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Atmospheric Measurement Techniques Discussions

Interactive comment on "Performance of a low-cost methane sensor for ambient concentration measurements in preliminary studies" *by* W. Eugster and G. W. Kling

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Final response – our replies are given in blue color between the reviewer statements.

Anonymous Referee #1

General Comments

This manuscript describes the evaluation of a low cost, low performance sensor for methane to determine the suitability for its use in a survey application. The intention is to identify some means of surveying extended greenhouse gas emission sources, such as arctic tundra. A network of low cost sensors could be designed to cover the areal extent of the source efficiently. Higher performance (and therefore higher cost) sensors could then be deployed to characterize the most representative points of the source. The manuscript evaluates the performance of a low cost, solid state, metal oxide (MO)-based sensor for methane. Two such sensors were deployed for a season on a floating platform in an Alaskan lake, along with a tunable diode laser (TDL)-based high performance sensor. The MO sensor output is known to have a dependence on both ambient temperature and humidity, and cross sensitivities to other potentially important species, such as CO. The authors have presented their arguments clearly. The supporting figures and tables are sufficient and the language is clear and concise.

Thanks for this positive assessment of our manuscript.

The paper provides a somewhat rare look at some of the issues inherent in designing a network to measure extended natural emission sources. This includes the perspective of determining if less capable and less expensive sensors have any value in surveying for the best sites to locate more capable sensors. The characterization of the individual sensor capabilities in the field is quite useful and the analysis of the initial field data is valuable. In this effort though, there seems to be a need for definition of the metrics for usefulness. For example, in general terms, how much performance can be sacrificed and still identify major trends; can diurnal excursions be measured with what signal-to-noise ratio?

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Reply: In our present text we only examined the mean diurnal cycle over a full season. It remains an open question for which projects a cheap sensor is suitable (hence our terminology with "preliminary" studies) and where a full-fledged (optical or similar) sensor is needed. We will do our best in the revisions to clarify this open question in terms of the "signal-to-noise" ratio.

The performance of the MO sensors were evaluated with respect to one site and not generalized. It might also be useful to determine the economic boundary conditions of the problem for a given site. . . for example. . . how many less-capable sensors can one afford in order to cover the emission source ? Can the required survey measurements still be made even with inexpensive sensors?

Reply: We agree that this would be a next step in the application of such a sensor.

The authors may want to include mention of other highly capable but still economical, laser-based sensors coming available. For example, S. So, A. A. Sani, L. Zhong, F. Tittel, G. Wysocki, "Laser Spectroscopic Trace-Gas Sensor Networks for Atmospheric Monitoring Applications", The 8th ACM/IEEE International Conference on Information Processing in Sensor Networks, Proc. of ACM, vol. ESSA 2009, (2009) as well as others.

Reply: We will clarify the term "low-cost" in our revised version (see also our reply to Reviewer #2) and will also add this reference to the list of other economical sensors. We were unfortunately not able to obtain an estimate of the costs of the sensor by So, Wysocki etc. but it is certainly worth mentioning despite the fact that the price tag will need some extra zeros compared to the solid state sensor.

Specific Comments

There are several points in the manuscript that the authors could provide some additional clarification:

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*Page 2570, line 24 . . . Cross sensitivities. . . Are there cross sensitivities to any hydrocarbons or other volatile organics that might also be emitted from the lake or tundra wetlands?

Reply: We address this question later in the discussion on page 2578 and 2579. We can add a statement in Material and Methods (Section 2.1) to indicate that we are going to address this question in our discussion.

*Page 2574, line 24. . . Does the sensor drift in one direction only so that initial and final calibration measurements will provide a linear interpolation? What is the drift mechanism?

Reply: The manufacturer does not specify any details on this question. Our interpretation is the following: the sensor is actively heated, has a large reactive surface due to the granular micro-crystals of the reactive material (see page 2571, line 14). Most likely during the use of the sensor there will be some loss of sensitivity either via the aging of the heated granular micro-crystals or by some minor dust or reaction product deposition that may reduce the efficiency of the reacting surface of the sensor. This should increase Rs over time, which translates to an apparent increasing trend in CH_4 concentrations (see Eq. (5)) even if CH_4 concentration is kept constant. We however do not have any separate long-term data of such a sensor kept at constant CH_4 over a longer period, but strongly encourage such additional measurements.

*Page 2575, line 12 The correlation coefficients obtained for the two MO sensors are quite low compared to that obtained from the manufacturer's data plot. Is there any understanding of why this is the case?

