Responses to Reviewer comments for "Aerial Estimates of Methane and Carbon Dioxide Emission Rates Using a Mass Balance Approach in New York State" by Catena et al.

Reviewer comments are in black and line numbers are referenced to the original submission. Author responses are in bold as indented bullet points below each of the comments and line numbers are referenced to the revised clean version.

Reviewer 1 Comments

The description of the mass balance method using Gauss's theorem is overly brief and lacks critical details. I only understand the method by reading Conley et al. (2017). I recommend the authors expand the methods section to include a detailed explanation of the mass balance approach, explicitly addressing the storage term, data interpolation, and altitude binning, and explain the similarities and differences compared to the study of Conley et al. (2017), clarifying any deviations from their method. Conley assumed that the vertical mixing at the upper boundary is zero, and their schematic in Fig. 1 illustrates that the tracer mixing ratio vertical gradient at the upper boundary is close-to zero, which defends their assumption. However, this paper does not explicitly mention this assumption.

- **We have included extra language explaining the methodology starting on line 139. We also added Figure 2, which shows how the upper boundary of the plume reduces to background levels.**

The authors do not present their flight data (i.e., spatial structure of the data) at all. It is difficult to visualize data and confirm if these data accurately capture the plume. I suggest that the authors present their data in SI. In addition, a schematic in the main text would help to understand the method.

- **We added Figure 2 to help to visualize the flight path.**

The city-level emission in Table 5 might be problematic. It is unclear if the measurements adequately sample the downwind region of the entire city. It would be better to show the flight track and wind fields.

- **We ended up taking out the urban emissions due to the high uncertainty.**

Methods:

The uncertainty analysis is limited to turbulent flux variability between flight loops, ignoring other sources such as plume extrapolation below the lowest flight altitude, instrument

precision, wind measurement errors, or seasonal representativeness. The assumption of a constant methane mixing ratio, from the surface to the lowest flight level, is poorly justified, especially given the potential for strong near-surface gradients in urban or landfill settings. It is unclear how the authors quantify the PBL height and whether lower measurements are all within the PBL. It is unclear how PBL diurnal cycle alters the results. Overall, it is not clear what the sources of uncertainty are. It would be better to provide a high-level summary here rather than only pointing out to other papers.

- **We added in a new section on uncertainty starting on line 180. This discusses other possible sources of uncertainty and what was included in the uncertainty estimation. The uncertainty method used in this paper was also used in several other publications (Conley et al., 2017; Erland et al., 2022; Ravikumar et al., 2025). The convective boundary layer height was determined by the temperature inversion profile.**

The reliance on single-day measurements (or two days for some facilities) introduces potential bias, as emissions may vary significantly due to operational or meteorological changes. The paper acknowledges seasonal differences but does not assess whether one single-day measurement is representative of annual emissions. We can see that the authors mention this potential caveat at the end of the paper, but this should be highlighted very early in the paper. In addition, it is not clear whether the reported 2021 EPA GHGRP emission should be interpreted as annual average or at the time that can be directly compared with airborne measurements. It seems like all the measurements are collected during the daytime. Would the missing of nighttime measurements bias the flux quantification, or in other words, is there a diurnal cycle in the emissions?

- **We added more language that highlights the limitation of single day measurements [lines 193 and 334]. There is diurnal variation in emissions, with a peak in concentrations during the daytime in the later afternoon (Delkash et al., 2022; Gollapalli and Kota, 2018). We mention this potential bias on line 197.**

Facilities with non-detectable emissions (e.g., some WWTPs) are inadequately explained, possibly due to sampling limitations in urban areas or insufficient plume capture.

- **Refer to paragraph about WWTPs starting on line 245. We clarified a sentence on line 248 about the WWTP observations.**

Minor comments:

Table 3 rows are not lined up.

- **Fixed a bit. We edited the tables that way to group facilities with multiple measurements together.**

L143: For each loop, are you flying at the same height?

- **We added the average standard deviation of the height change within each loop on line 166.**

L190-192: This statement might be significantly weakened as you only have one day of data per season. Unless you can somehow show that the emission is consistent over the course of entire season, and your measurement is not sensitive to any sorts of synoptic scale disturbances.

- **Added some information on this starting on line 241.**

L193: How does ambient pressure affect CH4 emission?

- **Methane emissions are negatively correlated with ambient pressure, especially at landfills (Delkash et al., 2022; Xu et al., 2014).**

L194: Wind speed and direction would change the redistribution of GHG emissions. However, your method accounts for the wind to inversely derive flux from concentration.

- **Good point, we decided to take wind out of that sentence.**

L194: It is unclear what you mean by soil moisture. Soil moisture of the landfill?

