1 Response to Reviewer Comments 1 (RC1)

Link to Original Submission: A New Technique for Airborne Measurements to Quantify Methane Emissions Over a Wide Range: Implementation and Validation – Dooley et al. (2024)

(RC 101) The title is a bit vague in referencing a "wide range" – it is referenced that the UAS can sample scales of up to 1 km, but aircraft can sample at much wider ranges... would "A new UAS-based technique for quantifying and attributing methane emissions from small and distributed point sources: [...]" or similar be more relevant?

(AC 101) ***Title.*** We appreciate the reviewer's suggestion on the title and have updated it to "A New Aerial Approach for Quantifying and Attributing Methane Emissions: Implementation and Validation". This removes the ambiguous "wide range" statement and better summarizes the system's goals and development.

(RC 102) In general, the manuscript is organized appropriately, but structuring within sections could be either reconfigured, renamed, or provide more detail. In general, it could be helpful to add a traditional "Methods" section, which encompasses Sections 2-3, whereas "Results" can include section 4.

(AC 102) ***Section 2. Section 3.*** The original sections 2 (System Design) and 3 (Deployment) were combined into a single "Methods" Section (2). Original section 4 was renamed from "Analysis" to "Results" (Section 3).

(RC 103) The information in L110-116 seems more like "onboard data logging and transmission".

(AC 103) ***Section 2.1.5. L162-168.*** This section was renamed as proposed and moved to the "Methods" (Section 2).

(RC 104) Section 2.5: please explain why this section is needed; L193-195 provides the motivation for this paragraph, but this motivation (and perhaps L196-203) should be described and presented at the beginning of this section.

(AC 104) ***L241-247.*** A paragraph motivating the plume simulations was added to the beginning of Section 2.4.

(RC 105) Section 4: this section is difficult to follow with the many flights that occurred at different times, with varying purposes (e.g., October/November 2022 controlled release experiments; Socorro, NM MWF, Spring 2022-Summer 2023; Orphan Well, April 2023; WWTP Summer 2023). A table outlining the flights, locations, or purpose, or even more descriptive section headers and a few sentences describing each might help. To me, the distinction between (a) controlled-release experiments and validation of the system and (b) smaller, point source emissions detection via case studies is important.

(AC 105) ***Section 3.1. Section 3.2. Table B1.*** This section was split into clearer sections and subsections to highlight the (a) controlled release validation flights (section 3.1) and (b) the smaller targeted source case studies (section 3.2). Additionally, a table with information on source type, environmental conditions, and estimated emission rates was added to the manuscript (table B1)

(RC 106) Background determination: L165-169 could be expanded upon to detail the procedure that is plotted in Figure 3. Please also describe the gradient method you use in (3b) within the text (and not just the caption) and how this is used in (3c).

(AC 106) ***L218-234.*** These paragraphs were rewritten to better describe the background estimation process and how each of the steps follows from the previous.

(RC 107) Figure 7: This can be described more effectively, in general. If I am reading this correctly, this is the uncertainty in the background estimation from just two test flights, which ties to Section 2.4 and is better explained prior to the uncertainty estimate in Section 3.3. To me, this figure and its description would be better suited just following Figure 3, where readers can directly connect the uncertainty in background CH_4 to CH_4 emissions rate uncertainties.

(AC 107) ***Figure 4.*** This figure was moved to section 2.3, as suggested, since the error confidence intervals shown directly follow from the background estimation routine. Figure 4 was updated to include three different flights in order to highlight the variability in background estimation residuals. Additionally, the average confidence interval from 28 independent flights is now overlaid on this plot for reference.

(RC 108) Do you have estimates of how results in Figure 7 compare to all flights (i.e., are the two test flights representative of typical flights)? Why is the background uncertainty not incorporated on a per-flight basis?

