Biogeosciences

Discussions

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Interactive comment on "Drivers of diffusive lake CH$_4$ emissions on daily to multi-year time scales" *by* Joachim Jansen et al.

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Received and published: 9 October 2019

Response to reviewer 1:

The authors wish to thank the reviewer for their thoughtful comments and insightful suggestions.

RC1: L 33: Statement "A significant portion of sediment-produced CH4 reaches the atmosphere by turbulence-driven diffusion-limited gas exchange" is misleading and term "significant" is conveniently vague. The synthesis of CH4 fluxes from inland waters given by Bastviken et al (2011) and cited by the authors provides a total diffusive flux of CH4 of 9.9 TgCH4/yr that is much smaller than the total flux of 103.3 TgCH4/yr. I suggest that authors be more specific and introduce quantitatively the importance of

diffusive CH4 fluxes from inland waters.

Author's response: we have changed the introductory paragraph to include the estimated contribution of open water diffusive CH4 emissions from three regional and global budget studies. We note that the Bastviken et al. (2011) study separates 'diffusive' and 'storage' emissions. Because the latter is defined as the 'flux when CH4 stored in the water column is emitted upon lake overturn', and occurs via the diffusion-limited pathway, we counted storage fluxes as diffusive fluxes. We thus computed the contribution of diffusion from the pathway specific budgets in Table 1 of Bastviken et al. (2011) as follows: ('diffusive' + 'storage')/('plant flux' + 'ebullition' + 'diffusive' + 'storage') = 34.8%. This is within the range of values computed from diffusive and ebullitive flux estimates in DelSontro et al. (2018) (21-24%) and Wik et al. (2016b) (46%).

RC1: L36: Chambers also "traditionally" capture CH4 ebullition fluxes in addition to diffusive fluxes.

Author's response: as described in the method section (L. 124-125) our chambers were equipped with plastic shields to prevent bubbles from entering the chamber headspace.

RC1: L44: DelSontro et al. (2018) estimated global (and not regional as stated) CH4 emissions based on a statistical (and not "process-based" as stated) approach.

Author's response: we have removed the reference to DelSontro et al. (2018).

RC1: L 52: the formulation of equation (1) was given by Liss and Slater (1974) well before Wanninkhof (1992).

Author's response: the reference has been changed.

RC1: I have the impression that methane oxidation is the main process "that dissociate[s] production from emission rates", it's odd this is not mentioned in section L69-83.

Author's response: we have included oxidation as one of the dissociating factors.

RC1: L141-143: Can you please elaborate this section? It's unclear how the effect of

artificial enhancement of turbulence was discarded, and how the citation of the Ribas-Ribas et al. paper is relevant in this context, since this technical paper describes an apparatus to measure fluxes with chambers.

Author's response: we included a more detailed description of the analysis of the cited paper: "Ribas-Ribas et al. (2018) compared acoustic Doppler velocimeter measurements inside and outside the perimeter of a chamber of similar design, size and flotation depth as those used in this study, and, based on a comparison of measured TKE dissipation rates and computed gas transfer velocities, concluded that the chambers did not cause artificial turbulence."

RC1: L164: It's strange that only one standard was used to calibrate the GC-FID (a multipoint calibration curve is recommended, Wilson et al. 2018), and the value of standard is so low compared to the sample values, as pCH4 in the headspace was Âż 2 ppm, as shown in Figure 2. Authors should provide an accuracy and precision of the CH4 measurements and propagate this into an error analysis of the CH4 fluxes, as well as for the computed k600 values.

Author's response: detector FID's with N2 carriers are known to be linear over several orders of magnitude (e.g. Colson, 1986). The linearity of the detector is better than the uncertainty in the gas mixtures. The instrument precision is discussed in Section 2.4 of the paper. 10 standard measurements before and after each run were used to assess instrument precision and drift. The precision – defined as the relative standard deviation of the 10 standard measurements - was generally <0.25%. This converts to negligible deviations in the surface concentration and derived fluxes, and would not affect any of the binned or multi-year mean values or functional relationships discussed in the paper. For example, relative standard deviations of the air-water concentration difference binned by time, temperature and wind speed (Fig. 4e-g) are generally >30%. Thus, uncertainty in this study is dominated by the spatiotemporal variability of the fluxes and surface concentrations rather than uncertainty in the concentration measurements.

RC1: L168: Could be useful to explain here how zmix was estimated from the temperature profiles.

Author's response: the mixing depth was estimated from a density gradient threshold, as described at L. 431. We have now written a few sentences about water density calculations and mixing depth in section 2.5. Here is the revised text: "Water density was computed from temperature and salinity (Chen and Millero, 1977), using lake-averaged specific conductivity and a salinity factor [mS cm−1 / g kg−1] of 0.57. The salinity factor was based on a linear regression of simultaneous measurements of conductivity and dissolved solids (R2 = 0.99, n = 7) in five lakes in the Torneträsk catchment (Miljödata-MVM, 2017). We defined the diel mixing depth (zmix) at a density gradient threshold (d/dz) of 0.03 kg m−3 m−1 (Rueda et al., 2007)."