- **Added "soil moisture of the landfill".**

Figure 2: It is unclear what each point represents, and what time of the year is shown. Please print the name of each site on x-axes. Are error bars assumed to be 1-sigma uncertainty?

- **The plot has been updated to include the colors and markers representing summer/winter measurements and the names of the sites. The error bars are the uncertainty levels listed in Tables 2-4.**

Figure 5: This figure does not provide much useful information. Your table is good enough to raise the point.

- **Figure 5 was taken out.**

L304: Is there a way you can showcase that the extra CO_2 emissions you observed comes from biogenic CO_2 emissions?

- **Unfortunately, we don't have the data to showcase that. Figure 2-1 from EPA's website (https://www.epa.gov/lmop/basic-information-about-landfill-gas) shows how carbon dioxide is emitted from aerobic decomposition in landfills for about a year after waste placement until conditions become anaerobic and methane is emitted thereafter. Based off of this, we're assuming that the carbon dioxide we measured was emitted from decomposition of relatively newer waste from within the landfill. However, the CO_2 emissions could also very well have been emitted from combustion sources within the landfill parameter as well. Because of this, we are not including the plot of landfill CO_2 emissions.**

Although it may be beyond the scope of this study, have you considered using a Lagrangian transport model (i.e., STILT, etc.) to translate concentration to flux, which I believe is much more robust?

- **We have considered using STILT. However, the goal and focus for this paper was to publish the observations and emission results from the Scientific Aviation flights done for this study.**

Maybe it is beyond the scope of ESSD, which focuses on presenting the data, but I am curious about the implications of the results, such as why some landfills show larger discrepancies than others. The paper mentions operational differences and waste quantities but lacks specific data or analysis to support these claims.

- **Since this is a data paper, we did not go into too much detail on why the discrepancies were so large between the landfills. We assume operational differences could be the reason for this just because there are so many options for varying results from different management practices, such as what types of covers are used, leachate management, are flares present, types of waste, etc.**
- **If you're referring to why some landfills had larger discrepancies between observations and the self-reported inventory more so than others, that could possibly be due to how they estimate the emission rate for the self-reported inventory and the assumptions or defaults used for the methane generation. Once again, there are several data inputs and default options that could lead to various results. Because of this, we included in line 323, "This inconsistent comparison with the inventory likely results from the assumptions employed by each landfill operator about the methane captured (all the landfills sampled employ methane capture technologies)." We also don't have all the details of what assumptions were used by the operators to calculate emissions, but it does appear that different operators can and do make different choices.**

The tables (2–5) are comprehensive but you could put in them SI to enhance clarity.

- **We decided to keep the tables in the main text for conveniency.**

A lot of the references are not correctly formatted.

- **Updated the references to match the correct format.**

Reviewer 2 Comments

First, it seems there is some inconsistency in the language around whether the reports are for methane only or also CO2. Clearly the authors are reporting emissions of both but in some parts of the paper they refer only to "emissions" when referring to methane specifically, as if perhaps initially they only calculated methane and then added CO2 in a later revision. I suggest going through the text with an eye toward consistency once more, and I point out a few places below in the minor comments.

- **Went through and made sure to decipher between the two. We also added a sentence on line 95 stating that this whole study was focused on methane with CO_2 reported as a co-pollutant.**

Second point is that in the introductory material there are references to the mass balance method and how one may compare to an annual average emission reported in GHGRP, but the language favors the former. The EPA is more interested in annual emissions, not snapshots, so it is not a fault or failure of the GHGRP that it only reports annual emissions -- this is the goal. Rather, it is important for the mass balance measurements to be scaled up to the annual scale. The authors correctly point to this as one possible reason for discrepancy but somehow the language makes it sound as if there is some blame to be shouldered by the annual inventory.

- **Good point! We added a few lines (Line 56, 61, and 78) in the introduction to make it less accusatory. Also note similar comment from Reviewer 1 and our responses.**

[Note: This review was written before looked at the other reviewer's notes, so here I will comment on a point of agreement with that review. As a Data paper, I do not think that this manuscript needs to include much interpretation and is satisfactory in that regard.

However, I agree that the data does not include several sources of uncertainty that should be discussed. The main issue I had already mentioned in my review was with the representativity of the emissions but that uncertainty is obviously difficult to quantify

without more information. As I have already noted in my review, this should be mentioned more up front. However, I do agree with the other review that some of the other sources could and should be estimated and included.]

- **We added a new section on line 180 including more details on how uncertainty is calculated and other possible sources of uncertainty.**

Specific:

L18: why is meteorology influencing emissions? Are the authors saying that landfill emissions are seasonal or just variable in time in general (other literature points to management practices being the source of temporal variability, i.e. what is going on on the workface -- see Scarpelli et al. recent paper, for example)?

