

We appreciate the thorough, meticulous, and detailed review of our manuscript, and for the most part we agree with the reviewer's comments and have made revisions accordingly. In particular, the reviewer's focus on the manner in which we reported our regression results is thoughtful and helpful, and we are grateful to their contribution in improving our presentation of our work.

We respond to each comment below. For convenience, we reproduce the reviewer's comments in italics, and our responses are given in normal text.

Dear Editor,

This manuscript tests a low-cost sensor node containing a Figaro TGS 2600 and a Figaro TGS 2611-E00 metal oxide sensor, alongside a high-precision reference instrument. The sensor was tested both outside and inside. The outside dataset could not be used due to a lack of methane enhancements, despite efforts to artificially enhance the background. The inside dataset could however be used. The correlation of both sensors to various environmental conditions and methane concentration were presented, using the TGS 2611-E00 as the main methane sensor. This work took the approach of deriving a baseline resistance corresponding to methane concentration below 2.3 ppm, to incorporate non-methane environmental effects, which included a time component. A second baseline was also devised which included TGS 2600 measurements, as this sensor was assumed not to be methane-sensitive. The ratio between measured resistance and baseline resistance compared to methane concentration was poor for both approaches. Instead, the baseline was split into 10 pieces for which individual baseline coefficients were derived. This approach was slightly better. A sensitivity analysis was conducted on piecewise baseline variability components over time. In another analysis, the TGS 2600 was also treated as the main methane sensor, but this did not yield results better than treating the TGS 2611-E00 as the main sensor. In any case, any analysis on methane concentration is slightly dubious as most methane concentration measurements were obtained in background conditions. The greater value of this work is in baseline resistance modelling. This paper is a welcome addition to the growing body of literature on this subject. I was particularly impressed by the meticulous nature of this work. The standard of English and the presentation is very good. I suggest the following editions to the work, in addition to the two paragraphs below, where I address two key issues.

Using the TGS 2600 resistance measurements to derive a baseline fundamentally requires for TGS 2600 methane sensitivity to be negligible. The theory is that this (assumed) non-methane sensor can be used to characterise non-methane effects. Yet, if methane effects are not negligible, then methane sensitivity may be reduced in the resistance to baseline ratio, thus being counterproductive. Based on visual inspection of figure 7, there is not much improvement when comparing an eq 1 (no TGS 2600) baseline to an eq 2 (with TGS 2600) baseline versus methane concentration. The authors do not provide R^2 values for each fit and only provide values for their favoured fit (eq 2 piecewise). Unless the authors can prove that the eq 1 baseline approach is significantly better than the eq 2 baseline approach, I do not think using TGS 2600 measurements within a baseline is necessarily better. The authors' justification for using eq 2 instead of eq 1 is that it results in a better baseline fit (i.e. modelled baseline versus measured baseline). This is hardly surprising as this simply shows that two similar TGS units behave in a similar way. But the authors must also be sure that there is no TGS 2600 methane effect.

To test the TGS 2600 methane effect, the authors do apply a baseline correction to the TGS 2600 data (in the same way for the TGS 2611-E00 data), observing poor methane correlation. Yet, the authors fail to provide an R^2 value of the methane fit for comparison. Furthermore, most TGS 2611-E00 baseline attempts also failed, when compared to methane concentration. I think the 10 ppm limit and the number of data points may have been insufficient to reveal sufficient correlation for this sensor. I therefore question the conclusion from this manuscript to dismiss the utility of the TGS 2600 from further research. The authors must clearly state these caveats and limitations when discussing their views and outlook on the use of the TGS 2600. This work is not sufficient to conclude that the TGS 2600 is not sensitive to methane below 10 ppm.

Thank you for your comments! We have added R^2 and RMSE values to the various fits to add context and support our claims. We believe that our claims about the unsuitability of TGS2600 for methane monitoring in this range are also supported by our previous laboratory work (Furuta et al., 2022), as we have stated in the manuscript. We have attempted to more clearly state the caveats and limitations in our revisions to your specific comments below.

