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Geoscientific Instrumentation Methods and Data Systems Discussions

Interactive comment on "Continuous In Situ Measurement of Dissolved Methane in Lake Kivu Using a Membrane Inlet Laser Spectrometer" *by* Roberto Grilli et al.

Anonymous Referee #2

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This paper seeks to demonstrate the reliability of a membrane-based system to continuously measure methane in aquatic environments other than oceans. The authors place their research in stratified lake Kivu, in which water can have very high methane concentrations, and span several orders of magnitude. It is shown how a manipulation of the sensor system can accommodate this large dynamic concentration range, making the presented sensor system a potentially interesting and useful addition to the biogeochemists' toolbox. Such an in-situ sensor system, usable in methane-rich freshwater environments, is of course interesting and relevant, and some of the results are indeed very encouraging, but at the same time the manuscript suffers from some conceptual and structural weaknesses that in my opinion undermine its suitability for

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Discussion paper

publication in the present form.

General points: Introduction: The authors choose with Lake Kivu an interesting, but also particular case. To me, it is unclear, why the manuscript introduction focusses on lake Kivu (given the choice of journal) instead of focusing on the need for continuous methane profiles in aquatic systems to answer a large array of very interesting and pressing questions, including e.g., transport, production/consumption etc.. While this would require substantial changes to the introduction, it would make a much more useful and strong manuscript. The method presented here is different from a previous paper (Grilli 2018) mostly in the altered measurement mode that allows for much higher concentration to be quantified. That this extended range is still not extensive enough to cover the range in CH4 concentrations of lake Kivu is unfortunate but not critical. A review of the concentration range in freshwater systems would help the reader to understand the relevance of the presented instrument modifications.

Methods: Even though much of the general aspects of the sub-ocean instruments have recently been described (Grilli 2018), the general setup must be presented here in detail. Further, it is unclear to me how the authors arrive at a systematic error of 10% for the HydroC. I am missing information for the use of the instrument (e.g. lowering velocity, frequency of measurement), which should be moved away from the results section.

Results: The authors often mix “discussion” and “results” elements which leads to confusion and unnecessary repetition (up to the point where they mention that the “discussion of a certain value can be found in the results section, L269). Any discussion around the underlying reasons for the measurement uncertainties surely do not belong in the results section. Many of the aspects on lake Kivu methane concentrations are unnecessary, and the comparisons of past CH4 measurements in Kivu through in time and space should be better linked to each other (making clear when technical and when ecological reasons drive differences), and synthesized, of course, in the discussion section. To me, it seems odd to compare the average results measured in the surface

waters with other values that, depending on situation (e.g. season and mixing), span 3 orders of magnitude (L 236).

Discussion: basically missing. Much of the discussion (actually found in the results section) oddly focusses on Kivu-specific observations (e.g. temporal mixing dynamics) instead of methodological aspects. The actual discussion section is mostly a repetition of introduction elements alongside some of the uncertainty numbers mentioned in the results. The latter were derived based on the assumption of homogenous methane concentration across the lake (L263), although the distance from the sediments is known to have large influence on local methane concentrations (del Sontro 2018). While this observation is mostly true for epilimnetic waters, it is unclear how if the hydrodynamic features of Lake Kivu allows for omission of this important control of the spatial distribution of CH4 in lakes. Further, as a reader or potential user of such an instrument, I'd be highly interested in performace metrics (e.g. opposed to other techniques and instruments), or reasons for particular performance-related issues (e.g. regarding the carrier gas flow), and these aspects therefore require much more elaboration.

Specific points: Figure 5: panel TDGP, point 70-90m, how can there be a nonlinearity in the interpolation? What value is interpolated?

Figure 6: I understand the quantification of σ for precision and repeatability. However, it seems overly simplified to express the error quantified at one concentration in percent, and then extrapolate the relative error across all orders of magnitude (making very small absolute errors in small concentration).

L90: It would be interesting to know how long the instrument could run with "between 2 and 40 bar" of carrier gas.

L185: general information about complementary sampling campaign and their published results sshould be shortened

L214 N2 calculations from other measured gases is used in a figure and should therefore be part of the method section.

L221 Orange lines in what figure?

L232 I don't understand this piece of information

Technical corrections

Sloppiness with the references does not increase the joy of reading this manuscript.

I am no native English speaker myself, but many terms and expressions seem awkward (e.g., L86 "at the price of", L118 "electromechanical cable", L176, L191 "than", L236 "higher edge", L261 "seeing a background").

Typos

L246 "O2 completely vanished"

L249 reasons

L244 e.g.

L433 replace "retrieved"

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