

## ***Interactive comment on “Continuous methane concentration measurements at the Greenland Ice Sheet-atmosphere interface using a low-cost low-power metal oxide sensor system” by C. J. Jørgensen et al.***

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Dear Anonymous Reviewer #1. Thank you very much for your help in improving the manuscript. Please find our detailed point-by-point to your constructive criticism of our manuscript in the included file "Combined point-by-point responses to reviewer's comments"

"General comments This paper presents laboratory and field calibrations of a low-cost MOS for the measurement of methane concentrations in air. Whilst laser spectroscopy

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is currently the state-of-the-art solution for high-precision measurements of trace gases such as methane, this technology is expensive and ill-suited to remote, hostile environments such as the Greenland Ice Sheet, which is the study site of this paper. There is a great need to develop low-cost, low-power, rugged sensors capable of operating autonomously in remote locations and this is particularly critical for Arctic ecosystems where the effects of climate change on greenhouse gas emissions are believed to be much larger than at lower latitudes. This paper compares a low-cost MOS with a state-of-the-art Picarro cavity ringdown spectrometer (CRDS), using the latter as a benchmark, and demonstrates the suitability of the prototype for real-time, in situ measurements. The proof-of-concept study is well-designed and generally adequately documented, and the subject matter is a good match for the scope of the journal. The technology is interesting and I hope that it will be developed further. This manuscript should be considered for publication provided that all the comments listed below are addressed."

⇒ Reply 1: We appreciate the constructive criticism by Reviewer #1, #2 and #3. We have prepared a point-by-point response to each of the raised issues below, and incorporated appropriate changes to the manuscript, accordingly:

"Specific comments: The quality of English is acceptable but efforts should be made to shorten sentences throughout the manuscript."

⇒ Reply 2: Ok.

"Line 61: the MOS would not directly inform on climatic feedbacks. Please shorten the sentence to ": : sensing element for future studies into CH<sub>4</sub> emissions from the subglacial domain under the Greenland Ice Sheet."

⇒ Reply 3: Suggestion has been followed. The revised sentence is: "This was done to assess the MOS's potential for serving as a sensing element for future studies CH<sub>4</sub> emissions from the subglacial domain under the Greenland Ice Sheet. "

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“Line 80: what is an open-ended enclosure? (A)”

“Lines 85-91: Where/how was the air fed to the MOS sampled from? Through a 50 m tube, independently of the CRDS? If so, the sampling rate, and hence flushing rate of the MOS enclosure, would have been > 3 times that of the CRDS. My interpretation of this is that the autonomous setup would have been different from the calibration one and you would no longer compare like for like (direct comparison?). Please explain. (B)”

“Line 131: also refers to comment above. “Parallel measurements: : .”; the setup is still unclear to me. Did you use separate sampling lines for the CRDS and the MOS? (C)”

⇒ Reply 4: A combined reply has been prepared for the three above reviewer comments (A,B,C)

Two different configurations were used depending on the measurement period:

1) Field calibration period where parallel measurements were done with the CRDS and MOS connected in series. In this configuration, a 50 meter plastic tube connected the subglacial sampling point to the inlet of CRDS. Here, the sample gas passed through the internal pump of the CRDS to the measurement cell before exiting the outlet port of the CRDS. The outlet port was connected via 1 meter tube to enclosure where the MOS was placed. 2) Autonomous measuring period where the CRDS was replaced by a small 12 volt diaphragm pump (inlet of pump connected to the sampling point and outlet of pump connected to bottom of enclosure).

In order to make this more clear as well as to accommodate the general advise of shortening sentences. the 2nd and 3rd paragraph of section 2.1 has been revised to the following:

“Real-time reference concentration measurements of CH<sub>4</sub>, carbon dioxide (CO<sub>2</sub>) and water vapor (H<sub>2</sub>O) was obtained using a CRDS (Ultraportable Greenhouse Gas Analyzer, Los Gatos Research, USA). The inlet port of the CRDS was connected to the

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subglacial sampling point via a sampling tube (50 m length, inner diameter of 4 mm and total volume of 630 mL) which was zip-tied to the aluminium pole. Flow of sample gas from the subglacial sampling point to the measurement cell in the CRDS was obtained via the analyzer’s internal diaphragm pump (800 mL min<sup>-1</sup>). The outlet port of the CRDS was connected in series via a 1 m plastic tube to a metal can enclosure (400 mL), where the lid had been removed (Fig. 2b). The prototype CH<sub>4</sub> sensing system (MOS) was placed in the metal enclosure, where the short serial tube connector ensured a rapid flushing of the headspace in which the CH<sub>4</sub> measurements with the MOS were made. Due to the non-destructive sampling principle of the CRDS and the rapid flushing of the headspace volume in the enclosure with the MOS system (2 times per minute), the concentration of CH<sub>4</sub> is estimated to be virtually identical at the same time step for the MOS and the CRDS during the entire field calibration period (22nd to 26th July 2018).

