

## *Interactive comment on* "Drivers of diffusive lake CH<sub>4</sub> emissions on daily to multi-year time scales" *by* Joachim Jansen et al.

## Joachim Jansen et al.

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Response to reviewer 2:

The authors wish to thank the reviewer for their thoughtful comments and detailed suggestions, which helped improve the paper and clarify the narrative.

RC2: This manuscript documents almost a decade of weekly-monthly resolution methane concentration and flux data from 3 sub-Arctic lakes. They found Arrhenius-type temperature relationships with flux and concentration, which has been found before and suggests a strong coupling to methane production rates. They also found that wind shear drove the gas transfer velocity, but on timescales of less than a month while temperature was a driver on timescales longer than a month. They also found

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that stratification only played a small role in storage/accumulation and emissions in general from their systems. The methods are sound and the results are well-detailed, perhaps a bit on the long side. The dataset is quite unique as it is so long. The authors need to use the length of their dataset to substantiate their results more. They find a temperature relationship that has been shown before in quite a few other datasets, but perhaps ones not as long as theirs. Also, they find that convection does not play as large of a role in surface turbulence as has been found in other lakes. How do those datasets compare to theirs? I also strongly suggest the authors structure the discussion to highlight the main takeaway messages from this work.

Author's response: The concerns raised in the reviewer's initial statement have been addressed in our response to individual comments below.

General comments: RC2: 1. The title seems broad as if you are referring to all lakes, but you actually point out in the manuscript many differences between your findings and those of other lakes, for example, in terms of convection contribution to k. I suggest you narrow down your title slightly. You could even highlight more in the title the amount of data that you have. This multi-year dataset is quite unique.

Author's response: We wanted to avoid a long and complicated title. Instead, we chose specify the length of the dataset and the studied lake types in the abstract. The basic physics that control diffusion-limited emissions from water surfaces are common to all lakes though, of course, specifics such as depth and geomorphological setting will be unique to each. We feel that this is a contribution that is broadly useful.

RC2: 2. I think the discussion could do with some restructuring and more concisely define the main points of your findings. The subheadings closely follow the results structure, but this doesn't help the reader easily identify your main points. I like the way you summarized your findings in the first paragraph of the last section (summary and conclusions). I would suggest laying out the discussion with subheadings similar to the structure in that paragraph, at least to start and then edit from there. You also may not

need all the information in the discussion if you find it does not highlight one of your main points.

Author's response: We believe the dataset and the variety of the analyses merits a detailed and thorough discussion. We hope the sections and subheadings as they are currently structured would allow for easy navigation to topical discussions of interest. Noting your point, we added summarizing sentences to some of the paragraphs, restructured Section 4.2 and removed section 4.7 as it does not add to the discussion. Thank you for having us revisit the organization.

Specific comments: RC2: Line 50- should read ', of which the upper boundary.'

Author's response: We changed the sentence in accordance with the reviewer's suggestion.

RC2: Line 72 – did you not include Aben et al. 2017 because it is about ebullition? You don't specifically mention diffusive only in this sentence.

Author's response: Yes, correct. We considered the Aben et al. paper to not be directly relevant to the diffusion–limited emissions focus of our paper.

RC2: Line 101 - 'stochastic tools' sounds too vague here

Author's response: Thanks for pointing this out. We changed line 101 to "We then estimate the importance of these and other flux controls on different timescales."

RC2: Line 129 – I would say 'During the 24 hr period...' to avoid confusion. But why 2-4 samplings? What resolution and why?

Author's response: This information is detailed in section 2.8, where we write: "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." Also the use of a short and longer time sampling provided information on those manual fluxes that might have been more episodic (i.e. affected by sub-daily changes in the

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gas transfer velocity) than the more regular increases that we might expect given our assumptions about diffusion-limited emissions.

RC2: Line 135-136 – you need to define Fch,unsh and Fch,sh here in this sentence (i.e., place the variables after 'shielded' and 'unshielded')

Author's response: Thanks for noting the confusion. We removed Fch,unch-Fch,sh from the equation, as we clearly state that we talk about the difference.

RC2: Section 2.2 – Do you flush the chambers between samplings or leave them the entire 24 hrs?

Author's response: This is clarified in section 2.8. We use the accumulation rate of gas in the chamber headspace to compute the flux, so we don't flush the chamber within the 24 hour deployment period.

RC2: Section 2.3 – Do you flush or mix the 4m long tube before sampling?

Author's response: Yes we do and we clarified this point in the text as follows: "the tubes were flushed by extracting a sample volume equal to the tube's volume at each location and depth."

RC2: Line 196 – do you mean 'offshore' instead of 'nearshore' here since you are differentiating between the littoral zone and another zone?

Author's response: Yes, we consider the shallow littoral zone to be near-shore, and the deeper, pelagic or profundal zone to be offshore.

