

We thank the reviewer for taking the time to review our manuscript and for some very useful and constructive comments and suggestions. We have responded (red text) to each comment (black italic text) in turn below. We have used these comments and suggestions to update and improve a revised version of the manuscript.

Shah et al. employ two different UAV platforms to quantify known sources of CH₄ during a series of release experiments. The deployment of a lighter prototype Microportable Greenhouse Gas Analyzer (pMGGA) is new and may be interesting to other potential users as well. The authors have done a reasonable job to characterise the sensor in the laboratory; however, testing of the sensor in a harsh environment with varying temperature and pressure is missing. The near-field Gaussian plume inversion methodology presented in previous work was applied to the release experiments, and the estimated fluxes and associated uncertainties were compared with the known sources. The paper is well structured and well written, and can be published at AMT after taking into account the following comments.

We agree that the importance of temperature and pressure variability were not well described. We hope we have now strengthened the manuscript by addressing this concern. We have added a new table (Table 5), which shows that variability in cell temperature for the MGGA and pMGGA was no more than 1.3° C and 2.7° C, respectively, within each UAV flight, compared to a variability of 2.9° C noted during our 19-hour MGGA Allan variance characterisation. During the MGGA Allan variance test, there was no discernible systematic effect of either cell temperature or cell pressure on measured methane mole fraction (see section S1 of the supplement, newly added, which now describes this). With regard to pressure specifically, all measurements were made at ambient atmospheric surface pressure, which changes only very slightly up to 50 m in height and between flight surveys. Furthermore, the pMGGA was closely controlled to a fixed cell pressure. Regarding temperature and pressure conditions during sampling, we would argue that they were not “harsh” (see Table 5) and that we should have made this clearer in the original manuscript. During UAV sampling, recorded cell temperature was on average 23° C across all UAV flights compared to calibration temperatures for the MGGA and pMGGA of (31.4±0.7)° C and (24.6±0.1)° C, respectively. We thank the reviewer for raising this point. On a wider point, a truly harsh environment, such as volcanic caldera sampling, would (we agree) likely require further characterisation in extreme temperatures, but this is out of scope of this work.

General comments:

1. A weak point of the calibration in Section 3.2 is that the temperature and pressure dependence of CH₄ measurements by both the MGGA and the pMGGA is not characterised, which may be potentially much larger than the gain factor uncertainty and the offset uncertainty. In the case that the field characterisation was not performed, why not characterize it in the laboratory?

This is a very useful and valid point. To test the effect of changes in cell temperature, we measured a test gain factor of 0.9979 (at 12.7° C lower than the main calibration) and for cell pressure, we measured a test gain factor of 0.9967 (at 37.2 mbar lower than the main calibration), in the MGGA (noting, as above, that cell pressure is fixed and controlled for the pMGGA). Neither test revealed a discernible change in MGGA gain factor within uncertainty (this point is now made in the final paragraph of section 2.3). To put this into context, if sampling 12.7° C above the cell temperature conditions of the main calibration, mole fraction enhancements (above the background) of at least 2.3 ppm would be required for the temperature effects on gain factor to be larger than the instrumental noise (characterised by

the 10 Hz Allan deviation). Such an enhancement is relatively large compared with typical ambient sampling of plume emissions at a distance from a source.

In addition to these specific tests, we utilised sampling from the 19-hour MGGGA Allan variance test, where variability in cell temperature and pressure was characterised. This is also now described in the supplement. Though the cell pressure and temperature variability ranges were limited to a “reasonable” range of typical ambient conditions, these parameters showed no obvious methane mole fraction correlation, with Pearson’s correlation coefficients of -0.3835 and 0.4849, respectively. Unfortunately, housekeeping data were not recorded by the pMGGGA during the Allan variance test, but in principle, any correlation should be similar as the same spectroscopic fitting is used in both instruments. These results are now summarised in the final paragraph of section 2.1.

2. What’s the reason behind the exponential decay of H₂O with CH₄ mole fraction? Is it due to line interferences? It is difficult for readers to judge when the wavelengths of H₂O, CH₄ are not given. Is there an interference between CO₂ and H₂O as well? Notice that the exponential fits in Figures S3&S4 are based on very limited data points. What’s the air matrix of the 100 ppm CH₄ cylinder? Could the dependence of H₂O measurements be caused by other species?

We realise that the choice of exponential fit is arbitrary, but we chose this as it provided a suitable fit the available data points. However we also now note that that the WMO calibration scale only extends up to 5 ppm of methane, so attempting to model the water baseline any higher than this is problematic and potentially meaningless. We have therefore changed our approach and we now instead use a linear fit across the methane mole fraction range.

There is a difference between the water and methane absorption peaks of 0.2 nm which is now stated in paragraph 1 of section 2.1. This line separation is sufficient that we do not expect spectral line overlap to have a significant impact here, as the reviewer rightly points out.

