Response to RC2's comments

We would like to thank Reviewer 2 for the careful reading of our manuscript and the many comments below. Below we address each comment specifically. The reviewer comments are written out in **black** text. Our responses are indicated in **blue** font, with text added to the manuscript indicated in **red** font.

The manuscript describes the field deployment of an dual comb spectroscopy (DCS) system along two simultaneously operated open measurement paths with the goal to measure CH4 emissions from cattle. This capability is tested and demonstrated in a controlled release experiment simulating cattle emissions while measuring along approximately 200m long paths upwind and downwind of the release area. The paper describes the experimental setup, hardware, the data processing, and spectral analysis which in the end produce path averaged mixing ratios. It further describes the analysis to infer flux estimations from these mixing ratios and their differences. This work provides a contribution in the ongoing challenge of measuring methane emissions from ruminants and on the open question of how open-path measurements of greenhouse gases (GHGs) are best employed in practice. I recommend publishing this work after addressing some minor and some potential major comments below.

General remarks:

I think you do not clearly and transparently present the extend of your measurement campaign (i.e. line 153 ff.). For what timespan was the DCS setup deployed? How many release experiments did you perform and on which days? The data you present in Figures 5 to 8 spans at least 3 months, yet you do not show for example the emission estimates for the day in Figure 5 which you used to show the enhancements during the release. Did you operate the open-path system during the mentioned grazing period (May to mit July) and try to measure real cattle emissions? If so, what were the additional challenges compared to your controlled release experiment? Since your stated goal is demonstrating the capability of such a system to monitor emissions, I would encourage you to state transparently how often you had high quality results. In its current state, the manuscript generates the impression that the extend of the dataset is intentionally vague and potentially data presented very selectively.

Our objective was to do a limited number of releases in the best wind conditions for the field beam geometry. This was done in part to reduce the amount of methane we would release in the atmosphere. The number of days with favorable winds was smaller than what we expected based on past meteorology data for the site, so the overall time we were at the site was longer than expected. DCS data were taken for those times, but since no cattle was present at the site only background emissions were measured and that data does not add anything to this manuscript. The release data presented in the paper is nearly all the data measured, except for one run where an error in the gas manifold prevented us from getting the proper release rate. Real cattle emissions were measured in the spring of 2023 where enhancements were measured. However, our inability to properly track animal positions prevented us from calculating flux from these measurements, and thus the data are not presented here. Tracking animal positions is an additional challenge, one we have not solved at this point, to doing real grazing measurements.

Detailed remarks:

Line 23 f.: I do not see how the provided materials show that only optical power limits the measurement. The controlled release experiment had quite accurate knowledge of the release area and the manuscripts does not provide a systematic analysis of the impact of source distribution uncertainties and transport uncertainties,which typically contribute significantly to the uncertainties of fluxes estimated from concentration measurements.

The ability to detect small changes in molecular absorption (thus gas concentration) depends on the measurement signal-to-noise ratio (SNR), where the SNR depends on the optical power that gets back from the alignment between transceiver and retroreflector. If the SNR is poor, then there is more error in measuring small changes in the molecular absorption with respect to the background. This may not be clear in the abstract for a more general audience so the phrase was removed from the abstract.

Line 61 f.: To my knowledge, McGinn et al. (2011) did not use an FTIR system.

This has been fixed in the text.

Line 71: I assume with "square-law photodetector" you mean a photodiode operated in a linear (power to current) regime. If so, calling it that way might make this more accessible to a wider audience. If not, I do not understand the point you are making here.

The reviewer is correct on the meaning of "square-law photodetector", written this way to be more accessible to a wider audience.

Line 82 f.: I appreciate you citing LMFIT but think, if you do it, the doi should be included in some form: https://dx.doi.org/10.5281/zenodo.11813

The doi was somehow dropped by the code that generated the reference. This link has been added to the text.

Line 83: You did not cite the most recent version of HITRAN (HITRAN 2020, Gordon et al. 2022). If you did not use the most recent version, you might want to mention which version you used and your reasoning behind that. You also might want to mention which information (i.e. line shape model) you used.

