

## Response to RC1's comments

We would like to thank Reviewer 1 for the careful reading of our manuscript and the clear and thoughtful comments. 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.

### **Review of “Using Open-Path Dual-Comb Spectroscopy to Monitor Methane Emissions from Simulated Grazing Cattle” by Weerasekara et al., submitted to Atmospheric Measurement Techniques**

#### **General Comments**

This manuscript examines the potential of a line-averaging gas sensor (based on dual comb spectroscopy, DCS) for use in calculating methane (CH<sub>4</sub>) emissions from grazing cattle. This is an important topic as grazing cattle are significant CH<sub>4</sub> emitters, and a sensitive and robust CH<sub>4</sub> sensor is needed to measure emissions in that environment. The subject is appropriate for Atmospheric Measurement Techniques.

The paper is interesting but there is a lot going on in the short manuscript. What are the objectives of the paper: a modelling study to estimate CH<sub>4</sub> enhancement levels downwind of cattle; a gas release trial to determine the enhancement; an examination of DCS sensitivity relative to the cattle signal; an examination of the accuracy of the IDM technique for calculating emissions? The manuscript touches on all of these, but generally without enough description and discussion to address each adequately. The authors likely intend this work as giving a short-overview of the potential of DCS, but it becomes an overly simplified paper that tries to do too much. My main recommendation to the authors is to revise the manuscript to follow a simpler objective(s), so that an expanded explanation and discussion can be added (for the more focused objective). Along these lines:

1. There should be a clear objectives statement in the introduction. There is such a statement deep in the manuscript at line 168: *“The main goal of this study is to determine if the DCS can detect small CH<sub>4</sub> concentration enhancements downwind from the area of interest, equivalent to those caused by a typical herd of cattle grazing on an extensive pasture.”* This is a reasonable objective, but it does not require using the DCS measurements to estimate emissions (using the IDM technique). Other verification trials have shown that IDM can accurately give emissions when provided an accurate concentration measurement. So the authors could drop the emission calculations to simplify the paper.

We moved the modified the objective to make the main goal of the paper more clear and included the statement in the introduction, specifically on lines 61 to 62 as

The main goal of this study is to determine if the dual-comb spectroscopy (DCS) combined with an IDM can precisely infer CH<sub>4</sub> fluxes from a typical herd of cattle grazing on an extensive pasture.

We kept the emission calculations because, as noted by the reviewer below, the DCS concentration measurements will be utilized with an IDM to infer trace gas fluxes. So, we believe it's important to show how the two techniques perform at estimating fluxes from a small herd as the one simulated in this study. This will make the results of the paper more appealing to a broader audience that is often more interested in the final product, namely emission estimates, than the concentration measurements.

2. Despite the above comment, the paper is potentially more interesting when the DCS measurements are used to estimate emissions (with IDM). I would be OK making this task the main objective. But the description of the tracer release verification needs improvement (comments below).

We have added more details to the emission estimates.

3. In terms of dropping material ... what is the value of the dispersion model calculations in the section "*Sensitivity and precision required for grazing measurements*"? If it is to estimate concentration enhancement levels, can these instead be determined from the tracer release results (e.g., Fig 5, 7, 8)? I prefer to see real-world measurements used for this task. Another reason to drop the modelling work is that it is inadequately described (comments below).

We believe the forward modeling section is important to demonstrate how the methane plume concentration varies with distance from the herd. These simulations were also useful to test the placement of sensors in the field. Furthermore, the scaling of the CH<sub>4</sub> precision in the section '*Sensitivity and precision required for grazing measurements*' provided us the minimum signal-to-noise ratio needed to obtain during the measurement to see the estimated small enhancements. The controlled-release results confirmed these WindTrax simulations which we believe are important results to show.

We have improved the description of this section and lines 111 to 123

Wind orthogonal components and temperature data were measured at 10 Hz using a sonic anemometer (CSAT3, Campbell Sci, Logan, UT) deployed 5 m above a grazing unit on the Rannells' Flint Hills Prairie Preserve (full site description below) near Manhattan, Kansas USA. The wind dataset selected for these simulations consisted of about 30 days in June, 2021 during the grazing season. The wind raw data files were processed using the software Eddy Pro (Licor, Lincoln, NE) and means, variances and covariances for wind velocity and sonic temperature data were calculated for 30-min intervals to be used as input variables for the Windtrax simulations. To investigate if the DCS system can resolve the expected increase in CH<sub>4</sub> concentration due to the presence of cattle above the typical CH<sub>4</sub> atmospheric background level (2000 nmol mol<sup>-1</sup>), two expected CH<sub>4</sub> emission scenarios were evaluated: 100 g head<sup>-1</sup> day<sup>-1</sup> and 300 g head<sup>-1</sup> day<sup>-1</sup>. These values were selected based on the reported IPCC Tier 1 emission values for grazing cattle in North America of 208 g head<sup>-1</sup> day<sup>-1</sup> (Eggleston et al., 2006). The simulated herd consisted of a fixed grid of point sources spaced 20 meters apart (Fig. 2). The height of gas release was set to 1 meter above the ground to mimic the height of the animal mouth and a total of 50,000 particles were released for each point source. Three beam lines were used in this simulation located at 45, 160 and 310 m from the geometric center of the herd.