RC2: Line 198 - make sure the year is correct on the reference

Author's response: Thanks, we have corrected the year.

RC2: Line 205 - define and give units for 'kch'

Author's response: kch is now defined and units given.

RC2: Line 211-212 - why were there some water measurements not taken and which

ones and how many?

Author's response: The design of the initial water sampling program was not intended to facilitate computation of gas transfer velocities. Simultaneous and co-located sampling was introduced in years 2016 and 2017.

RC2: Line 239 – should be 'kmod' specifically in this sentence, no?

Author's response: In our usage here, k refers to the gas transfer velocity in general.

RC2: Line 245 – why do you need to do this qualitative comparison? Why is it important? Author's response: We added a note of explanation with the following sentence: "In this way, we can assess whether the flux relations with wind speed and temperature are reproduced by the model."

RC2: Line 338 – definte ' $\sigma$ init'

Author's response: This term has now been defined in the text as follows: "To allow for comparison between variables we normalized each  $\sigma$ -series by its initial, smallest-bin value:  $\sigma$ norm =  $\sigma/\sigma$ init."

RC2: Line 420 - include in the caption the panel letters for the histograms in parentheses too

Author's response: Yes that will help our explanation, panel letters have been added.

RC2: Figure 4 caption – you need to describe the squares, triangles, and diamonds in the caption itself – all the variables that you are presenting here.

Author's response: Thanks for noting our oversight, symbol descriptions have been added to the figure caption.

RC2: Figure 5 caption – what are the curves you speak of in line 500? Are you sure that e and f are the right panels when you discus the white lines on line 499? What is the resolution in panels c and d?

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Author's response: Thanks for catching this; the white mixing depth lines are indeed displayed in panel f-h, not e-f. We replaced the word 'curves' with 'lines' at line 500. The resolution of the chamber flux and water concentration measurements was approximately weekly. We hope this is evident when looking at the monthly tick mark intervals.

RC2: Table 3 title - need to describe N here

Author's response: The table title has been adjusted to reflect all variables.

RC2: Figure 6 caption – add '(a-c)' after 'residence time' and '(d-f)' after 'storage'. You mention the regressions for residence time but not for storage. Also, it looks as if there could be a trend between temperature and storage (panel e) for at least 2 of the lakes. Was there not?

Author's response: We have included the panel indicators, and fit storage quantities to Arrhenius-type exponential functions in panel e, which describe the data reasonably well ( $R2 \ge 0.70$ , p < 0.001).

RC2: Line 560-561 – the sentence starting with 'On diel timescales.' needs rewording. I don't understand it.

Author's response: Thanks, we rewrote the sentence as follows: "On diel timescales  $\Delta$ [CH4] and kmod were out of phase;  $\Delta$ [CH4] peaked just before noon, when kmod reached its maximum value (Fig. 7b,d)."

RC2: Figure 7 – put a complete legend in panels a and c and state that they apply to panels b and d.

Author's response: We preferred to keep the legend as is to avoid crowding in the left panels, but we changed the symbol colour of the 1-hour fluxes to improve the clarity of the figure.

RC2: Line 612 - what is 'Twater/ice'?

Author's response: Our surface temperature sensors were frozen in the ice in winter. Because we use the whole-year temperature timeseries in our spectral analysis, we specify that this variable reflects both summer and winter variability. In the caption, we now specify "temperature of the surface water and ice".

RC2: Section 4.1 – The subheading 'Magnitude' doesn't explain much. Magnitude of what?

Author's response: We have changed the section title to 'Magnitudes of fluxes and gas transfer velocities'.

RC2: Line 632 – you obtained lower k-values by nearly a factor of 2 compared to what?

Author's response: This is in comparison to literature models. This has now been specified in the text.

RC2: Line 636 – who had the offset at 0 wind speed? You or the literature? Be specific as this sentence is a bit confusing.

Author's response: Thanks for pointing out this omission. We meant that several models in the literature have a default offset at 0 wind speed. We have amended the text as follows: "Part of the difference with the models of Vachon and Prairie (2013), Cole and Caraco (1998) and Soumis et al. (2008) was caused by the offset at 0 wind speed."

RC2: Line 637 - 'Another explanation' for what?

Author's response: Thanks for noting our oversight. This refers to the other explanation for the low k-values found in our study. We changed the sentence to specify this.

RC2: Line 639-640 - how was the atmosphere stable?

Author's response: We consider a stable atmosphere to be those periods when the tropospheric boundary layer being stably stratified, i.e. when the air temperature exceeds the surface water temperature.

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RC2: Line 644-645 – I am confused because you have an equation in Figure 9 caption that has an exponent for u10 with 95% Cis.

Author's response: The equation in the caption was a linear equation, while we discussed a power-law equation in the text. We've now changed the equation in the caption to the power-law equation of Table S1.

