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 2

5 Response to the Reviewer's Comments

General comments

- **Reviewer Point P 2.1** The Introduction/references, scope of work, and scientific approach of the work are good. Some issues I found already mentioned by Reviewer 1 include finding L13 of the abstract confusing about the stretching by 0.06 seconds, wondering whether the methane data in all the plots are already background-subtracted, and suggesting
- 10 improvements to the general readability of figures (other than Fig 1).

Regarding the novel cluster kriging approach adapted here, the paper cited by van Stein et al. (2020) concluded the method is designed to 'reduce the time and space complexity of the kriging method'. While dividing into elevated and background clusters makes sense, I do wonder how the above statement fits in. Specifically, if the difference between cluster kriging and ordinary kriging shown here has less to do with the theoretical basis of the method, and more a

- 15 difference in the parameters used given that Fig 5d on left has a significantly different appearance (length scale or search radius?) than that of Fig 5c. Please explain. In general, while the math is presented if Section 4.5.2, I think some of the more practical details could be mentioned. Does the cluster kriging python package mentioned in the code availability statement also perform ordinary kriging, or that comes from elsewhere? Maybe add an example of the semivariogram or kriging parameters in the supplement to better illustrate the method?
- 20 **Reply**: We have revised the abstract to reduce the ambiguity in the text. All reported CH₄ mole fractions in the plots are enhancements above a background and we changed the labels of the plots to clarify this. Additional information on how background measurements were identified were added in Sect. 4.4. All plots were redone to improve readability and Fig. 5 was divided into two plots to reduce the amount of information in one single figure.

The average runtime to reconstruct the methane plume using the cluster kriging approach was around $150 \,\mathrm{s}$ on a single

- core Intel Xeon W-2175 CPU @ 2.5 GH for 80x20 m² grid at a 0.5 mx0.5 m resolution, while it took around 270 s to do the same using ordinary kriging—supporting the statetment of van Stein et al. (2020) on time-reduction. We agree, however, that the main motivation for applying the method was to account for the structural differences between the background field and the plume enhancements rather than computational aspects. The code also performs ordinary kriging. Regarding the significant difference between the cluster kriging vs ordinary kriging, the advantage of using the cluster kriging approach
- 30 is the presence of the membership probabilities of the background and the elevated clusters. We have added Fig. S7 in the supplementary material illustrating the difference between cluster and ordinary kriging. The figure presents the reconstructed emission plume using ordinary kriging with length scales similar to that of the cluster kriging approach. It shows that the absence of membership probability of the background and elevated cluster in the ordinary kriging tends to produce noisier emission fields resulting in a 10% decrease in emission flux estimate.

35 Specific comments

Reviewer Point P2.2 — L27: Gurney et al. (2021) in Nature Communications is likely the wrong reference here. That paper is focused on FFCO2 from cities, not CH4 from oil and gas.

Reply: We have changed the reference and cited Alvarez et al. (2018); Omara et al. (2018); Zhang et al. (2020) for the text.

40 **Reviewer Point P2.3** — L48-50: Shaw et al. (2021) and / or Hollenbeck et al. (2021) could be also considered adding here, as they are recent reviews on the subject of UAV methane quantification.

Reply: These are indeed relevant new publications, which are now cited in the manuscript.

Reviewer Point P 2.4 — L114: 'capturing raw streams' – This is perhaps too vague. I think it is not so much a raw stream as a different stream (the carrier phase).

45 Reply: We edited the text to be more specific

... by capturing measurements of carrier phase signals from the GPS satellites ...

Reviewer Point P 2.5 — Figure 2

- 1. The 14 on the x-axis tick labels is unneeded (see presumably matplotlib.dates.DateFormatter)
- 2. Isn't 0 AGL [m] defined as the takeoff altitude for UAV-GPS by the Matrice? Also, are takeoff and landing locations
- 50 here different or the same?

- 3. The pressure altitude is impressively consistent comparing against RTK altitude. The spikes seen in the bottom panel of Figure (2) could be a little misleading since they appear to be caused simply by small differences in timing relative to the RTK during ascent and descent where altitude is changing quickly.
- caption the meaning of subscript m in bottom panel legend could be mentioned (slope from linear regression).
 They must also have some impact on the pressure altitude drift estimate unless robust regression was used

55

Reply:

- 1. Figure 2 has been replotted to remove the day "14". Labels have also been made bigger for better readability.
- 2. The 0 AGL [m] is defined as the takeoff altitude of the RTK-GPS. The takeoff and landing locations are not exactly at the same point but location of the two points is $\sim 2 \text{ m}$ radius of each other.
- To have a better representation of how the pressure altitude and UAV altitude differs from the RTK altitude, we have removed the data points related to the sudden ascent and descent of the UAV.
 - 4. The caption text was adapted and we added a sentence at the end:

Dashed blue and orange lines are fits representing a linear regression with the subscript m referring to the slope of the line.

