Authors replies to the interactive comments of anonymous referee #2 (5 June 2019) on "High-resolution under-water laser spectrometer sensing provides new insights to methane distribution at an Arctic seepage site" by Pär Jansson et al.

RC: denotes referee's comments

AR: denotes authors' reply

MC: denotes manuscript changes

RC: General comments The manuscript describes how a new technology/sensor can improve our knowledge on the distribution and the dynamics of CH4 over an Arctic seep area. This technology uses a laser spectrometer and a membrane inlet to extract the gas from the aqueous phase. The manuscript is clearly written, results and discussion are well presented, although a bit confusing when it gets to the description of the models (a schematic/conceptual model would have been appreciated). Without any doubt, the lack of in situ, high-resolution measurements of methane in marine environments makes difficult to fully understand their role as a source and/or a sink of methane. This is probably for this reason the contribution of the oceans to the global methane budget has been underestimated. So every effort to develop and test new sensors and technologies must be encouraged. In that regard, the manuscript does represent a significant contribution towards a better comprehension of the marine methane cycle, and therefore, deserves to be published in OS, upon minor revision.

However, I would not say this is a first. Yes it is the first time that this particular technology is deployed in operational conditions – with satisfying results – but this is not the first attempt to get a high-resolution map of CH4 distribution in marine environments. Just to name a few studies on the subject: Sommer et al 2015 (10.1016/j.marpetgeo.2015.08.020), Gentz et al 2014 (10.4319/lom.2012.10.317), Wankel et al 2009 (10.1016/j.dsr2.2010.05.009)... Perhaps, this new sensor has better performance in terms of detection limits and response time, but it's very hard to find them in the manuscript. How does the MILS compare to them?

The development of CH4 sensors has been the holy grail for decades now, and a few technologies emerged from this effort. Each of them were considered as the new solution but I think the main mistake is to believe that one instrument can address the full range of concentrations encountered in the ocean – from 0.1 nM to several mM. This is of course not possible and the instrument must be adapted to the scientific question. In that regard, the MILS seems to be very well adapted to the environmental conditions in which it was deployed. Can the MILS be deployed in oligotrophic waters, i.e. at very low concentrations? And can it measure very high concentrations like in the Black Sea or in the Baltic Sea? One big question at the moment is the role of phytoplankton blooms on the emission of methane to the atmosphere. There are many areas in the open ocean that are characterised by methane anomalies in the upper layer (i.e. the ocean methane paradox). Concentrations are not necessarily very high (up to 5 nM) but enough to oversaturate the upper layer, and therefore create a positive flux to the atmosphere. Is the MILS able to measure concentrations in this range? I think the effort must be now pointed to low concentration measurements. Anyhow, if one can adapt this instrument to lower concentrations, and then if it can be deployed on AUVs (or any other autonomous platforms), then we will definitely advance the knowledge on the marine CH4 budget. The ideal would be to use this kind of instruments for process studies, i.e. in situ measurements of production/ consumption rates, which will further advance the comprehension of the biogeochemical cycle of methane.

AR: Thank you for taking the time and effort to read and comment on our study. You have acknowledged the importance of this type of investigations. We feel confident that we will see more high-resolution surveys of the same type in the future. In your general comments, you specifically asked for a graphic describing the numerical models, which also reviewer #1 asked for. We added an illustration along with a caption as a part of a new supplementary document.
Regarding the instrument capability and how it compares to other instruments, we refer to the study of Grilli et al. (2018). We would like to avoid an explicit comparison of the MILS to other instruments in this study, and leave that to an impartial instrument comparison study. On page 3, we already mentioned the work of Gentz et al. (2014). Additionally, we now mention the work of Sommer et al. (2015), Wankel et al. (2010), and Boulart et al. (2017) in lines 53–57.

The instrument has a specific range of concentrations as you mention, but for instance, the optical spectrometer can be differently tuned or even replaced to improve its sensitivity or to sample more CH_4 enriched waters. The SubOcean (which we call MILS in our study) was deployed in March 2018 at Lake Kivu, measuring up to 3 mM of CH_4. The report from the Lake Kivu campaign is found here: https://www.dora. lib4ri.ch/eawag/islandora/object/eawag%3A18541/datastream/PDF/Schmid-2019-Intercalibration_campaign_for_gas_concentration-%28published_version%29.pdf.
We believe the MILS would be an excellent tool for evaluating CH_4 related water column processes. Grilli et al. (2018) reported a sensitivity of ±25 ppbv in air, translating into ±0.03 nmol L−1 at 20 °C and a salinity of 38, which is low enough for investigations of atmospheric exchange and CH4 production/ consumption rates.

