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Atmospheric Measurement Techniques Discussions

Interactive comment on "Assessing a low-cost methane sensor quantification system for use in complex rural and urban environments" *by* Ashley Collier-Oxandale et al.

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COMMENT: "This work presents a detailed analysis of the performance of one type of low cost metal-oxide sensor for methane detection. The study involved the deployment of multiple sensors both in the Colorado Front Range (for a period of ∼1 month) and in urban Los Angeles (for ∼2 months). These experiments were well designed within the constraints of larger studies, providing multiple opportunities for infield comparison of the sensors with established methane measurement technologies. The analysis presented provides a thorough comparison of several calibration strategies and possible explanations for the observed discrepancies. The authors show that the sensors are

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capable of providing useful information on spatial variability, whilst not overstating their capabilities. Overall I feel the paper is well written and a valuable contribution to the growing body of work assessing the potential of low cost sensor technologies. I therefore recommend publication after the following minor comments have been addressed."

RESPONSE: The authors appreciate the overview provided by the reviewer and would like to thank the reviewer for their thoughtful comments.

Minor Comments:

COMMENT: "1) It would be beneficial to the readers if the authors could provide more detail on how well the sensors agreed when they were co-located. Previous work (e.g. Jiao et al. 2016; Smith et al. 2017) has shown that variability between sensors can be significant. As Figs. 16 and 18 attribute observed differences between spatially distributed sensor signals to variations in methane concentrations at the different locations, an idea of the observed variability between co-located calibrated sensors would be useful. This could potentially be added as an extra panel to one or both of these figures?"

RESPONSE: The authors agree that more detail regarding the inter-sensor variability would be a valuable addition to the manuscript. The attached figure and table (Figures 1 and 2 here, Figure 19 and Table 9 in the revised manuscript) have been added in an appendix to the manuscript, along with additional text noting where to find this figure and table as well as text discussing the inter-sensor variability. These additions have been made to Section 3.5 as they expand on the information already provided in Table 8 (Table 7, prior to manuscript revisions).

New text added to Section 3.5: "The details of each individual sensor versus P1 is available in Figure 19 and Table 8, in Appendix A. These details demonstrate the extent of inter-sensor variability for co-located sensors and the increased variability for deployed sensors. While there is some variability among correlation coefficients, for nearly all sensors the periods of enhanced methane fall along the 1:1 line and most

offsets occur at lower methane concentrations. Additionally, all RMSE's for co-located sensor falls below our expected uncertainty, while the RMSE's for deployed sensors is larger than this uncertainty (with the exception of the P1/P2 pair, which was co-located during the deployment)."

COMMENT: "2) As acknowledged by the authors, the analytical method used by the sensors in this study is sensitive to hydrocarbons other than just methane. As oil and gas operations co-emit a variety of hydrocarbons along with methane it is possible that the sensor response attributed to methane could instead be due to other hydrocarbons. Although the authors say that this will be covered in a future publication, I feel the issue does require some further discussion in this manuscript.

The authors state that the calibration parameters derived for the two study locations are significantly different, a problem well documented in the literature, but it would be useful to know which of the parameters in the equation show the most difference between the locations. Figure 6 shows that temperature and humidity observed in Los Angeles are within the range seen in Colorado, so one would expect the parameters associated with sensor response to these variables to be similar? It is likely that the hydrocarbons co-emitted with methane in Los Angeles and Colorado are different (geological basins often show characteristic hydrocarbon fingerprints). This would be visible as a difference in the parameters associated with the sensor sensitivity to methane if the sensor was responding more to the co-emitted hydrocarbons than the methane. The authors should comment further on this source of uncertainty."

RESPONSE: The authors agree that the difference in optimal calibration model parameters between the two locations is likely the result, at least in part, of different hydrocarbon mixtures. Not only are Los Angeles and Colorado likely to have different hydrocarbon fingerprints associated with the geologic basin, but also differences in traffic and other sources will like to affect the background hydrocarbon mixture as well. To address this, the authors have added an additional analysis of variance on the sensor signal that includes some hydrocarbon species. This analysis supports the point in

the paper that other hydrocarbons do help to explain the sensor signal, but they do not displace methane. The text below was added to provide context for and discussion of this analysis, and the table has been added as Table 7 to the revised manuscript (attached as Figure 3 here). An additional note is that methane is generally present in the atmosphere at much higher levels than other hydrocarbons, such as ethane. Therefore, while this sensor is likely responding to other hydrocarbons, methane is likely a big driver of sensor response.