Following the field calibration test of approximately 100 h, the MOS system was left in the field as an autonomous monitoring system. For this autonomous measurement period, the CRDS was replaced by a 12 volt diaphragm pump (Thomas pumps, 1410VD DC) with a constant air-flow of approximately 3 L min<sup>-1</sup> attached to the common sample tube with similar connection of the pump inlet and outlet as the CRDS ports. During this period the MOS system was powered by 12V LiFePO<sub>4</sub> batteries connected to solar panels and a voltage regulator, placed in a water-proof case and buried under a pile of rocks to minimize the impact of sunlight induced temperature variations of the sensor system. ”

Figure caption of Fig.2 has also been updated for improved clarity.

Also, the wording “Parallel measurements” has been changed throughout the manuscript to “simultaneous measurement” to avoid the potential ambiguity of whether the CRDS and MOS were connected in series using a common sample tube (as were the case) or in parallel using different sample tubes (which were not the case).

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“Line 147: I don’t understand why 0.042 is more complicated than 0.05. What uncertainty does rounding up (why not round down to 0.04 which is nearest?) add?”

⇒ Reply 5: In a sense, the reviewer could be right that it defies its own purpose to do an optimization for a best values, and then round it up afterwards. We have revised the data smoothing with 0.42 for both dataserie, and updated the figures accordingly.

“Line 165: a graph illustrating the differences in model parameters would be useful. How significant are the differences between lab and field calibrations? In line 115 (lab calibration) you mentioned that the temperature was kept constant at around 22 °C. Was there a temperature effect in the field calibrations? Please, comment.”

⇒ Reply 6: The environmental conditions between the controlled atmosphere of the laboratory and the uncontrolled field conditions in Greenland are of course significantly different, which is one of the reasons why field calibration of the MOS seems necessary, unless we work out a better way to do a generic standard calibration. During the field measurements used for the calculation of the  $R_0^*$ , air with a CRDS confirmed CH<sub>4</sub> concentration of the atmospheric background (approx. 1.9 ppm CH<sub>4</sub>) was sampled within 10 meters of the ice margin where meltwater and CH<sub>4</sub> emission was absent. The exact temperature and relative humidity of this air mass is unknown, but likely within the range between 1-4 °C and above 90 % RH. The text in section “2.4 Field calibration of the MOS” has been revised to include this information.

“Lines 187-189: I do not understand the relevance of discussing the response time of a similar MOS, unless by similar you mean same model, different unit. Furthermore, the response time range (1-30 minutes) is massive compared to the CRDS (< 1 Hz). Considering the large differences in response times, you would have to take into consideration the temporal buffering introduced by the pumping rate, particularly where the Picaro is concerned (≈ 47 seconds to flush the 50 m sampling tube @ 800 mL/min).”

⇒ Reply 7: Since we cannot be absolutely sure that the model used in the reference is identical to the TGS2611 used in this study, we have followed the advice to remove

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this part of the discussion.

“Lines 220-222: Re. filtering out the fluctuations attributed to micro-turbulence/ dilution of cavity air by influx of ambient air. If the purpose of the exercise is to study the emissions of CH<sub>4</sub> from the cavity, then filtering out such perturbations is justified.

However, this paper is concerned with a field assessment of a MOS sensor, and in this context, characterizing the response of the 2 sensors to these perturbations is of great interest. This ties in with the comment above (response time and temporal buffering). Looking at Fig. 7a, the outliers in the turbulent period are further from the smoothed line for the CRDS than for the MOS. This might be an effect of the faster response time of the CRDS. It would be interesting to choose a longer averaging time ( $\geq$  sample line flush rate + sensor response time) and plot the time series of Fig. 7a and b again. I would like to see this analyzed and discussed rather than just smooth it out.”

