

# ***Interactive comment on “Long-term reliability of the Figaro TGS 2600 solid-state methane sensor under low Arctic conditions at Toolik lake, Alaska” by Werner Eugster et al.***

## **Anonymous Referee #1**

Received and published: 17 January 2020

### **1 General comments**

The manuscript summarises the findings from a 6-year field deployment of a small, low-cost methane sensor under low Arctic conditions. Given the interest of the community in small, low-cost sensors on the one hand and the measurement of atmospheric methane on the other hand, this work is of high relevance to the readers of Atmospheric Measurement Techniques.

The manuscript is written and structured well. The methods are described in appropriate detail. However, I see some shortcomings in the data analysis and presentation of

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results, detailed below, that should be addressed before publication.

## 2 Specific comments

It is unclear which quantity the Authors use when they report the abundance of methane. The sentence 'We report all gas concentrations in mixing ratios by volume (ppm or ppb dry mole fractions)' (ll. 78–79) is contradictory, as it uses three distinct quantities as if they were synonymous. In a mixture of two components A and B, concentration  $c$  is defined as  $c = q_A/(V_A + V_B)$ , where  $V$  is volume and  $q$  one of the quantities mass, amount, volume or number concentration [1]. Mixing ratio by volume  $r$  is rather uncommon and defined as  $r = V_A/V_B$  [2]. Mole fraction  $x$  (IUPAC recommends  $y$  for gaseous mixtures, but this is not common in atmospheric science) finally is defined as  $x = n_A/(n_A + n_B)$ , where  $n$  is amount of substance [3]. Given that the WMO scale for methane abundance is a mole fraction scale [4], the reporting of mole fractions would be desirable. While the use of the term 'concentration' for mole fraction is accepted for communication with the general public [4], a publication in a scientific journal should in my eyes favour exact terminology. In any case the Authors must make clear which quantity is reported.

Two TGS 2600 sensors were deployed at the site, referenced to as #1 and #2. However, in several instances in the manuscript 'TGS 2600' appears without a number when I think TGS 2600 #1 is meant. Also, 'sensor', 'TGS' and 'TGS2600' are used. This should be made more consistent. Results from TGS 2600 #2 are presented exclusively in l. 104, ll. 185–186 and Fig. 9. Explaining the minor role of TGS 2600 #2 around l. 59, l. 104 or l. 162 might prevent confusion of the reader.

ll. 74–77 How often and to which scale were the reference analysers calibrated?

l. 153 '[...] relative humidity (which is a ratio and not a physical variable of atmo-

spheric water content)' – I think I have an idea of what the authors mean, but I find the wording not quite right. Would the authors say that the refractive index of a material is not a physical variable because it is a ratio? In general, I miss some thoughts about the temperature dependence of the quantity used for expressing humidity. The Figaro TGS 2600 has a heated sensing element, so the relative humidity at the sensing surface is different from the relative humidity in the environment. The temperature dependence of relative humidity makes this quantity a less than ideal choice for this type of correction. Unfortunately, both alternative quantities chosen by the authors to express water vapour content depend on temperature as well. Mixing ratio (by mass) or specific humidity would be temperature-independent alternatives [6]. Using the ideal gas equation, the terms in Eq. 2 that contain the product  $T_a \cdot \rho_v$  would also cancel out the temperature dependence of absolute humidity if  $T_a$  was absolute temperature (in K) – but in the manuscript a Celsius temperature is used. Hence, my suggestion to the authors is to try out either mixing ratio by mass or specific humidity as an independent variable in Eq. 2. Using absolute instead of Celsius temperature might be advisable as well.

- II. 161–162 Using the entire dataset for estimating the parameters of Eq. 2 is a comprehensive test of how well the model can describe the dataset, but is of limited relevance for field deployments where calibrations are performed during limited periods of time and the main interest is in the uncertainty of independent measurements. For this reason, splitting the dataset into a calibration and a validation part yields important insights. The caption of Table 1 explains that the authors have in fact performed analyses of a split data set. This fact should also be mentioned in the main text around the lines given.
- I. 167 For the reasons given before, the results presented in the columns 'Linear

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Model - Calibration' and 'Linear Model - Validation' in Table 1 should be discussed here, even more so because the results for the validation period are substantially worse than for the period used for calibration.

