Review

Testing the near-field Gaussian plume inversion flux quantification technique using unmanned aerial vehicle sampling

Adil Shah et al.

The authors use a near-field Gaussian plume inversion (NGI) technique to determine the strength of methane emission sources. In that context, they describe, characterize and deploy two commercial near-IR off-axis integrated cavity output spectroscopy (ICOS) spectrometers from ABB/Los Gatos Research. One instrument is ground-based and samples through a tube from the UAV, while the other, more lightweight instrument is deployed onboard the UAV. The authors rightly claim that more reliable techniques are needed to determine emissions from diffuse methane sources. The paper has two main technical sections: chapter 2 "Methane instrumentation and calibration" and chapter 3 "Method Testing". After struggling hard with chapter 2, I decided not to continue the review, and I suggest that the paper must be fundamentally rewritten, with respect to both language and content, before publication in AMT.

Chapter 2 has four subsections:

2.1 Instrument inter-comparison (20 lines) Jonas, please check # lines

2.2 Water vapour correction (61 lines)

2.3 Calibration (28 lines)

2.4 Methane enhancement and uncertainty (20 lines)

Section 2.1 d contains very little actual intercomparison, and thus the title is misleading The inter-comparison consists mainly of one table containing the manufacturer's specifications and the key numbers of one Allan Variance plot (based on laboratory measurements), which is given in S1. In the context of this paper, which attempts to use the instruments under harsh field conditions, much work could and should have been done to provide useful, real-world characterization under field conditions, beyond what the manufacturer reports. This may include (but should not be limited to) performance characteristics and comparison (see section title) of long-term stability, stability of calibration factors, temperature and pressure dependence, sensitivity to vibration (UAV), linearity, selectivity, (measured) response time, concentration range, etc.

Section 2.2 is the core part of the instrumental section, which is reflected by its length. It covers an in-depth description of water vapor correction but sadly contains a number of fundamental flaws. Some examples are below, but this list is not exhaustive:

- The largest correction to obtain dry CH_4 values ($[X]^{dry}$), needed for flux calculations, is water vapor. While the authors do many tests and calculations (see below) to get this value, they never show how accurate the H_2O measurements of the instruments are. They likely have the information (based on the measurements with a dew point generator), but the corresponding performance is never shown. For such a detailed water vapor study, it would be key to deduce this effect first and then discuss (and correct) the remaining influencing parameters in a second step.

- Figure S3 shows the effect of CH_4 on apparent water, due to cross sensitivity of the analyzers. Equation 1 has no physical meaning or motivation, but it produces an exponential curve, which fits the data points nicely. If there is no clear motivation, such a weak set of data may as well be described by a linear interpolation (see S3). Furthermore, applying the same parameter (ω) for the other analyzer, which shows clearly different absolute and relative behavior (S4), is not justified.
- Somewhat similar is the issue in S5 and S7. It is not surprising to find a (nearly linear) dependence between ν and water vapor. It is also not surprising that the residuals become a bit smaller if using a second order polynomial. However, since there is no information on the long-term stability of this system (e.g. repeatability and reproducibility), these fit characteristics are of very little use (a larger order polynomial would give even smaller residuals).
- The authors call ν the water correction factor. This, again, is misleading because it contains normalization to dry conditions, which is a standard procedure, and corrections for non-perfect spectroscopy (which is specific to their analyzers).
- Based on these badly justified fits and corrections, the authors deduce a water uncertainty factor (σ_ν). However, σ_ν is the standard deviation of the mean of the residuals in S6/7. This parameter carries information about the quality of the fit of the polynomial, but no information about the quality of the water correction for a specific measurement at a given CH_4 and H_2O mole fraction.

Sections 2.3 and 2.4 describe the calibration procedure and derive measurement uncertainty.

- Calibrated enhancement in methane mass density (E) is calculated based on parameters that are obtained in the above experimental and fit procedures. The procedures themselves are correct (but see some remarks above and below). The authors then state that their approach is especially useful because it can be used to determine measurement uncertainty (equation 11). However, this uncertainty calculation is based on laboratory measurements and badly motivated fit procedures. It does not contain non-linearity nor any changes (drifts) in time, e.g. of G and ν. Furthermore, it does not include any field (in)stability considerations, nor the fact that one parameter is just assumed to be the same in MGGA and pMGGA. A realistic uncertainty budget would be highly welcome, but the approach chosen here is useless.

Some less fundamental comments are below, illustrating that the whole paper should be strongly revised:

<u>Abstract</u>

Page 1 – Line 12

Many parameters contribute to the uncertainty. Are they large or small, and is the contribution due to the "maybe" poorly quantified sources important or neglectable?

Page 1 – Line 14

This is not a NEW near-field Gaussian plume inversion (NGI) technique – it has been published before by the same authors.

Page 1 – Line 20

Modified with respect to what?

Page 1 - Line 23

Simplify sentence, e.g. the uncertainty was between ... and ...

Page 1 – Line 25

The term "range" implies that it is the range between 17 and 218 %, but this is not meant here.

Page 1 – Line 27

"flux approaches": check language

Page 1 – Line 27

Replace "may perform well" by "may be a valuable alternative"

Page 1 – Line 28

"applied to UAV sampling" should read "combined with UAV sampling"

Introduction

Page 2 – Line 53

precision should read accuracy or precision and accuracy

Page 2 – Line 54

have not yet

Page 2 – Line 60

This suggests that controlled release is the only acceptable method. Make statement more general, e.g. the method was not validated, and its uncertainty was not quantified (e.g. with controlled release experiments).

