https://doi.org/10.5194/bg-2023-43
Preprint. Discussion started: 23 February 2023
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Biogeosciences Discussions
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EGU

Temporary stratification promotes large greenhouse gas emissions in a shallow eutrophic lake

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Abstract

Shallow lakes and ponds undergo frequent temporary thermal stratification. How this affects greenhouse gas (GHG) emissions is moot, with both increased and reduced GHG emissions hypothesised. Here, weekly estimation of GHG emissions were combined with high-resolution temperature and oxygen profiles of an 11 hectare shallow lake to investigate how thermal stratification shapes GHG emissions. There were three main stratification periods with profound anoxia in the bottom waters occurring quickly upon isolation from the atmosphere. Average diffusive emission of methane (CH_4) and nitrous oxide (N_2O) were larger and more variable in stratified phase, whereas carbon dioxide (CO_2) was on average lower. CH_4 ebullition was an order of magnitude greater in the stratified phase. In addition, there was a large efflux of CH_4 and CO_2 when the lake mixed after periods of extended (circa 14 days) thermal stratification. These two turnover events were estimated to have released the majority of the CH_4 emitted

https://doi.org/10.5194/bg-2023-43
Preprint. Discussion started: 23 February 2023
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between May and September. These results highlight the role of turnover emissions resulting from temporary thermal stratification and also the need high frequency measurements of GHG emission in order to accurately characterise emissions from these temporarily stratifying lakes.

Keywords: Climate change; lake stratification; methane; carbon dioxide; nitrous oxide; climate feedbacks

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1. Introduction

Fresh waters are key sites for the processing of greenhouse gases (GHG), methane (CH_4), carbon dioxide (CO_2) and nitrous oxide (N_2O). Shallow lakes in particular, have been identified as hot spots of CH_4 release, particularly when ebullition is taken into account (Davidson et al., 2018; Aben et al., 2017). The certainty that fresh waters are large emitters of GHGs contrasts sharply with the uncertainties associated with the quantities emitted and this is in large part due to historical paucity of measurements (Cole, 2013). A recent study identified the highly variable emissions from lakes and ponds which make a large proportion of total emissions (Rosentreter et al., 2021). A dearth of measurement combined with these highly variable emissions makes determining the drivers and controls of those emissions a challenge, which in turn makes predicting future emissions difficult.

The current and future effects of climate change on lakes in general and on their GHG emissions are relevant questions as there is potential for positive feedbacks and synergies with other human impacts such as eutrophication (Davidson et al., 2018; Beaulieu et al., 2019; Delsontro et al., 2016; Meerhoff et al., 2022). Taking a broad metabolic theory of ecology approach temperature increases should promote methanogenesis and shift the balance from primary production to respiration increasing CO_2 emission at cellular and ecosystem scale (Yvon-Durocher et al., 2010). However, empirical and experimental data indicate that temperature is not the sole control of primary production and methanogenesis. In particular eutrophication, and the promotion of large algal crop, has been associated with increased emissions of CH_4 and N_2O (Delsontro et al., 2016) both by diffusion and ebullition (Zhou et al., 2019). Furthermore, in what is globally the most abundant lake type, small shallow lakes, where macrophytes can colonise large areas of the lake bed, the identity of the dominant primary producer may be more important than temperature in shaping GHG dynamics (Davidson et al., 2015; Davidson et al., 2018; Bastviken et al., 2023).

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Climate change effects on lakes are not limited to increases in average temperatures and lengthening of the growing season. Increases in both the frequency and intensity of heat waves are predicted, which will promote the warming of surface waters and in turn make permanent and temporary thermal stratification of lakes more likely (Woolway and Merchant, 2019), even in lakes typically classified as non-stratifying (Kirillin and Shatwell, 2016). Emissions of gases, in particular CH_4, that accumulate in the isolated bottom waters of a stratified lake, occurs upon mixing and can make very significant contributions to cumulative emissions (Schubert et al., 2012). High-resolution studies of sites that undergo temporary stratification are, however, rare (Søndergaard et al., 2023). In terms of its effects on GHG dynamics, there are potentially antagonistic processes at work in a stratified lake. On the one hand the 'shield effect' results in lower temperatures at the sediment surface slowing down metabolic processes that scale with temperature, i.e. methanogenesis and mineralization of organic carbon, reducing emission and promoting carbon burial. On the other hand, anoxia at the sediment surface may shift processes towards fermentation, increasing the proportion and total amount of CH_4 produced and perhaps reducing C burial (Bartosiewicz et al., 2019). Recent work combining empirical observations and models has suggested that shielding effects are larger than the anoxia effects and that stratification, in general, increases C burial and reduces GHG emissions. The stratification induced isolation of bottom waters was reported to lead to reduced ebullition of CH_4 and a shift to diffusive pathways (Bartosiewicz et al., 2015). It might, however, be predicted that in shallow lakes stratification would lead to much larger CH_4 release as anoxic conditions would limit CH_4 oxidation by CH_4 oxidizing bacteria (MOBs) (Bastviken et al., 2008). There may also be other factors with the potential to increase GHG emission, such as sediment organic content and lake trophic status (Delsontro et al., 2016), which may interact with stratification patterns in shaping GHG emissions.

In this study, we used data from a shallow lake with high frequency measurements of temperature profiles combined with weekly measurements of dissolved gas concentrations in the surface and bottom waters and continuous measurement of ebullitive emissions of CH_4 to track the effects of lake stratification on

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GHG emissions. The key question was how ebullitive and diffusive fluxes of the key GHGs: CH_4, CO_2 and N_2O respond to temporary thermal stratification.

2. Materials and methods.

2.1 Study site

Ormstrup lake (lat 56.326°, lon 9.639°) (Fig.1) (depth map with GHG sampling locations) is an 11 ha, shallow lake (average depth 3.4 m), with a maximum depth of 5.5 m, with a relatively long hydraulic retention time (> 1 year). The lake is eutrophic with high TP and chlorophyll-a (Table 1; Søndergaard et al., 2022) with very sparse occurrence of submerged plants.

2.2 Depth profiling and high frequency measurements

In June 2020 a Nexsens (NexSens Technology, Fairborn, OH, USA) CB-450 data buoy system (https://www.nexsens.com/pdf/CB450_datasheet.pdf) was deployed at the deepest point of the lake equipped with a Nexsens TS210 thermistor string https://www.nexsens.com/pdf/TS210_datasheet.pdf) with temperature nodes measuring temperature at 4 levels; one sensor “in air”, ca. 5 cm above the water surface, at the buoy and three sensors at -1, -2, -3 meters, respectively relative to the water surface. In addition two Aqua TROLL 500 (In-Situ, Fort Collins, CO, USA) multi-sondes were mounted near the surface (-1.0 meters) and at deeper water depth (-3.8 meters). The near surface and deeper water sonde were configured with sensors to measure dissolved oxygen (DO) and temperature (Tw). The optical sensors were calibrated according to manufacture guidelines and checked on a weekly basis.

