Review of "A New Technique for Airborne Measurements to Quantify Methane Emissions Over a Wide Range: Implementation and Validation" – Dooley et al.

General comments:

The manuscript describes the use of an Unmanned Aerial System, equipped with a fast-response methane and ethane sensor and ultrasonic anemometer, for estimating methane emissions from various point sources. The UAS payload configuration, sampling and flight, and analysis and verification techniques are outlined in the manuscript alongside estimates of methane emissions from various point sources, including controlled releases. Due to the availability of coincident ethane and methane measurements, ratios of ethane to methane enhancements above background levels are also evaluated for these various point sources. Overall, there is a need to bridge the gap between aircraft and/or satellite-based emissions estimates and ground-based, bottom-up estimates (and limitations therein for both). The novel use of UAS to provide the capability to bridge spatial and detectability gaps is important to describe for estimating emissions from smaller or distributed leaks and distinguishing biogenic versus anthropogenic sources. Overall, a great deal of testing has been done with this system and onboard payload and I would recommend publication in AMT after addressing the comments below that highlight places where clarification to better describe the technique and its uncertainties is needed, as well as potential modifications to the manuscript structure to improve readability.

Specific comments:

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?

- L294 and L376 mention a limited operational distance and flight time being potentially prohibitive, so these comments also support changing the title to be more descriptive of the technique and its capabilities.

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.

- When first describing the system and onboard payload, these sections jump around a bit, with 2.2.1 seeming a bit out of place (and perhaps fitting better under 2.1, Onboard sensors). Consider changing for clarity.
- The information in L110-116 seems more like "onboard data logging and transmission"
- 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.
- 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.

- L274-276 can be moved to the methods as this describes the flight strategy.

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).

- 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 CH4 to CH4 emissions rate uncertainties.
- 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 baseline uncertainty not incorporated on a per-flight basis?
- The word "baseline" is used interchangeably with "background" and it would be clearer in the text to just use one or the other.

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 CH4 and C2H6 enhancements, or uncertainties in (u *n). All of these propagated uncertainties should be incorporated in the lower LOD of the flux estimate, correct?

- L96: wind speed magnitude uncertainty is 0.35 m/s, but Table 1 states 0.2 m/s. Please describe how the wind speed uncertainty is derived.
- The MIRA CH4 uncertainty is stated as 10 ppb here, but the error in background derivation is stated as 20 ppb. Is the propagation of MIRA CH4 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 C2H6.
- It should be stated somewhere how the MIRA is calibrated prior to each flight for CH4 and C2H6
- Please state what is meant by "standard flight conditions"

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… 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.

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?

Technical comments:

- Abstract and throughout: "UAS" is used, but also "UAV" – please choose one or the other to be consistent
- Figure 1, caption, and L126: What does "dual-opening" mean with respect to the sampler inlet?
- L84: precision on CH4 and C2H6 is not what is specified in the abstract – please confirm which is correct.
- L86: below, the response time was ~ 2 s – please state one or the other for consistency
- 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.
- L141: symbol for 'yaw' different here than it is in Eq. 1.
- L145: Please punctuate and define Eq. 1
- L173: …as shown in 'Figure' 3
- L174: Sentence fragment starting at '+\-'
- Equation 2: please describe as Eq 2 in text and punctuate.
- Equation 3: same as above
- Equation 4: please punctuate within paragraph
- Equations 5 and 6: same as above
- L189: 'volume mixing ratio' as described in mol/mol is actually a mole fraction
- L194: 'ppmv' is described here but CH4 is described in ppm elsewhere
- Figures 11 and 12: please correct spelling of municipal
- 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?
- Figure 6: X-Xo 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.
- L283: please change ground truth-methane to 'ground-truth' methane
- L302: … was expected 'to' emit …
- L306: please delete 'and'
- L316: 400 ppb is roughly the range of plume enhancements shown in Figure 6, but not Figure 9.

- L325-326: How can you assess a correlation between C2H6 and (delete ‘&’) CH4 when C2H6 enhancements cannot be discerned from background levels due to a low signal to noise ratio?
- L365: “with the error associated for other low-emission” needs to be revised for clarity