Page 2 – Line 62

Make it clear that "Shah et al" is not any other researchers but the same group, e.g. our previous study...

Page 2 – Line 65

Language: the sampling does not develop the technique.
e.g.: ... sampling ... used in combination with ...

Methane Instrumentation and Calibration

Page 3 – Line 97

σ_{AV} is a very unusual parameter. Contains very little information because it depends on averaging time. "Allan deviation uncertainty factor" is confusing.

Page 3 – Line 108

"These three effects have a net effect of decreasing $[X]_0$ in both instruments". You probably mean "decreasing [X]". Furthermore, this is only useful information if you state before which effects have a positive/negative impact. Whether the sum is a net negative effect depends on the conditions (usually dilution dominates, so the statement is not wrong).

Page 3 – Line 110

If the pressure broadening coefficient should account for effects two AND three, then state so explicitly.

Page 4 – Line 122

Has a calibration for water vapor been performed, and what are the results?

Page 4 – Line 132

"more points" means: 1.9, 2.1, 5.0 and ca. 104 ppm. It does not make sense to derive parameters of a (complicated) exponential function if there is no physical concept that supports this function.

This H_2O baseline is very large; your data in S3 suggests that 100 ppm (dry) CH_4 results in ca. -5500 apparent ppm H_2O. Is this analyzer malfunctioning and should be returned to the manufacturer, or is it within the specifications for water vapor measurements? Using the MGGA H_2O baseline function "as-is" for another analyzer does not make sense, especially since the experimental values for the pMGGA (S4) are significantly different from those of the MGGA, indicating that the two functions may well not be the same. Alternatively, one may just acknowledge this fact and do a corresponding uncertainty budget. In the context of the large uncertainty of flux estimation with NGI, this may be appropriate. However, it would fundamentally change the arguments and line of thought in this paper.

Page 4 – Line 135

"Having established a well characterised water baseline..."

This baseline may be well characterized. However, it corresponds to a very large correction, which cannot easily be redone in the field. Therefore, it is necessary to measure the longterm stability of the baseline and instrument-to-instrument variability. Otherwise the "well characterized" is not helpful.

Page 4 – Line 136

What was the mole fraction of CH_4 in this cylinder?

The H_2O baseline has a huge dependence on CH_4. Nevertheless, this experiment was only done at one CH_4 concentration only. This may not be representative and add uncertainty to the retrieval of the "real-world" measurements.

Page 4 – Line 150

The difference between wet and dry corresponds to humidity, thus typically 1-3 %. This is not "almost equal". If it has little impact on the result, then this is because the retrieved apparent H_2O does not very strongly depend on CH_4 under the conditions used to test the simplification.

Page 5 – Line 156-157

The uncertainty of water measurements cannot be derived using "the water correction residual (R) from Eq. (2)". The calculated value is not the uncertainty of water measurements, nor the uncertainty after water correction. It is the standard deviation of the mean of the residuals. This parameter carries information about the quality of the fit of the polynomial, but no information about the quality of the water correction for a specific measurement at a given CH4 and HO mole fraction.

Page 5 – Line 160

"For example our correction was used to increase 160 the MGGA measurement accuracy of $[X]_0$ (at 2 ppm) by +0.27%, at a humidity of 0.001 mol water mol^{-1}, and by +1.8%, at a humidity of 0.01 mol water mol^{-1}". This is a meaningless statement. It does not consider that any correct measurement includes the correction of the dilution effect. You compare apples with rotten pears.

Calibration

Page 5 – General Comments on Section/Calibration

Useful:

The alternating measurement of two standards. This gives the repeatability of measurements under laboratory conditions. A simple average and standard deviation would characterize this. Alternatively (x - average x) vs. N (in bins) as a figure would illustrate that the values show a normal distribution.

Not useful:

The concept of uncertainty in C and G because it neglects the fact that the linearity in response is most frequently the key factor determining the uncertainty of the measurement.

Needed:

Determination of linearity.

Page 5 – Line 168-169

This does not make sense. Water vapor in reference gas cylinders should be very low if filled dry (which I assume was done here). Even if filled wet, then 60 bar/20°C should be significantly below 500 ppm. Thus, H_2O from a pressurized bottle (if treated correctly) is so low that it has very little impact on the measurements. An additional trap (as used here) may well lead to water contamination if one is not very careful.

Page 6 – Line 195-196

C deviates by about 12 ppb from 0. Whether that is good or bad (and under what conditions) depends on the application. However, to say that this is "almost equal to 0" for an analyzer with an Allan Minimum below 1 ppb is bizarre.

Page 6 – Line 195

ν is ca. 0.98 at 1 % H2O. This is not "almost equal to 1". Furthermore, ν in S6 contains all water-related corrections, including normalization to dry conditions. Stating that this is "almost equal to 1" does not make sense, because the correction to dry conditions should be done in any case.

Additional remarks

"A lightweight wind sensor (FT205EV, FT Technologies Limited) was mounted on-board UAV1, on a carbon fibre pole 305 mm above the plane of the propellers. It recorded wind speed and direction at 4 Hz. This data was used to model the change in wind speed with height above ground level (z)."

- Was this sensor validated?
- Are the measurements independent of the propellers? How was this validated?
- How reliable were the direction and speed when the drone was moving? How was this determined?

One analyzer was using an inlet just underneath the drone. The corresponding sample is representative of the air above the drone because of downwash. Was this considered? How?

Flux of one drone --> the other just sampled further away.

The Allan Variance plot should be in main section of the paper. Its v-shape is relevant for interpreting the measurements during flight (i.e. 10 – 30 minutes, typically).