The optical sensors of the Aqua TROLL 500 have a built-in wiper mechanism to clean sensor heads to hamper bio-fouling. The wiper function was enabled to perform cleaning in sync with sensor measurements, hence every 15 minutes. In addition, manual cleaning of sensor heads was done every

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week, while routine manual field monitoring was carried out at the lake. Prior to the deployment of the buoy, and as a validation exercise for the buoy data, weekly manual profiles of DO and Tw were collected at the deepest point.

Periods of stratification were defined by a greater than 2 °C difference between the surface and bottom waters and DO below 0.5 mg l^{-1} at the time of the weekly manual profiling of the system. The high frequency measurements were used to confirm the patterns. During periods of stratification, there may be partial mixing events where some of the water column mixes, but the bottom waters remain undisturbed.

2.3 Water chemistry

Water samples for the analysis of Chlorophyll-a were collected weekly from the 20. April 2020 from surface (-0.5 m) water at station 3 (Fig. 1) and analysed according to Danish standard procedures (Søndergaard et al., 2005). Depth profiles of temperature, electrical conductivity (EC) and dissolved oxygen (DO) were measured manually with an Aqua TROLL 500 probe from every -0.5 or -1 m down to -5 m depth).

2.4 Greenhouse gas sampling

2.4.1 Dissolved concentration

Samples of dissolved concentrations of CH_4, CO_2 and N_2O were collected weekly from the 20. April 2020 from surface waters and weekly from surface and bottom water from the 26. May 2020. The samples were taken using head-space equilibration after (Mcauliffe, 1971), where 20 ml of water was collected from just below the water surface and 20 ml of N_2 was introduced as a headspace in a 60-ml syringe and then shaken vigorously for one minute. The 20 ml headspace was then transferred to a 12-ml pre evacuated glass vial.

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133

134 Samples were collected between 12. May 2020–15. October 2020 which is 126 days and cover the majority
135 of the growing season.
136 Gas concentrations in the headspace were determined on a dual-inlet Agilent 7890 GC system interfaced
137 with a CTC CombiPal autosampler (Agilent, Nærum, Denmark) (Petersen et al., 2012). For the GC,
138 certified CO_2, CH_4 and N_2O standards were used for calibration and validation. Aqueous concentrations in
139 N_2O, CH_4 and CO_2 were calculated from the headspace gas concentrations according to Henry's law and
140 using Henry's constant corrected for temperature and salinity (Weiss, 1974; Weiss and Price, 1980;
141 Wiesenburg and Guinasso, 1979).
142 The fluxes of N_2O, CH_4 and CO_2 between the water and the overlying atmosphere were estimated as

143 $$f_g = k_g\left(C_{wat,g} - C_{eq,g}\right)$$

144 Where f_g is the flux of a specific gas g, k_g is the piston velocity of the gas and $C_{wat,g} - C_{eq,g}$ is the
145 gradient of concentration between the concentration of gas dissolved in the water ($C_{wat,g}$) and the
146 concentration of gas the water would have at equilibrium with the atmosphere ($C_{eq,g}$).
147 We calculated a gas transfer velocity k_{600} for each sampling occasion using the relationship based on
148 windspeed described in (Cole and Caraco, 1998).

149 $$k_{600} = 2.07 + 0.215U_{10}^{1.7}$$

150 U_{10} is the mean daily windspeed at 10m (m s^{-1}) obtained from the Danish meteorological institute
151 (DMI;20x20 km grid data)

152

153

154 $$k_g = k_{600}\left(\frac{Sc_g}{600}\right)^x$$

155

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156 Sc_g is the Schmidt number(Wanninkhof, 1992) of the specific gas g. We chose x =-2/3 as this factor is
157 used for smooth liquid surface (Deacon, 1981).
158
159 Daily flux rates were calculated using linear interpolation of the weekly surface measurements from each
160 of the sampling points. Total diffusive surface water fluxes were calculated by taking an average of the
161 daily flux rate from the 12. May 2020 to the 13. October 2020. Then an average was taken for each location
162 and then an average of the 3 locations was multiplied by the area of the lake and the number of days covered
163 by the study, here 126 days was chosen to match the period over which ebullition was measured.
164
165 The total content of the gases in the lake's bottom waters were calculated from the concentration of the
166 gases per litre multiplied by an estimate of the volume of the water in the hypolimnion. The volume of
167 water in the hypolimnion was estimated from the lake profiles manually conducted on the day of
168 sampling. The top of the hypolimnion was determined by the depth below which oxygen was less than 0.5
169 mg l^{-1} A detailed bathymetry of the lake allows the calculation of the area and therefore volume of water
170 that lies below a given depth. For the purposes of this study, it was assumed that all the gas in the
171 hypolimnion was released on turnover. This release was calculated from 30. June 2020 and from the 25.
172 August 2020, as they are the periods where complete mixing occurred and all the gases accumulated in
173 the bottom waters was likely to be released.
174
175
176 2.4.2 Ebullition

177 The ebullitive flux of CH_4 was estimated using at total of 40 floating chambers placed on 4 transects of 10
178 chambers each (Fig. 1). As the existing literature indicated that ebullition is lower as water depth
179 increases (Wik et al., 2013) the transects were placed to maximise the measurement of the low end of the
180 depth gradient on the shallower slopes of the western end of the lake (Fig. 1). The averge and maximum

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depth of each transect was T1: 293 cm and 472 cm; T2: 181 cm and 267, T3: 223 cm and 300 cm and T4 166 cm and 220 cm.The chambers were set on the 14. May 2020 and sampled every two weeks from that date, and on one occasion after one week. Twenty ml of sample was taken from the floating chamber and injected into a pre-evacuated 12 ml vial (exetainer, Labco). Gas concentrations were determined on the same GC than described above (Petersen et al., 2012)

Ebullitive flux of CH_4 was estimated as:

$$\frac{p_{gas} \times Vol_{bub}}{t \times A}$$

Where p_{gas} is the concentration of CH_4 in the gas that was trapped, Vol_{bub} is the volume of the chamber (i.e. 7L), t is the time during which the samples was collected and A is the area of chamber (i.e. 0.075 m^2). A fraction of the CH_4 released via ebullition in the chamber will have re-dissolved in the water or might leak through the chamber walls, thus underestimating the ebullitive flux. A study (Delsontro et al., 2016) attempted to correct for this underestimation assuming that the CH_4 gas present in the bottle of the trap is in equilibrium with the CH_4 dissolved in the water present in the bottle. This correction increased their estimate of ebullitive flux by 2-5%. The chambers used here are even more vulnerable to the diffusion of the bubble emissions from the chambers back into the water, due to their relatively large surface area of water for the exchange of gas. In addition, there will be diffusion of CH_4 out of the chambers, estimated at up to 100 ppm per day (Davidson & Audet, unpublished data). As the point at which the bubbles enter the chamber is unknown it is difficult to estimate the degree of underestimation. The method is therefore a suboptimal way of accurately estimating ebullition, as it is almost certainly an underestimate of the true flux. However, the method is easy to apply and so can be used for longer periods, as it was here, and extend the temporal resolution of the data providing information of variation in ebullition in space and time. The estimates of absolute emissions of ebullition must be treated with caution, we can be confident that they are underestimates, just not the degree to which they are underestimates.

