A tracer release experiment to investigate uncertainties in drone-based emission quantification for methane point sources

Randulph Morales et al.

Response to the Reviewer's Comments

We thank the three reviewers for their positive comments, critical assessment, and useful points to improve the quality of our paper. In the following, we address their concerns point by point. Changes in the paper are shown in blue.

Reviewer 1

General comments

Reviewer Point P 1.1 — I recommend swapping th phrase 'tracer release' for something more appropriate such as 'controlled release', especially in the title. So-called tracer techniques rely on the concurrent measurement of the target gas (in this case methane), and a tracer gas, and are generally referred to as tracer ratio, tracer dispersion or tracer release methods in the literature (see e.g. Mønster et al., 2014; Yacovitch et al., 2017, and various references in the authors' own introduction) . Such an experiment, involving a tracer gas, was not performed here. Validation of plume mapping and flux methodology using a source with a known emission rate (as done in this work) is usually referred to as a controlled release experiment (see e.g. Thorpe et al., 2016; Heltzel et al., 2020; Shah et al., 2020).

Reply: We agree with the reviewer and changed all the existing 'tracer release' to "controlled release', including the title of the publication.

> A controlled release experiment to investigate uncertainties in UAV-based emission quantification for methane point sources

Reviewer Point P 1.2 — Drone and UAV are used interchangeably throughout the text. It should be made more clear, somewhere early on, that the two refer to the same thing and one term (probably UAV) used consistently.

Reply: Changed as suggested

Reviewer Point P 1.3 — Figure text is often very difficult to read without considerably zooming in. All figure text needs to be much larger.

Reply: All figures are replotted and all font sizes are changed accordingly.

Reviewer Point P 1.4 — Calibration of the three instruments, with respect to measurements of methane, is not mentioned at all in the text. Was calibration to a methane standard (World Meteorological Organization) performed at all, and if so what was the calibration procedure? If no calibration was performed, this throws the validity of the results into question.

Reply: This is an important point. We added descriptions of the calibration for the three instruments in their respective sections.

1. Sect. 2.1: QCLAS

   The instrument's precision, linearity, and calibration were described in detail elsewhere (Tuzson et al., 2020). Briefly, the instrument was calibrated by inserting it into a custom-built small volume ($60\,\mathrm{L}$) climate chamber. This chamber was then hermetically sealed and continuously purged with a certified calibration gas with high CH4 concentration ($200\,\mathrm{ppm} \pm$ 1%; PanGas, Switzerland). Furtheremore, the gas was dynamically diluted with dry nitrogen ($\mathrm{N_2}$) in a stepwise fashion using calibrated mass flow controllers. The overall uncertainty was estimated to be $\pm$2%. Repeated experiments showed that the instrument preserves its linearity and only a marginal drift may appear in the offset. This, however, is fully accounted for, when applying the background $\mathrm{CH_4}$ subtraction step (see Sect. 4.4).

2. Sect. 2.2: Active AirCore-CRDS:

   A single-point calibration was used to correct the potential drift of the CRDS measurements. Measured methane mole fraction obtained using the AirCore system was linked to a known calibration standard that is traceable to the WMO X2004A $\mathrm{CH_4}$ scale (Vinkovic et al., in review)

3. Sect. 4.2:LI-COR-OTM-33A

   The analyzer was calibrated before and after each measurement on the field and can be linked to at least two certified standards: the atmospheric $\mathrm{CH_4}$ value ($2\,\mathrm{ppm} \pm$ 5%), $5\,\mathrm{ppm}$ standard ($5.05\,\mathrm{ppm} \pm$ 5%), and a 25 ppm tank ($24.98\,\mathrm{ppm} \pm$ 5%).

Reviewer Point P 1.5 — The OTM-33A method usually uses instrumented vehicles and mobile sampling to quantify a flux, although examples do exist in the literature of stationary measurements (see e.g. Foster-Wittig et al., 2015; Shaw et al., 2020). Could the authors' comment on the possibility of applying the mobile OTM-33A method to the UAV-based measurements (has this possibility been explored)?

