Review of "Technical note: Isolating methane emissions from animal feeding operations in an interfering location" by McCabe et al.

December 5, 2022

## Reviewer Recommendation:

This paper is suitable for publication in ACP in structure and content but requires revisions to clarify key scientific conclusions.

## Summary:

This method paper describes two approaches to isolate CH4 emissions in the NCFR where both ONG and feedlots contribute to the total emissions. The data is obtained from airborne in-situ measurements. CH4, NH3, and C2H6, with NH3 and C2H6 used as tracers for feedlots and ONG. The two methods that are compared for one CAFO from one research flight are a subtraction method and a multivariate linear regression method. MVR has lower uncertainty and fulfills the determined selection criteria in determining a CH4 emission from the feedlot. The final emission results of both CH4 and NH3 are higher than previous studies and inventory estimates and there is spread especially in the NH3:CH4 ratio compared to previous studies. The flight pattern during F2 in Nov 2019 was not the optimal pattern determined for the remainder of the TRANS2AM campaign.

## Specific Major Comments:

1) Concern about chosen data set:
The publication by Pollack et al. (2022) utilized the exact same flight (F2) and performed similar analyses as reported in this manuscript such as the MVR and comparison of enhancement ratios. Pollack et al. (2022) also reported that the optimal flight track pattern would be racetrack or box patterns, whereas F2 used an approach that was determined to be less ideal (Lines 110-112, 113-115f of this manuscript). It is my understanding that TRANS2AM flights took place in 2021 and 2022 with such racetrack patterns flown (observed in UCAR field catalog). If this understanding is correct, I would suggest that flight data with a more optimized sampling strategy should be used as the basis for this technical note or an analysis of the errors resulting from non-ideal sampling should be included.
My concern is that the results are not transferrable to analyses where the background of mixing ratios is more carefully determined from the racetrack patterns, which will cause unquantified biases, see comment 2) below.

2) Concern about secondary sources:
You consider the background of the mixing ratios to be constant and use the transect that is 12 km downwind of the targeted CAFO to support that conclusion. This assumption, if incorrect, is substantially problematic. One way this assumption could be invalid is if you observe 2 plumes. In my understanding, the ideal flight pattern would be racetrack or boxes where you fully capture any upwind influence along a full transect that is transported towards the horizontal transect downwind, which would capture any secondary plume passing through

your region of study. It appears to me looking at Figure 4 that you may have indeed measured two distinct sources contributing peaks in NH3 (and CH4), one near -10000m from center of the F2 transect and one that is likely the targeted CAFO at +/- 5000m from the center of the transect. Additionally, C2H6 (and CH4) is enhanced from -10000 to the far end of the transect (on the right of the plot) but is not enhanced at the end of the transect towards the left (<-10000m from center). Your conclusion in Line 384 is that the emissions per head from this study are higher than previous estimates. If there was another CAFO contributing to the second, smaller peak in Figure 4, then the per head emissions are higher than the actual per head emissions. Without this more ideal sampling strategy, how can you support your conclusion that you are observing an isolated source?

3) Concern about constraint for CH4 attribution:
In Line 199 is stated that F2 is all data within the MBL during the flight – but your flight started and ended in Laramie, Wyoming (see Line 83), which is in a very different area than the observed CAFOs, separated from the NCFR by a mountain range. All flight data that is outside of a certain boundary of the NCFR should be excluded instead of only those outside a certain altitude range.

## Minor Comments:

Line 4: I suggest using present tense instead of past for "relied" unless the current method is something else. Then current method should be stated as well.

Line 20: It is unclear whether the 8.4% growth is for the US or globe or a specific region within the US. Assuming it is the US based on second half of the sentence, I suggest "grew 8.4% in the US"

Line 22f: Remove second mention of EPA reference

Line 64f: This sentence is somewhat true but also misleading and weakening your conclusion in that MVR is the optimal method. I believe you are trying to say that using more variables in the MVR for additional sources can falsify the results and is not appropriate. On the other hand, using fewer variables in the MVR and only those you are certain that are sources will not falsify the results for those known sources – additional possible sources will be treated in the extra term or background. I would suggest adding to the sentence first a quick focus on how MVR is appropriate, then what situations make MVR inappropriate.

