ~~be~~ potentially be biased due to a lack of turbulence and gustiness, no such issues were ~~not~~ observed ~~in~~during the investigated period ~~investigated here~~. On the contrary, all flux data exhibited sufficient turbulence and stationarity. This observation was also confirmed by spectral analyses ~~performed in parallel~~, which showed clearly developed power and cospectra.

2.3 Dissolved Gases

To detect eventually occurring dissolved CO_2 concentration gradients at the water surface, we deployed two CO_2 probes (Contros HydroC, -4H- JENA engineering, Germany) at different depth from September 18 to September 25 in 2022 (hereinafter referred to as *study period*). These probes, featuring diffusive membranes with a diameter of 8 cm, were positioned so that the central points of their membranes were situated at depths of 0.05 m and 0.25 m, respectively. We used a frame that consisted of two aluminium bars, which were connected by two cross bars in the middle. Aluminium extensions on the cross bars were used to mount the CO_2 probes horizontally and allow adjustments of the measurement depths. Buoyancy floats in the four corners were used to make the frame float below the surface (Figure S 1). The CO_2 probes were powered by 24 VDC provided by the floating platform. To maintain the integrity of surface water stratification, we left the membranes of the CO_2 probes uncovered. CO_2 was measured every minute and data was logged internally by the probes. The probes performed internal zero baseline corrections once a day.

CO_2 probe performance was validated before deployment. After deployment, internal data required post processing (as described in Fietzek et al., 2014) because the measurements were out of the factory calibration range. In brief, the internal zero measurements using CO_2 absorbents generate frequent calibration points for $pCO_2 = 0$. Daily zero measurements were used to calibrate signal outputs between the last and the next zero measurement. Finally, a modified polynomial function was used to determine corrected CO_2 concentrations (Supplement, S1).

The equilibrium concentration for CO_2 was derived from the EC CO_2 analyser (LI-7200, LI-COR Environmental, Inc., Lincoln, NE, USA).

Oxygen concentrations were measured every 15 minutes by optical O_2 loggers. Surface O_2 concentration at 0.05 m depth was measured with a miniDOT logger equipped with a miniWIPER (Precision Measurement Engineering, USA, wiping frequency 12 hours) mounted at the same depth as the surface CO_2 probe. A D-Opto oxygen logger (ZebraTech, Nelson, NZ) was installed in the bottom water at 1 m above the sediment using a separate mooring. Both oxygen sensors were calibrated using a two-point calibration at 0 % and 100 % oxygen saturation and corrected for potential drift.

2.4 Data and Statistics analysis

The platform data was prepared and exported in Python. For this study, the platform data, CO_2 probe data and oxygen data were compiled in R (R version 4.4.1, R Core Team, 2024). All subsequent analyses, statistical computations, and visualizations were performed in R (Supplement, S2). CO_2 and oxygen measurements were averaged over 30-minute periods. Gas fluxes were calculated from in situ water and air p_{CO2} and k (calculated from U_{10} after (Cole and Caraco, (1998)) ~~(Cole and Caraco, 1998b)~~using equation 1.

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3 Results

In 2022, Bautzen reservoir was thermally stratified from the beginning of May. During the stratified period (mean mixed layer depth = 4.4 m), the maximum T_w at the surface was 30 °C, while T_w above the ground reached a maximum of 13 °C (Figure A 1). On September 9th 2022, a thunderstorm with strong winds hit the Bautzen region, leading to a shutdown and subsequent 5-day outage of the measurement platform. This storm also marked the end of the stratified season and started mixing of the reservoir. On September 18th, which marks the beginning of our extensive GHG measurements, T_w was 16 °C at both the surface and bottom. The oxygen C_{O2} was 9.1 mg L^{-1} at both the surface and the bottom.