General comments

Abstract

- This is concise and easy to follow. However, this section is written slightly vaguely. The abstract should include more specific details on the work presented in the manuscript and not be afraid to use a bit of technical language. For example, a crucial advancement in this work is the development of a baseline resistance model. This should definitely be in the abstract. The authors could also add some more details on the enclosure (such as size), if appropriate.*
- I also think that the duration of testing time should be included here. • I am interested by the decision not to specify that a Figaro TGS is being used either here or in the title. I do not insist that the authors do so. However, I would appreciate some rationale please, as I do not understand the value of this.*

We have added more details to the abstract as noted in our response to specific comments below, including specifying TGS models. We have attempted to balance detail and conciseness,

Introduction

- This section provides a very good summary of recent research on the TGS in methane concentration measurements, with good referencing. However, this first paragraph could provide a better overall motivation for the necessity of this work in the context of increasing methane emissions, with some citations.*
- The way in which the narrative of this section is conveyed is sometimes confusing as it is not clear whether it is written in the context of methane or more generally. Based on the first sentence, this is a paper on methane measurements. It should therefore be emphasised throughout this section that points are being made in the context of methane sensors and not more generally.*

Thank you for the comments. In the interest of not repeating the same content often found in

other papers, we have added a reference to a paper that effectively summarizes the motivations and applications for low-cost methane sensing. Please also see our responses to the specific comments.

Methods

- *Overall, this section is well-constructed and covers the methods very well. The calibration section was particularly well-written and easy to follow.*
- *Regarding the timestamps, was the lag time of the TGS enclosure ever tested? The 30 s lag time of the LI7810 was assumed to be negligible, which is fine. However, the lag time of the TGS logger is a bigger issue. If a spike of methane was emitted just beneath the TGS enclosure, I wonder how long it would take TGS resistance to peak in response. It would not be short (a few seconds), as air is carried to the sensors by diffusion. I do not think 10-minute averaging will overcome this problem, unless TGS peak time is less than 1 minute. The averaging is however still necessary to account for the nature of diffusion, which is not instantaneous like a LI7810. The overall solution is both a lag-time correction in combination with averaging to smooth any emission spikes.*

Thank you for your comments. We have added a figure to Appendix A illustrating the sensor lag with a series of large methane spikes - the sensor responds reasonably quickly to the spikes (within minutes), but takes considerably longer to decay back to baseline. It is not immediately clear to us that this can be accommodated through data processing, and we believe that the addition of a pump will likely be necessary to resolve this issue.

We have added a description of this lag to this section as well.

Results

- *The investigation of various causes of baseline disparity is a welcome inclusion in the research on this topic. The correlation plots and the baseline analysis are both good. However, the work on a methane response using different baseline approaches lacks detail.*
- *In addition, this section must include an analysis of methane correlation excluding periods used to produce a baseline fit. Otherwise, it is mostly an analysis of the ability of the baseline to predict the baseline as the vast majority of datapoints were below 2.3 ppm, i.e. the baseline threshold.*

Thank you for your comments. We believe we have addressed your points in our revisions to your specific comments below.

Discussion

- *The sensitivity analysis of the time component on the baseline is a very useful and robust way of identifying the nature of temporal variability.*
- *Testing the ability to derive a TGS 2600 baseline for methane correlation is a very good idea. However, the caveats must be discussed more clearly here. Furthermore, a plot of methane correlation would be welcome. It is not clear how much worse the TGS 2600 is than the TGS 2611-E00, when detecting methane.*

We believe we have addressed the caveats in our revisions to your specific comments below, and have added R^2 values to help give additional context to the relative performance of the sensors (we had initially omitted these as they are negative, which we thought might confuse

readers unfamiliar with the notation).

Conclusion

- *This is a nice overview of the work, but lacks a few key values and outcomes from the work, that could be included.*

Please see our revisions to the specific comments.

Specific comments

Line 11: We deployed the prototype sensor alongside a reference methane analyzer in two sites - one outdoors, one indoors - for several months each of data collection across a range of environmental conditions and methane levels.

- *This sentence is difficult to follow.*
- *Also, please specify which types of environmental conditions. Variations in temperature?*

Revised.

Line 13: calibration models

- *Please add a few very brief details on what sort of calibration models. What is the crucial basis of the calibration approach? Were linear models used? Was machine modelling used?*

Revised.

Line 14: background monitoring and enhancement detection

- *Monitoring and detection of what? Presumably this is methane concentration.*

Revised.

Line 14: performance

- *What does “performance” mean? Does this mean accuracy compared to the reference instrument? Please clarify.*

Revised.

Line 14: 2 to 10 ppm range

- *Please specify that this refers to methane concentration.*

Revised.

Line 17: these and similar inexpensive MOx sensors

- *Of which sensors? It is not specified which sensors have been used. The authors cannot refer to “these” sensors and compare them to similar sensors, if the sensor has not yet been introduced.*

Revised; we have added model numbers to the beginning of the abstract.