Reply: From the manufacturer's documents it is not clear, how the relationship of the sensor on temperature and rH should be explained. Namely, since the sensor is actively heated one would rather expect that the wind speed or turbulent conditions at the sensor itself might be an issue since the sensor surface may not heat up to the

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desired temperature under strong wind and high turbulence. At the same time the heat loss from the sensor is also driven by the difference between ambient temperature and the unmeasured sensor surface temperature. In our understanding, the only way to address such issues would be to have the manufacturer produce a combined sensor where the gas sensor and a temperature and rH sensor measuring the conditions at the gas sensor's surface is measured simultanteously. As a first step in this direction we'll start a follow-up experiment this summer where we will place a temperature and rH sensor next to the gas sensor.

*Page 2576, line 17 The authors state they found no consistent lag between the MO and TDL sensors but reproducible lags seem to have been observed with the diurnal data trend (Fig 5). A little more discussion here might help.

Reply: We'll address this issue in the revision, but it is important to take note that we were unable to clearly relate each single data 30-minute average point from the MO sensor with the corresponding TDL concentration values (see page 2577, lines 19/20 and our reply to Reviewer #2 on the same issue), but found a realistic mean diurnal cycle if averaged over the entire measurement period.

*Page 2576, lines 19-22 The magnitude of the observed MO sensor drift is twice that of the seasonal change. What is the smallest seasonal change that can reliably be recovered?

Reply: Good question! We argue in Section 3.4 that an initial calibration and a terminal calibration is required to remove the seasonal trend. So our specification of the trend given on page 2576, lines 18–22 is the result obtained via this approach (difference between two values) and hence is not suited to address this question. If one wanted to use the data in a different way (e.g. not calibrating at the end of the period and simply correct for the drift values we published here), one would need to explore the temporal variation of the sensor drift. Currently we do not have the long-term experience to quantify the minimum trend that could be reliably recovered

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without the calibration suggested in Eq. (6). To be honest, we do not actually recommend to rely on the assumption that the sensor drift is a constant value and the same for all individual sensors. One would need to test this before making a qualified statement to address this question.

*Page 2577, lines 19-20 The R^2 values obtained for the concentration data of the MO sensors with respect to the TDL sensor data were poor. . .supposedly because of time lags. . ..any idea as to mechanism for the lag? If the lag could somehow be removed, the R^2 values would presumably increase. Could a correlation calculation be done that would identify a constant time lag that could then be removed?

Reply: With time series we always use the cross-correlation procedure to explore time lags. In the case presented here the conventional result of synchronized time series was found with the best correlation at lag 0, hence we did not dwell into this issue in more detail. Unfortunately, there is not an easy solution to find a better correlation by using lagged correlation analysis, as can be seen in the figure on the last page of this document which shows the cross-correlation between MO and TDL concentrations (each smoothed by a 60-minute running average filter). The red vertical lines are drawn at 1-day intervals to show the effect of the diurnal cycle, which is also seen in the cross-correlation plot.

*Page 2579, lines 12-13 CO contamination from forest fires could be well above normal ambient, depending on the distance of the observation point from the fires

Reply: Yes, we thought the same and hence included lines 5–9 on page 2578 saying "there is some risk of confounding effects in areas where wildfire or other burning is present. However, during summer 2011, it appears that the uncertainty of the behavior of the TGS 2600 at cold temperatures (around freezing and below) led to the largest discrepancies with the reference concentration measurements". Since we could not find any evidence that there was any fire influence at the time where we had the largest discrepancy between the LGR FMA 100 methane concentration

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measurements and those obtained from the Figaro sensors, we believe that our data are minimally biased by CO contamination.

Technical Corrections

*Page 2572, line 2 RL should be Rs
*Page 2575, line 10 R2 should be Rs

Reply: Thank you very much for pointing out these typos, we will rectify this in our revised version.

We thank Reviewer #1 for detailed and helpful feeback on our manuscript.

Referee #2

J. Kutzner (Referee)
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General Comments

This work reports on the evaluation of a non-specific solid state sensor for the measurement of methane concentrations in air. Measurements of this kind are important and have the potential to initiate a new direction in sensor developments; it would be a breakthrough if low cost solid state sensors could be used to measure the low atmospheric concentrations with sufficient precision. However, if the issues of gas species cross-sensitivity, temperature dependence, and relative humidity sensitivity of these solid state sensors (and others) cannot be solved or if their performance leads to unacceptable results, it might encourage the development of alternative low-cost optical sensor technology.