Line 66: Missing an "a" or "the" prior to "methodology"

Line 78: Missing a space in "Greeley,CO"

Line 86: "COtextsubscript2" should be "CO2"

Line 87: Suggest "This Picarro and other Picarro models" instead of "This model and other Picarro models" to avoid confusion with atmospheric model

Figure 1: I suggest adding numbers for the middle two circles as well, as 1 and 125000 are the two extremes and it is nearly impossible to envision what capacity the intermediate circles represent.

Figure 1 caption: Are animal units determined by yourself or is there a reference? Are the equivalents with respect to CH4 or NH3 emissions or are the NH3:CH4 ratios between different animals identical? Please clarify.

Figure 1 caption: I suggest shortening the second sentence in the caption to "CAFOs are colored by animal type and sized by max animal units" as the color info is found in the legend.

Line 125: Do you mean 6.5° variability in the wind direction? Which instrument was used for wind and what is its measurement uncertainty (0.65° as currently stated seems too low in my experience)?

Line 126: "at t 4 km- 14 km downwind" should be "at 4-14 km downwind"

Line 191: Remove comma from "(2019),"

Line 193, 196: Once you use ppb, once you use ppbv. Pick one and check throughout paper and figures for consistent use of units.

Line 201: "(ratio x 100)" should be "(ratio x 100%)"

Line 228: "performed sensitivity analysis" should be "performed sensitivity analys**e**s"

Line 237: "regions for MVR analysis" should be "regions for MVR analys**e**s"

Line 238: I suggest moving Eq. 4 behind "as shown in Eq. 4." in Line 224 where it is described in the following sentence. Currently it seems to be floating without context prior to the next Section.

Line 246 and Figure 4: In Line 246 you state that you "screened out points that were <5 ppbv of NH3" but in Figure 4 I certainly seem to see cyan colors further to the left in the layer between 1500-2000 m AMSL as well as along the entire transect in the layer above 2500 m AMSL. The color bar indicates 5 ppbv is a darker shade than the cyan color.

Figure 4: How were the vertical curtain bins determined? Why does the curtain not start at 1500 m AMSL, since you stated in Line 81 the lowest flight altitude was 100 m AGL and state in the caption that the surface is at 1400 MSL?

Table 1: Currently missing the CI superscript in the table header

Line 272: Wind speed of 8.4 +/- 2.7 m/s was stated in Line 123. The wind speed has uncertainty of 32%, what do you quantitatively mean by "smaller contribution to the overall uncertainty"?

Line 284: Correct the spelling of "Sec.t"

Line 317f: Your argumentation about removal of NH3 seems invalid for two reasons: 1) Your downwind transect is 12 km downwind of the target CAFO, so with the reported wind speed the plume age is <24 minutes. NH3 has a lifetime on the order of hours and can be expected to still be present at that distance. 2) Your NH3:CH4 ratios span a wide range but are larger than reported in literature from NCFR studies (which are generally <50%), which would indicate that your NH3 background was set too low and/or CH4 background set too high.

Line 324f: I don't understand the conclusion here that it fails criteria 2. The SM ratio is the only one that compares to previous literature values that are based on observations for the NCFR reported in Table 2 in Kille et al. 2019.

Figure 5: I suggest shortening the caption. For panel a) descriptions are already included in the Figure and repetitive in the caption. For panel b), the two sentences in "Box plots of residuals for the four different methods. Residual boxplot of the different methods" are repetitive.

Lines 341-345: The wording is unclear, as line 344f states that Transect MVR is not the result with the lowest uncertainty. Then in the following sentence you state both MVR methods have lower uncertainty than SM. I suggest changing "This is, however, not the result with the lowest uncertainty" to "Transect MVR has slightly greater uncertainty than F2 MVR and both have lower uncertainty than SM because they do not require subtraction."

Line 351: "absolute errors and in quadrature" should be "absolute errors add in quadrature"

Line 370: Do you mean "of both CH4 and NH3" in this sentence?

Line 384: Missing grams in "2-12 g head-1 hr-1"

Line 386: Missing emissions in "The NH3 emissions are highly affected"

Data availability: Needs to be updated with link to dataset or explanation of how to access the data from the home page.