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Total ebullitive flux from the lake was calculated by taking a mean of the emissions from each transect over the 126 day period. Then taking an average of the means of four transects and multiplying this by the time of deployment of the chambers in days, which was 126 days, and by the area of the lake. This gives a total ebullitive flux of CH_4 for the lake over the period of measurement.

The three different flux types, surface diffusion, ebullition and turnover emission were then converted in comparable units of total lakes emissions (as g or kg of gas) over the studied period and also converted into CO_2-equivalents using a conversion factor related to their 100 year global warming potential (GWP) of 28 for CH_4 and 298 for N_2O.

3. Results

3.1 Lake physical and chemical characteristics

Depth profiles measured weekly from April show that stratification was initiated by the 26. May 2020 this may have broken down briefly and established again, visible in the temperature sensors for the buoy on the 5. June 2020 (Fig, 2). There were then 12 days of mixing followed by stable period of stratification with onset the 14. June 2020 and a duration of 16 days until a mixing event around the 30. June 2020. The following two weeks had cooler water and a mixed water column, herafter a ca. 6 day period of stratification from the 15. to 21. July 2020. A mixed phase of two weeks then followed until stratification reestablished on 4. August 2020 and persisted until the end of August, partial mixing is indicated by the buoy data from the 21. August 2020, but the weekly manual profile to deeper water inidcate that full mixing did not occur until after the 25. August 2020. The effects of the stratification and mixing events on the high frequency DO data measured at -3.8 m are clear, with rapid deoxygenation occuring after the onset of stratification and oxic bottom waters returning when the lake mixed (Fig, 2). The pattern in chlorophyll-a also follow, to some degree, those of stratification, with the exception of early spring.

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Chlorophyll-a values were extremely high in spring peaking at the start of June 2020 and falling gradually (Fig. 2). During the periods of stratification chlorophyll biomass was lower, likely limited by nitrogen (Søndergaard et al., 2023) and when a mixing even occurred the values increased, which is particularly evident in the July mixing periods (Fig. 2).

3.2 Concentrations of dissolved gases and fluxes from the surface waters.

The concentrations of the dissolved gases showed great variation from near or below atmospheric concentrations in some cases and up to an extremely high concentration (over 5 mg CH_4 C l^{-1}) in the bottom waters on the 30. June 2020. There was some spatial heterogeneity in the surface waters, with the more littoral locations showing the greatest variation and the highest values (Figs. 3,4,5). In particular the most littoral zone, where the water was shallower around 1 m in depth, showed the highest values just prior to, or coincient with, the stratification turnover. Table 2 shows the mean diffusive flux of each gas over the sampling period along with the mean flux in mixed and stratified phases. For CO_2 there was a lot of temporal variation in flux dynamics, though not a large difference between mixed and stratified phases in terms of mean values (Table 2). There were some periods of CO_2 influx in spring and later summer and these tended to coincide with the end of a mixed phase and the start of the stratification phase. N_2O concentrations were generally low (Figs 4 & 5) with the lake being a source of N_2O in the spring period and a sink or a very small source thereafter. The CH_4 concentration in the surface waters (Fig. 3) and the calculated diffusive emissions are relatively low, but did increase in the stratification periods with higher average values (Table 2 & Fig. 6). There was also some spatial variation with higher CO_2 and CH_4 diffusive emissions in the shallower sampling locations, both in stratified and mixed conditions (Fig. 6).

The most marked patterns in GHG concentration were evident in the bottom waters sampled at -4.5 m, which accumulated to very large concentrations of CO_2 but particularly CH_4 in the periods of stratification (Fig. 3 & 4). The ratio of CO_2 to CH_4 is illustrative in highlighting how stratification has

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altered the biogeochemical processes in the hypolimnion with CH_4 production becoming more prevalent. . For example on 30. June 2020 after 16 days of stratification the the ratio CO_2:CH_4 in the bottom waters was 0.8, whereas 7 days later after the mixing event it was 187 at the same depth.

3.3 Ebullitive fluxes

The CH_4 bubble flux is presented here as mean values for each of the 4 transects ranged from 0.303 to 81.1 mg CH_4 C m^2 d^{-1} for the individual transect over the growing season measurement. There is a very clear impact of stratification on the ebulltive efflux of CH_4 with stratified periods showing markedly higher levels of emission (Fig. 7 and Table 2). In addition there was a difference in average emissions among the different transects, with those with lower average water depth (T2 & T4) having lower emission than the transects with chambers over deeper water (T1 & T3) (Fig. 7).The samples collected from the chambers reflect two weeks of bubble and diffusion collection and the quantification of the flux is therefore an average of the period of chamber deployment, which was two weeks, or in one case a single week (Fig. 7). This two week period on occasion covered both stratified and mixed phases and on these occassions efflux was intermediate between purely mixed and stratified periods (Table 2 and Fig. 7).

3.4 Total lake fluxes

Scaling up the results to total flux of gases from the whole lake over the period of study and including the estimated emissions from two turnover events show a very different effect of stratification on the balance of types of emissions for the three gases. The majority of CH_4 emission (56%) result from the two short-lived turnover events (Fig. 8), whereas their contribution to CO_2 and N_2O emission was 5% and 1% respectively. Fluxes of CO_2 and N_2O were mostly diffusive, which represtented 95% of emissions of both gases. CH_4 diffusive flux was 14% of total emission whereas CH_4 ebulltion was more than twice and much representing 29% of total CH_4 emission. In terms of global warming potential CO_2 and CH_4

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emission were comparable, but the contribution of the turnover efflux was the dominant factor for CH_4 emissions.

4. Discussion

The emission of CH_4 reported here (Table 2) showed great variation between the mixed and stratified phases. The mean of the total emissions from Ormstrup in the stratified phase (58 mg CH_4-C m^{-2} day^{-1}) correspond relatively closely to the mean of the total emissions (ebullition plus diffusion) reported for lakes in this size range (47 mg CH_4-C m^{-2} day^{-1}), though they are well above the median value 15 mg CH_4-C m^{-2} day^{-1}(Rosentreter et al., 2021). The data provide a clear answer to the question of how thermal stratification effects GHG dynamics in shallow eutrophic lakes with an increase in total emissions (diffusion, ebullition and turnover) during the stratified period (Table 2, Fig 9). Previous work, combining observations and modelling suggested the opposite patterns (Bartosiewicz et al., 2019) as their study suggested that the shielding effect results in cooler bottom waters and this reduce CH_4 production due to the temperature dependence of CH_4 production (Bartosiewicz et al., 2016). This strong shielding effect may apply in deeper lakes experiencing more stable stratification, or less eutrophic lakes, the result here show, however, that stratification causes massive increases in GHG emissions.