New text added to Section 3.4.3: "As previously stated, the FRAPPE/DISCOVER-AQ campaigns offered many opportunities for co-location with high-quality instruments and there was also a PTR-MS sited at the PAO, providing speciated VOC measurements (Halliday et al., 2016). Future work will provide a more in-depth analysis of VOC sensitivity and selectivity for different metal oxide sensors; however, we have included here a preliminary look at this cross-sensitivity to other hydrocarbons. Table 7 provides the results of another sensitivity analysis in which the explanatory power of a few speciated hydrocarbons is examined. For simplicity, one hydrocarbon from different correlated groups was selected (e.g., benzene was selected from the aromatic group, which exhibited high correlation among species). This analysis illustrates that VOCs (particularly acetaldehyde and benzene) do help to more fully explain the variance in the sensor signal, but they do not displace methane. This is most apparent for Parameter Sets 5 and 6, in which we see the variance explained by residuals increase slightly and the variance explained by temperature increase quite a bit as this factor compensates for the missing methane. When methane is added back in for Parameter Set 7, along with all three VOCs and CO, the variance explained by the residuals is at its lowest and the variance explained by methane is at 10.1% and higher than the percentages for the individual hydrocarbons. Thus, as demonstrated by our analysis the Figaro TGS 2600 sensor is cross-sensitive to carbon monoxide and hydrocarbons, along with methane."

COMMENT: "3) I am not convinced by usefulness of the methane baseline filtering

approach shown in Fig. 5 (A.P.). Removing data points that are below a known background methane would surely introduce a negative bias into the sensor data and unlike the other filters does not seem to be a test of the calibration model, but just a method of improving the agreement statistics. These signals below background levels could be indicative of other sensor dependencies not captured by the calibration model, (e.g. changing hydrocarbon mix (see comment 2) and thus contain useful information. I would suggest omitting this filter or providing more justification of why it is a valid approach."

RESPONSE: The authors appreciate the reviewer's observations regarding the inclusion of this baseline filtering and have chosen to add further justification for its inclusion. The authors feel that this filtering is valuable to leave in as it highlights the need for discussions around data processing for different purposes. Data processing such as this may be a necessary part of using data from low-cost sensors as these anomalies are likely to occur. That being said, the authors agree that this filter will likely introduce a negative bias and might remove valuable information about sensor behaviour and response. Therefore, to address this comment, the authors have added further justification in the text highlighting the benefits and drawbacks of using this filtering – highlighting that this type of processing may be useful for sharing sensor data. We have also added another figure (Figure 4 here, Figure 20 in the revised manuscript) to the appendix illustrating that for every instance where these underestimations were removed by this filter, the dynamic range of methane for was less than our expected uncertainty (RMSE = .18). Therefore, this relationship does seem to indicate that the underestimations are related to environmental factors, or it might possibly be a limit of detection issue. This has also been pointed out in the text with additional discussion and a note to see the additional figure added to the appendix.

New text added to Section 3.4.2: "The final filtering approach, utilizing atmospheric composition, should only be applied to sensor data selectively. Removing improbable values from sensor data that fall below zero or a known baseline may be a useful or

even necessary strategy in certain situations. In dealing with air quality data, there are examples of additional processing being used to reduce negative values (Hagler et al., 2011), and examples of guidelines to remove negative values below a given threshold (US EPA, 2016). For work with sensor data, if the focus of the analysis is to understand enhancements over background captured by sensors, then removing improbably low values can elucidate these results. If preliminary data is being shared with the public, then flagging and removing improbable values can reduce confusion. Given the challenges in sensor quantification, this second example in particular warrants consideration by those using sensors in partnership with communities and the public. However, it is also likely that these underestimations contain valuable information about sensor behaviour and sensitivity; removing these values will also introduce a negative bias to the data. Accordingly, when using this type of processing, researchers will need to be clear about why this approach is useful and valid for given situations. For this data set, every instance where underestimations are removed coincides with days having a dynamic range of methane less than the expected uncertainty for the sensor data, which indicates that these underestimations may be connected to a limit of detection issue. Figure 20 (Appendix B) demonstrates this association."

New Sources:

Hagler, G. SW., Yelverton T. LB., Vedantham, R., Hansen, A. DA., Turner, J. R.: Post-processing method to reduce noise while preserving high time resolution in aethalometer real-time black carbon data, Aerosol and Air Quality Research., 11(5), 539-546, doi: 10.4209/aaqr.2011.05.0055, 2011.

United States Environmental Protection Agency (US EPA): Technical Note âĂŘ Reporting Negative Values for Criteria Pollutant Gaseous Monitors to AQS, information available at: https://www.epa.gov/sites/production/files/2017-02/documents/negative_values_reporting_to_aqs_10_6_16.pdf (last access: April 2018), Oct. 2016.

COMMENT: Typographical errors: “Pg 15 line 12 “it is important explore” should read “it is important to explore” Pg 19 line 4 “temperate” should read temperature”

RESPONSE: The authors have corrected this typographical error in the revised manuscript and have reviewed the entire manuscript once more for other errors.