65 **Reviewer Point P 2.6** — L144: Later, the make/model of 3D sonic anemometer is mentioned (uSonic-3 Scientific). What was the type of 2.5D anemometer?

Reply: The 2.5D anemometer is a TriSonic Mini from Anemoment. We have included the make and manufacturer of the 2.5D in the manuscript.

Reviewer Point P2.7 — Table 1 stability here is presumably based on equation from L215, not the Pasquill stability 70 classes, which are also mentioned (L180)

Reply: Stability classes listed in Table 1 were indeed not determined according to Pasquill but by computing the turbulence parameters such as the Obukhov length and friction velocity. The Pasquill stability (PS) classes mentioned in L180 is a note to specify that the quantification procedure of OTM-33A was developed using the PS wind classes. We added this information in Sect. 3.

Stability classes listed in Table 1 were determined by calculating a dimensionless height, $\zeta = z/L$, where *z* is the height of wind measurement and *L* is the Obukhov length. The dimensionless height is used as a stability parameter where $\zeta < 0$ indicates unstable, $\zeta > 0$ unstable, and $\zeta = 0$ for neutral conditions. **Reviewer Point P 2.8** — L250: It's a little unclear if the 3S algorithm is new to this manuscript, or if it is presented in the two manuscripts cited on L247 that are in preparation / review. With being able to read those, the writing here is a is a

80 little hard to follow. A simple 1D Gaussian smoothing function need only have one parameter – a standard deviation. How does F(x,b) accommodate three parameters?

Reply: The 3S algorithm is not written in the two manuscripts that are in preparation and review. Thus, we have revised and added some clarifying text in the section.

We approximate the active AirCore measurement as, y, defined as

85
$$\mathbf{y} = \mathbf{f}(\mathbf{x}, \mathbf{b}) + \mathbf{e}$$
 (1)

where f is a model function that fits the high-resolution QCLAS and projects it onto the low-resolution AirCore measurement. The model function consists of x which is the independent variable where the QCLAS is measured (i.e., timescale) and the fit parameters b containing three elements describing the shift, stretch, and smoothing (i.e., 3S) of the AirCore. The error e represents the instrument's error as well as the error from the model function. We used a 1st-order Lagrange polynomial interpolation and applied a Gaussian filter to parametrize the shift, stretch, and smoothing of the AirCore. Starting with an arbitrary initial guess, the optimal parameters $\hat{\mathbf{b}}$ was determined using a nonlinear least squares fit solved iteratively using the Gauss-Newton method.

Reviewer Point P 2.9 — Figure 4

90

105

- 95 1. Legend : 'Stretch' is written twice. Is one of them supposed to be shift? Also, the 0.06 s/s stretching is mentioned in abstract, but the other two numbers (12.81 and 17.90) are different?
 - 2. Frankly, the algorithm mainly just seems to correct for the shift, also called time lag by some other authors. Are the other two parameters really helpful?

Reply: Figure 4 was redone to correct the legend

- 100 1. The numbers written in Fig. 4 are specific for that quantification flight. We derived the shift, stretch, and smoothing parameters for every individual flight where the QCLAS and the AirCore were present and took the average. The values written in the abstract are the average values.
 - 2. We think that all three parameters are essential: The signal of the AirCore is smoothed out considerably due to mixing in the sampling tubes. Furthermore, the time of the AirCore measurement can only be indirectly determined, and our results showed that the time is not only be shifted by a constant value ("shift") but that the time lag may change over the duration of the flight ("stretch"). A reconstructed plume without using the stretch parameter for flight 312_03 is shown in the figure below.

4



Figure 1. Reconstructed methane plume for flight 312_03. The figure on the upper left shows a reconstructed plume from the QCLAS measurements using the cluster-based kriging approach. The figure on the upper right is the reconstructed plume without applying a proper time correction (i.e., no shift and no stretch) for AirCore measurements, whereas, the figure on the bottom left is a reconstructed plume obtained after applying the proper time correction (i.e., with both shift and stretch). The figure on the lower right is a reconstructed plume but only applying the time-lag and excluding the stretch (i.e., with shift and no stretch). By not accounting the stretch parameter, the methane plume split into two spatially.

Reviewer Point P 2.10 — L361 16.04 kg should be g for the molar mass of methane

Reply: Correction applied

110 Reviewer Point P 2.11 — Table 2: In footnote about optimal conditions, suggest mentioning they are defined in Section 5.2.2

Reply: We added this information in the caption, but we moved the full table to the supplement and only retained a summary of the results as suggested by Reviewer 1.

Reviewer Point P2.12 — Table 5: Suggest putting 'This study' (or similar) in the column next to Airborne CKPW massbalance. Or somehow clarify, since the studies mentioned here - Golston et al. (2018); Yang et al. (2018); Shah et al. 115 (2020) - do not use CKPW.

Reply: We have adopted the suggested change.