⇒ Reply 8: We understand the point raised by the reviewer, and agree that a better understanding of especially the response of the MOS to fluctuation conditions would be great. In the current dataset, we unfortunately have no data to actually quantify the amount and rate of dilution by microturbulens to support such an analysis. Since the ultimate aim of this study is to develop a low-cost low-power system specifically designed to study subglacial CH<sub>4</sub> emissions, we feel more comfortable with proceeding the data smoothing as presented, and thereby avoiding the risk of over-analyzing a dataset with respect to parameters which are uncontrolled.

“Line 284: “very close”; please quantify this statement.”

⇒ Reply 9: See revised sentence under reply 24.

“Lines 295-297: Please tone down this statement. Your study evaluates a low-cost sensor for the measurement of CH<sub>4</sub> in a hostile environment with the potential to lead to a better understanding and quantification of CH<sub>4</sub> emissions from GrIS and similar locations.”

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⇒ Reply 10: OK. The sentence has been removed in the revised MS.

“Technical comments paper Line 57: “: : : and in sensor network grids.” This might require clarification.”

⇒ Reply 11: text has been changed to “...sensor networks.”

“Line 57: change “we have in situ tested: : :” to “we have tested in situ: : :”

⇒ Reply 12: Corrected.

“Line 67: “: : :southern flank: : :” of what?. Terminus does not seem to be the right term.”

⇒ Reply 13: “... at the terminus...” has been removed in the revised MS.

“Line 74-75: this sentence is clumsy and needs re-structuring. Suggestion “Humidity and temperature of the subglacial air were measured every 10 s using a combined sensor (: : :) mounted at the tip of the aluminium pole inserted into the cave. The data were recorded using: : :”

⇒ Reply 14: Good suggestion. Text has been replaced as suggested.

“Section 2.2: could you specify whether the MOS setup was built by your lab?”

⇒ Reply 15: The following has been added: “The final prototype was assembled in the laboratory at Aarhus University.”

“Line 99: “electrical circuit converts” not convert.”

⇒ Reply 16: Corrected.

“Line 100: “were powered” not was.”

⇒ Reply 17: Corrected. “Line 127: “are inversely” not is.”

⇒ Reply 18: Corrected.

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“Lines 132-136: long sentence, difficult to read. Split into 2 parts.”

⇒ Reply 19: Sentence now reads: “Access to a controlled and humidified zero gas was not available in the field. Instead the atmospheric background concentration of CH<sub>4</sub> of the air (approximately 1.9 ppm) close to the ice sheet was used to calculate the average ambient sensor resistance (R<sub>0</sub><sup>\*</sup>) using Eq. 1. The output value of the MOS under these conditions was then used to establish the resistance ratio (R<sub>S</sub>/R<sub>0</sub><sup>\*</sup>) vs. CH<sub>4</sub> concentration field calibration function for the MOS (Fig. 6).”

“Line 168: “which has been reported to scale linearly: : :”?”

⇒ Reply 20: Sorry, incomplete sentence. The revised wording is:

The reason for this difference is unknown, but a possible explanation could be the potential difference in input heater voltage for the MOS sensor (i.e. pin 1 and 4 in Fig. 1), since variations in the input heater voltage have been reported to affect the CH<sub>4</sub> concentration measurements (van den Bossche et al., 2017).

“Line 177: “at the margin of the: : :” not if.”

⇒ Reply 21: Corrected.

“Line 211: “Measurements: : : show: : :” not shows.”

⇒ Reply 22: Corrected.

“Line 268: “: : : while being undetected: : :”

⇒ Reply 23: Corrected.

“Line 284: “departs” means leaves. Use a more appropriate verb.”

⇒ Reply 24: The sentence has been reformulated: “CRDSsmooth data for period 2 fills the data gap between the MOS measurement of period 1 and 3, where the start concentration data of the MOSsmooth concentration data are similar to the concentration level where the CRDSsmooth measurements end

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“Line 313: “: : : which could significantly improve: : :”

⇒ Reply 25: Corrected.

“Lines 314-317: this sentence is too long. Please divide it into two.”

⇒ Reply 26: Corrected.

“Line 325: Remove “very clean” unless you can substantiate its meaning.”

⇒ Reply 27: Corrected.

“Fig. 7: please indicate the temporal resolution of each plot.”

⇒ Reply 28: Temporal resolution is 10 seconds. Info has been added to the figure caption.

“Fig. 8: as in Fig. 7, what is the time step?”

⇒ Reply 29: Temporal resolution is 10 seconds. Info has been added to the figure caption.

Please also note the supplement to this comment:

<https://www.atmos-meas-tech-discuss.net/amt-2019-468/amt-2019-468-AC1-supplement.pdf>

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Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-468, 2019.