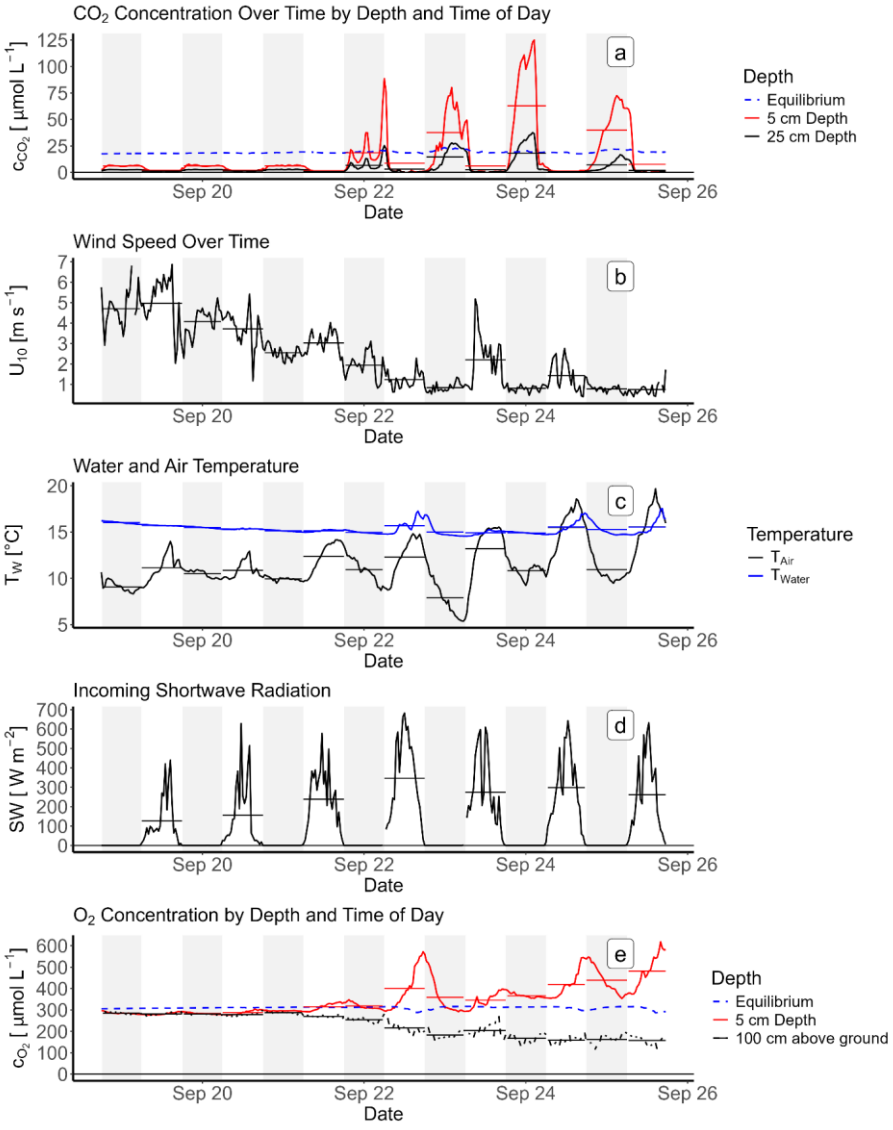
CO2 Concentration Over Time by Depth and Time of Day
a
Depth
Equilibrium
5 cm Depth
25 cm Depth
Wind Speed Over Time
b
Water and Air Temperature
c
Temperature
Incoming Shortwave Radiation
d
O2 Concentration by Depth and Time of Day
e
100 cm above ground
Sep 20
Sep 22
Sep 24
Sep 26
Date

Figure 1: CO_2 concentrations (a) measured at 0.05 m and 0.25 m depth (red and black line, respectively), and equilibrium concentration with the atmosphere (blue dashed line). Wind speed at 10 m height (U_{10}, b), Water temperature at 0.25 m depth + air temperature (c), and incoming short-wave radiation (SW, d). Oxygen concentrations (e) at 0.05 m depth and at 1 m above ground (red and black line, respectively), as well as equilibrium concentration with the atmosphere (blue dashed line). In all plots, horizontal bars show mean values of days and nights and grey shading indicates night-time.

Our measurement period can be divided into two parts with differing weather conditions. The first period from September 19th to 21st was windy with U_{10} mostly above 3 m s^{-1}. In contrast, during the second half of our sampling period, wind speeds were mostly below 3 m s^{-1}, with 63 % of those times even falling below 1 m s^{-1} (Figure 1 b).

The CO_2 concentrations near the water surface showed a fundamentally different behaviour during those two periods (Figure 1 a). During the windy period, CO_2 was permanently undersaturated and just above zero. In contrast, high CO_2 concentrations up to 125 µmol L^{-1} were observed during the calm period. Notably, nightly CO_2 concentrations in both depths were ten to twenty times higher during the calm period compared to the windy period. Starting on September 22nd, nightly CO_2 concentrations exceeded the equilibration concentration, leading to temporary supersaturation in calm nights. While the nightly mean CO_2 concentration at 5 cm depth exceeded the equilibration concentration in the nights of September 23rd to 26th, the mean concentration at 25 cm depth never surpassed equilibrium. The CO_2 concentration at the water surface showed a consistent diurnal pattern during the entire study period. At night, the CO_2 concentration at 5 cm depth was generally higher than at 25 cm depth. Thus, a gradient of CO_2 concentration near the water surface developed every night of our study period. In contrast, no such gradient was observed during the day. At both depths, CO_2 concentrations increased with the disappearance of short-wave radiation at sunset and decreased after sunrise again (Figure 2d).

