

Atmos. Meas. Tech. Discuss., referee comment RC2
<https://doi.org/10.5194/amt-2022-200-RC2>, 2022
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Comment on amt-2022-200

Anonymous Referee #2

Referee comment on "Reconstruction of high-frequency methane atmospheric concentration peaks from measurements using metal oxide low-cost sensors" by Rodrigo Andres Rivera Martinez et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2022-200-RC2>, 2022

General Comments:

Monitoring methane (CH₄) emissions from industrial sources is an observational challenge due to the highly episodic and localized nature of such emissions. In this study, the authors aim to address this problem using low-cost metal oxide sensors (MOS), which are cost-efficient to deploy over a large network. The authors conduct several laboratory studies to calibrate the long-term accuracy and stability of several Figaro TGS sensors against a research-grade cavity ring-down spectrometer (CRDS) with well-established accuracy and precision. The authors test several parametric and non-parametric models in reproducing a series of randomly generated CH₄ spikes from the MOS voltage and ancillary measurements of ambient environmental conditions. The authors find that a second-order polynomial model and a multilayer perceptron model achieve the best results as measured by the RMSE of reconstructed vs. observed CH₄ peaks (the CRDS measurements being the independent variable). The authors conduct further parsimonious training experiments by clustering the generated CH₄ peaks into self-similar categories. They demonstrate that a small subset (25%) of the data adequately trains the models (as measured by RMSE) when an appropriate representation of peaks are selected. In addition, training one set of sensors reproduces the data from other sets of sensors, provided the peaks fall into the same broad clusters. Overall, the authors have conducted a thorough and careful evaluation of different calibration approaches for the Figaro TGS sensors that meets the accuracy and drift requirements for detection of CH₄ leaks in industrial monitoring.

I recommend this paper be accepted with minor revisions. The methods and approach are thorough, but some points could use further clarification, as detailed below.

Specific Comments:

- L23-24: The statements read as if natural gas accounts for all the anthropogenic CH₄
- L65-66: How could the influence of other VOC on the measurement be addressed?
- L89-90: It would be worth mentioning here how the three Figaro TGS sensors differ from each other, and why only Types C and E were used for the analysis.
- L101-103: Does weather during the sampling time frame have any impact on subsequent analysis? For example, Figure A4 shows the humidity and temperature ranges during the experiment. During the summer months with substantially higher water vapor mixing ratios, do the H₂O vapor influences ever become nonlinear? As an aside, what causes the temperature spikes in Fig A4? Is this heating from the MOS?
- L128-129: Does the time constant differ for the TGS C and E sensors? Is this one reason for the higher phase mismatch when training the models with the Type E data?
- L144-145: As in point 4, does the H₂O-voltage relationship ever become nonlinear at high enough humidity?
- L224-225: Will the spikes that end up being influential have any dependence on the structure in the data? In other words, is this method applicable when your dominant spike structure for a real sampling site may be unknown ahead of time?
- L309-311: Why does the interquartile range increase when more training data is used?
- L315-317: I think this is an important result from a policy perspective, where monitoring emission magnitudes may still be of value, even if the spike phase is not exact.
- L352-352: This sentence is unclear. The MSD is larger for Case than Case 11.
- L383-385: This result speaks to point 7 above. If your peak clustering in your training data set differs (by some threshold) from your observational set, the parsimonious training often fails to meet the acceptable RMSE. Does this imply that you would need to bring a CRDS instrument to the field site for short-term sampling? It would be worth clarifying an optimal strategy for field deployment/field calibration.

Technical Corrections:

- Figure 1: Make image a larger so chamber set up is more easily distinguished.
- Figure 1: Is chamber D mentioned in the text anywhere?
- Figure 4: Mislabeled as Type E, not Type C
- Figure 9: It may be helpful to label the input data on the panels directly for ease of interpretation, if it's not too wordy.