Reply: We have tested the OTM-33A approach on several drone flights during the ROMEO campaign (Röckmann and team, 2020). The method suggests the anemometer to be placed in approximately the same height and location as the analyzer. As explained in P1.8, we tried to mount an anemometer (TriSonic Mini, Anemoment) on top of the drone but failed to isolate the wind data. Moreover, due to the limited flight time of the UAV, we could not fly the drone long enough to be able to obtain sufficient data necessary to provide an emission flux using the OTM-33A approach.

Reviewer Point P 1.6 — The plots appear to show CH4 mole fractions of 0 ppm (Figure 3 and 5). Unless these plots are actually showing ΔCH_4 (CH_4 – background), this is impossible. The tropospheric background mole fraction of methane is roughly 1.9 ppm (Lan et al., 2021). Could the authors explain these results?

Reply: All reported CH_4 measurements in the manuscript are already above the background. We have changed the figure text into 'CH_4 - $CH_{4\mathrm{bg}}$ [ppm]' for easier comprehension. We added a paragraph discussing the background methane mole fractions as a response for P 1.16.

Reviewer Point P 1.7 — Abbreviations for the six methods (CK, OK, PW, LW and combinations thereof) are used inconsistently. It would be useful to the reader for them to be introduced more distinctly in the methods section and then used consistently throughout the results and in figures/tables.

Reply: We added a few sentences in Sect. 5 to establish the six methods and their corresponding abbreviation.

> ... A total of six quantification approaches were applied to all flights and evaluated for their ability to reproduce the true releases. These approaches arise from the combination of two different treatments of methane measurements and three different treatments of wind measurements. The treatments involved in mapping the discrete methane points into the measurement plane are the standard ordinary kriging (OK) and the cluster-based kriging (CK) interpolation schemes. The three different ways of estimating wind-speeds during each quantification flight involves the scalar wind (SW), logarithmic wind (LW), and projected wind (PW) as discussed in Sect. 4.

Reviewer Point P 1.8 — Did the authors consider measuring wind speed and wind direction in-situ from the drone by attaching an anemometer? Concerning wind speed estimation/interpolation, would there be any improvement using a combination of methods two and three (LW and PW)? Further, in Section 4.3, I would recommend definitively stating the abbreviated definitions used to refer to the three approaches later in the text (for example "Proj. wind" Table 2, or "PW" later).

Reply: Prior to the controlled-release experiment, we tried to mount an anemometer (TriSonic Mini, Anemoment) on top of the drone to test whether wind measurements obtained from this set-up is a viable quantification approach. However, our tests suggested that wind measurements obtained this way are too noisy due to the interference from the UAV, and we were not able to isolate the wind data. Regarding the combination of two different methods, the emission estimates between the two methods are very close to each another, especially between the PW and LW, thus we don't expect a significant improvement by using a combination.

Reviewer Point P 1.9 — How representative is the range of controlled release rates used here (0.2 – 0.7 g s^{-1}) of true emissions from oil and gas facilities, or other methane sources? I would expect real emissions to have a much greater range, and that the release rates used are at the lower end of that range. Are the authors' conclusions (for example, on wind speed, wind direction, and distance from plume limits) therefore only applicable to the controlled release rates used in this work, or are they equally applicable to emissions tens, or hundreds, of times stronger? If this is not the case, then the conclusions should be caveated by stating that these results are for a limited range of emission strengths.

Reply: A recent study by Omara et al. (2018) investigated over a thousand natural gas production sites in the US. In their study, 85% of the sites belonged to a low- to mid-level natural gas production site category with emissions in the range of 0.13 – $0.58\,\mathrm{g\,s^{-1}\,site^{-1}}$. Although emissions per site are quite low, the sheer number of low–mid-level production sites accounted for almost two-thirds (63% [CI:45–83%]) of the $\mathrm{CH_4}$ budget in the US. "Super-emitters", producing an average of $2.31\,\mathrm{g\,s^{-1}\,site^{-1}}$, only accounted for 13% [CI:7-21%] of the total $\mathrm{CH_4}$ budget. Thus, we consider our results to be representative for the quantification of emissions from low–mid-level oil and gas wells. We have added this information in Sect. 3: Control Release Experiment to put in context the chosen release rates during the experiment:

> The release rates used in this study are a good representation of emissions from normal operating (i.e., excluding super-emitters) natural gas production sites in the US which produces 0.13–0.58 $\mathrm{g\,s^{-1}}$ (Omara et al., 2018).