(AC 108) ***Figure 4. Table 1. L235-238.*** The original plot was updated to include a third representative flight as well as the average background estimation confidence interval from 28 independent flights. Comparison with the average confidence interval shows that these individual flight residuals, while different, are representative of the mean. The reviewer is correct that all background uncertainties are applied to the flux quantification error analyses on a per-flight basis, Thus, the average uncertainties due to background estimations in Table 1 are provided only as representative values for these quantities. The manuscript has been revised to better emphasize these points.

(RC 109) The word "baseline" is used interchangeably with "background" and it would be clearer in the text to just use one or the other.

(AC 109) ***Throughout.*** For consistency, the word "background" is now used throughout the manuscript as opposed to "baseline".

(RC 110) Section 3.3: this section needs to be expanded upon so that it is clear where uncertainties in each term in Equations 4-6 come from and how they contribute to the overall uncertainty in F_{tot}. For example, Table 1 seems to only describe onboard UAS sensor precision and/or accuracy, but not include other sources of uncertainty like the uncertainty in CH_4 and C_2H_6 enhancements, or uncertainties in $(\boldsymbol{u}{\cdot}\hat{n})$. All of these propagated uncertainties should be incorporated in the lower LOD of the flux estimate, correct?

(AC 110) ***Section 2.7. L342-365. Section B1.*** The reviewer is correct that all the propagated uncertainties are incorporated into the final flux uncertainty and lower limit of detection. The paper was unclear on this point and more complete details are now given in. section 2.7 of the updated manuscript.

(RC 111) L96: wind speed magnitude uncertainty is 0.35 $\mathrm{m/s}$, but Table 1 states 0.2 $\mathrm{m/s}$. Please describe how the wind speed uncertainty is derived.

(AC 111) ***L120. Section 2.7. Table 1. Section B1.*** The individual vector wind speed uncertainty is $0.2\,\mathrm{m/s}$, and the text (L120) and Table 1 are now consistent.

(RC 112) The MIRA CH_4 uncertainty is stated as 10 ppb here, but the error in background derivation is stated as 20 ppb. Is the propagation of MIRA CH_4 precision and the background uncertainty taken into account in F_{tot}, which likely adds to the overall uncertainty and lower LOD? It is unclear how the background uncertainty from each flight is incorporated into the total flux uncertainty. All of this would be very advantageous to outline in Section 3.3, and similarly, for C_2H_6.

(AC 112) ***Section 2.7. Table 1.*** The uncertainties in table 1 have been updated for clarity and the per-flight error propagation is further detailed in Section 2.7.

(RC 113) It should be stated somewhere how the MIRA is calibrated prior to each flight for CH_4 and C_2H_6.

(AC 113) ***L265-272.*** The response of the MIRA Pico is evaluated prior to each flight by releasing a small volume of processed natural gas approximately $1\,\mathrm{m}$ upwind of the gas inlet tube. We verify that pulses are recorded in both methane and ethane, and check for time coincidence and consistency in ethane/methane ratio. This procedure is now described in the paper.

(RC 114) Please state what is meant by “standard flight conditions”.

(AC 114) ***L366-369.*** We define standard flight conditions as periods of high solar insolation and steady windspeed between 2-6 $\mathrm{m/s}$. This is discussed on lines L366-369 in the context of calculating the Limit of Detection (LOD) for the system. The plume evolution is dependent on multiple environmental conditions, but a cross-sectional area of 100 m2is used in the LOD calculation.

(RC 115) The uncertainties for each of the various source emission rates calculated in section 4 should be stated alongside the measured emissions rates. Some are stated, but others are not (e.g., in 4.1.2). This is important when assessing how well the technique might fare with one type of source versus another.

(AC 115) ***Throughout. Table B1.***

(RC 116) Why is the upper uncertainty in 4.1.3 for the WWTP up to 250%, and why does the lower uncertainty differ? Because this is an AMT manuscript, it would benefit the reader to offer explanations for the calculated uncertainties in each of the various cases to assess limitations and capabilities of this technique.