Line 17: near-background methane monitoring

- *What does this mean? Perhaps state that this refers to up to 1 ppm (or whatever) concentration enhancement.*

Reworded for clarity.

Line 19: scarcity of high-resolution ground-level data

- *Please provide some references for this statement.*

Although we believe this to be true, it is difficult to cite a lack, and an in-depth discussion would be tangential to our work; we have chosen to remove the statement. We agree with your comment.

Line 21: variety of sensor mechanisms

- *Please specify that this refers to methane sensors. It is better to write this section in the context of methane detection, rather than general low-cost sensor use.*

Revised.

Line 29: sensor array or “e-nose” configurations

- *Please briefly explain how using a sensor array can overcome issues of selectivity.*

Revised.

Line 38: when calibrated in a laboratory setting

- *Please specify that it was both calibrated and tested in a laboratory setting. This 1.7 ppm value refers to an indoor laboratory test.*

Revised.

Line 41: above 100 ppm concentrations

- *In the laboratory or in the field?*

Revised.

Line 41: 2 to 100 ppm range

- *Again where? In the laboratory or at a glacier?*

Revised.

Line 42: but found that quantitative emission estimates had poor accuracy • I don't understand the relevance of this. This is a manuscript on methane concentration measurements and not flux measurements.

The cited study estimates fluxes from concentration estimates derived from TGS2611-E00 sensors, and so we believe there is some relevance; however, we agree with the reviewer's comments and have removed this clause to reduce confusion.

Line 48: positive results

- *Please clarify, positive results in what? In methane concentration measurements? It is previously stated that this sensor can be used to measure many gases, so this must be clear.*

Revised.

Line 40: Cho et al. (2022) find

- *Please write this in the past tense.*

Line 48: Eugster and Kling (2012) report a

- *Write this in the past tense.*

Line 49: Other papers find

- *Write this in the past tense.*

Revised.

Line 83: We burned in the sensors and regulator for a week prior to data collection. • Why

Revised.

Line 91: Our system performed well in both sections of the experiment, with 95% of all data showing a sensor supply voltage within ± 0.25 mV of the mean and 99.99% of all data showing a supply voltage within ± 0.80 mV of the mean across the full dataset.

- *This is a very good demonstration of good practice and an excellent result. It is impressive.*

Thank you for your comment! We hope that the system design given in the appendix will be helpful for future research.

Line 119: Our first site was an urban yard in Minneapolis, USA

- *Perhaps provide the coordinates, if possible.*

Due to privacy concerns we are unable to provide the exact address; we have added the neighborhood, however, to give a better sense of the location.

Line 125: The background methane concentration at our research site

- *How is this known? Is this based on the LI7810? Please specify.*

Revised.

Line 127: 2.5% methane gas cylinder

- *What was the rest of the cylinder filled with Synthetic air? Natural air? Argon? Nitrogen?*

Revised; the cylinder was balanced with air, but we are unable to find further details on the composition (the manufacturer datasheet and the calibration certificate just say “Air” without specifying natural or synthetic).

Line 129: These releases produced a maximum 10-minute averaged methane value of 5.8 ppm, with most of the releases producing methane concentrations between 3 and 4.5 ppm.

- *Again, please specify how this is measured. Is this based on the reference gas analyser?*

This is correct, revised.

Line 133: Our second site was indoors in the Biosystems Engineering building at the University of Minnesota, Twin Cities campus.

- *This is a little confusing. Quite simply, one test was conducted was outside and one test was inside the building. However, it reads as if there were two totally different sites in different locations, which adds to the complexity. “Our second site” implies that there were two totally separate testing locations.*

- *In reality, there was only one site, not a first and second site.*

Revised; we added context that the two sites are approximately 5 km apart.

Line 136: We also expected possible emissions of methane and other gasses from surrounding labs

- *This could be a crucial point. The lab was filled with potential interfering gas sources, which could influence the TGS response. I think the authors may elaborate here on potential gases that could be present or, at least, provide some more specific details on the activities taking place in the vicinity.*

Revised to add some more context on possible things in the vicinity.

Line 153: we removed two hours of data after each reboot

- *Can the authors confirm that 2 hours was sufficient for the sensor to stabilise after power loss? I suggest a plot showing this.*

We have added a figure to appendix A showing the startup behavior for each time the unit was powered up during the experiment. The curves are difficult to interpret, but it appears that the initial “warm-up” period for the sensors is rather short, possibly 10 minutes. Evaluating the full power-up behavior would be a useful contribution for future work.

