Authors replies to the interactive comments of anonymous referee #3 (11 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: The manuscript by Pär Jansson et al "High-resolution under-water laser spectrometer sensing provides new insights to methane distribution at an Arctic seepage site“ describes the application of a new methane sensor to methane seeps off Svalbard. As the sensor measures methane in situ with a high temporal resolution a very accurate methane inventory of this probably highly variable area is given. Overall the Ms is well written and straightforward. However, the figures contain too much information, which is either not well explained or not necessary for the specific message, and thus are sometimes rather confusing. For a “non-modeller” I found it sometimes difficult to follow the outline of the applied models. In the discussion, both the technical and the scientific aspects should be discussed, But both are rather short. I would be interested in the comparison with the other commercial methane sensor which was attached to the device. . ... Also an estimation on which temporal resolution is really necessary to dissolve the methane distribution would be appreciated, and what influence has the towing speed on the pattern?? More detailed comments can be found in the attached pdf-file

AR: Thank you for taking the time and effort to read and comment on our manuscript. You acknowledge the need for this kind of research, and recognize the hardship of acquiring useful data, even with the new advanced technology. We appreciate your comments, which we believe improved the manuscript.

Regarding your comment that some of the figures contain too much information, Figure 3 has been split into two parts and the information about concentration gradients have been moved to a supplementary section. We also improved the resolution of figure 4. We prefer to keep the “MILS all” in the figure, since it shows the general vertical distribution, which is not shown anywhere else in the manuscript. We put the inset maps in the figure to visualise the origin of the different data points so it would be easier to see the spatial separation between them.

We now supply a visualisation of the model domains in the supplementary information for a better understanding of the control volume and 2D model, as also requested by two other referees.

The technical details of the MILS has been largely omitted in this manuscript, because the instrument has already been thoroughly described in a previous publication (Grilli et al., 2018) . Here, we evaluate its functionality in this particular environment, with a focus is on what we can learn from the high-resolution measurements. This is why the technical discussion is short.

We agree that a comparison with the reference sensor would be interesting. However, for this publication, we decided to refrain from a direct sensor comparison, as we would rather leave it to a non-biased future publication to compare the MILS, the reference sensor and other CH_4 sensors in a more technical publication.

Regarding the resolution, towing speed and the sampling frequency, we believe that we have good enough resolution, since the MILS picks up the concentration gradients along the ship track. See lines 278–280 and the new Fig. SI 2 in the supplementary information. If we want better resolution in three dimensions, we would need denser surveys, and so it will always be a trade-off between costs and data resolution.

Hereafter is a list of comments and suggestions from the referee, which was posted as pop-up notes in a supplement to the comments. Care has been taken to include all comments and suggestions, and answers given to the best of our ability.

RC: Line 99: At which speed was the ship moving ??

AC: The ship's position was logged continuously and can be found in the file in the data repository. During Line 3, the average speed was 0.79 m/s (1.5 knots) with a standard deviation of 0.065 m/s (0.13 knots).

MC: We added a note on the speed on line 105.

RC: Line 101: Can you give an estimate on the accuracy of this distance ??

AC: We added this information on lines 254–255.

RC: Line 151: What is meant with control volume ??

AR: In engineering literature, a control volume is a region fixed in space and its surfaces are called control surfaces. (e.g. Kundu et al., 2008).

MC: We now clarify it on line 164

RC: Line 186: In October 2015, ...

AR: We rephrased this sentence

MC: Line 200 was changed accordingly.

RC: Line 186: I find the two "depth" or "height" definitions confusing, I suggest to use only one of them…

AC: We changed the phrase so it is easier to read, and are now avoiding usage of two different abbreviations.

MC: Lines 200–201 were changed accordingly

RC: Line 222–225: That is too much information in one figure…I suggest to shift the inlet and additional infos on line 3 into an extra figure… The gradient story is not mentioned in the text, thus if rather confusing here…

AR: We followed your advice and split this information into two figures. Figure 3 now focuses only on the concentrations along the five main trajectories. The caption has been truncated accordingly. A new figure, visualising the gradients, is included in the supplementary document (Figure SI 2). The gradients are discussed on lines 272–283.

MC: New figure 3. Caption of figure 3 truncated. New figure in supplementary document (Fig. SI 2).

RC: Line 226–229 : This technical information should go either M&M or

AR: We now describe this in the methods section, and mention the results in the appropriate section.

MC: Lines 139–141 and lines 236–238

RC: Line 230: Again, there is too much information in the figure 4, which is then not mentioned in the text... please refrain to the important facts. If you only want to compare the vertical casts of CTD 616, 618,619 than all other informations are not needed and are more confusing....

AR: We believe that visualising the different measurements together with their spatial separation is key to understanding the heterogeneity of the CH_4 distribution. It may take some effort to appreciate this figure, but we think it is valuable to show the spatial separation together with the concentration data, in order to realise the distribution of dissolved CH_4.

RC: Line 247: But there are also areas with strong bubble streams but with low methane concentrations ?? For example at the very left side of the figure ??

AR: From our experience with echosounder data, no bubble streams are visible in the echogram on the left side of the figure. Conversely, there are methane peaks without visible flares, which we discuss at some length on lines 374–385. On the right hand side of the figure, there are flares without increased concentrations. This may be due to the fact, that the echosounder swath width is ~40 m at the seabed, while the MILS measures locally. Therefore, it is possible that we passed nearby a flare, which was acoustically identified but that we were too far away to see the CH_4 plume in the MILS data.

RC: Line 249: Again too much information here: what for are the upper inserts needed? And MILS all ?? What about the other DS from the casts along line 3, 1623, 1621, ff ??