We have updated the specific HITRAN versions used, added a reference to HITRAN Application Programming Interface (HAPI) including the Voigt lineshape used. That being said, we use HITRAN 2008 for methane which is trusted and tested for multiple outdoor

measurements done by NIST and our collaborators.

Line 108 f.: I think the concept of "molecular time" would be worth a one line explanation somewhere in the manuscript if you need it.

'Molecular time' refers to the picosecond timescale associated with the period of molecular oscillations. This timeframe occurs too fast to be observed directly by a detector; the optical heterodyne maps these picosecond oscillations to microsecond oscillations which we detect with a fast photodetector. A clearer one-line explanation has been added to Figure 1 caption.

'Molecular time' is the timescale associated with the period of molecular oscillations which is typically picoseconds.

Line 114: What does "Turbulence data" include? Which parameters were measured and are available for analysis?

We have addressed this comment see reviewer #1 specific comment #7.

Line 138 f.: Your estimation of your measurement sensitivity of 5 nmol/mol is really interesting and I think it would be worth a bit more thorough explanation. I for my part found it challenging to follow you and am still not certain if I understood it correctly.

The purpose of this discussion was to provide an estimate based on relationships between minimum gas concentrations and signal-to-noise-ratio described in Newbury et al., 2010. However, this discussion led to much confusion and relies heavily on the relations in the cited paper, which is not clearly expressed in that paper and is not a discussion accessible to a wider audience. Additionally this estimate does not add value to the paper since the actual precision is determined experimentally by the Allan-Werle analysis. To make things clearer we have removed the estimate.

Line 190: It seems like you did not show or state your exact path lengths anywhere and not in Figure 4.
The one-way path distances were 202 m for the North beamline and 203 m for the south beamline. We have modified the caption of Fig. 4 to include this information.

Line 208: How did you calculate your mole fractions? What did you use to estimate your total airmass?
We generate the molecular lineshape using HITRAN molecular parameters and Voigt lineshape generated by HITRAN Application Programming Interface (HAPI). The mole

fraction is an independent variable in that function and the molecular gas parameters include both self and 'air' line broadening factors. Thus by fitting to this lineshape using look-up tables and LMFIT, we directly get the mole fraction of the gas. To make that more clear, we have added a reference directly to the HAPI documentation.

Line 249: While this is not wrong in any way, I feel that using the combination of arrows and the convention to give the wind direction by where the wind is blowing from generates an unnecessary potential for misunderstanding.

We understand the possible confusion with the direction of the lines, but this convention is commonly done in meteorology and the convention is described in the caption.

Line 254 f.: I do not understand the difference between your estimate of 1 +- 217 g/day using the IDM and your WindTrax estimation of +-974 g/day. I thought, that WindTrax is your IDM (Line 220).

This statement is written incorrectly and has been corrected, as described above in response to Reviewer 1 comment 17.

Figure 8 b-d: I would again encourage you to show a bit more context around the presented data. If you have the data from 13:00 to 14:00 available, your plots here would provide a better impression on how well the signal can be distinguished from the background.

Unfortunately, we do not have earlier data for that particular day to show. The background CH_4 variability can be seen in Figure 5.

Line 291 ff.: As in Line 23 f. I do not see where you presented data on the transmitted power over time and how this is limiting your performance.

This statement needs more context. The limit is seen by performing Allan-Werle analysis of data sets with different signal-to-noise ratios, whose results are not presented here. However, getting any light back to the detector is a limitation of the measurement, especially in a windy environment on a prairie in Kansas. So we have reworded the statement, getting rid of SNR and focused on how to improve through better alignment.

Line 298 ff.: Again, I do not see where you discussed the technical aspects you list in point 1).

This was discussed in the experimental section around lines 170. We added more text to that section and the conclusion to make this more clear.

Line 335: You might want to mention how contributed to drafting/writing up the manuscript.

We added the phrase ‘All authors contributed to the writing of this manuscript.’