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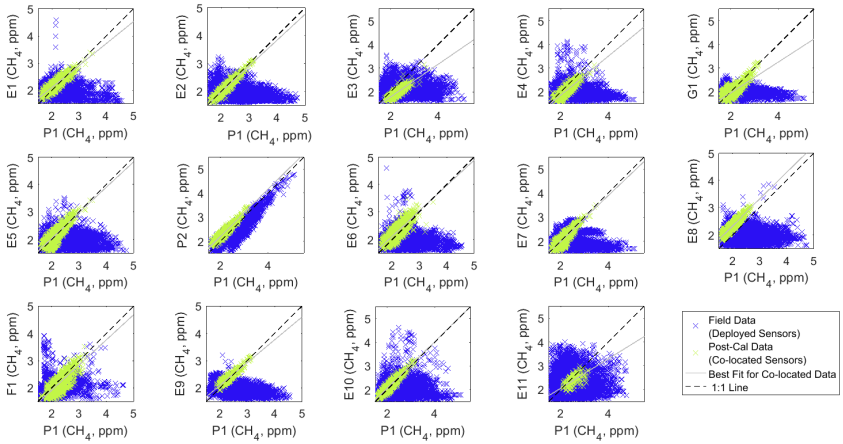

Fig. 1.

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	Co-located		Deployed	
	R	RMSE	R	RMSE
E1	0.914	0.117	-0.159	0.581
E2	0.940	0.084	-0.095	0.594
E3	0.909	0.284	0.168	0.619
E4	0.863	0.201	0.050	0.591
G1	0.737	0.200	-0.179	0.565
E5	0.849	0.132	0.054	0.529
P2	0.961	0.210	0.904	0.241
E6	0.847	0.166	-0.061	0.535
E7	0.864	0.124	-0.067	0.572
E8	0.931	0.336	-0.186	0.599
F1	0.819	0.182	0.221	0.550
E9	0.931	0.095	-0.454	0.655
E10	0.929	0.107	0.053	0.585
E11	0.624	0.208	0.104	0.571
Average	0.866	0.175	0.025	0.556
Std Dev	0.092	0.073	0.306	0.096

Fig. 2.

Source of Variation	Set 1	Set 2	Set 3	Set 4	Set 5	Set 6	Set 7
Temperature	9.6%	17.4%	10.1%	10.9%	33.7%	28.9%	16.3%
Abs. Humidity	10.2%	11.7%	14.2%	7.5%	6.2%	9.0%	12.6%
Time	8.9%	4.5%	9.2%	4.4%	3.0%	2.8%	5.8%
CH_4	21.8%	12.3%	14.2%	18.5%	-	-	10.1%
CO	15.0%	13.6%	14.4%	19.7%	-	9.8%	11.4%
Acetaldehyde	-	7.5%	-	-	13.4%	8.9%	6.5%
Benzene	-	-	4.6%	-	8.4%	6.4%	4.0%
Methanol	-	-	-	0.9%	0.4%	0.1%	0.3%
Residuals	34.5%	33.0%	33.4%	38.2%	34.9%	34.1%	33.2%
Total	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%

Fig. 3.

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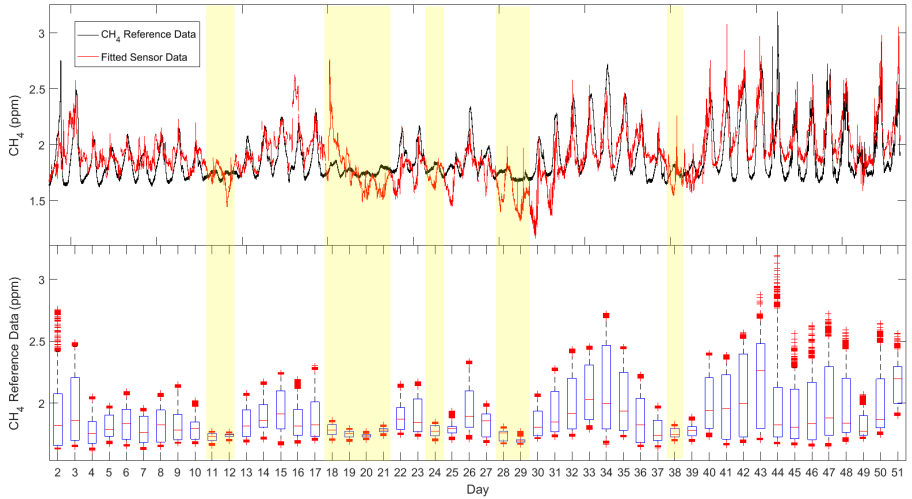

Fig. 4.

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