Diffusive emissions did not on average show a strong stratification effect (Table 2). There were peaks in emission of CH_4 and CO_2 at the end of stratification periods, particularly in the shallower water sampling points (Fig. 6). Littoral zones can have markedly different GHG dynamics to deeper zones due to shallower water having lower pressure (Wik et al., 2013), less time for CH_4 oxidation (Bastviken et al., 2008) or abundant plants which influence a range of biogeochemical processes (Davidson et al., 2018; Esposito et al., 2023). It is therefore possible that littoral zone dynamics could cause these differences. However, the increase occurred at all three sampling points at the end of June 2020, which indicates a more lake-wide driver and the peak may represent the start of mixing after stratification. Strong winds were measured on the 29th and 30th June 2020 (Søndergaard et al., 2023) coincident with these increased

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littoral emissions. These winds would have caused lateral movement of the surface water causing an upwelling of bottom water, rich in CH_4 and CO_2, in the littoral margins at the opposite end of the lake. Thus, whilst we do not have direct evidence it seems more likely that these increased emissions in the littoral zone were not driven at least in part the partial mixing of the GHG rich bottom waters. In contrast to the diffusive flux, the ebullitive emission of CH_4 shows a very clear response to stratification with an order of magnitude difference in emissions between periods where the sampling reflected purely mixed or stratified periods (Table 2 & Fig. 7). The two-week resolution of the sampling meant that some samples covered both stratified and mixed phases and these samples had intermediate fluxes, as they cover both low (mixed) and high emission (stratified) periods. The spatial variation in ebullition is also illustrative of the impacts of stratification and the role of anoxia in shaping CH_4 fluxes. The two transects with the largest mean and maximum depths (T1 and T3) had the largest emissions, with the deeper of the two (T1) having the highest emissions and they saw the greatest relative increase during the stratification phases. This pattern is the obverse of that found in other studies where bubble emissions were larger in shallower water as higher pressure in deeper location means production rates of CH_4 need to be higher for bubbles to form (Wik et al., 2013). Deeper water at Ormstrup experiences anoxia earlier and this appears to cause locations with deeper water to have higher ebullition rates than shallower areas. This is at odds with ideas stemming from the metabolic theory of ecology stating that temperature (Yvon-Durocher et al., 2014) in particular at the sediment surface (Bartosiewicz et al., 2019) can be used to predict CH_4 efflux. Whilst it is a fact that CH_4 production is temperature dependent at the cellular level, CH_4 emissions were rather independent of the sediment temperature, for example in the first two weeks of July 2020 emissions were low and the sediment surface temperature was relatively high. Thus, temperature alone is a poor predictor of ecosystem scale CH_4 emissions.

It should be noted that the methods used to estimate bubble flux here, where floating chambers are sampled every two weeks is a “less than perfect method”, which in most cases will underestimate ebullitive flux. Logistical and financial constraints make continual sampling difficult and here we

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330 balanced these constrains against the greater time required to apply more accurate methods, such as
331 bubble traps (Wik et al., 2013) or automatic flushing chambers (Bastviken et al., 2015). Such is the
332 variability of bubble flux in space and time that sampling campaigns covering days or weeks would
333 potentially give an even more inaccurate picture of emissions than the method used here. Thus, the
334 continuous monitoring of ebullition using chambers with known biases was deemed the least worst
335 method available, but we acknowledge the caveat that ebullitive emissions may be underestimated.
336
337 In addition to diffusive and ebullitive emissions the turnover flux, which consists of the gases
338 accumulated in the hypolimnion being released on turnover, was also estimated. There were two major
339 turnover events at the end of June and in late in August 2020, which were preceded by 16 and 22 days of
340 stratification, respectively. It was not possible to directly measure turnover flux, as they are relatively
341 discrete events where the efflux likely occurs over the course of just a few hours. Evidence of this is that
342 the complete oxygenation of the water column during the mixing event at the start of July 2020 took just a
343 few hours (Søndergaard et al., 2023). Thus, the efflux is estimated from the bottom concentration with the
344 assumption that all the CO_2 and CH_4 in the bottom water was released at turnover, which is potentially an
345 over estimate. Notwithstanding this uncertainty we can be confident the turnover flux represents a very
346 large proportion of the total emission of CH_4 emissions from Ormstrup Lake over the growing season. We
347 estimate it contributed more than 50 % of growing season CH_4 emissions and 5 % of CO_2 emissions. This
348 highlights a very significant, and generally unmeasured, contribution to GHG emissions from lakes
349 undergoing temporary stratification, which are among the most common lake type in Denmark.
350 Furthermore, stratification is increasingly recognised as prevalent in ponds and shallow lakes (Holgerson
351 et al., 2022) and it is likely to become more common with continued climate change impacts (Woolway
352 and Merchant, 2019). Predicting the response of GHG emissions in a warmer world is, however, not
353 straightforward, as stable stratification over the whole summer may have different impacts to the more
354 transient stratification patterns as observed here. Conversely, more frequent summer storms in climate
355 change predictions could also result in mixing events that could cause large turnover emissions.

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356

357 The results here suggest that GHG dynamics were driven both directly and indirectly by the stratification
358 patterns and the anoxia it induced in the bottom waters. At Ormstrup Lake the thermal stratification of the
359 water column quickly led to anoxia, with only a matter of hours to days for the oxygen to be consumed
360 once the bottom waters were isolated (Fig. 2). The ratios of CO_2:CH_4 evidence how this promotes CH_4
361 over CO_2 production in the stratification phase (see Fig 9). In addition to promoting CH_4 production such
362 conditions would preclude, or severely limit, CH_4 oxidation, which has the potential to consume a large
363 proportion of CH_4 produced in the anoxic sediments (Bastviken et al., 2008). The raw emission data do
364 not provide any direct information on the balance of production versus oxidation, but the CO_2:CH_4
365 suggest there was marked shift to conditions where methanogenesis was the dominant process and there
366 was reduced CO_2 production. Studies have shown that CH_4 oxidation can consume large proportions of
367 the CH_4 produced under hypoxia (Saarela et al., 2019) and it is possible that there is intense CH_4 oxidation
368 occurring at the thermocline during the periods of stratification at Ormstrup lake , but this was not directly
369 measured at the lake. In addition to the more direct effects of anoxia there may be some indirect effects of
370 the patterns of stratification and mixing that promote greater GHG emissions. Søndergaard et al. (2023)
371 recently reported how nutrient dynamics at Ormstrup Lake were altered by the lake stratification and full
372 details can be found there, of relevance here is the impact on chlorophyll-a which saw a large spring peak
373 after which the abundance tracked the stratification and mixing regime, with a lag time. There was a
374 general reduction, or at least no increase as the stratification period progressed, perhaps due to nutrient
375 limitation in the epilimnion. Upon mixing there was generally an increase in chlorophyll-a, though the
376 weekly sampling resolution makes this difficult to assess. Chlorophyll-a and the labile dissolved organic
377 carbon (DOC) that result from abundant chlorophyll-a have been shown to be associated with higher
378 diffusive and ebullitive CH_4 emissions (Davidson et al., 2015; Beaulieu et al., 2019; West et al., 2012;
379 Zhou et al., 2019). It is not possible to say here whether a stable summer long stratification would have
380 led to decreased chlorophyll-a as nutrients became limiting due to their isolation in the bottom waters and
381 reliable high frequency chlorophyll-a data are required to convincingly demonstrate this phenomenon.