II. 193–194 Is there any conclusion that can be drawn from this finding of a -1:1 relationship?

Sect. 3.4 The discussion in Sect. 3.1 leans heavily on the coefficient of determination  $R^2$ . In Table 1, each  $R^2$  for the ANN approach is higher than the corresponding  $R^2$  for the Linear Model. Considering just the validation period, the ANN approach outperforms the Linear Model by a factor of 3–10 by this measure. Similarly, in Fig. 6 and 8 the ANN approach outperforms the Linear Model (comparing  $R^2$  of 'ANN' and 'c/v', 'all' is irrelevant in this respect); in Fig. 5 and 7 they perform nearly equally well. None of these comparisons is made here. Instead, the authors state that the root mean square error (RMSE) does not improve substantially with the ANN approach. While I generally appreciate the reporting of RMSE together with  $R^2$ , its interpretation here is questionable. On the one hand, RMSE is reported in Table 1 with one significant digit only, potentially masking up to ~30% differences for an RMSE of 0.03 ppm (0.025 ppm vs. 0.0349 ppm). On the other hand, the RMSE should be seen in the context of the variability of the data, specifically the root mean square difference between the reference measurements and their mean value over the whole dataset, which is not stated. Overall, the discussion in this section appears negatively biased with regard to the ANN approach. This also manifests in the last paragraph of this section, where 'understand[ing] the physical response of TGS sensors' is prioritised over 'technically nicer fits to data', a stark contrast to the lack of a physical interpretation of the terms in the empirical model (Eq. 2). Section 3.4 must be revised to reach the level of neutrality expected from a scientific publication.

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- II. 229–231 To make such an argument, the reader must be informed about the amplitude of all input variables, especially  $S_C$ .
- I. 237 I might be mistaken, but as far as I understand the term homoscedasticity it would in this case mean that the variance of the deviation in CH<sub>4</sub> abundance is the same for every temperature bin. The authors do not report variances, but both interquartile range and 95% confidence interval suggest that the variance is higher at low temperatures than at high temperatures, i.e. heteroscedasticity.
- II. 253–257 ‘laboratory conditions simplify the real world too much’ – What could be the simplification that makes laboratory calibrations problematic? The input variables used in the empirical model (Eq. 2) can – practical difficulties taken aside – be controlled in the lab. Any other variable that might prevent transfer of lab results to field conditions is not included in the empirical model, so the problem would not be a simplification of the lab environment but a model deficiency. The last sentence of the paragraph seems to go in this direction (‘relevant factors’), but is unclear. Please explain better or leave out.
- II. 258–261 Suggesting to move the first sentence to I. 150 and to remove the other one (repetition).
- Fig. 5–8 The graphs are squeezed in horizontal direction, making comparisons between the lines difficult. A shorter period, e.g. 14 days, would give more insight.
- Fig. 5 and 7 The collected in 2012 and 2015 are both part of the calibration period, not the validation period, which is important to know for the reader to correctly interpret ‘TGS 2600 c/v’ and ‘ANN’. I therefore strongly suggest a note in the figure caption.

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- Fig. 5 Suggesting to replace '(TGS 2600 - Reference)' with '(TGS 2600 all - Reference)' in the caption
- Fig. 9 A plot of the difference of the methane abundance calculated from the measurements of the two sensors would be of high interest for the readers. With such a new panel it is also important to state if the parameters derived for TGS 2600 #1 have been used when applying Eq. 2 to the measurements of TGS 2600 #2. In my opinion the new panel could replace panel (b), as the signal difference seems of little relevance.
- Fig. 10 If the main text in ll. 189–190 is correct, 'and the reference' is missing at the end of the first sentence of the figure caption.

### 3 Technical corrections

- l. 1 Suggesting to remove “weak” to avoid misunderstanding. Alternatively, it could be written in parentheses like it the conclusions.
- l. 8 Insert a space between value and unit of temperature. This correction is necessary wherever “°C” is used [5].
- l. 76 replaced
- l. 140 typeset 'Ta' as  $T_a$
- l. 305 'cross-sensitivities [...] **are** of no concern'
- Fig. 2 There seems to be a non-displayable glyph at the beginning of the label for the vertical axis, possibly a  $\Delta$ . This is also the case in Fig. 3, 5 and 12. 'CH4' specifies a substance, not a quantity. Use ' $x_{CH_4}$ ' or another

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appropriate quantity symbol. The same applies to Fig. 3 through 9 and Fig. 12.

## 4 References

- [1] IUPAC Gold Book, term 'concentration',  
<https://goldbook.iupac.org/terms/view/C01222>
- [2] IUPAC Gold Book, term 'mixing ratio',  
<https://goldbook.iupac.org/terms/view/M03948>
- [3] IUPAC Gold Book, term 'amount fraction',  
<https://goldbook.iupac.org/terms/view/A00296>
- [4] GAW Report No. 242 19th WMO/IAEA Meeting on Carbon Dioxide, Other Greenhouse Gases and Related Measurement Techniques (GGMT-2017),  
[https://library.wmo.int/doc\\_num.php?explnum\\_id=5456](https://library.wmo.int/doc_num.php?explnum_id=5456)
- [5] SI Brochure: The International System of Units (SI),  
<https://www.bipm.org/en/publications/si-brochure/>
- [6] WMO Guide to Meteorological Instruments and Methods of Observation,  
[https://library.wmo.int/doc\\_num.php?explnum\\_id=4147](https://library.wmo.int/doc_num.php?explnum_id=4147)

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