Specific comments

Reviewer Point P 1.10 — L13: It is not clear to me what "stretched by 7 s and 0.06 seconds for every second of QCLAS measurement, respectively" is referring to here. This phenomenon is better explained in Section 5.3 and the authors should consider amending the abstract text to avoid confusion.

Reply: We have revised the abstract which now reads:

> ...smoothed by $20\,\mathrm{s}$ and had an average time lag of $7\,\mathrm{s}$. AirCore measurements were also shifted linearly with time at an average rate of $0.06\,\mathrm{s}$ for every second of QCLAS measurement.

Reviewer Point P 1.11 — L105: For comparison, it would be useful to include the instrument measurement precision for the Picarro CRDS (as mentioned for the QCLAS system on L83).

Reply: We modified the text accordingly and added a sentence.

> The precision $(1\sigma, 0.25\,\mathrm{Hz})$ of the CRDS was determined to be better than $0.7\,\mathrm{ppb}$.

Reviewer Point P 1.12 — L129: Could “not too strong winds” be quantified here e.g. greater than X m s-1? I also assume this was due to the limitations of the UAV system used? Explaining the reason behind this limitation would be useful for guiding others.

Reply: We modified the sentence as follows:

> ... ,i.e. days with no precipitation and a sufficiently large wind speed but smaller than $8\,\mathrm{m\,s^{-1}}$ which is the maximum value given by the UAV flight specifications.

Reviewer Point P 1.13 — Figure 3

1. The wind rose is exceptionally small and doesn’t add much information to the figure in its current form. The wind rose might be better viewed in a separate panel, adjacent to the top-down view of the CH4 data. The wind rose is also not mentioned in the figure caption.
2. Is the orange line showing the source-transect distance? This is not clear and should be made clear in the figure caption.
3. The figure may benefit from an additional arrow illustrating the average wind direction for this flight (which ties in with the wind rose).

Reply: Figure 3 has been replotted.

1. The wind rose has been placed adjacent of the top-down $\mathrm{CH_4}$ data and caption was revised to include windrose
2. Yes, the orange line is the source-transect distance. Now, it is explicitly mentioned in the caption.
3. From the wind rose, one can infer the average wind speed and direction for this flight.

Reviewer Point P 1.14 — L195: As for the Picarro instrument, it would be useful to include instrument characterisation (measurement precision etc.) here for the Li-COR instrument.

Reply: We have added some specifications of the instruments and added it on Sect. 4.2

> The $\mathrm{CH_4}$ analyzer has a portable footprint ($12\,\mathrm{kg}$, 51 × 33 × 18 $\mathrm{cm^3}$) and can measure methane mole fractions up to $50.0\,\mathrm{ppm}$. It operates between -25 and 45 °C and can reach a precision (1σ) of $0.6\,\mathrm{ppb}$ at $1\,\mathrm{s}$ and $0.25\,\mathrm{ppb}$ at $5\,\mathrm{s}$ averaging time.

Reviewer Point P 1.15 — L219: Extra “to” in “matching the timestamp of the anemometer to the to GPS location”.

Reply: Correction applied.

Reviewer Point P 1.16 — L230: For clarification, were background CH4 mole fractions measured upwind of the emission source, or from either side of the emission plume? It might be useful to present the measured background mole fraction value(s) and uncertainty somewhere.

Reply: Background $\mathrm{CH_4}$ mole fractions were measured on either side of the emission plume. A discussion of measured background mole fraction was added and is now discussed in Sect. 4.4.