RC1: L206: This equation assumes that Caq remains unchanged during the 24h chamber deployment which seems unrealistic. Please clarify what does Caq correspond to. Was Caq measured each time Ch was measured?

Author's response: Caq is defined at L. 51 and corresponds to the measured CH4 concentration in the surface water. When we compute gas transfer velocities from chamber fluxes and the air-water concentration difference (Caq−Cair,eq), we only use water samples that were collected simultaneously with, and in close proximity to the floating chamber observations. We took one water sample at each chamber location. Thus when we compute kch from Eq. 2, we assume that the flux at the time of the concentration measurement was equal to the 24h flux. It is likely that the flux varied over those 24 hours (Fig. 7b,d). However, a quantitative bias assessment would require continuous, 24h observations of the diffusive fluxes and of the surface concentration, which were unavailable for this study.

RC1: L207: specify if T is the average during the 24h chamber deployment. In Eq [3] explain how dx/dt was computed. Linear regression over all points? Difference between end and start? Difference between each of the samples?

Author's response: we have now specified how T and dx/dt were computed. That is, the average over the flux integration time (this is the 24h chamber deployment time for most of our analyses) and OLS linear regression of concentrations onto time, respectively. Here is the revised text: "$\partial x_h/\partial t$ is the headspace mole fraction change [$10-6$ ppm $d-1$] computed with an ordinary least squares (OLS) linear regression (Fig. 2), M is the molar mass of CH_4 (0.016 mg mol-1), P is the air pressure [Pa], T_{air} is the air temperature [K]. Scalar c_1 corrects for accumulation of CH_4 gas in the chamber headspace and increases over the deployment time. Comparing both chamber flux calculation methods we find $c\_1 = 1.21$ for 24 hour deployments (OLS, $R2 = 0.85$, n = 357). Chambers were sampled up to 4 times during deployment (at 10 minutes, 1–5 hours and 24 hours) which allowed us to compute fluxes at time intervals of 1 hour and 24 hours. P and T_{air} were averaged over the relevant time interval."

RC1: The use of a single value for scalar c1 is surprising because the accumulation of CH_4 in the chamber should depend on the flux intensity itself, so I would expect this value not to be constant.

Author's response: c1 is based on a linear regression of fluxes computed with Eq. 3 (simple linear regression in the time vs chamber headspace concentration plot) and Eq. 2, which corrects for the headspace effect. The good linear fit ($R2 = 0.85$, L. 216) indicates to us that the headspace effect did not change significantly within the range of fluxes observed. If the headspace accumulation effect would increase significantly with the flux, we would expect a highly non-linear correlation.

RC1: In equations 2 and 3, the same symbol (T) is used for water temperature and air temperature, when separate symbols should be used for distinct variables.

Author's response: we have specified the symbols in the equations.

RC1: L 241: It's odd that both a symbol and an abbreviation are used for turbulent kinetic energy

Author's response: they refer to different quantities. As stated at L. 241, we use TKE as an abbreviation for 'turbulent kinetic energy' and epsilon as the symbol for the dissipation rate of turbulent kinetic energy. Both the abbreviation and the symbol are commonly used in the literature.

RC1: L307: Please explain how the residence time of a CH4 molecule in the lake was estimated.

Author's response: this computation is described in section 2.7 at L198-199. The sentence reads: "We computed the average residence time of a CH4 molecule by dividing the amount stored by the lake mean surface flux."

RC1: In Figure 6 the relation between storage time and water T seems significant for I Harrjon and M Harrsjon.

Author's response: we infer that the reviewer refers to Fig. 6e, which shows an increase of storage with water temperature. We fitted Arrhenius functions, added lines of best fit to the plot and included the following sentence in the figure caption: "Arrhenius-type functions (Eq. 7) adequately described the relation between storage and temperature in each lake ($R^2 \geq 0.70$, $p < 0.001$)."

RC1: L637-639: why would damping of turbulence by near-surface stratification affect particularly your lakes but not those reported by Cole & Caraco (1998) and Wanninkhof and Crusius (2003)?

Author's response: we moved the discussion of turbulence damping to a separate paragraph, which now reads: "Damping of turbulence results from near-surface stratification and can reduce the gas transfer velocity (MacIntyre et al., 2010, 2018), however, such stratification was intermittent in our study (Fig. 5f-h). It may also result from our typically having a stable atmosphere in the day for much of the summer which reduces momentum transfer to the water surface."

RC1: An alternative explanation could be fetch limitation (Wanninkhof 1992) in the very

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small sampled ponds, and this effect could be more marked at high wind speeds than at low wind speeds.