- **Updated the abstract and moved a couple sentences. Landfill emissions are seasonal due to changes in temperature, pressure, and soil moisture (Delkash et al., 2022; Gollapalli and Kota, 2018; Maurice and Lagerkvist, 2003; Rachor et al., 2013). We suggested that there are seasonal influences due to the trend we saw in our data between the seasons. Besides High Acres landfill, all landfills with measurements in both seasons showed higher emissions in the winter than in the summer.**

L60-65 This paragraph is a bit awkward between the first sentence and then the second half talking about cities, perhaps just re-read and edit to smooth out the transition?

- **After further discussion, we decided to leave out the urban emissions.**

L75-76 Similar to above, the sentence beginning with "However" is out of place somehow.

- **Took out everything regarding the urban emissions.**

L91 again just awkward language, perhaps change to "across these source sectors and from all the sectors in the cities of Buffalo and Rochester"?

- **See above.**

L113 "is captured" should be "was captured"

- **Updated this.**

L119 - here only the specs for methane are given when CO2 is also part of this work.

- **Updated the specs to include CO_2.**

Table 1: What is the surface fraction? It is not mentioned in the caption at all.

- **Decided to take this out.**

L153, 154, I believe there is an assumption of constant air density here, since the equation is focusing on concentration and not mole fraction or mass fraction. Perhaps this should be noted. Esp. in the case of the urban studies this may be a factor where the flights are longer and cover more area.

- **Updated equation and text on line 149.**

L268 - methane should be mentioned before "emissions" at least the first time, because I think all these rates are methane, not co2.

- **Added methane before emissions.**

L283 - Here this paragraph is overstated, and I would argue the first sentence is not correct. These results do not themselves show the underestimation of an annual inventory -- perhaps they "suggest that the self-reporting… may be underestimated". As the authors discuss, the snapshot results shown here are just snapshots and do not represent an annual mean.

- **Updated the sentence.**

L284 - why does an underestimation in GHGRP mean an underestimation in the NYS inventory? Do they use the same methods or data to determine emissions, or perhaps do they use the GHGRP data directly in the NYS inventory? Also why is there a specific sentence about why the GHGRP may be higher, but that reason is different from the ones where it is lower? Errors are errors so it seems weird to attribute them differently depending on their direction? Looking at Fig. 4(a), it really does not seem to be the case that all landfills are under-reporting, even when just comparing with this snapshot data -- several landfills' reported values are higher than the mass balance estimate. Seneca Meadows is driving the mean bias up as an outlier. I realize this is discussed more in depth in L270+ for each landfill.

- **An underestimation in the GHGRP could mean an underestimation in the NYS Inventory due to how each calculates the emission estimates. While there are subtle differences between the two estimates, the methodology is similar between the two with some slight modifications to specifics such as input data, the model used to estimate emissions, the oxidation factor, etc. between the GHGRP and the NYS Inventory. We ended up taking that part out and said that the inventory values may be underestimated on line 321. We also reworded most of the paragraph.**

L287 - Sentence starting "The difference in emissions…" is confusing. What difference in emissions? If this says what I think (which is not clear), this is saying that the inventory is wrong because of seasonal variations but I would think the inventory calculation does not make actual emissions measurements, do they? They calculate based on annual average data like captured percentage and amount waste etc. So isn't it more likely that the variability is actually causing an error in the upscaling of the flight data rates reported here, when trying to determine annual values?

- **Ended up rewording most of the paragraph. Switched out "The difference in emissions…" with "This inconsistent comparison with the inventory…" on line 322.**
- **What we were trying to convey was that since there are seasonal variations in methane emissions at landfills, the methodology used to estimate methane for the inventory isn't completely comprehensive since it does not account for seasonal variation. However, you make a valid point, so we added some language to clarify it starting on line 330 onward.**

L290- this is well-stated here, so I think the previous few sentences just need to be clarified - perhaps just reworded because what they are trying to say is confusing and is almost laying the blame on the inventories for the fact that there is variability they don't account for.

- **See above.**

Perhaps there should be at least another sentence or two devoted to figure 4(b) and Figure 5. Figure 5 is not discussed at all -- these are not landfills, right? L303-305 could refer to a figure - I think this is referring to Figure 5 results, but these are combustion facilities, not landfills? A little confused, so some clarifying text would be good.

- **We decided to take out the last figure. We also added in the reference to (now) Figure 5b on line 313.**

Conclusion seems well-stated.

- **Great!**

I also note that I downloaded and checked the accompanying data files.

- **Thank you.**

References

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