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Reply: We definitely agree

Although the presented research has several shortcomings it might stimulate discussion about suitable sensor technology for atmospheric research.

Reply: We hope so and will work on the shortcomings in our revision of the manuscript.

The authors report on atmospheric methane measurements. Methane is an important greenhouse gas. Unfortunately the authors do not discuss this importance in sufficient detail and therefore fail to provide the basis for the important contribution that their work might have for measuring this greenhouse gas. For example, methane could be compared to carbon dioxide and concentrations versus heat trapping capabilities could be briefly mentioned. Another important aspect is the short chemical lifetime (about 12 years) of methane in the atmosphere. Methane might have significant implications for the Earth's climate future because rising concentrations have been reported from different monitoring stations over the last few years. Increased release from Arctic regions or tropical wetlands could drive a feedback mechanism. Methane released from hydrates on the ocean floor and the implications of such an event might also be shortly addressed. These points might be referenced in addition to those already mentioned by Dr. Peter Werle.

Reply: We understand the critique, and certainly such a general description of the dynamics of methane could be helpful for some readers. However, we were under the impression that Atmospheric Measurement Techniques rather focuses on measurement techniques and less on all aspects why one would want to use these techniques. We may have been mistaken and will add a short (but not exhaustive) paragraph on the aspects mentioned by Dr. Kutzner in our revised introduction.

Another important aspect missing in the paper is the clear outline of requirements for atmospheric methane measurements. Atmospheric methane concentrations are low; in the 2 parts-per-million range. The challenge for atmospheric methane sensors is to

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measure the concentration accurately but also to measure small changes in methane concentration, i.e. changes in the 10-parts-per-billion range (or below). This is a challenge even for high performance optical sensors. The “signal-to-noise characteristics” of an optical sensor must be in the 1:1000 range to be suited for atmospheric methane measurements.

Reply: Maybe we should have compared such a sensor with the concept of passive samplers used for O_3, NH_3, NO_2 etc. Although one can sample such a sensor at relatively high temporal resolution, we may leave a wrong impression by even thinking of achieving the quality of an optical measurement principle with such a low-cost sensor. What we will do in our revision is to add a paragraph that more clearly refers to the concept of passive samplers (where rough longer-term averages, e.g. weeks or months are targeted, not the short-term fluctuations). Still, we agree that it is a challenge to measure CH_4 with high accuracy.

The authors report decent results that they achieve with “low-cost” solid state sensors operating in the field. These sensors are not intended to measure extremely small concentration changes of methane. Therefore the presented study represents a first approach to evaluate such sensors for such demanding tasks. Only relative measurements are possible with the solid state sensors and calibration measurements require a high performance methane analyzer. However, the argument using many low- cost sensors for large area coverage is attractive. Two solid state sensors based on the Figaro TGS 2600 were run parallel and the results compared to a high performance optical methane sensor from Los Gatos Research. Main problems of the solid state sensors are outlined as cross-sensitivity to other gas species as well as temperature and relative humidity sensitivity. Cross-sensitivity to other gas species is discussed in detail. However, the measurement location in Alaska essentially eliminates cross-sensitivity. Temperature and relative humidity sensitivity of the sensor seems critical. This is addressed and the data have been corrected for both. An important aspect here is that the “methane sensor” used for the

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presented work consists of the Figaro TGS 2600 sensor head and also other components. How was this sensor operated? Important details are missing in the description of the experimental setup. How is the temperature/humidity sensor placed? How well does it reflect the temperature of the sensor head? What are the specifications for the temperature and relative humidity measurements? Are they relevant at the sensor head location? What are expected uncertainties? The expected achievable measurement precision based on the determined temperature and relative humidity sensitivity should be discussed.

Reply: We have similar questions, but some of these cannot be easily answered. The operation principle is only vaguely described in the documentation of the manufacturer and it is hence not trivial to claim that the user of the sensor has obtained full physico-chemical understanding of the working principle (the authors included); because of the limited detail available from the manufacturer, our work has a somewhat empirical character. With respect to the location and accuracy of temperature and relative humidity sensor we come to the same conclusion that maybe it is more important to measure the temperature and relative humidity next to the sensor (and hence compromise on absolute quality of measurement for ambient conditions), but even then it is clear that since the sensor is actively heated, the temperature of the sensor surface is always different from the temperature of the air sample. Therefore we wonder why this type of sensor is not constructed with a built-in temperature and relative humidity sensor in one single housing. Such questions remain, but we will address the issue in our revisions to clarify to the reader in which direction the next step of improvements should go. We will also add a sensitivity analysis (boot-strapping) to quantify how uncertainties in temperature and relative humidity measurements translate to methane concentrations derived in the way we suggest in our paper.