Line 156: dataset averaged to a 10-minute timescale

- *Was it a 10-minute running average or were 10-minute averages taken next to each other. Please clarify this both here and throughout the manuscript.*

Revised to clarify that we used consecutive 10-minute averages.

Line 172: We chose to include sensor run time to incorporate any effects of sensor aging; • This is a very good idea. Is this the absolute time or the time of the sensor being switched on to account for data gaps?

Revised to clarify that we used the cumulative time the sensor was turned on from the beginning of the experiment (the long data gap was due to an issue with the reference analyzer, and did not affect the sensor).

Line 177: Relative humidity is dependent on water vapor concentrations and temperature • And also, pressure.

Revised.

Line 179: we decided a priori to include water vapor concentrations and not relative humidity as a possible term in our analysis.

- *I totally agree with this rationale. Perhaps cite some other research that preferred to use water concentration instead of relative humidity.*
- *Another important detail is missing here. Is this water concentration from the LI7810 or is it derived from the SHT? If it is derived, the authors must state how it derived.*

Revised to clarify we used water vapor concentrations from the LI7810, and added a citation to Shah et al. (2023) which discusses this issue in greater depth.

Line 193: The TGS2611-E00 sensor response is expected to deviate from the predicted baseline response as a result of methane levels as shown in Equation 3a. • Why was this equation chosen? It is based on any previous work? I have never seen a purely linear fit applied to a resistance ratio. This is no criticism, but I am interested to know where it comes

from.

Previous work, such as Fig. 8 in Shah et al. (2023) or the curves in the sensor datasheets (e.g. Figaro USA, 2013), suggests a power function or logarithmic fit. However, this work also suggests that a fit in the limited methane range we exam may be approximately linear (similar to the small signal model in electronics work). We chose to use a linear equation for simplicity, and our results shown in Fig. 5 do not appear to suffer from bias as a result.

We have added a note that a power function will likely perform better over a wider range.

Line 212: We observed a diurnal cycle in methane levels

- *How? Using the reference gas analyser?*

That's correct, revised.

Line 213: soil processes

- *What does this mean?*

We have removed this as speculative.

Line 240: humidity

- *What does this mean? Is this water concentration? The term "humidity" is used regularly throughout the manuscript. However, I do not think this is wise as it could mean either water concentration or relative humidity. I suggest that the authors replace all "humidity" terms in this manuscript with a more precise term (either water concentration, relative humidity or something else) to avoid ambiguity.*

This is an excellent suggestion. We have revised the paper throughout accordingly, replacing "humidity" with "water vapor concentration" in cases where it was ambiguous.

Line 248: relative humidity

- *I'm not too sure in the value of including relative humidity here in this list and in the previous analysis. As the authors themselves state, relative humidity and water mole fraction are related. So, it is hardly surprising that both influence TGS resistance.*

This is correct. We chose to include relative humidity in the list and in the next sentence to remind the reader that we have chosen a priori to use water vapor concentration rather than relative humidity; we feel that restating this decision is helpful to the reader, but are open to revision if the reviewer feels strongly that it adds confusion.

Line 252: by including TGS2600 in the baseline response, we can possibly remove influence from nontarget gasses and other unexpected factors.

- *The authors also remove the baseline from the TGS2600 dataset? This should be mentioned in this section somewhere.*

Our apologies, we're uncertain exactly what the reviewer is asking with this comment. We use the averaged TGS2600 in our baseline Equation 2 without removing data, if this is the question. We have added a citation to the previous sentence to support the claim that TGS2600 does not respond to methane in this concentration range.

Line 260: 1.46, 1.56, and 2.81 k Ω

- *These values should include a plus-minus sign as the square root has a negative and positive solution. The same for RMSE below.*

We respectfully disagree with this suggestion. RMSE is equivalent to the standard deviation of the regression residuals, and accordingly should be represented as a positive magnitude (as the standard deviation conventionally is) rather than as a +/- . Furthermore, of the previous work in Atmospheric Measurement Techniques we cite, four papers (Eugster et al., 2020; Collier-Oxandale et al., 2018; Jørgensen et al., 2020; our previous work in Furuta et al., 2022) report RMSE as positive magnitudes, and one (Shah et al., 2023) reports +/- values. We accordingly prefer to keep RMSE values as positive numbers, both in keeping with the underlying statistical argument and in accordance with what appears to be typical convention.

Line 293: The fit is the best quality

- *Which fit?*

Revised.