The oxygen concentration patterns also differed between the two periods. During the windy period, O_2 concentrations at both 5 cm depth and at the bottom were slightly undersaturated. When the water column started stratifying again (Figure 1 e), O_2 concentrations at the two depths started to diverge. The O_2 concentration at the bottom continuously decreased over the rest of the sampling period. The concentration at 5 cm increased to supersaturation, showing clear diurnal patterns of oxygen production and consumption (Figure 1 e).

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Mean air temperatures (T_a) ranged between 9 °C and 15 °C, with distinct diurnal patterns. The water temperature at 25 cm depth (T_{w25}) decreased slightly from 16 °C to 15 °C during the measurement period, except on September 23rd, 24th, and 25th, when T_{w25} increased by 1 °C during the day (Figure 1 C). During the days of September 23rd, 24th, and 25th, T_a was higher than the water temperature. On all other days and nights, T_{w25} consistently remained higher than the air temperature.

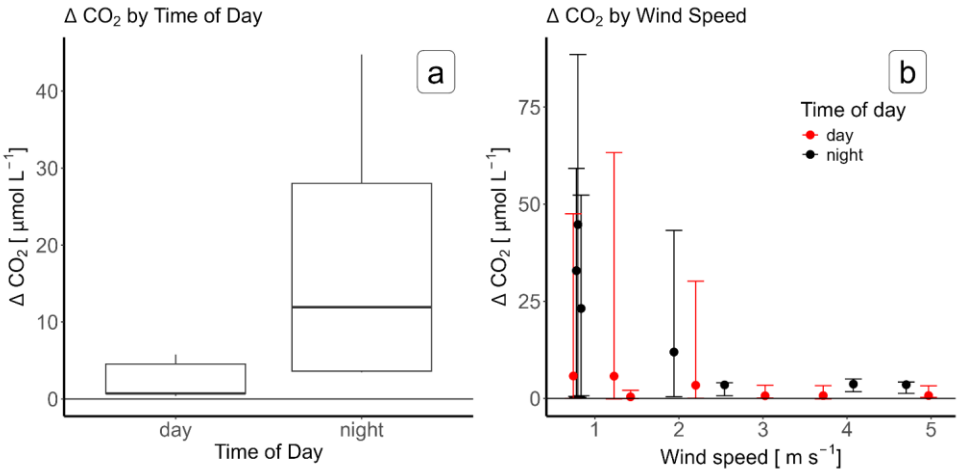

Figure 2: Dependence of the absolute difference in CO_2 concentration between 5 cm and 25 cm depth on the diurnal period (a), expressed as ΔCO_2 in µ mol L^{-1}. Panel (b) shows the relationship between ΔCO_2 and wind speed (U_{10}), with daytime observations highlighted in red and night time observations in black. Whiskers show minima and maxima of the respective period.

To gain a more detailed understanding of the CO_2 gradients at the surface, we calculated the difference in CO_2 concentrations between 5 cm and 25 cm depths (ΔCO_2). The mean concentration gradient was significantly steeper during the night compared to during the day (t-test, $p < 0.05$, Figure 2 a). The magnitude of ΔCO_2 depended on wind speed (Figure 2 a). Generally, the mean CO_2 gradient was lower at high wind speeds for both day and night. However, while low wind speeds did not lead to higher CO_2 gradients during the day, the gradients in calm nights were 5 to almost 10-fold steeper (Figure 2 b). This diurnal development of a CO_2 gradient at the surface should have affected CO_2 emissions.

