

RC1:

We thank the expert reviewer for the very fruitful comments and remarks, which helped us to improve the manuscript.

All the remarks been addressed below, and changes in the manuscript have been done accordingly using word track changes. Some grammatical improvements have also been done. The instruments have been named more consistently over the manuscript as SubOcean and HydroC HP.

We hope that the manuscript is now acceptable for publication in *Geoscientific Instrumentation, Methods and Data Systems*.

Reviewer(s)' Comments to Author:

The manuscript describes the deployment of a new sensor for the in-situ measurements of dissolved methane concentrations in Lake Kivu, known for its very high concentration of CH₄, especially in the deep waters. The sensor is based on a membrane inlet to extract the gas from the water, which is then analysed using a laser spectrometer. This technology has been already described in other papers (Grilli et al 2018 (ES&T), Jansson et al 2019 (OS)) and has proven reliable for low concentration measurements (down to the nM) mainly in oceans.

The challenge here is to evaluate the performance of the technology in very high concentrations (several mmol/l). Lake Kivu offers indeed the perfect conditions for this. It appears that some tuning of the instrument was necessary and yet, the authors concluded that the upper limit for concentration measurement was 3.5 mmol/l even after reducing the sensitivity, while concentrations in the lake can reach 18 mmol/l.

One can see the interest of monitoring the dissolved methane concentrations in Lake Kivu for safety reasons but, it may not be necessary to use a too sensitive technique. As presented, the technique based on laser spectrometer cannot respond to the (important) question of the accumulation rate of CH₄ in the deep layers (highlighted in the introduction). Maybe the same technique could be used in a 'high-concentration' mode by using a less sensitive laser spectrometer. Is it possible to use the near IR absorption bands of CH₄? i.e. 1.3/1.6 μm?

Thanks for this interesting and pertinent remarks. As mentioned in the manuscript, we agree with the reviewer that the high sensitive spectrometer working at 2.3 μm (generally used for trace gas detection) is not well adapted for the measurement in Lake Kivu. Moving to 1.6 or even 1.3 μm could indeed help to decrease the sensitivity by one and two order of magnitude, respectively, but it would have required a dedicated development and in this case it would have been preferable to switch to a less complex technique based on direct or multipass absorption probing strong fundamental transitions. The spectrometer used in this work was developed for study the fate of methane in the ocean (see the work Jansson et al 2019 (OS)) for which the high sensitivity was a key feature. The campaign at Lake Kivu was for us more an opportunity to test the adaptation of our extraction technique (which provides fast response time) in a very different and harsh environment (high dissolved gas concentrations and pressures, anoxic environment...). With this work we proved that the extraction technique is well suitable for such environments, and that in the future in situ sensors based on this technique could be used for fast monitoring of those type of lakes.

Another way of development could be to reduce the exchange surface of the membrane. I also wonder if going for a Teflon membrane was a good change as Teflon is known for a better permeability to methane (although there are different types of Teflon but the authors do not give

any precision on this). In fact, I don't think I properly understood this modification of the instrument. A scheme of the membrane block would help the reader.

The membranes used here are made in PDMS. This is now specified in the manuscript (line 90). A drawing of the membrane block was reported in the SI of Grilli et al 2018, and therefore not further added in this work. We decreased already the membrane surface by replacing one of the two membranes with a gas-tight Teflon film (this is now more clear in the text (line 110)).

Regarding the surface measurements, I would have liked some GC measurements as reference (from samples taken in the same time as MILS measurements). I don't think we can use the data from a commercial sensor as reference.