Author's response: we thank the reviewer for this suggestion.

RC1: Figure 9: abbreviations given in the plot should be defined in the figure legend.

Author's response: abbreviations of literature references are now included in the figure caption.

RC1: In Figure 9, the binned data value at highest wind correspond to a wind speed that is higher than highest wind speeds of individual Kch measurements. How is this possible? The binned value should be below the highest individual wind speeds measurements.

Author's response: we chose to plot the symbols for binned quantities at the center value of each bin. The center value of the bin may be higher than or lower than the mean value of the datapoints contained in that bin.

RC1: L668-670: While I agree with the idea that CH4 is formed in the sediment, as this seems the most likely process in this type of environments, I do not see why the Arrhenius relation proves this. All biological processes follow Arrhenius-type relations, so the occurrence of this relation only shows that CH4 might be biologically produced, but does not allow to pin-point it as sedimentary. Please rephrase. Since it's not explained in the text how the residence time was computed it is not clear how this proves or disproves a sedimentary CH4 production.

Author's response: we explain how the residence time was computed at L. 198-199. Sediment production of CH4 is well-known in aquatic systems and we do not mean to prove or disprove it with our observations. We restructured the sentence to put more emphasis on redistribution rather than sediment production: "The Arrhenius-type relation of CH4 fluxes and concentrations (Fig. 4b,f) together with short CH4 residence times (Fig. 6) suggest that efficient redistribution of dissolved CH4 strongly coupled

emissions from the Stordalen lakes to sediment production."

RC1: L671: Why do high CH4 in the stream suggest this is of "terrestrial" origin? CH4 is also produced in-stream in sediments. Do you mean that CH4 comes from soils then to streams? Or that the stream CH4 production is fueled by terrestrial organic matter? This statement is very vague and confusing, please clarify.

Author's response: we have clarified this sentence as follows: "High CH4 concentrations in the stream suggest that external inputs of CH4 âĂŤ produced in the fens and transported into the stream with surface runoff, or produced in stream sediments âĂŤ may have elevated emissions in Mellersta Harrsjön (Lundin et al., 2013; Paytan et al., 2015)."

RC1: L677-679: or alternatively from dilution with water with low CH4 from surface runoff and rain?

Author's response: we thank the reviewer for this suggestion.

RC1: L723: methane oxidation is also an important removal process that should contribute to imbalances between production and emission.

Author's response: One could argue that because methane oxidation rates tend to change with concentration and temperature, they would influence the flux on timescales similar to those of production (that is, timescales of a week or more). Changes in storage occur within the short residence times of CH4 gas (1-5 days). This suggest that dissociation occurs on shorter timescales – i.e. those governed by wind speed. However, in this paper we do not present a quantitative assessment of methane oxidation. Following the reviewer's earlier comment we have mentioned oxidation as a process that dissociates production from emission in the introduction.

RC1: L730-740: Wave breaking and bubbles also explain why the relation between the gas transfer velocity and wind speed is non-linear in the ocean (e.g. Wanninkhof 1992), while here you report a linear relation between gas transfer velocity and wind

speed.

Author's response: the processes obtained in large water bodies are not necessarily operative in small lakes. Moreover, it is not clear from our data whether the wind-k relationship is linear or non-linear. At L. 643-645 we state "Due to the large spread of the chamber-derived gas transfer velocities (small rhombuses, Fig. 9) a power-law exponent to U_{10} (1.0 (0.0-1.8); exponent and 95% CI) and thus the nature of the wind-k relation could not be determined with confidence."

RC1: L763: Is thermocline tilting expected to occur in small ponds?

Yes, wind forcing can cause the thermocline to tilt in small water bodies. The extent of tilt is computed from the Wedderburn number (Imberger and Patterson, 1989). This dimensionless index takes into account stratification, wind speed, and basin dimensions. The lakes in this study are larger than ponds, albeit small, and for the relatively high winds found at these arctic sites, thermocline tilting is expected.

RC1: L797-811: Methane oxidation affects CH_4 concentrations, so it's very obscure why methane oxidation should affect the alpha term. This is a scaling between gas transfer velocity that is measured and modelled, and gas transfer velocity depends on physical processes (mainly turbulence)

Author's response: in this study we infer kch from measurements of the chamber concentration increase and surface concentrations. The formulations of Eq. 1 and Eq. 2 implicitly assume that all CH_4 measured in the water column is emitted to the atmosphere. However, if a fraction of CH_4 is removed by oxidation, this would lead to an overestimation of $\Delta[CH_4]$ and an underestimation of kch. This in turn affects the alpha term. So oxidation does not impact gas transfer velocities directly, but may bias the gas transfer velocity high if one uses the two-layer model (Eq. 1), as is common. We have added a few sentences to the paragraph to clarify this point.

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