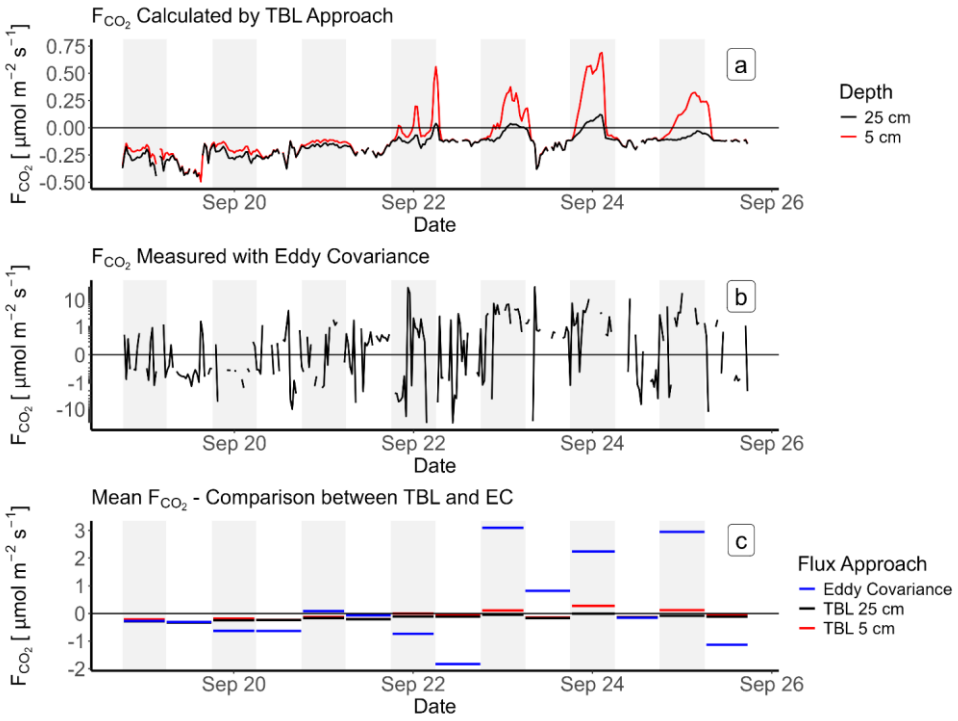

Figure 3: Comparison of fluxes (F_{CO2}) derived from Eddy Covariance (EC) measurements and the Thin Boundary Layer (TBL) approach. TBL fluxes (a) derived from $c_{CO2\_0.05}$ and $c_{CO2\_0.25}$ are shown as red and black lines, respectively. F_{CO2} measured by EC (b) is shown as black lines. F_{CO2} averaged over every day and night period is shown in (c), where red bars are fluxes derived from $c_{CO2\_0.05}$, black bars derived from $c_{CO2\_0.25}$, and blue bars derived from EC measurements. All fluxes shown have the unit of µmol m^{-2} s^{-1}. Grey highlights show night-time in all subplots.

We used our CO_2 concentration data to calculate F_{CO_2} by the TBL approach and compared those fluxes with measurements done with Eddy Covariance (Figure 3). During the windy period, TBL fluxes were negative regardless of which CO_2 probe data we used and comparable to the fluxes measured by EC (Figure 3 a, b, and c). In calm nights, supersaturation of CO_2 led to positive TBL fluxes with data from both depths. Because at 25 cm depth oversaturation was only reached at the end of the night, mean night fluxes derived from 25 cm depth remained negative (Figure 3 c). EC fluxes during those nights were positive too, but significantly higher than TBL fluxes (Figure 3 b, c). In 5 out of 14 cases, the flux direction differed between the TBL approach and EC measurements, when using the measurements of the 25 cm probe for the TBL approach. In contrast, when using CO_2 measurements from the 5 cm probe, only 2 out of 14 instances show different flux directions. During our study period of 7

days, the total CO_2 emissions were -2.2 g m^{-2} when calculated using the 5 cm data, -4.2 g m^{-2} based on 25 cm data, and 4.6 g m^{-2} as recorded by the eddy covariance system.

4 Discussion

Our data revealed how changes from windy to calm weather and from a mixed water column to stratification within a week influenced microstratification and its effect on greenhouse gas exchange between lakes and the atmosphere. Our detailed CO_2 concentration measurements clearly showed the temporal development of CO_2 gradients at the water surface. This observation is consistent with previous studies like Hari et al. (2008), whose results indicate CO_2 gradients within the upper 50 cm of the water layer.

During the windy period, CO_2 concentrations were almost zero at daytime, probably caused by photosynthetic CO_2 consumption by abundant phytoplankton. CO_2 undersaturation in high productive lakes is a common observation (Balmer and Downing, 2011; Zagarese et al., 2021). In that period, CO_2 concentrations were consistently lower than the atmospheric equilibrium concentration, turning the reservoir to a CO_2 sink during that time. The oxygen concentrations measured at the surface and above the bottom as well as the temperature profiles suggest that the entire water column was mixed in that period. This likely lead to a homogeneous vertical distribution of CO_2 too. The fact that there was no thermal stratification in that time means that the phytoplankton's CO_2 uptake stripped CO_2 from the entire water columns volume, rather than the smaller volume of the upper mixed layer only. Consequently, more CO_2 would be required to increase the concentration at the surface, compared to a stratification scenario. This observation is supported by the fact that no CO_2 gradient was measured during the day, while a slight gradient was detected at night. This occurs because CO_2 is consumed quickly during daylight hours, whereas at night, the uptake of CO_2 from the atmosphere leads to slightly higher concentrations at the water surface. This was reflected by our 5 cm probe measuring concentrations closer to atmospheric equilibrium than the 25 cm probe.