RC2: Section 4.2 – delete 'the' in the subheading

Author's response: Thanks for noting this. 'Drivers of flux' sounds better.

RC2: Section 4.2 – this is a very important part of the discussion but I feel it needs a little more work to really bring out your main points. It reads a bit like a bunch of ideas thrown into a paragraph but without linking them all together nor highlighting why these ideas matter. For example, the first sentence states that the temperature relationship with flux and concentration suggests a strong coupling to sediment [methane] production (need that word 'methane' in there). I agree with this statement and it's an important one because you did find some nice relationships there. But the next sentence talks about stream inputs (from your own data, correct?) and then the following sentence is back to how sediment methane production could be enhanced. They seem out of order. Then the last thought about the decrease in CH4 after cold rain events is actually still in line with the temperature relationship you saw but you start this sentence off attempting to state that that shouldn't be the case if there was runoff from fens. This fens part goes more along with the streams sentence from above.

Author's response: We have revisited the organization and added two introductory sentences to the paragraph to add context to the discussion: "Methane emitted from lakes in wetland environments can be produced in situ, or be transported in from the surrounding landscape (Paytan et al., 2015). The distinction is important because some controls on terrestrial methane production, such as water table depth (Brown et al., 2014), are irrelevant in lakes.". We also replaced "cold and rainy" in the final sentence of the paragraph with "rainy", to emphasize that were are discussing horizontal transport processes here. We removed the sentence about terrestrial inputs of nutrients.

RC2: I feel the same for the second paragraph of the section. I think you clarify your point about the difference between your results and those of Read et al. I am actually not sure who had lake in the warmer, lower humidity regions – you or them? Also need to put the 50 w/m2 value in context. At the end, I wouldn't use the word 'expect' because I think you showed this. And I believe in this whole section you should already elude to the fact that these drivers work on different timescales.

Author's response: We have rewritten this section. Read et al. (2012) did not consider Monin-Obukhov similarity scaling in their analysis. When computing dissipation rates with it, wind shear is raised to the 3rd power and divided by depth whereas the contribution from buoyancy flux is only to the first power. With that constraint, buoyancy flux only drives near-surface turbulence when winds have ceased. Figure 4k shows this for our model. Thus, differences in the meteorology between temperate and arctic lakes are not relevant here. On average, the 50 W/m2 represents the value of the net long wave radiation (Lwin - LWout) we've computed during the ice-free season in the Toolik area. We normally measured Lwin and computed LWout as a function of the surface water temperature. For reference, in our arctic work at other sites, net long wave radiation applies to periods with cloudy conditions, as often occur in the Stordalen Mire.

RC2: Line 716-728 – The first sentence of this paragraph reads more like a summary sentence. It's confusing to hear about the feedback before you describe how you got to that point. I would try restructuring this paragraph a bit. I would start with the second sentence and state it like so: 'Higher temperatures led to elevated CH4 concentrations, which in turn increased emission rates, but high wind speed was correlated with high emission rates and low concentrations. In this way,...'

Author's response: We agree and we rewrote the paragraph as suggested by the reviewer: "Higher temperatures led to elevated CH4 concentrations (Fig. 4f) which in turn increased emission rates (Eq. 1, Fig. 4b) but high wind speed was correlated with high

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emission rates and low concentrations (Fig. 4c,g). Degassing prevented an unlimited increase of the emission rate with the gas transfer velocity. In this way,  $\Delta$ [CH4] acted as a negative feedback that maintained a quasi steady state between CH4 production and removal processes throughout the ice-free season."

RC2: Line 744 – add the range of binned means in those parentheses of 0 - 10

Author's response: The ranges have been included.

RC2: Line 784-791 - This is actually one very long sentence. Consider splitting it.

Author's response: Thank you, the sentence has been split per the reviewer's suggestion.

RC2: Line 798-799 – missing a word or something here '....but can limit surface exchange could be responsible...'

Author's response: We have split the sentence to clarify its meaning: "The observed variability in  $\alpha$ ' could be explained by chemical or biological factors that limit surface exchange. Such processes do not affect turbulence in the actively mixed layer, and are thus not accounted for in kmod."

RC2: Line 834-837 – So you don't completely degas the lake, despite shallowness and frequent mixing, but you also don't have storage/accumulation of methane. I am finding a hard time reconciling those two results. I feel this needs more explanation here but also in the discussion where you mention it.

Author's response: Of course there are dynamics in the water column methane concentrations as a result of variability in the loss and input terms. Accumulation is transient – it changes on a timescale of days – and is the result of an imbalance between production and emission rates. Storage increases during long periods of stratification are not due not only due to the reduction in turbulence-driven emissions but also, in the ice-free seasons especially, to higher production rates as a result of elevated water temperatures. We rewrote section 4.3 to provide a more intuitive explanation of these

## processes.

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