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382 Notwithstanding these uncertainties it may be the case that the temporary stratification, interspersed with
383 mixing events, observed here represents a 'sweet spot' providing both the resources, i.e. chlorophyll-a and
384 the labile DOC it produces, and optimal conditions (anoxia) for CH_4 production.
385
386 The combination of high frequency data on water temperature and dissolved oxygen combined with
387 weekly measurements of GHGs increase the reliability of the findings presented here. Up until relatively
388 recently it has been assumed shallow lakes, such as Ormstrup lake, stratification is not an important
389 feature. Sampling has therefore focused on surface waters, using dissolved concentrations of gases or
390 floating chambers to characterise flux, e.g. (Davidson et al., 2015; Audet et al., 2020; Peacock et al.,
391 2021). Thus, most studies have overlooked bottom waters and do not have the temporal resolution
392 required to capture turnover flux emissions from surface measurements. Furthermore, whilst many studies
393 now include estimates of bubble emissions of CH_4 e.g. (Bergen et al., 2019), the necessary temporal
394 resolution to accurately characterise ebullitive emission is not well established. The finding here indicated
395 that in such dynamic systems near continuous measurement is desirable and that short term collection
396 over one or two days could provide massive, over or under estimate of CH_4 ebullition.
397
398 Our results show very large temporal variation in emissions of all three gases, but in particular CO_2 and
399 CH_4, and this highlights the need for high frequency measurements to accurately characterize emissions
400 from lakes. Even the weekly frequency of the sampling in this study was not sufficient to directly measure
401 all the emission pathways and turnover flux had to be inferred from bottom water calculations. These data
402 show that to capture the extent of GHG emissions from lakes it is vital we include all forms of flux,
403 including ebullition and turnover flux. Recent work has highlighted the fact that most emissions (over
404 50%) from fresh waters come from highly variable systems (Rosentreter et al., 2021), with the mean and
405 median emission rates of CH_4 differing greatly indicating a few large emitters are responsible for a large
406 proportion of emissions. The sampling frequency applied here is rare, if a more standard resolution of
407 monthly measurements was applied the emissions estimate of all the gases, but in particular CH_4, would

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be highly dependent on what phase of the stratification was captured. As an example, a monthly sampling frequency could potentially miss all the stratification peaks - consequently massively underestimating emissions, whereas a different sampling frequency could catch a number of peaks and giving a much higher estimate. Thus, the same sampling frequency on the same lake, but timed differently could lead to conclusions of highly variable emissions. Thus, it seems in these highly dynamic systems that if temporarily stratification may occur in summer, high frequency measurements are required to accurately estimate emissions. This is logistically challenging but the current advances in the use of automatic flushing chambers (Bastviken et al., 2020) may provide the potential for affordable high spatial and temporal resolution measurement of GHG dynamics. This is a requisite for understanding the drivers of GHG dynamic, which is required for being able to predict how they will respond in a range of scenarios related to land use, climate change and management interventions.

Code/data availability

The datasets generated during and/or analysed during the current study are not publicly available as they form part of ongoing research projects but are available from the corresponding author on reasonable request and will be made publicly available later in the research project.

Author contributions

MS secured the funding for the wider lake restoration research project supplying the data. TAD, MS and JA conceptualized the gas study. TAD and AN established the buoy and sensor system. EL, CE, TAD, JA collected and analysed the data. TAD wrote the paper and all authors commented on earlier versions and read and approved the final draft.

Competing interests

The authors declare that they have no conflicts of interest.

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Acknowledgement

We thanks our crack technician team of Lene Vigh, Malene Kragh, Dorte Nedegaard and Dennis Hansen for their extreme competence on the lab and the field. We acknowledge Theis Kragh for the depth map of the lake already published in Søndergaard et al. 2022. We are very grateful to the Poul Due Jensen Fonden for providing great support for this work and the Ormstrup project generally. TAD and CE were also supported by GREENLAKES (No. 9040-00195B) and The European Union's Horizon 2020 research and innovation programmes under grant agreement No 869296—The PONDERFUL Project.

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567 Table 1. Summary lake information, summer mean values of a range of variables

Variable	Year 2020
Secchi depth (m)	0.88
Chlorophyll a (µg/l)	52
pH	7.7
Total phosphorus (mg/l)	0.58
Total nitrogen (mg/l)	1.50
TN:TP (by weight)	2.6

568

569 Table 2. Mean greenhouse gas flux (units CO_2: mg CH_4-C m^{-2} day^{-1}, N_2O: mg N_2O -N m^{-2} day^{-1} CH_4 both
570 diffusive and ebullitive in mg CH_4-C m^{-2} day^{-1}) from the lake from spring to Autumn 2020. The emissions
571 are divided in diffusive, ebullitive emissions. The mean values for all the surface water stations and all
572 four transects of chambers are given. Mixed stratified versus phases and there SD are also given.
573 Ebullition is collected for a period covering two weeks so on a number of occasion covered both mixed
574 and stratified periods and so for ebullition a third category where both mixed and stratified conditions
575 occurred is given.

576

Emission type	gas	mean	mixed	Stratified	Strat and mixed
	CO_2	506.14	616.6	446.1	

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Diffusive		(*520.24)*	*(399.5)*	*(571.7)*	
	CH_4	9.05 *(16.1)*	5.9 *(3.84)*	10.7 *(20.40)*	
	N_2O	0.10 *(0.097)*	0.083 *(0.082)*	0.12 *(0.10)*	
Ebullition	CH_4	16.99 *(19.39)*	4.84 *(3.27)*	47.26 *(21.67)*	12.78 *(10.34)*

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579 Figure legends

580 Figure 1. Ormstrup lake bathymetry and sampling stations for surface water greenhouse gas sampling
581 (St1, St2, St3) bottom waters were sampled at S3. Transects of 10 bubble traps were placed on T1- T4.
582 Adapted from the Søndergaard et al. 2023.