The reviewer is correct, GC measurements at shallower depths would have been very profitable, but unfortunately this was not done during the campaign. However, two other techniques were used at depths below 150 m: an "on site" mass spectrometer sensor analyzed water pumped from different depths developed at EAWAG (Switzerland) and a water sampling followed by GC measurements performed by the group at UFZ (Germany) (Bohrer et al, HESS 2019). The HydroC commercial instrument well agreed with those other two methods up to a depth of 250 m, afterwards a discrepancy was observed with a systematic underestimation of 12% by the HydroC sensor. Further details can be founded in the official report of the campaign [Schmid et al, 2019](#). Here, an extract of the figure presented in the report (section 3.5 showing an intercomparison of the four techniques used during the campaign). Red circles corresponds to the HydroC HP measurements, while the green line corresponds to our continuous measurements.

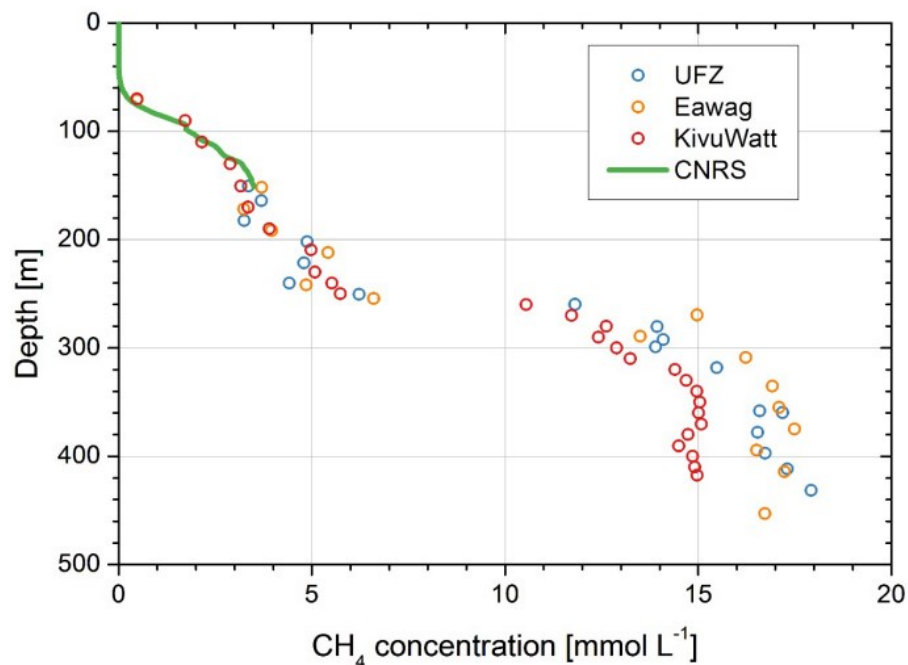


Figure 3.5: Summary of methane concentrations measured in the intercalibration campaign.

This discussion has now been added in the manuscript:

« During the campaign the HydroC HP sensor also showed a good agreement with the other discrete techniques (on site mass spectroscopy and discrete sampling followed by GC analysis) between 150 and 250 m, while at larger depths, the HydroC HP values were lower by ~12% (Schmid et al., 2019). »

One thing we can conclude from this deployment is that in situ sensors must be carefully chosen according to the environmental conditions and the scientific question, and adapted accordingly. Because this paper is a good illustration of the constraints of in situ sensor development and also because the technique is promising despite its limitations when deployed in high concentrations, I would recommend its publication in Geoscientific Instrumentation. However, I have some comments on the form of the manuscript. My main problem is the presentation of the results and the discussion that follows. I would go for a 'results and discussion' section followed by a conclusion instead of the current structure. This would avoid the discussion of some of the results in the results section (line 240-255) and repetition in the Discussion section (line 270). Otherwise, the manuscript is clearly written, figures are well described and clear although I would put the units into brackets, e.g. CH₄/% changed to CH₄ (%) to avoid any confusion.

We thank the reviewer for supporting the publication of our work. The organization of the manuscript and the units have been changed accordingly. Thanks for these suggestions which improved the structure of the manuscript.

Finally, I have only a few typo corrections:

Line 232: 'therefore not...' therefore no...

Corrected

Line 263: 'not spatial variability...' no spatial variability

Corrected