

Interactive comment on “A simple calculation algorithm to separate high-resolution CH₄ flux measurements into ebullition and diffusion-derived components” by Mathias Hoffmann et al.

Anonymous Referee #2

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This paper proposes a simple algorithm to derive the diffusion and ebullition components of CH₄ fluxes in open-water systems measured with high temporal resolution using autochambers. This would be a useful contribution to enhance our current understanding of CH₄ dynamics in wetland systems, but the paper needs to be further revised before being considered for publication in this journal. As this paper aims to describe a method in determining ebullition and diffusion from the concentration trace of autochamber measurements, the authors should spend more time in describing the methodological details of the calculation (e.g. determination of diffusive flux) and justifying the use of this approach compared to other existing ones (e.g. Goodrich et

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al 2011) in estimating ebullition. The authors spent a substantial proportion of time examining the temporal variability of diffusive, ebullition, and total CH₄ fluxes – but these does not prove that the algorithm is working successfully. There is a need to provide further validation of this method in separating diffusion and ebullition through field testing, e.g. the use of bubble traps. Such comparison should be done in this paper to provide a more affirmative testing of the algorithm, rather than in future studies as suggested by the authors in the last paragraph. Also, are there any drawbacks of using IQR of concentration change to detect ebullition events? If ebullition occurs continuously through the measurement period, would the proposed method fail to identify ebullition events due to a consistently, large magnitude of concentration change? Also would the proposed method be able to identify both “major” and “minor” ebullition events? The lab test does not provide a definitive answer to these questions, and further tests with a greater variety of conditions (e.g. pulse vs. continuous injection) are required. Since the algorithm only estimates ebullition as the difference between total CH₄ flux and diffusive flux, accurate quantification of the diffusive components becomes very important. What is the minimum detectable flux of this system? In open-water systems, CH₄ flux is expected to be lower than that in vegetated wetlands – if CH₄ concentration change within the 15-second interval is not observable, the calculation of diffusive flux might be biased that further causes inaccuracies in the estimation of ebullition. Why not determine the ebullition component directly from the time series of headspace CH₄ concentration? These issues should be addressed before the algorithm could be trusted and applied in other autochamber systems for separating the CH₄ flux components.

Specific comments

L41-42 – Were the quartiles and IQR referring to concentrations within one measurement period?

L70 – In open-water systems, I assume vegetation is absent. If this is the case, plant-mediated transport will not be one of the CH₄ release mechanisms.

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L108 – How does one define “medium” and “major” ebullition events? Are there any objective criteria for such categorization?

L134-136 – I think the hypothesis can be further refined. The flux separation algorithm might help tease out the contribution of diffusion and ebullition to overall flux, but itself could not be used to reveal the spatial and temporal dynamics – this is rather achieved by the AC system.

L161-165 – Only 4 chambers along the transect? No replications? Also, what is the shape of the chamber?

L171-173 – What is the rate of gas flow within the AC system? Do the chambers equipped with a vent tube for pressure equalization?

L173-175 – What did “overcompensation” exactly mean? Did you refer to the drop in CH₄ concentration in the chamber headspace – this looked strange to me. Would this mean that the fan is not effective enough in homogenizing the headspace air?

L192-196 – The equation deriving CH₄ flux does not look right – the unit of CH₄ flux shown is $\mu\text{mol C m}^{-2} \text{s}^{-1}$, but in the calculation molar mass of CH₄ is used?

L200-204 – Would appreciate a more in-depth description of the protocol here, as this is the crucial part of the paper. How did the variable moving window work? Were fluxes calculated for various durations of MW within the 10-min deployment period, as long as the rigid outlier test was passed? If this was the case, which one would be chosen to represent diffusive fluxes?

L226 – Were these the volume and area of the chamber or the tub? How much water was added into the tub?

L231-233 – This assumed that all the added gases would be released as gas bubbles without any CH₄ being dissolved in water. How would the authors ensure the absence of dissolved CH₄ in water?

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L237-239 – How long was the chamber closed, and how was CH₄ ebullition converted to amount (mg m⁻²) as shown in Fig. 4? Would be useful to show the time series of CH₄ concentration as well in the lab test.

L242-246 – However, increasing the frequency of concentration measurement might make it harder to detect significant concentration changes for quantifying diffusive fluxes, which could be low in open-water systems.

L251-257 – Would need some elaborations on why this method is better than other existing methods of quantifying ebullition (e.g. Goodrich et al. 2011). Gas traps should still be able to work in shallow water systems?

L257-260 – It is a bit far-fetching to suggest that this method is “applicable to a broader range of different manual and automatic closed chamber systems, instrumental setups, study designs, and ecosystems” without other solid evidence.

L280-281 – Not sure about the claim that diffusive flux shifted to a daytime maximum was valid. Higher CH₄ flux was still observed during the night period between midnight and 6 am.

L281-286 – This might be tested by measuring CH₄ concentration at different water depths. While thermal mixing might be weaker during daytime, this might be compensated by stronger wind and mechanical mixing. Low wind speed at night might contribute to lower diffusive fluxes owing to poor mixing of air above water surface. Also, temperature in July and September did not differ that much – why was the diurnal pattern different between these two months? Further discussion is needed.

Table 1 – No details about these statistical tests were given in the methodology section. How were differences among chambers tested – which post hoc test was used? Please justify the choice of $p < 0.1$ in detecting statistical significance – the norm is to use $p < 0.01$.

Technical corrections

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L80-81 – “if aiming to identify relevant environmental drivers of CH₄ emissions” is grammatically incorrect. Modify as “if relevant environmental drivers of CH₄ emissions are to be identified”

L139 – “Exemplary” field data? Not sure about the purpose of this heading.

L161 – Change “installed as transect” to “installed along a transect”

L175 – Fig. 3 appears below Fig. 2?

L209-210 – The phrase “smaller or larger than the upper and lower quartile \pm 0.25 times IQR” is confusing – do you mean something like outside of the range between the upper and lower quartile \pm 0.25 times IQR?

Figure 3 – This was not exactly a scatterplot of concentrations – perhaps a time series plot would be more appropriate.

Figure 5 – Why would the bars (I assume is CH₄ flux) in the top graph have different colors? What do gray and black colors represent (the bars, not the pie chart)?

Figure 6 – Please show the error bars for the data points. Also, change “Juli” to “July”, and “montly” to “monthly”

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