583 Figure 2 Temperature profile from June 2020 when the buoy was deployed and surface and bottom water
584 oxygen from June to the end of September 2020. Manual chlorophyll-a (µg L^{-1}) values are also given in
585 the top panel.

586 Figure 3. Dissolved CH_4 concentrations from surface and bottom waters – thermal stratification periods
587 highlighted in grey and the white background indicate mixed waters

588 Figure 4 . Dissolved CO_2 concentrations from surface and bottom waters–thermal stratification periods
589 highlighted in grey and the white background indicate mixed waters

590 Figure 5 Dissolved N_2O gas concentrations surface and bottom thermal stratification periods highlighted
591 in grey and the white background indicate mixed waters

592 Figure 6. Omstrup lakesurface fluxes of the CH_4, CO_2 and N_2O gases based on dissolved conentration ,
593 thermal stratification periods highlighted in grey and the white background indicate mixed waters

594 Figure 7. Plot of CH_4 ebullition averaged for each transect (10 chambers per transect), data collected from
595 40 traps every two weeks. thermal stratification periods highlighted in grey and the white background
596 indicate mixed waters.

597 Figure 8 – Total lake emissions per gas over the growing seasson in CO_2 equivalents. The emissions are
598 divided different emission modes: Diffusive, ebullitive and turnover flux.

599 Figure 9. Summary of the quantities of the gases present in the water and the volumnes emits from the
600 different pathways. The size of the arrow is proportional to the emissions from each pathway and with the
601 startified state on the left and the mixed state on the right, with the turnover flux in the centre.

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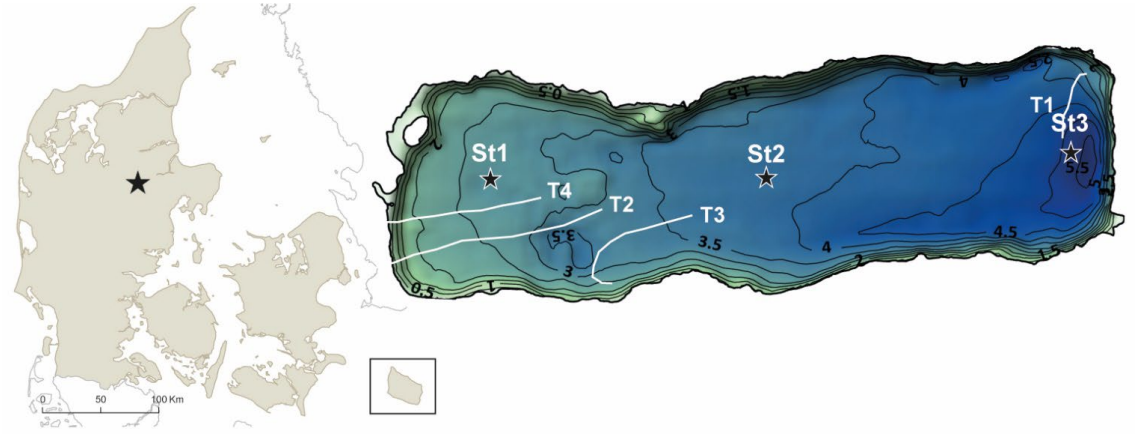
602 Figures and legends

603 Figure 1.



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605 Figure 1. Ormstrup lake bathymetry and sampling stations for surface water greenhouse gas sampling (S1,
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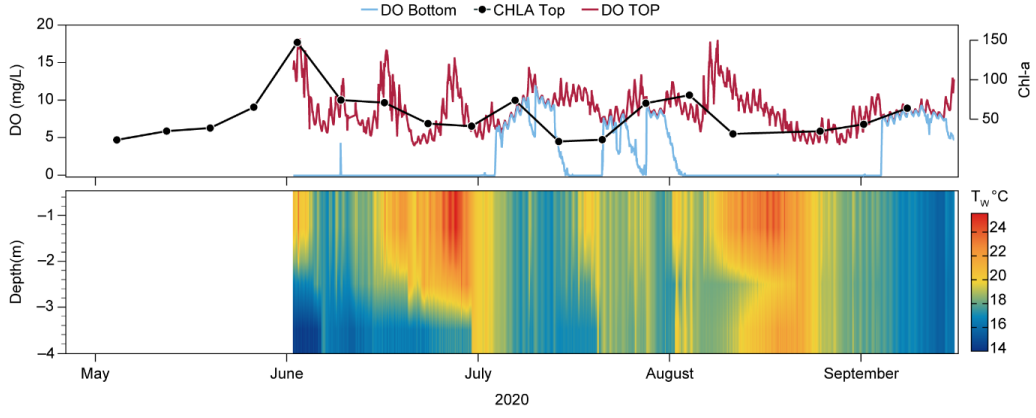
607 from the Søndergaard et al. 2023.