When the wind ceased at September 21st, the vertical distribution of gas concentrations changed. Starting on that day, oxygen concentrations at the surface and at the bottom began to increase and decrease, respectively. This observation indicates that the reservoir slowly underwent stratification again. Because the mixing depth was much greater during the windy period compared to the calm period, the water volume available to dissolve CO_2 was much greater also. Consequently, the lower windspeed in the second half of the week decreased both the mixing depth and the available volume. During these calmer nights, respiration in the shallow mixed layer led to significant CO_2 accumulation at the surface. Additionally, microstratification within the top 25 cm of the water column could further restrict the volume available for CO_2 accumulation, potentially leading to even higher concentrations (Figure 4). While there was no difference in CO_2 gradients during calm daylight hours compared to windy days, we observed notably high CO_2 concentrations within the top 25 centimetres during calm nights. Our findings are consistent with a study of Åberg et al. (2010), who found that short term CO_2 variations at the surface were best related to thermal dynamics within the upper mixed layer, whereas parameters like wind and radiation did not influence CO_2 concentrations. In our study we found negative effect of incoming solar radiation on CO_2 concentrations

during the day, but a positive feedback during the night. Further, varying CO_2 concentrations have been related to changes in lake metabolism after storms before (Vachon and del Giorgio, 2014).

Interestingly, the 5 cm probe, despite being closer to the surface, recorded higher CO_2 concentrations than the underlying water. This requires a CO_2 producing process in the surface layer, likely ~~biological~~ respiration. There are various groups of organisms that could increase CO_2 in the 5 cm layer, such as neuston, which comprises organisms living at or even within the surface micro layer. Phytoplankton was found to float at the water surface at wind speeds lower than 3 m s^{-1} (Zhang et al., 2021). Further, some species migrate from the lower boundary of the epilimnion to the surface during night. This behaviour is called diel vertical migration and is used to access food while avoiding predators that need light for hunting (Ringelberg, 1999). Both neuston and migrating species are respirating during the night, thereby producing CO_2. Furthermore, the vertical mixing during windy periods likely increased nutrient availability for phytoplankton, potentially triggering growth and accumulation of phytoplankton at the water surface (Nürnberg et al., 2003). That phytoplankton switches from net photosynthesis during the day to net respiration at night, thereby increasing CO_2 concentration at night. In the calm period, mean gross primary production and respiration as calculated from oxygen data were 10.4 mg O_2 L^{-1} d^{-1} and -9.0 mg O_2 L^{-1} d^{-1}, respectively, which is comparable to other hyper-eutrophic lakes, especially after storm events (Williamson et al., 2021). During algal blooms, the water surface is often covered by a mat of floating phytoplankton. Although we lack chlorophyll data, satellite images taken before and after our measurement period suggest rapid algal growth following the storm events that occurred just before our study (Figure B 1). Such mats can become very dense, possibly increasing CO_2 production and accumulation even more.

Besides biologic ~~activity, convective~~activity, convective mixing within the top water layer can play an important role in the distribution of CO_2 concentrations. Convective mixing, driven by water density differences due to cooling at the water surface, can enhance the vertical movement of water, thereby influencing the distribution of CO_2 in the water column. While we did not find signs of thermal microstratification at the surface during nights with strong CO_2 gradients, we contrastingly observed conditions favoring convection (Figure **1**~~Figure 1~~ c). Further, meteorological parameters such as atmospheric stability, emitted long wave radiation, or sensible heat flux, indicate unstable conditions in the air above the water. From this, we infer that the biomass accumulated at the water surface could either produce CO_2 at rates that exceed its transport in the upper water layer, or the algal mats acted as a physical barrier between the water and air, leading to increased stability of the water beneath the mats which reduces the influence of atmospheric instability on the water below. In our case the high CO_2 concentrations at the surface were not driven by convective upward mixing of CO_2 rich water but by biological activity at the surface.

