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

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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 3

General comments

Reviewer Point P 3.1 — In general I've tried not to repeat comments already made by the other reviewers, but I do agree with Reviewer 1 that a brief discussion of calibration is required. I also think it would help to clarify things if the term "$\mathrm{CH_4}$ enhancement" were to be used in cases where background values have been subtracted from the data (which I think is pretty much everywhere). Perhaps a couple of extra sentences briefly summarising the application of the REBS algorithm would be useful too (especially with regard to the Reviewer 1 question concerning where the background measurements were taken - I assume the answer is anywhere on the downwind measurement plane that the REBS algorithm identified)?

Reply: We agree that a brief discussion on the calibration of the instrument is needed to validate the measurements. We included a short text briefly discussing the calibration method of the instruments in their respective sections (Sect. 2.1, 2.2, and 4.2). The $\mathrm{CH_4}$ molar fractions reported in the manuscript indeed refer to "$\mathrm{CH_4}$ enhancement". To address this aspect, we adapted the text and referred to measured $\mathrm{CH_4}$ as enhancements. We also changed all plot labels and used "$\mathrm{CH_4}$ - $\mathrm{CH_4}_{\mathrm{bg}}$ [ppm]" instead. Additional clarification was also added in Sect. 4.4 regarding how the background values were identified.

Specific comments

Reviewer Point P 3.2 — L27: Alvarez et al. (2018) would be a more appropriate reference here (Gurney et al. (2021), is definitely wrong), although there are more recent options that would do the job too.

Reply: We replaced Gurney et al. (2021) by Alvarez et al. (2018) and also added the work of Omara et al. (2018); Zhang et al. (2020) in the citation.

Reviewer Point P 3.3 — L202:the star on the friction velocity should be a subscript (as in Equation 4).

Reply: We have corrected the typo.

Reviewer Point P 3.4 — L226:I'm a bit confused as to why this step was necessary. If both the QCLAS and RTK-GPS received GPS signals, why were they not already synchronised on GPS time?

Reply: This was just done to make sure that all the clocks across all systems, including the AirCore, have the same clock.

Reviewer Point P 3.5 — L230:in addition to my general point above, it would probably be clearer to say that background CH4 mole fractions were "subtracted" instead of "removed".

Reply: We have adapted the word "subtracted".

Reviewer Point P 3.6 — Equation 5: I understand that this approach is based on previously published work, but the application is sufficiently different that it would be useful to provide some more information here. I suggest explicitly stating the form of F. Have I got it right that the parameter vector b consists of the QCLAS measurements? If so I would also state that explicitly.

Reply: We have revised the text surrounding Eq. 5 to add further detail on the process of matching the AirCore data to the QCLAS data (see response to P2.8).

Reviewer Point P 3.7 — Figure 4: Somewhere in either the caption and/or the associated main text it should be explicitly stated that these values were optimised separately for each flight.

Reply: We have fixed the text in Sect. 4.4.1: Processing of AirCore measurements

Reviewer Point P 3.8 — L315 : maybe I missed something, but is it explained anywhere how the data are hard-clustered prior to performing ordinary kriging?

Reply: We have revised the text to explain this:

> Hard clustered data-points are obtained by rounding the probability obtained from the GMM to either belong to the background or the elevated cluster.

Reviewer Point P 3.9 — L322 : I have no doubt that the Matèrn covariance kernel is a valid choice here, but as a general comment I feel that the choice of kernel should be based on an examination of the specific dataset on which kriging is being performed (although of course it can be guided by previous studies/experience). I'm sure that such examination was performed (i.e. someone checked to make sure the optimised function was a reasonable fit to the data for each flight) - I'm happy to leave it up to the authors as to whether stating this explicitly would be useful or not.

Reply: We did indeed try to use different covariance kernels (e.g., spherical, exponential, and gaussian among others) for our dataset and the best results were obtained with the use of a Matérn covariance kernel. The choice of using this kernel was reinforced when we came across the study of Stachniss et al. (2009) which also tested different covariance kernels in predicting a concentration field.

Reviewer Point P 3.10 — L324 : was anisotropy in the hyper-parameters considered? My prior assumption would be that the vertical and horizontal length scales could be quite different, but perhaps that was found not to be the case here?

Reply: Anisotropy was not particularly considered in the optimization of the hyperparamaters.

Reviewer Point P 3.11 — Equation 15 : this is a really minor point, but just to make sure I've understood things - is y not already included in the set X?

Reply: Yes.

Reviewer Point P 3.12 — Figure 5 : I agree with Reviewer 1 - this would be best split into two separate figures. Also, the grey outline on the circles in Fig. 5a needs to be removed, as you currently have to zoom in a lot in order to see the fill colours of each point.

Reply: Figure 5 has been divided into two separate figures as suggested by reviewer 1. The grey outline on the circles are removed and the markers were also made bigger.

Reviewer Point P 3.13 — L431 : I'm not sure if this is the best place for it, but I think it is worth mentioning somewhere that there are alternative ways to deal with this smoothing problem. One approach is to select a variogram model that results in nearby points being assigned large weights (e.g. a linear model). Such a model must obviously be supported by the experimental variogram, but in any case a subjective choice must always be made regarding how the model parameters should be optimised to "best fit" the data. Therefore it can reasonably be justified that the model variogram should be chosen with a particular focus on representing the experimental data at small separation distances (see Kitanidis, 1997), for further discussion). A moving neighbourhood approach can also be adopted; in fact this is the default approach in the frequently used EasyKrig MATLAB package (see e.g. O'Shea et al., 2014; Pitt et al., 2019). The cluster-based approach presented here has some advantages over these alternatives; in particular it removes many of the more arbitrary subjective choices associated with them. I think they are worth mentioning in this context, probably just a sentence or two would do.

Reply: Thank you for this information. We revised the text and moved it in Sect. 4.5.2: Kriging estimate

Although other kriging option modules are available such as a moving neighborhood approach where only data-points within a certain radius are considered in the kriging process (Mays et al., 2009; O'Shea et al., 2014; Pitt et al., 2019), the cluster-based kriging approach offers the advantage of removing many arbitrary subjective parameters present in other approaches.

Reviewer Point P 3.14 — L527 : Needs rephrasing. Could go for "As a general guideline, performing drone-based emission quantification of emission sources requires. . . "

Reply: We have adapted the changes.

Reviewer Point P 3.15 — L529 : Would it be clearer to say "at a downwind distance of less than 75 m"?

Reply: We have adapted the suggestion of the reviewer.

References

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