608

609 Figure 2 Temperature profile from June when the buoy was deployed and surface and bottom water
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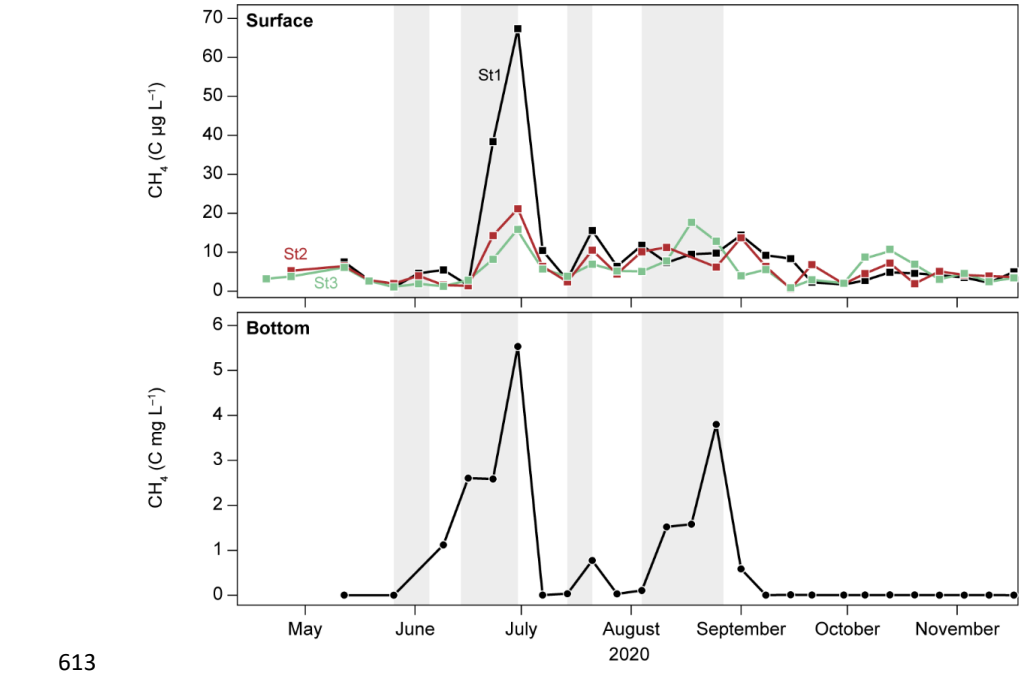
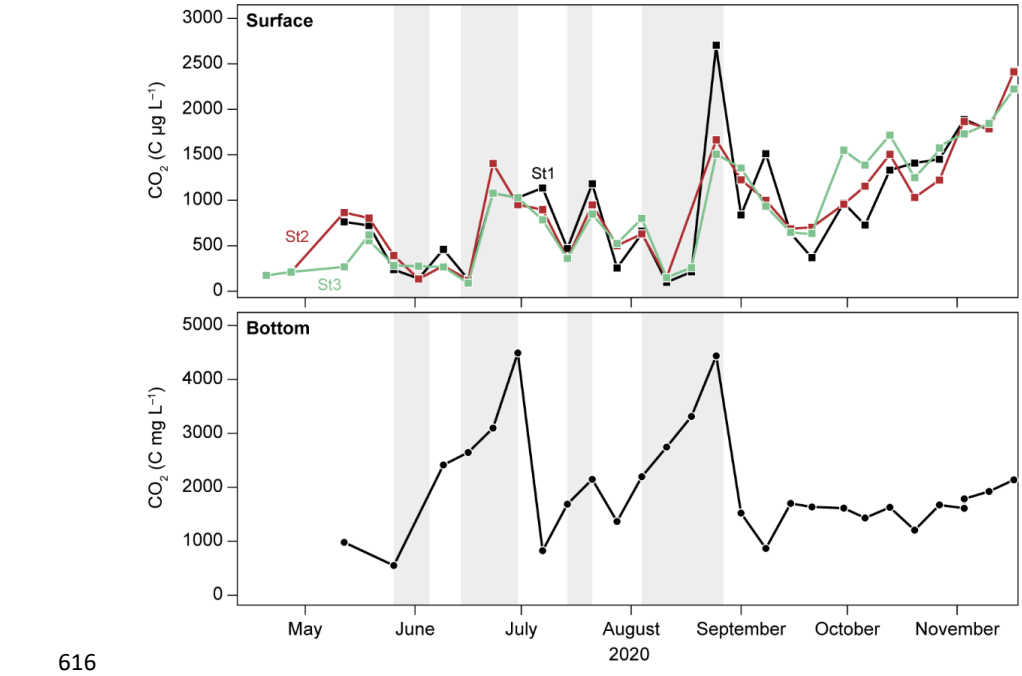


Figure 3. Dissolved CH_4 concentrations from surface and bottom waters – thermal stratification periods highlighted in grey; white background indicate mixed waters



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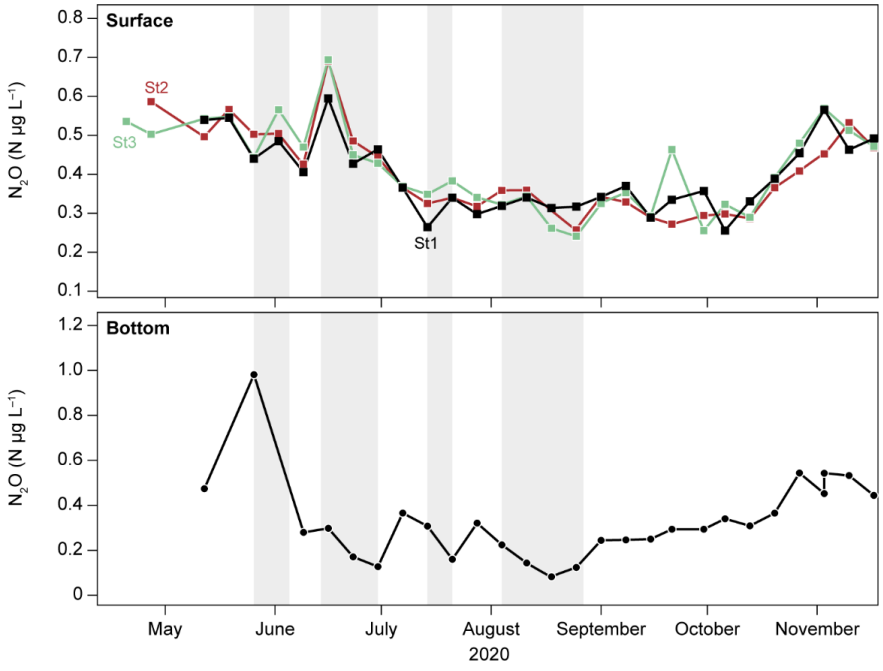
617 Figure 4 . Dissolved CO_2 concentrations from surface and bottom waters–

618 thermal stratification periods highlighted in grey; white background indicate mixed waters



619

620 Figure 5 Dissolved N_2O gas concentrations surface and bottom thermal stratification periods highlighted

621 in grey; white background indicate mixed waters

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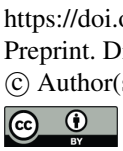

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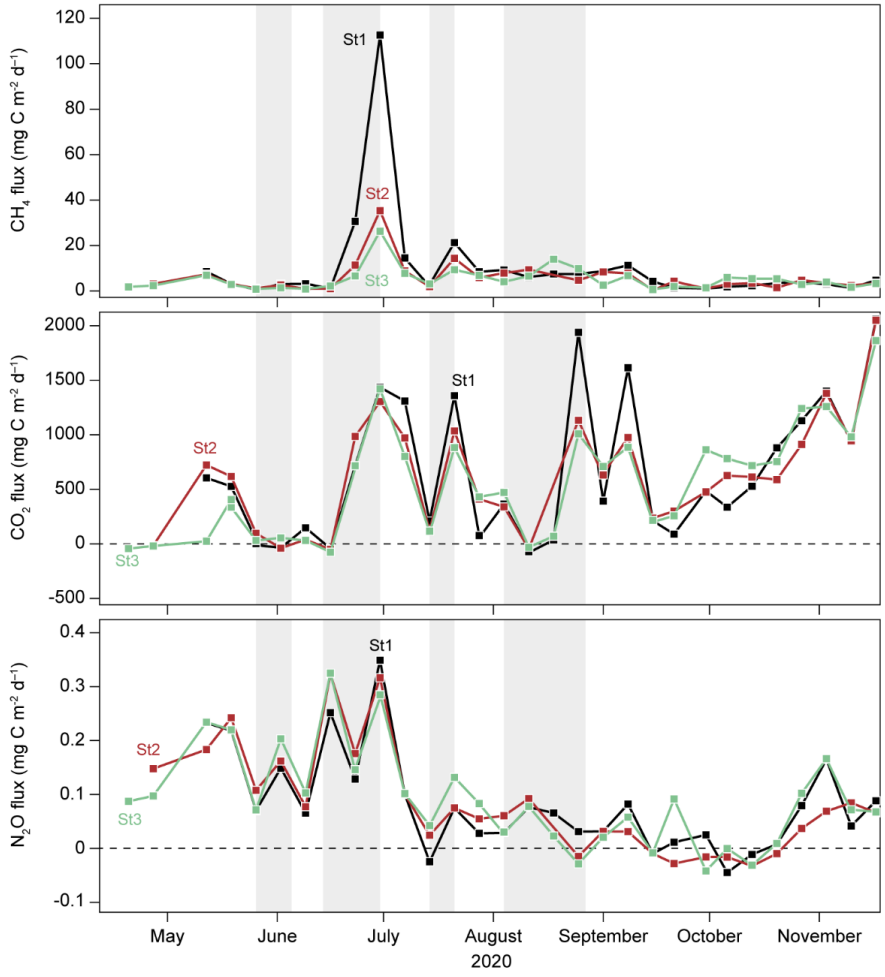