The CO_2 gradient measured during calm nights was fundamentally influencing the fluxes calculated with the TBL method. Fluxes derived from concentrations in 0.05 m depth were closer to the fluxes measured by EC than fluxes derived from 0.25 m depth. However, TBL fluxes derived from 0.05 m were a magnitude lower than those of EC. This can be due to either an unprecise parametrization of k, or by an inaccurate CO_2 measurement. Many previous studies showed that water-side convection has a significant contribution to the gas transfer coefficient under calm conditions (Eugster et al., 2003; Rutgersson and Smedman, 2010; Heiskanen et al., 2014; Podgrajsek et al., 2015). However, these studies reported increased fluxes up to 200 %, which would still underestimate TBL fluxes in our experiment, compared to EC fluxes.

While it is generally acknowledged that daytime measurements do not reflect the concentrations at night (Erkkilä et al., 2018), the depth of CO_2 measurements has not been questioned so far, as long as measurements were done in the upper mixed layer and close to the surface. Our measurements provide evidence that representative measurements of the CO_2 concentration in the water strongly depend on depth and time of measurements. In our results, the depth of measurement even determined whether the TBL method would result in efflux or influx (Figure 3b~~a~~). This was especially visible at night when the flux calculated from the CO_2 concentration measured at 25 cm depth was negative, while both EC measurements and TBL based on the measurements in 5 cm showed significant positive fluxes. The fact that the EC fluxes were higher than the TBL fluxes based on the 5 cm probe could be explained by the CO_2 gradient which probably continued towards the water-atmosphere interface.

Our uppermost probe measured the mean CO_2 concentration between 1 and 10 cm depth and it is plausible that the concentration at the water surface was even higher than measured by that probe. Based on our results, the highest CO_2 concentration is expected to occur at the very surface of the water, driven by night-time respiration and restricted mixing. However, current measurement methods are not able to resolve such small-scale vertical gradients within the SML. While floating chambers have been used to measure CO_2 in the surface by deploying them as closed systems, therefore equilibrating the chamber volume with the surface water, this method is limited by slow equilibration times and insufficient temporal resolution (Rudberg et al., 2021). Therefore, advanced measurement techniques with higher vertical and temporal resolution are necessary to accurately capture steep CO_2 gradients and to understand the dynamics of CO_2 exchange at the water-atmosphere interface.