Line 306: with $R^2=0.46$ and $RMSE=0.65$ ppm

- *The authors should also provide RMSE and R^2 values for periods not used to derive a baseline (i.e. above 2.3 ppm). This would help to evaluate the capability of the approach when not applied to the same data used to derive a baseline fit.*

We agree with the reasoning of this suggestion. We have added R^2 values (0.39 with the outlier points, 0.58 excluding them) and RMSE values (0.73 and 0.59 ppm with and without outliers, respectively) above 2.3 ppm.

Line 317: we calculated the absolute change in methane concentration with the data immediately before and after each point

- *What is the time distance between each point? Are they 10-minute running averages of averages next to each other?*

Revised.

Line 338: occurrence after a data gap (D)

- *What are the red dots? I think this figure could be improved with the different colours given as a legend in each subplot. The figure caption is not clear as all of the points are plotted.*

Line 371: Figure 7: Sensor calibration results using the different baseline regression approaches.

- *Please change the axis from “actual CH₄” to “LI7810 CH₄” or similar, as “actual” doesn't mean much.*

We appreciate your helpful comments on figure clarity. As reviewer 2 has also made some comments on the figures, we will work on improving the figures in our later response to their comments, with your suggestions in mind as well.

Line 439: However, neither possible fit (with the Equation 1 or Equation 2 baselines) for methane had R^2 better than simply predicting the mean, nor RMSE better than 4.5 ppm. • Unless R^2 values are provided, corresponding to the plots in Figure 7, it is not clear whether the TGS 2600 is truly worse than the TGS 2611-E00. The non-piecewise baseline attempt for the TGS 2611-E00 also failed and only one model worked.

We have added R^2 values for these regressions, as well as for the fits with different baselines in section 3.4. We had previously omitted these values out of concern that the reader would not know how to interpret negative R^2 values, but we agree with your point that specifying the values gives important context. We have attempted to explain that negative R^2 values indicate that predicting the data mean for all points is a better fit.

We believe that the drastically worse R^2 and RMSE values at this line as compared to those in section 3.4 helps support our argument that TGS2600 is a worse quality methane sensor at this low concentration range, an argument which is also supported by our previous laboratory work in Furuta et al. (2022).

Line 452: parts cost of under \$200

- *This is very impressive. It should be included in the methods section.*
- *Also, please provide the cost of telemetry data transfer in the methods section. Presumably there is a regular subscription charge of some nature.*

Thank you! We have added the parts cost to the first paragraph of the methods section, and added a note that the Particle Boron module we used as a cellular modem came with an included data plan sufficient for our needs without extra or recurring cost.

Line 457: We suggest that work finding TGS2600 to respond to methane in a low concentration range should consider possible co-emitted gasses, algorithmic overfitting, or other experimental factors.

- *I do not think that this sentence should be in the conclusion as they are not key findings from this work and are simply the opinion of the authors. We do not know for sure why the TGS2600 has worked in previous studies*

This is fair, we've removed this (and in the previous section as well).

Line 462: above 10 ppm

- *Why above 10 ppm? This work was below 10 ppm. All available data above 10 ppm were excluded from the analysis so I do not understand why this point is being made.*

Reworded. We feel it is useful to give an opinion on the concentration ranges in which our sensor (or similar) may be relevant, and where it is likely not the best solution.

Line 463: Our sensor response correlates with methane levels with moderate accuracy in the lower 2 to 10 ppm range

- *This is misleading for two reasons.*
- *First, the authors must be precise what they mean by "moderate accuracy".*
- *Second, most of the analysis was performed on data below 2.5 ppm. Only a small percentage of data was anywhere near 10 ppm. So, the sensor was mostly tested at background levels and not mostly over a range up to 10 ppm.*

As noted at the end of section 3.3, 45% of the inside data was 2.5 ppm or lower, while the majority was higher concentration - the data is certainly biased towards the low concentration range, but there was a range of concentrations represented. We also feel that this bias towards low concentrations most likely makes our results pessimistic - we would expect the sensor to perform better with a higher mean concentration.

We have revised the sentence to clarify “moderate” with RMSE.

Line 474: controlling water vapor levels and temperature in the sensing chamber • Why would this help if the baseline model already accounts for these effects? Surely the problem is unknown environmental factors. What is the use of controlling known variables?

This is a good point; we have removed this clause.

*Line 476: methane concentrations above the near-background range
• What does this mean?*

Revised to “above the 2 to 10 ppm range we examined.”

We have also corrected a typo in the caption for Table B1 and have made some minor changes to wording throughout for clarity.