The “methane sensor” presented here is the sum of multiple components and, due to the strong sensitivity of the TGS 2600 to temperature and humidity, the overall

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sensing system might be limited by the ability to measure temperature and relative humidity with sufficient precision. This is only speculation, but the TGS 2600 might not have been the limiting component of the sensor.

Reply: With our ANOVA we tried to address this issue and found that relative humidity explains 21.3% of the variance in the sensor signal, air temperature 12.7%, and methane 18.0%. So, methane and relative humidity seem to have similar weight in the signal strength. This suggests that despite all concerns about the strong dependence of the sensor signal on temperature and relative humidity, there is still a visible methane signal even at ambient concentrations. We think that it is worthwhile to try to improve concurrent temperature and relative humidity measurements (we actually have started with this already in a follow-up project). For the revised paper we will add a sensitivity analysis (see reply above) to help provide additional insight into this topic.

Specific Comments

This work reports the performance of a solid state based methane sensor during field measurements. These evaluations are important, however, before a sensor is ready for these evaluations thorough testing in the laboratory under defined conditions should be performed.

Reply: We see a shortcoming here that the sensor needs a minimum level of humidity to provide a signal that correlates with methane concentration (our Fig. 3, basically at least 35–40% relative humidity). In a previous project we had a similar issue with a photoacoustic analyser where we actually did such a thorough testing in the laboratory to only find out that although under laboratory conditions our photoacoustic sensor was partially able reproduce the CH_4 concentration, it completely failed under real-world conditions. This is a general problem of nonlinear sensors that respond to multiple influences. We do not believe that a simple linear combination can solve this issue, hence we have not done this, but we would welcome others to demonstrate that

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this approach does work with such a multi-gas sensor.

Questions of how stable the sensor is and what the limiting parameters are can be more easily addressed in the laboratory than in the field. For example the question of sensor pressure dependence is not addressed at all.

Reply: We have ambient pressure data and reran our ANOVA (Table 2) with atmospheric pressure as an additional factor. In this analysis the explained variance due to pressure variation is less than 0.04% and hence two orders of magnitude below the factors that we included in our analysis. We will hence expand Table 2 (where we actually address the question of CO_2) by also including pressure. Since this is a negligible component we will only briefly address this topic in the revised text.

What does low-cost mean? As mentioned above, a "complete" sensor should be discussed and not only a component of this sensor. What would be an estimate for a methane sensor based on a TGS 2600 element? How does it compare to an optical sensor, for example the Los Gatos Research FMA 100?

Reply: In the set-up we had, the Figaro sensor is approx. US$ 50 and the temperature/humidity sensor roughly US$ 700. We are currently experimenting with a low-cost temperature/humidity sensor that also only costs around US$ 50 but could be placed next to the Figaro. For data acquisition we used a Campbell CR1000 data logger, which is currently the most expensive component (order of US$ 1000), but we also have started to experiment with Arduino systems (http://www.arduino.cc/) that should allow us to build a system that costs on the order of US$ 200 (without labor). We realize that we get quite some feedback on the term "low-cost". We'll have to specify this more precisely in our revisions, since we learned that some of our readers consider a methane sensor that costs US$ 25,000 as a "low-cost" sensor already. We'll clarify this in our revision.

A paragraph where optical sensors are compared to solid state sensors would be helpful. The current trend is to replace solid state sensors with more reliable optical

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sensors. How well is the FMA from Los Gatos suited for atmospheric methane measurements? What are the specifications of this device?

Reply: It's probably one of the best and most reliable CH_4 sensors on the market, so we can easily elaborate on this issue and add additional references in our revised version. We have published quite a detailed investigation on this sensor in Eugster & Plüss (2010) (referenced e.g. on page 2572, last line) but in the meantime there are more studies and white papers that could be referenced.

I recommend avoiding the term 'cross-sensitivity' for temperature and relative humidity and reserve this expression to refer to 'different gas species'.

Reply: We can modify the terminology accordingly.

Sentence in line 19/20: This sentence should be reformulated. Not just increases in the atmospheric concentration of methane contribute to global warming; the current atmospheric concentration itself is also a contributing factor.

Reply: Yes this is correct, the current methane concentrations contribute to trapping heat near the Earth's surface, and any increases in methane will increase this contribution. We will reword this sentence to make it more clear.

