

You et al: Methane emissions from an oil sands tailings pond: A quantitative comparison of fluxes derived by different methods

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Response to reviewer's comment:

Reviewer's comments are in black, and authors' response are in blue.

The manuscript has improved considerably since the last version. The authors have implemented statistical tests to their flux comparisons, which makes the results and conclusions of this paper stronger. Figures and their captions have been improved and made more clear. Some mistakes/errors in flux processing have been found and fixed. I only have few minor points to correct before publication.

(1) I am still missing discussion on how the gradient flux method used in this study would benefit future flux measurements. If this gradient flux method relies on the EC setup, why is the gradient flux needed? In general, if one has an EC system running gas flux measurements, the gradient flux would not be needed because it is already directly measured with EC. Thus, gradient flux measurements are generally used in places where an EC setup is not possible. Can the results and methods of this study be applied to future gradient flux measurements? If so, when, where and how? For example, is the Sc function you have given in Eq. 6 applicable to future pond studies anywhere, or could it be used at the same site for gradient fluxes if the EC system is taken down? Add a couple of sentences about the wider use and application of this method to discussion.

Response: The objectives of the project were much broader than just methane fluxes; we quantified fluxes for 68 VOCs, 23 PACs, 12 organic acids, 7 reduced sulfur compounds and ammonia using gradient flux and inverse dispersion methods. These results will be disseminated in upcoming publications. For almost all of these gases, eddy covariance was not an option since the existing or available instrumentation did not have the required time resolution. Therefore we wanted to make sure that our gradient and inverse results agreed with the eddy covariance results, and designed the study to allow for a cross-comparison of all methods based on at least one gas with a clear, strong flux signal, and in this project that was methane.

Yes, the methods shown in this study can definitely be applied to future gradient flux measurements. We completely agree that if a flux can be measured using EC, then that is the preferred method, which is why we used it as our "benchmark". For most gases, that is not possible with current technology, and that is where gradient or inverse dispersion methods become necessary.

We see no reason why the Schmidt number relationship we derived would not be applicable quite broadly, since it is primarily a function of the characteristics of atmospheric turbulence. There may be dependencies that our comparatively large but still quite limited dataset cannot tease out, and for those, additional studies over different surface types and broader stability

scenarios would be required. Since the literature on atmospheric Schmidt numbers is very limited, even our average Sc estimate will be a useful starting point for future gradient CH_4 flux studies.

We have revised the manuscript as follows:

- a) The last sentence of the Introduction has been modified to more broadly reflect the applicability of this comparison study: “This manuscript describes the results of a comparison of flux chambers, EC, gradient and IDM approaches for estimating emission rates of CH_4 , to verify the suitability of these methods for quantifying fugitive emissions from such sources.”
- b) In the Discussion Section 4.4., line 320, we modified one sentence to: “The gradient fluxes of CH_4 agreed well with EC flux in our study, providing a basis for applying the derived K_c values to calculate gradient fluxes for a variety of other gases emitted by the pond (e.g. You et al. (2021)).”
- c) Also in Section 4.4. a sentence was added at the end stating “While the function derived (Eq. (6)) is primarily a function of the characteristics of atmospheric turbulence and should have broad applicability, it is based on a limited data set and should be verified in other settings in future studies.”

(2) Sometimes the results of the t-test are presented as “p-value=X” and sometimes as “p<Y”. Make it consistent.

Response: Thank you for pointing this out. We have corrected them to be consistent as “p” only.

Detailed comments:

L18-19: Suggest to reformulate as “The results show that the larger footprint together with high temporal resolution of micrometeorological flux measurement methods may result in more robust estimates of the pond greenhouse gas emissions.” since the whole pond is actually not measured and this study is about greenhouse gas emissions, not e.g. particle emissions.

Response: Accepted and implemented.

L75: eddy covariance = EC

Response: Replaced.

L112: “...from their means”

Response: Revised.

L231-232: This is stated already earlier in the text, suggest to remove.

Response: Removed.

L302: Replace “less” with “lower”

Response: Replaced.

L342: Mention that these are the chamber fluxes (although evident from the title). It comes as a surprise here that the measurements are done in a bubbling zone. Please mention it already in e.g. methods section when describing chambers, or in introduction.

Response: Chambers were not placed in bubble zones exclusively. We have modified the relevant statements to “in and around bubbling zones” in Section 3.4 line 212, and Section 4.6 line 344.

Sect 4.6: Chambers may indeed overestimate/underestimate a long-time emission estimate (like a yearly GHG budget) due to the lack of spatial and temporal resolution, but they can also be most useful in estimating smaller source areas (e.g. close to shore) and shorter timeframe emissions, especially in remote locations. Add a sentence or two about this in the discussion. At the moment it sounds like you are saying that chambers are not useful at all.

Response: We changed the last sentence of 4.6 to “In conclusion, while flux chambers present advantages in terms of finer spatial and temporal resolution for small sources or locations with high spatial heterogeneity, reliance on a limited number of flux chamber measurements can result in significant year-to-year variability, and spatially integrating methods such as eddy covariance or gradient fluxes will generally provide more representative averages.”

L422: Global warming potential is already defined in Introduction. Define the acronym GWP in Introduction and use it here.

Response: Implemented in line 39 and line 426.

L437: Suggest to replace “during other times of the year” with “throughout the year”

Response: Replaced.