Figure 6. Omstrup lakesurface fluxes of the CH_4, CO_2 and N_2O gases based on dissolved conentration , thermal stratification periods highlighted in grey; white background indicate mixed waters

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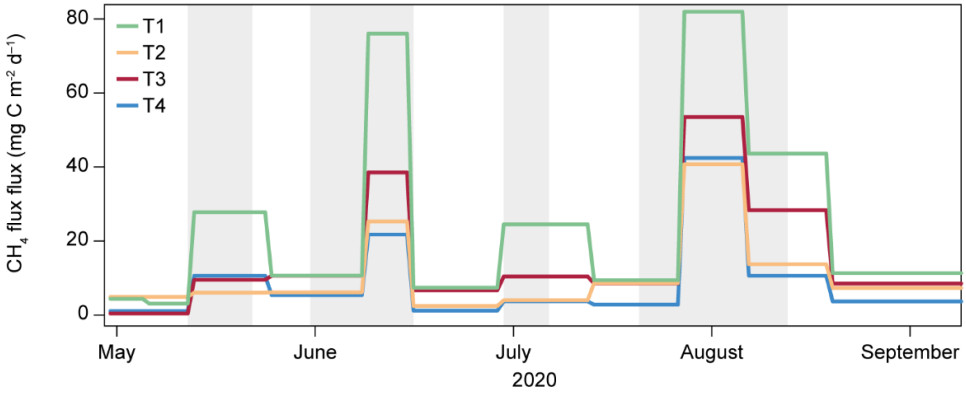

Figure 7. Plot of CH_4 ebullition averaged for each transect (10 chambers per transect), data collected from 40 traps every two weeks. thermal stratification periods highlighted in grey; whitle background indicate mixed waters.

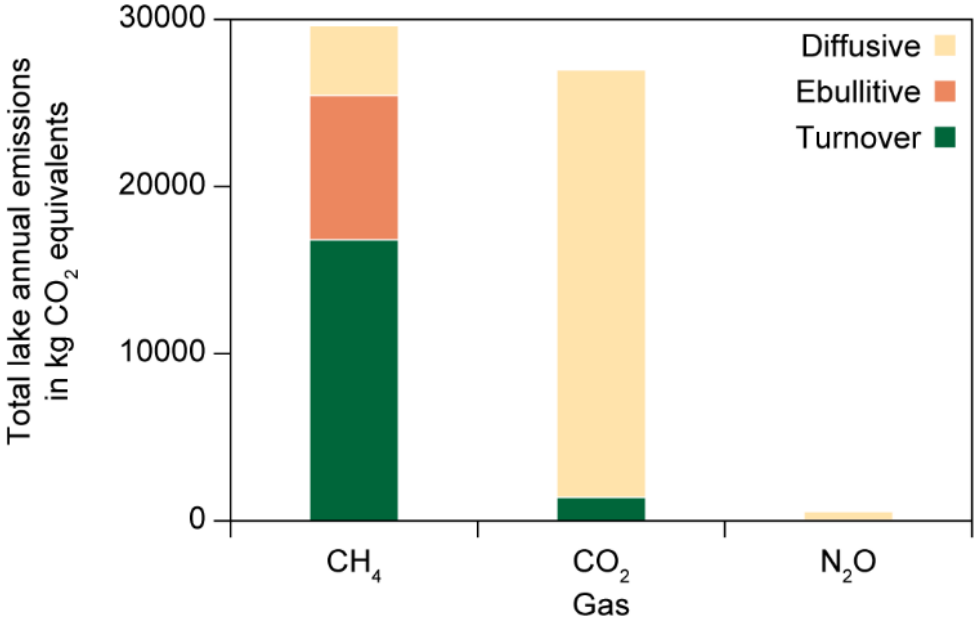

Figure 8 – Total lake emissions per gas over the growing seasson in CO_2 equivalents. The emissions are divided different emission pathways: Diffusive, ebullitive and turnover flux.

https://doi.org/10.5194/bg-2023-43
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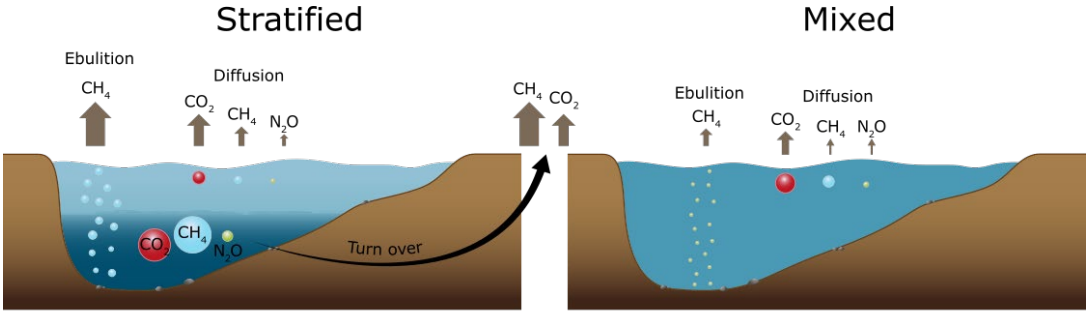

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640 Figure 9 Summary of different flux types (bubble, diffusive and turnover) for the main greenhouse gases
641 (CH_4 CO_2 and N_2O) observed between the stratified and mixed phases at Ormstrup lake patterns in the
642 stratified and mixed phase. The turnover flux of CH_4 and CO_2 is also represented. The size of the arrow
643 represents the relative amount of emission and the size of the circle in the lake represents the
644 concentration of dissolved gases in stratified or mixed water column.

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