AR: See our comment above about line 230. Comparison between the discrete samples from CTDs, and MILS data from line 3 data can be seen in Figure 7, which has been improved also after the request from another referee.

MC: Figure 7 has been improved

RC: Line 250: remove from figure and legend

AR: See our answer regarding line 230 and 249. We believe that this visualisation helps to understand the vertical distribution.

RC: Line 255–259: remove from figure and legend, I think it is sufficient to mention that the CTD casts were xx m away.

AR: See our answer regarding line 230 above.

RC: Line 262: The blue line is hardly visible, but propably also not necessary as already shown in figure 1.

AR: The blue line does not stand out very well in this document, but it looks good in the original figure. We think it will be clear in the final version without any changes. The instrument position is shown in Figure 1, but the depth is not indicated. The depth is shown in Figure 3, but has no reference. In this figure, it is presented to scale with the echosounder data, which we believe is important for the interpretation.

RC: Line 265: I do not understand what is meant with upstream and downstream gradient, and thus also can not follow your conclusions…

AC: We have removed the gradients from figure 3 and moved this information to the supplementary information. In the new figure (SI 2), the upstream/ downstream gradients are explained and visualised. Thanks for directing our attention to this. It was not very clear earlier.

MC: New Fig. 3 and new Fig. SI 2.

RC: Line 268–276: ???? I can not follow here and I am not sure if this information on the instrument characteristics is necessary here, as this should have been done in the previous publication and not its application now…

AR: Here we argue that the instrument has good enough resolution for this particular environment. It is not about the technical aspects of the instrument itself, but that we managed to resolve the CH_4 distribution by towing the probe with the right settings at the right speed, so it could pick up the heterogeneity.

RC: Line 292: could you indicate the stream / current in the figure ?

AR: Yes, of course.

MC: We added an arrow in this figure and in Figure SI 2.

RC: Line 297: But also the water depth of the instrument was more stable in this area, compared to the fluctuation before and after…

AR: After double-checking the data, we found that the relative standard deviations of the probe depth, salinity, and temperature is lower by factors of 3, 10, and 58, when compared to the upstream (later in time) data. This is consistent with the notion that the "flat profiles" between 10:50 and 11:15 are caused by enhanced turbulent mixing due to bubble streams. The standard deviation of the probe depth dropped by a factor of 3 which is not enough to explain the larger drops in salinity and temperature standard deviations. It is normal that temperature diffuses faster than salinity (see textbooks on ocean turbulence), so the fact that the temperature profile is flatter than the salinity profile has a reasonable explanation. We re-phrased the sentence to describe this feature more accurately. Thanks for noticing that.

MC: Lines 305 – 309

RC: Line 302: but is it a good match ?? the methane peaks on the left side are not resolved in the modell and at the right side the model seems to be shifted…

AR: We are not arguing that this is a good match. It is simply the best of the performed simulations, which are solely based on flare observations and the assumptions of a homogenous, steady water current along the domain, homogenous and constant diffusion etc. We do not expect a perfect match from such a simple model, but find it striking that the model does so well with so little information. The lack of sources for the downstream (left) peaks are mentioned in the discussion. We do not have an immediate explanation for the apparent shift on the right hand side of the figure. It could be due to undiscovered sources, imperfect time lag correction of the instrument data, wrong assumption of homogenous water current or it can be explained by the relatively large swath of the echosounder while the MILS measure locally (see our reply to the RC comment about line 247).

RC: Line 326: I find it difficult to understand how you calculated the average concentration of the specific area and being a non-modeller, when you compare a average shouldn't there be a standard deviation ?? To judge if 47 is about the same as 77 nM. ??

AR: The model domain is now visualised in the supplementary information and the improved Figure 7 shows the discrete and high-resolution data, which underlies the average calculations. The caption for Table 1 explains how the data was averaged. The point is that high-resolution data makes a better estimate for a CH_4 inventory, while sparse sampling can easily over- or underestimate the inventory.

Standard deviation does not make sense here, but one should keep in mind the uncertainties of the methods (4% for the discrete samples and 12% for the MILS data). The model builds on "flare quantification" with uncertainties related to bubble sizes and rising speeds, discussed at length in Veloso et al. (2015). The model has a correlation with the high-resolution data of 0.68, so should be evaluated with care.

RC: Line 353: but still below the pycnocline ... aha.. this was well below the pyccnocline, thus background levels of methane were reached below the pycnocline, which there fore could not act as barrier... Maybe you should re-phrase your argumentation here...

AR: That would be a way of saying the same thing, but it does not help to understand the mechanism. We believe it is appropriate to give a plausible explanation, rather than just stating the obvious fact that vertical transport is inefficient. Here we explain why it is not necessary to have a pycnocline to impede vertical transport of solvents (in this case dissolved CH_4). A small continuous stratification is enough. The argument that wintertime stratification-breakdown can cause sudden emissions of CH_4 to the atmosphere still stands.

Grilli, R., Triest, J., Chappellaz, J., Calzas, M., Desbois, T., Jansson, P., Guillerm, C., Ferré, B., Lechevallier, L., Ledoux, V. & Romanini, D. 2018. Sub-Ocean: Subsea Dissolved Methane Measurements Using an Embedded Laser Spectrometer Technology. *Environmental Science & Technology*, 52(18), pp. 10543-10551. doi: 10.1021/acs.est.7b06171.

Kundu, P. K., Cohen, I. M. & Dowling, D. 2008. Fluid Mechanics 4th. Elsevier.

Veloso, M., Greinert, J., Mienert, J. & De Batist, M. 2015. A new methodology for quantifying bubble flow rates in deep water using splitbeam echosounders: Examples from the Arctic offshore NW-Svalbard. *Limnology and Oceanography: Methods*, 13(6), pp. 267-287. doi: 10.1002/lom3.10024.