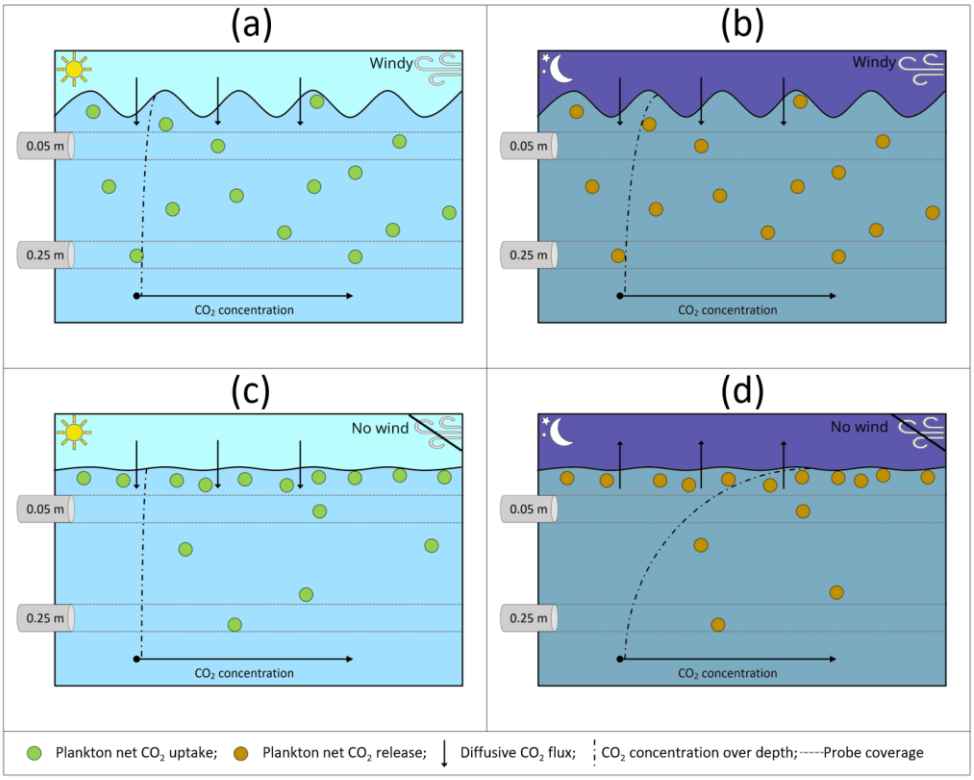

Figure 4: Schematic of CO_2 accumulation in the surface water, with CO_2 probes located in 5 cm and 25 cm depth. (a): wind-induced turbulence causes homogeneous distribution of plankton, resulting in CO_2 decrease in both measuring depths during the day. (b): wind induced turbulence causes homogeneous distribution of plankton. CO_2 is produced by respiration, but the CO_2 concentration across the whole water column is low, thus respiration is compensating strong undersaturation. CO_2 uptake from the atmosphere causes slightly increased concentration in 5 cm. (c): CO_2 concentration is low in the whole mixed layer, but calm conditions cause plankton to accumulate at the surface. (d): Calm conditions cause plankton to accumulate at the water surface. Respiration causes CO_2 increase in this layer, which is causing CO_2 emissions during calm nights.

Conclusions

We could show that, under the right conditions, a significant gradient of CO_2 concentrations at the lake's surface can develop. This gradient was largest in wind still nights. The gradient occurred at the same time as a phytoplankton bloom, from which we conclude that phytoplankton is the controlling source of CO_2. Only when limited mixing of the surface layer comes together

with high accumulation of biomass in the surface water, a thin surface layer with high CO_2 concentrations can develop. This effect can be large enough to turn a CO_2 under-saturated lake to a temporary CO_2 source. This result partly explains discrepancies of previous studies, where high CO_2 fluxes measured by Eddy Covariance could not be found with the Thin Boundary Layer method (Spank et al., 2020). We also show that uncertainty of the TBL method under calm conditions not only results from an incorrect parameterisation of the gas transfer velocity, but also by error-prone measurements of CO_2 in the water. Our findings imply that CO_2 emissions from eutrophic waters might be higher than previously thought – especially under calm conditions.

Appendix A: Detailed water temperature graphs

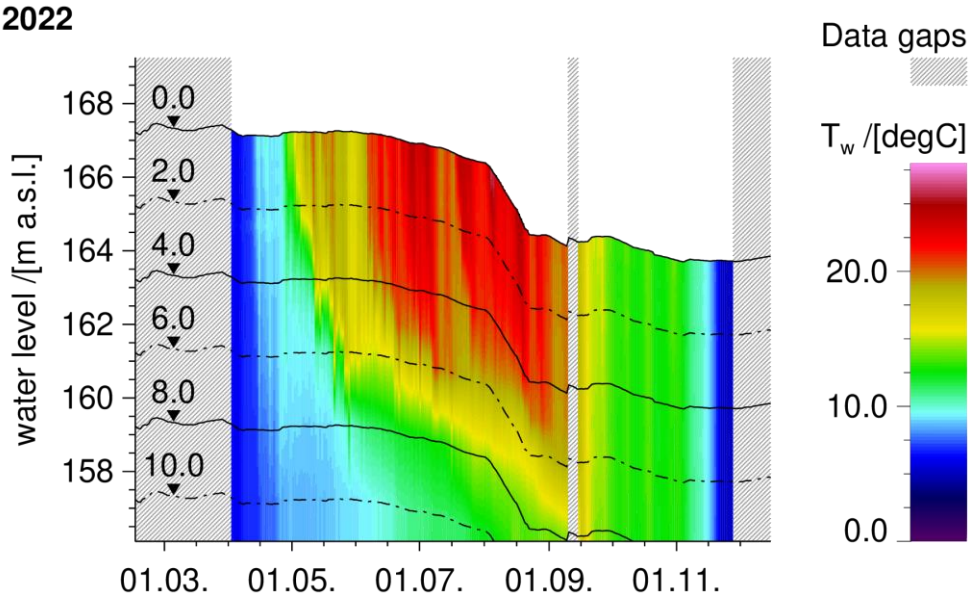

Figure A 1: Water level (meters above sea level) and water temperature profile from April 1st to November 28th in 2022 at Bautzen reservoir. Solid and dot-dashed black lines indicate water depths of 2 m, 4 m, 6 m, 8 m, and 10 m. Grey bars highlight missing data.