(AC 116) ***Figure 12. Table B1.***

(RC 117) L289: The results of the controlled release experiments indicate that there is a “systematic” underestimation, but this does not look systematic as some estimates are higher than the metered emissions rates. Can the setup of the experiment be explained in more detail here? The type of point source is mentioned, with wind conditions, but how was the UAS flown and does this contribute to this underestimation? There are no hypotheses provided for why emissions estimates from the UAS system are lower in general – can you provide some?

(AC 117) ***Section 3.1. L393-396. L321-334.*** Section 3.1 has been updated and possible causes of the underestimation are discussed at L393-396 and L321-334.

(RC 118) “UAS” is used, but also “UAV” – please choose one or the other to be consistent.

(AC 118) ***Throughout.*** Uncrewed Aerial Vehicle (UAV) is the mobile platform, the Matrice 600 Pro (M600P) alone. Uncrewed Aerial System (UAS) is the complete instrument including mounted payload and data acquisition hardware/-software. The authors recognize that this is a subtle difference and have therefore opted to use “M600P” and “UAS” to refer to the vehicle and the complete system, respectively.

(RC 119) Figure 1, caption, and L126: What does “dual-opening” mean with respect to the sampler inlet?

(AC 119) ***L149-150. Figure 1.*** "Dual-opening" refers to the y-shaped inlet on the sampler port which decreases the chance of damage to the MIRA pump due to clogging from dust and debris. This wording has been changed in 1 and better described in L149-150.

(RC 120) L84: precision on CH_4 and C_2H_6 is not what is specified in the abstract – please confirm which is correct.

(AC 120) ***Abstract. Table 1.*** This was an error and has been updated in the updated report.

(RC 121) L86: below, the response time was ~$2\,s$ – please state one or the other for consistency.

(AC 121) ***L112.*** This was an error due to changes in length of the sampler tubing and mast inlet position. The correct phase delay between inlet and MIRA is approximately 2 seconds due to the pump flow rate and tube length.

(RC 122) L90: mole fractions are presented here in ppm or ppb, which is a mol/mol. Please correct either mixing ratio or mole fraction determinations throughout.

(AC 122) ***Throughout.*** Mole fraction and mixing ratio were used inappropriately and interchangeably in the original submission. Units of ppm and ppb are $\mathrm{mol_{compound}/mol_{air}}$ and the more correct term "mole fraction" is used throughout the updated manuscript.

(RC 123) L141: symbol for 'yaw' different here than it is in equation 1.

(AC 123) ***L170.*** Typographic error in section 2.2 has been corrected in the updated manuscript.

(RC 124) Figure 12: What do the individual colors mean? If nothing, does it make sense to have controlled release vs. municipal vs. orphan vs. WWTP all be different colors?

(AC 124) ***Figure 8. Figure 12.*** The individual colors were unimportant and misleading. Thank you for the recommendation and figures 8 and 12 have been updated.

(RC 125) Figure 6: $\chi - \chi_0$ is used to show an enhancement, whereas in L338, a delta symbol is used, in addition to both being used in Figure 10 – please choose one or the other for clarity.

(AC 125) ***Throughout.*** $(\chi - \chi_0)$ is now used throughout all images and text in the updated manuscript.

(RC 126) L325-326: How can you assess a correlation between C_2H_6 and (delete '&') CH_4 when C_2H_6 enhancements cannot be discerned from background levels due to a low signal-to-noise ratio?

(AC 126) ***Figure 10. L429-442.*** The reviewer brings up a very good point which was not discussed in the initial submission. Figure 9 has been updated to include the expectation (modeled) C_2H_6 timeseries corresponding to the measured CH_4 for two different thermogenic mixtures.

(RC 127) L365: "with the error associated for other low-emission" needs to be revised for clarity.

(AC 127) ***Section 4. L491-493.***