Reviewer Point P2.13 — L529: 'Under these conditions, measuring at a downwind distance of 75 m ensures the true emission can be fully mapped both horizontally and vertically'. This is a little confusing, since it sounds like you need to be > 75 m downwind to fully capture the plume, while L523 indicates underestimation at those distances. 120

Reply: We have changed the text as suggested by reviewer 3.

...measuring at a downwind distance of less than $75 \,\mathrm{m}$ ensures the true emission to be fully mapped...

Reviewer Point P 2.14 — Figure 6, 8, and 9 show 'residuals' in %, which here must mean the percentage error of the estimate versus the known controlled release amount (but without calculating absolute values). Where does the 'range of residuals' come from?

125

Reply: "Range of residuals" shown in Fig. 6 was computed by taking the percentage error of the estimate-including the uncertainty of every individual flight-versus the known controlled release amount. In Figs. 8 and 9, the range of residuals only refers to the range of average residual (i.e., black dots in Fig. 6) computed for each quantification flight.

Reviewer Point P2.15 — L742 Suggest replacing the dead link to U.S. EPA with the new link

130 **Reply**: We have updated the U.S. EPA link

References

135

145

- Alvarez, R. A., Zavala-Araiza, D., Lyon, D. R., Allen, D. T., Barkley, Z. R., Brandt, A. R., Davis, K. J., Herndon, S. C., Jacob, D. J., Karion, A., Kort, E. A., Lamb, B. K., Lauvaux, T., Maasakkers, J. D., Marchese, A. J., Omara, M., Pacala, S. W., Peischl, J., Robinson, A. L., Shepson, P. B., Sweeney, C., Townsend-Small, A., Wofsy, S. C., and Hamburg, S. P.: Assessment of methane emissions from the U.S. oil and gas supply chain, Science. 361, 186–188, https://doi.org/10.1126/science.aar7204, 2018.
- Golston, L., Aubut, N., Frish, M., Yang, S., Talbot, R., Gretencord, C., McSpiritt, J., and Zondlo, M.: Natural Gas Fugitive Leak Detection Using an Unmanned Aerial Vehicle: Localization and Quantification of Emission Rate, Atmosphere, 9, 333, https://doi.org/10.3390/atmos9090333, 2018.

Gurney, K. R., Liang, J., Roest, G., Song, Y., Mueller, K., and Lauvaux, T.: Under-reporting of greenhouse gas emissions in U.S. cities,
 Nature Communications, 12, 1–7, https://doi.org/10.1038/s41467-020-20871-0, 2021.

- Hollenbeck, D., Zulevic, D., and Chen, Y.: Advanced Leak Detection and Quantification of Methane Emissions Using sUAS, Drones, 5, https://doi.org/10.3390/drones5040117, 2021.
 - Omara, M., Zimmerman, N., Sullivan, M. R., Li, X., Ellis, A., Cesa, R., Subramanian, R., Presto, A. A., and Robinson, A. L.: Methane Emissions from Natural Gas Production Sites in the United States: Data Synthesis and National Estimate, Environmental Science & Technology, 52, 12 915–12 925, https://doi.org/10.1021/acs.est.8b03535, pMID: 30256618, 2018.
- Shah, A., Pitt, J. R., Ricketts, H., Leen, J. B., Williams, P. I., Kabbabe, K., Gallagher, M. W., and Allen, G.: Testing the near-field Gaussian plume inversion flux quantification technique using unmanned aerial vehicle sampling, Atmospheric Measurement Techniques, 13, 1467–1484, https://doi.org/10.5194/amt-13-1467-2020, 2020.

Shaw, J. T., Shah, A., Yong, H., and Allen, G.: Methods for quantifying methane emissions using unmanned aerial vehicles: a

- 150 review, Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 379, 20200450, https://doi.org/10.1098/rsta.2020.0450, 2021.
 - van Stein, B., Wang, H., Kowalczyk, W., Emmerich, M., and Bäck, T.: Cluster-based Kriging approximation algorithms for complexity reduction, Applied Intelligence, 50, 778–791, https://doi.org/10.1007/s10489-019-01549-7, 2020.

Yang, S., Talbot, R., Frish, M., Golston, L., Aubut, N., Zondlo, M., Gretencord, C., and McSpiritt, J.: Natural Gas Fugitive Leak De-

155 tection Using an Unmanned Aerial Vehicle: Measurement System Description and Mass Balance Approach, Atmosphere, 9, 383, https://doi.org/10.3390/atmos9100383, 2018.

Zhang, Y., Gautam, R., Pandey, S., Omara, M., Maasakkers, J. D., Sadavarte, P., Lyon, D., Nesser, H., Sulprizio, M. P., Varon, D. J., Zhang, R., Houweling, S., Zavala-Araiza, D., Alvarez, R. A., Lorente, A., Hamburg, S. P., Aben, I., and Jacob, D. J.: Quantifying methane emissions from the largest oil-producing basin in the United States from space, Science Advances, 6, eaaz5120,

160 https://doi.org/10.1126/sciadv.aaz5120, https://www.science.org/doi/abs/10.1126/sciadv.aaz5120, 2020.