> Background $\mathrm{CH_4}$ mole fractions were determined from measurements outside of the emission plume. Each sampled vertical height was extended to pass both sides of the plume to ensure sampling of local background values. Local variation of measured background values were corrected by using the Robust Extraction Baseline Signal (REBS) algorithm developed by Ruckstuhl et al. (2012). Average $\mathrm{CH_4}$ background mole fraction during the whole release experiment was determined at 2.09 $\pm\, 0.19\,\mathrm{ppm}$. Take-off and landing times of the UAV were noted and all data before and after the flight were removed.

Reviewer Point P 1.17 — L237: Missing the word “to” between “due” and “the”.

Reply: Correction applied.

Reviewer Point P 1.18 — L271: Two identical references on this line - Tadić et al. (2015)

Reply: We corrected the identical reference.

Reviewer Point P 1.19 — L285: Missing the word “one” between “only” and “cluster”.

Reply: Change as suggested.

Reviewer Point P 1.20 — Figure 5

1. Could the caption include which instrument was used for the methane measurements shown?
2. The amount of panels here makes readability particularly difficult. I would recommend splitting into two separate figures: Fig. 5a as a single figure, and Fig. 5b, 5c, and 5d as a single figure. It may also be useful to have a direct side-by-side comparison of actual in situ measured CH4 (showing sparse spatial distribution on the vertical plane) alongside the ‘predicted measured’ Krigged CH4.
3. Fig. 5b: As in general comments above, here CH4 mole fraction is in a range of -0.06 to +0.06 ppm. These values are impossible (especially the negative values) in the troposphere. Could the authors explain these results?

Reply: As suggested, we split Figure 5 into two separate figures.

1. Measurements were taken using the in-situ UAV-based QCLAS and this information was added in the caption of Figure 5.

2. Figure 5 has been divided into two separate figures as suggested by the reviewer. A side-by-side comparison of measurements vs. predicted measured methane mole fraction is now shown in Fig. 6B and 6C.

3. As mentioned, all $\mathrm{CH_4}$ reported in the manuscript are already above the background. The background values are discussed in Sect. 4.4

Reviewer Point P 1.21 — I would recommend moving equations 17 through 23 (and surrounding text) to a relevant section(s) in the methods section, as this is more Methodology than Results.

Reply: We moved subsection 5.1 into subsection 4.6: Method - Example of quantification procedure

Reviewer Point P 1.22 — Table 2

1. This table is difficult to read due to the sheer amount of values. The information is much better visualised in a plot such as Figure 6. I would consider moving the full table to the Supplement, and only including the overall results (NMAE, Bias, RMSE) for all six methods in the main manuscript.

2. Abbreviations for the six methods (e.g. CKPW) are used throughout the text but not in this table.

Reply:

1. As suggested, we moved Table 2 in the supplement and only kept the overall results of the six quantification methods. We rewrote L387-388 to properly account for the changes in referencing the table:

   > The overall performance of each quantification approach is presented in Table 2 and estimated emission rates together with the true release rates for every individual flight are presented in Table S1.

2. The abbreviations for the six methods are included in the header of the table.

Reviewer Point P 1.23 — Figure 8: The caption should probably mention that these are residuals in flux estimates.

Reply: Changed as suggested.

Reviewer Point P 1.24 — Section 5.2.2: Could this section refer to Figure 8 as well, and the comparison of different meteorological regimes?

Reply: As suggested by Reviewer 2, we added a caption in figure 8 mentioning that optimal conditions and suboptimal conditions are defined in Section 5.1.2

Reviewer Point P 1.25 — Table 4: Would it be useful to present the NMAE, bias, and RMSE in this table, as done in comparisons of the AirCore with the QCLAS results (Table 3), and for the comparison of the six drone-based methods (Table 2)?

Reply: We have added the statistics in Table 4.

Reviewer Point P 1.26 — The link in the reference for US EPA 2014 goes to a page which states that "Emissions Measurement Center has Moved" – the link might need to be corrected.

Reply: The citation and the link have been updated.

Reviewer Point P 1.27 — Figure S6: Would it be worth showing the plume constructed from the QCLAS data too, for comparison?

Reply: The constructed methane plume for the QCLAS data is similar to the one shown in Fig. 6. Nevertheless, we added the constructed QCLAS methane plume in Fig. S6 to aid the comparison the difference between the two systems.

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