2.2 Principle of Operation: The TGS 2600 is mounted in a TO5 package. This package is well known in the laser community and might be a better description than used by the authors.

Reply: We'll change the wording accordingly.

Figure 5: How can the mean diurnal cycle data be so good with such small error bars so as to fall within the size of the marker? How were these data extracted? The Fig. 4 data suggest much larger errors because the data vary between 1.87 ppm and 1.98 ppm.

Reply: These are standard ERRORS (not standard DEVIATIONS), that's the

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difference. As stated in the text only the MEAN diurnal cycle (hence the standard ERRORS in the graph) can be extracted from the data, for individual days the scatter is too large as you correctly saw in Fig. 4.

Two TGS 2600 sensor heads were run parallel. The agreement shown in Figures 4 and 5 are impressive. Does that result from temperature and RH measurements being measured with the same device? In that case uncertainties in temperature and relative humidity determination would be propagated in the same way into the measurement results. This might indicate that the TGS 2600s are better than they appear in the presented measurements, i.e. when results are compared with the Los Gatos instrument. A different set-up where every TGS 2600 has its own temperature and RH measurement device might lead to completely different results.

Reply: This is an interesting thought, but consider that (a) if two Figaro sensors provide reproducible results in an inter-comparison, and (b) two independent temperature and relative humidity sensors produce reproducible results in an inter-comparison, then (c) also the methane concentrations measured in this way must end up being comparable (reproducible). Since temperature and relative humidity sensors are actually quite OK for the application of ambient measurements; hence we do not believe that one would yield "completely different results". But it will be interesting to test and we actually will add an additional T/rH measurement in a future study to address this question.

The traces in Figure 1 are too thick to reveal any sensitivity for methane in the low ppm concentration range. A look at Figaro's web site reveals that sensitivity.

Reply: We'll make the traces thinner.

The Figures 4 and 5 show data with surprisingly good agreement of the solid state sensor data with the Los Gatos instrument. On the other hand the authors report less favorable results for the pairwise agreement of all data points from the TGA 2600 sensor with the Los Gatos instrument. A graph with less favorable data would help

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understand the real performance of the solid state methane sensor.

Reply: On page 2577, lines 19/20 we stated that the R^2 is 0.195 and 0.191 for the two sensors. Of course one could add the random scatterplot with weak correlation, but we feel that it is clear from the low coefficient of determination stated in the text and the scatter seen in Fig. 4 that an additional graph would not lead to further understanding of the differences in the sensors.

Discussion of the temperature and RH dependence of the TGS 2600
From (5) it can be concluded that R0/Rs changes by 0.0288 per ppm CH4. The reported diurnal cycle difference is in the range of 0.01 ppm. For 0.01 ppm CH4 R0/Rs should change by 0.000288. According to (3) R0/Rs changes by 0.0072 per % RH. What can the CS215-L12 measure? Although the output resolution is specified as 0.03% the given short term hysteresis of $<$ 1% RH might indicate that this sensor cannot measure the relative humidity with better than 1% precision in the long term. That would imply that the sensor cannot resolve changes smaller than 0.5ppm based on the implemented measurement capability for the relative humidity. According to (3) R0/Rs changes by 0.0246 per °C. How precise and reproducible can the temperature be measured? Resolving 0.01 ppm methane would require determining the temperature with a precision of 0.01 °C in the long term. What are the specifications on the long-term temperature precision? The temperature in the sensor head is probably relevant. How realistic reflects the recorded temperature the temperature of the sensor head? These simple considerations show that very high precision in determining the temperature and the relative humidity are required. The question remains why the solid state methane sensor can measure effects of a 10 ppb concentration variation? Does the averaging compensate for temperature and humidity uncertainties?

Reply: We see the viewpoint of this reviewer who has a background in high-quality instrumentation where each single data point conforms to a high level of accuracy and precision. We also realize that the statistical concepts must be described in more

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detail, although they are standard: In our Fig. 5 we take the point that if the diurnal cycle is a recurring feature within one and the same statistical ensemble, then the replicated measurements of that cycle over many days helps to carve out the shape of the curve (hence the standard error of the mean of each hour in our Fig. 5). So, we think that more clearly introducing the reader to the concept of passive samplers as a proxy for accurate measurement obtained over longer time periods (weeks, months) would help to avoid such misunderstandings.

In conclusion, the presented results seem to indicate that simple solid state sensors are capable of monitoring the variation of very low concentrations of methane. Open questions remain which must be addressed in further detailed studies.