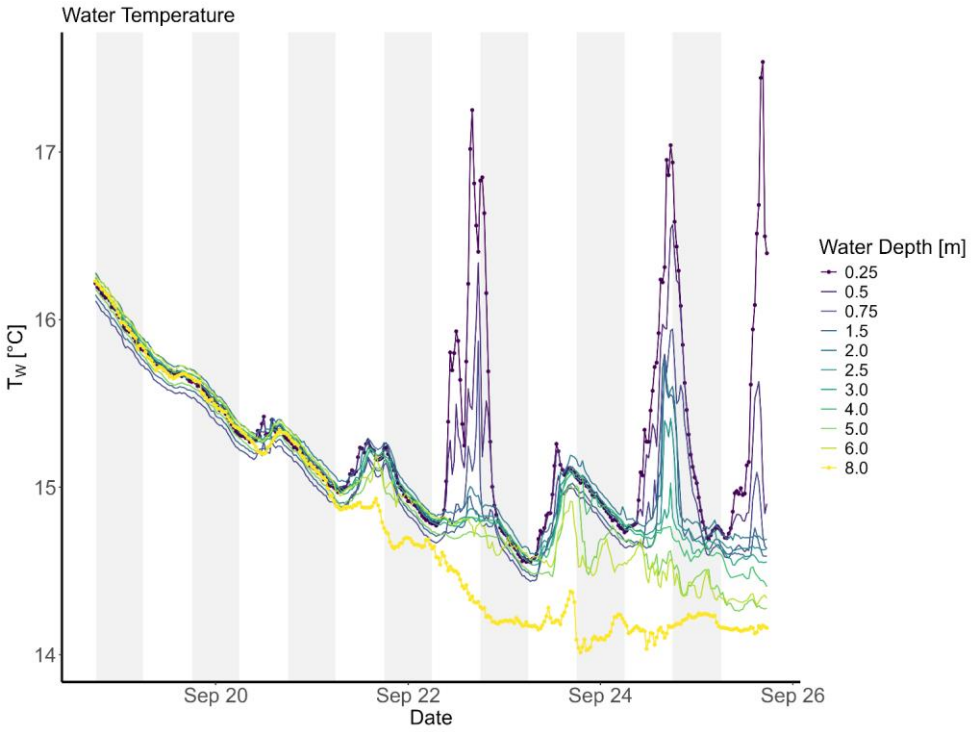

Figure A 2: Detailed water temperatures across the water column during the extensive sampling period. Dark purple and yellow dotted lines highlight surface and bottom temperatures, respectively. Grey highlights show night-time.

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Appendix B: Satellite images of the reservoir

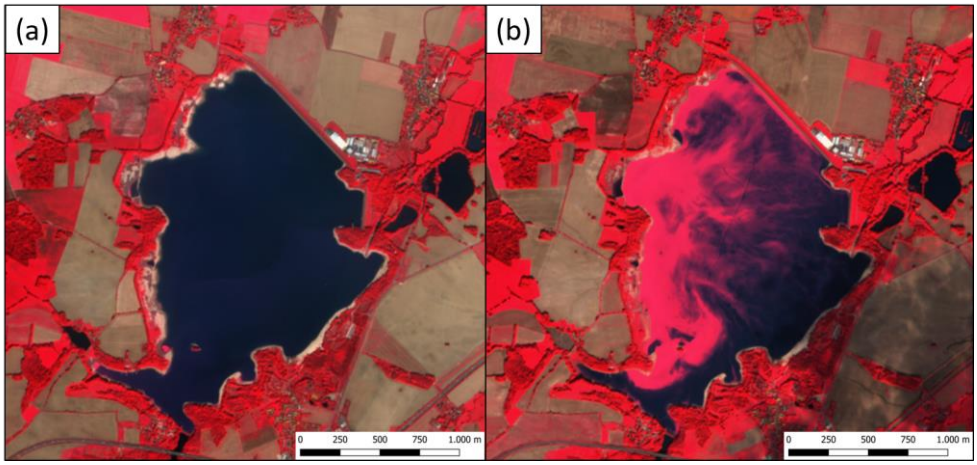

Figure B 1: False colour images from Sentinel-2 L2A taken on September 5th (a) and 25th (b), 2022, provided by the European Space Agency (ESA) and accessed via the EO Browser (https://apps.sentinel-hub.com/eo-browser/, Sinergise Solutions d.o.o., a Planet Labs company). September 5th was selected because it was the last day without cloud coverage before the sampling period. The images show the state of the reservoir during the algal bloom described in this manuscript. The false colour images, which are typically used to highlight specific features like vegetation or water, do not show the water surface properly on September 25th, due to algal coverage. This clearly highlights the intensity of the algal bloom, which was likely covering the water surface.

Code availability

Code is available upon request to the corresponding author.

Data availability

Data are available upon request to the corresponding author.

Author contribution

All authors contributed to the study's conception and design. Field measurements were carried out by PA and US. Meteorological data were processed by US, while limnological data were processed by PA. PA performed the data analysis. PA wrote the first draft of the manuscript, and all authors contributed to the final version. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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