MC: We added a graphic (Fig. SI 1) describing the numerical models in the supplementary document.
MC: The works of Sommer et al. (2015), Wankel et al. (2010), and Boulart et al. (2017) are now mentioned in lines 53-57
MC: In lines 408–411, we added a note on the suitability of the MILS for detailed charting of water column processes and ocean-atmosphere interaction.

RC: Specific comments Line 28: I would rephrase 'contributing to minimum oxygen zone formation, and possibly to ocean acidification, as a result of the oxidation of methane'. This last point is still under debate as it is impossible to evaluate precisely the contribution of methane oxidation to the production of CO2 (again because of the lack of in situ data). And yet, the dynamics of these 2 gases are very different.

AR: To our knowledge, the effect of CH_4 oxidation on ocean acidification is today still unknown, and has so far only been modelled. We have rephrased this sentence.

MC: Rephrased sentence in line 27–29.

RC: Lines 40 to 49. I would moderate the discussion here. I think we should view echosounding as a complementary technique to dissolved gas measurements. The big advantage of the echosounding technique is to locate seeping areas while measuring only dissolved methane cannot help deciphering the sources. As for example, in the Black Sea, concentrations are so high that it is impossible to detect the seeping areas other than using echosounding. One advantage I can see is to evaluate the dynamics of bubble dissolution in the water column as gas bubbles are a mean of transfer of methane from the bottom to the surface.

AR: Clearly, the methods described have their own advantages, and one does not exclude the other. We have edited this section and phrased it differently in order to give a more nuanced picture.

MC: Rephrased sentences in lines 40 - 50

RC: Line 53 I would not put in situ mass spectrometry away so quickly. It is commonly used in deep sea studies, especially in hydrothermal environments. Check Boulart et al. 2017, G3. It may have a slower response time but its main advantage is the ability to detect and measure several analytes in the same time.

AR: The MILS is by no means the only solution to in situ measurements of CH_4, and mass spectrometers has the advantage of measuring different dissolved gas species simultaneously. We now mention the Boulart et al. (2017) survey in the text.

MC: The work of Boulart et al. (2017) is mentioned in lines 57–60

RC: Line 101 What is the autonomy of the MILS? What is the power consumption?

AR: 12 h autonomy at 50W.

MC: We now mention the autonomy in lines 99–100.

RC: Line 101 So the MILS uses exactly the same sample introduction system as in situ mass spectrometers. I guess this is the same kind of PDMS membranes? As the authors wrote, membranes are sensitive to fluctuating water flow. I would add 'pressure of deployment' as well. Membrane's permeability is not the same when deployed at the surface and at 100m depth. How is the pressure effect calibrated? A table comparing MILS' performance with other instruments would be useful here, so the reader can see the advantages of using MILS rather than an ISMS or something else.

AR: Composite PDMS membranes were used. PDMS permeability is not significantly affected by the water pressure at these depths. (Robb, 1968)

RC: Line 114 What do the authors mean by 'careful positioning of the SBE5T'? How do they minimize the pressure change? Is the very close to the membrane inlet?

AR: The pump was positioned about 25cm away from the membrane inlets and connected with short ½" hose sections and a T piece. By shielding the inlet and outlets and mounting them at the same height with an open flow path pressure changes due to movement through the water column were minimised.

MC: We rephrased the sentences in lines 121–123.

RC: Lines 185-195 Was it obtained with the SBE or with the Anderaa?

AR: The vertical casts (Figure 2a) were obtained with the Seabird SBE CTD and the TS diagrams (Figure 2b, c) were obtained from the towed Anderaa CTD.

MC: This is now mentioned in the text (lines 200 and 208).

RC: Section 3.2 Why do the author use 'm above seafloor' as the vertical scale for their casts? This is unusual and can be confusing for the reader. Please use 'm below sea level' for all vertical casts.

AR: We agree that it may be a bit unusual to use 'm above seafloor' for the vertical scale, but we are specifically investigating seepage from the seafloor and found it natural to describe the flow from its source. This approach enabled us to evaluate the distribution resulting from seepage.

RC: When is the pump started during vertical or horizontal casts? I guess it is a continuous flow?

AR: The water pump (SBE5T) was started at the surface, and ran continuously for the duration of each deployment**.**

RC: Lines 349-356 The authors do not mention the possibility of methanotrophy (microbial oxidation), which is the main control of the vertical distribution of methane in the water column. They can refer to the studies in the Black Sea where concentrations close to the seafloor is up to 12 µmol/l. See Schmale et al 2011, BGS.

AR: Methanotrophic oxidation is an important sink on a larger scales and longer time scales, but is locally insignificant at sites with intense CH_4 bubble seepage and high water through-flow and therefore short residence times (Jansson et al., 2019).

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