All cylinders used during the water correction contained a synthetic air mix that included ambient levels of argon. While including argon in the mix is important for the calibrations (as it ensures the dry air broadening for the cylinder measurements is representative of dry air broadening for the sample measurement) we do not expect the air matrix to affect the water baseline because this represents the retrieved water vapour mole fraction for a dry sample. Therefore changes in the water baseline reflect changes in the baseline offset, not the line broadening, for this absorption line.

3. Regarding the uncertainties of the estimated fluxes σ_F , what are the fractional contributions due to individual components? This information may help reduce the uncertainties in future measurements.

This is a very useful idea. The σ_F uncertainties were derived from flux density uncertainties, which were in turn derived by combining individual statistical uncertainties in quadrature. Therefore fractional uncertainties (that sum up to form σ_F) cannot be derived. However it is possible to derive σ_F for individual uncertainty components, assuming other uncertainties to be zero. This revealed that uncertainty in wind speed was the dominant source of uncertainty. This conclusion is now given in the paragraph 3 of section 4, with individual σ_F results given in the supplement.

Detailed comments:

L92-93: The flow rate needs to be given when the e-folding time is discussed. Alternatively, the e-folding volume can be provided.

This is a very useful comment. The flow rate through both instruments is now given in Table 1.

L97-98: The unit should be ppb instead of ppm.

Thank you for spotting this. This has now been corrected.

L159-162: should make it clear that +0.27% and +1.8% are the differences between with and without the water vapour corrections, instead of an increase of measurement accuracy.

The suggested change has now been made.

L207: Equation 10 should use the molar density of dry air since CH₄ is given in dry mole fraction.

The reviewer raises a very valuable point here. We realise that we have thus far used wet methane mole fraction when this should have been converted into dry mole fraction. This requires the humidity to be taken into account when calculating molar density. We have renamed the “molar density of air” as “molar density of dry air” in paragraph 2 of section 2.4 to reflect this. We are thankful to the reviewer for raising this.

L234-239: what was the nominal flow rate? Was the flow rate recorded? What fraction of measurements on average were omitted from each flight?

Although the flow rate was not recorded, the average volumetric flow rate through the tubing can be calculated to be $(110 \pm 10) \text{ cm}^3 \text{ s}^{-1}$ from the volume of the tube and the measured lag time, which was determined as the time between a spike (cough) at the inlet and the instrument response. This has been added to paragraph 3 of section 3.3 alongside the measured flow rate through the instrument of $(27.90 \pm 0.05) \text{ cm}^3 \text{ s}^{-1}$.

16% of all sampling conducted by UAV1 was omitted due to kinks in the tubing. This now stated paragraph 3 of section 3.3.

L239-240: what was the flow rate through the pMGGA?

The flow rate was measured to be $(5.08 \pm 0.02) \text{ cm}^3 \text{ s}^{-1}$. This has been added to Table 1 and to paragraph 3 of section 3.3.

L326-330: Comparing T1.1 with T1.2 in Figure 3, I expect that larger emissions would be quantified for T1.1 and with larger uncertainties, however, the results showed the opposite. Why is that? Where are the centers of the plumes found?

The reviewer makes an interesting observation. While it may appear as though survey T1.1 should derive higher flux as there were more plume intersections, on close examination of where T1.2 intersected the plume near the ground, methane mole fraction enhancements in T1.2 were actually far higher than in T1.1. This can be seen more clearly in the time series in Figure 2, where mole fractions of up to 12.5 ppm were recorded for T1.1, whereas up to

25.3 ppm was recorded for T1.2. Both plumes were horizontally centred near the centre of the sampling plane, with T1.1 centred at -3 m and T1.2 centred at -5 cm from the centre line, according to the NGI method. The importance of taking into account the magnitude of flux density peaks, as well as the number of plume intersections is now described in a new paragraph (paragraph 4) of section 4.

L335-341: It looks that the crosswind distance is not sufficient to cover the plume, especially for UAV1. Why were the transects of UAV1 not centered? The current sampling tends to miss the center of the plume.

There may be some misunderstanding here, as there may be some ambiguity in the description of our flux quantification methodology, which we hope we have now improved. The sampling shown in Figure 3 and Figure 4 may appear as if the time-averaged plume was not centred. This was because the time-invariant plume was narrow (when nearer the source) and appeared episodically due to coincidental intersection with the UAV. When nearer to the source (approximately 50 m away), the time-invariant plume can be conceived to have had less time to disperse compared to sampling approximately 100 m away. Therefore it may appear as though the time-averaged plume was not centred in our sampling plane. We now explain this in the final paragraph of section 3.3. However, in reality, where the time-averaged plume is centred on our arbitrary sampling plane is irrelevant to the flux calculation, so long as the time-averaged plume morphology can be characterised (as is the case here).

During our sampling campaign, we ensured that we flew on a downwind sampling plane, with the UAV approximately centred downwind of the source using wind direction measurements. The flight strategy was decided on each day based on wind forecasts and on-site surface wind measurements to optimise downwind sampling of the turbulently advected emission plume. This point is now made in paragraph 2 of section 3.1 and its implications are discussed in paragraph 4 of section 3.3.