Reply: We thank Dr. Kutzner for this careful assessment of our manuscript. We completely agree with his concluding statement, and if this is the message that the reader takes home then we were successful to express the potential and limitations of such sensor, and how they could beneficially be deployed in combination with more expensive high-quality sensors in field studies.

Referee #3

H. Adam (Referee)
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General comments

A strategy of deploying low cost gas sensors for screening studies of Greenhouse Gases in remote locations is obviously desirable. It's impressive that the authors were able to demonstrate good correlation between the TGS 2600 and the LGR reference analyzer – even if only for a limited sample of the acquired data set.

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Specific comments

1. The significant amount of data manipulation and resulting corrections seem limited in their utility to temperature and humidity ranges which exist during only a minority of the time at the high latitudes of the field location. Perhaps these are the only times of the year during which CH_4 variations are important and require to be monitored; but this point is not emphasized in the discussion.

Reply: Yes, we agree that we only covered part of the year as it is typical in arctic field research where the ice-free season or vegetation period is of interest. Because CH_4 is produced by biological activity which in turn is a function of temperature, knowing the summer season methane concentration developments is important. We accept that this should be emphasized in more detail in the discussion and we will add a short explanation on the seasonal evolution of CH_4 concentrations in the arctic environment. Of course one idea of using low cost low power consumption sensors in the future would be the measurements over longer times (including winter, if the sensor really works under cold conditions) to investigate, whether unexpectedly large CH_4 emissions (which translate to high CH_4 concentration peaks) can also occur during periods where one would normally not perform measurements.

2. The cost-benefit of deploying the specific TGS 2600 gas sensor is neither discussed nor obvious. The balance of equipment and logistics costs associated with such screening studies at locations like Toolik Lake is considerable. Are the incremental savings produced by using the low cost gas sensors, with the associated data corrections and resulting limited data availability worth the effort compared with using admittedly more expensive CH4 sensors that would require less data manipulation and provide greater data availability (including over greater range of temperature and humidity)? Quantitative analysis of such economics would be desirable.

Reply: Reviewer #2 expressed a similar critique which in our understanding is related

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to the lack of clarity on what "low-cost" actually means. Please see our reply to this point of Reviewer #2. We think that we can address this issue by explictly referring to the passive sampler concept used for other components and specify the costs for reference.

We thank Dr. Adam for this feedback.

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Interactive Comment

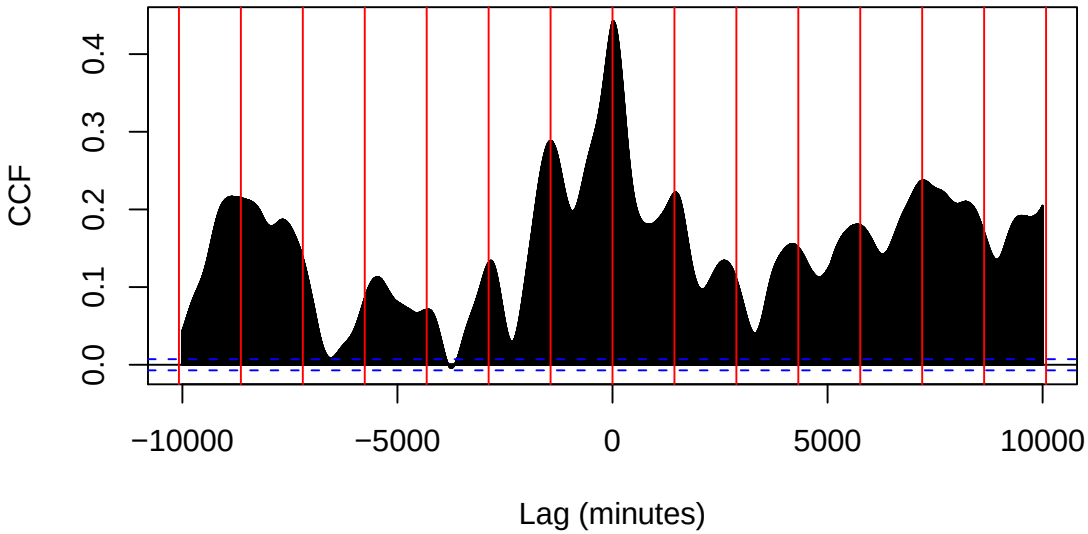

Fig. 1. Cross-correlation between MO and TDL concentrations (each smoothed by a 60-minute running average filter). The red vertical lines are drawn at 1-day intervals to show the effect of the diurnal cycle.

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