## Specific Comments

4. Ln 57: “*In typical IDM applications, open-path Fourier Transform Infrared (FTIR) sensors are setup upwind and downwind for the source of interest*”. I would not refer to use of an FTIR as the “typical” IDM application -- line-averaging lasers (TDLAS) have been more commonly used. I suggest substituting “line-averaging” for “FTIR” here. The association of IDM with “FTIR” also wrongly suggests the McGinn et al. (2011) study used an FTIR (they used a laser).

We thank the reviewer for pointing out this error. We rewritten the statement in line 55 as

**In typical IDM applications, open-path line-averaged concentration sensors are placed upwind and downwind from the source of interest.**

5. Ln 63: “*The limited path distance, bulky apparatus, multi-component retroreflectors (Bai et al., 2022) make employing open-path FTIR challenging in agricultural environments.*” Assuming the authors substitute “line-averaging” for “FTIR” as suggested above, they will have to either remove this list of FTIR disadvantages, or introduce the FTIR as a preferred (?) instrument in order to justify including this list.

We have removed the sentence.

6. Ln 112: “*A forward Lagrangian stochastic model (Windtrax, Thunderbeach Sci.) was used to simulate the concentration field downwind from a hypothetical herd of 20 head of beef cattle grazing in an area of 25 ha ...*”. Give more details about the simulation: How were the point sources spatially distributed (randomly, evenly, moving)? Where were the laser lines located relative to the sources (e.g., what does 45 m away from the herd mean)? What was the height of the release? How many model particles were used? Clarify these details.

We have added more details about the simulation in Section ‘Sensitivity and precision required for grazing measurements’. See Reviewer #1 general comment 3.

7. Ln 114: “*Turbulence data for the simulations were measured ... at a grazing unit adjacent to the measurement site. The wind dataset selected ...*” Explain which wind parameters were used in the simulations. Was the wind direction allowed to vary?

We have added more details about the simulation in Section ‘Sensitivity and precision required for grazing measurements’. Yes, the wind direction was allowed to vary similarly. See Reviewer 1 comment 3 for the details on changes to the text.

8. Ln 125: “*The forward model predicted that a herd of 20 cattle grazing in an area of 25 ha would produce a CH<sub>4</sub> enhancement of 16 nmol mol<sup>-1</sup> above a 2000 nmol mol<sup>-1</sup> background for a beamline 45 m away from the ...*”. The text implies the concentration enhancements were calculated from 30 days of wind measurements (line 115). Over time the enhancement will change as the wind conditions vary. So what does the single enhancement value of 16 nmol

mol<sup>-1</sup> represent (average value, median)? What do the enhancement values in Table 1 represent? What about the variability in the enhancement?

The enhancement refers to the average and the standard deviation has been added to table 1. We have also modified the table caption to explain the statistical parameters shown in the table.

Table 1 now reads:

Table 1 - Grazing system methane emission WindTrax simulation results showing the expected average and standard deviation (SD) of CH<sub>4</sub> concentration measured by line sensors positioned downwind from a cattle herd with two CH<sub>4</sub> emission rates. The CH<sub>4</sub> background level was assumed to be constant at 2000 nmol mol<sup>-1</sup>.

	Cattle CH <sub>4</sub> emission rate (g head <sup>-1</sup> day <sup>-1</sup> )					
	100			300		
<b>Distance (m)</b>	45	160	310	45	160	310
<b>[CH<sub>4</sub>] (nmol mol<sup>-1</sup>)</b>	2005	2002	2001	2016	2006	2002
<b>SD [CH<sub>4</sub>] (nmol mol<sup>-1</sup>)</b>	12	4	2	36	12	7

9. Ln 162: This paragraph (or a variation of it) would be good in the introduction. It gives important background information and contains a clear objectives statement: “*The main goal of this study is to determine if the DCS can detect small CH<sub>4</sub> concentration enhancements downwind from the area of interest, equivalent to those caused by a typical herd of cattle grazing on an extensive pasture.*”

We have moved the goal to the introduction and modified it following the suggestion made by Reviewer #1 in the general comments.

10. Ln 201: “*The CH<sub>4</sub> tank was weighted in the beginning and end of the gas release campaigns and the mass of gas released was determined gravimetrically ...*” For what purpose? To verify the release rate given by the mass flow controller? If so, did this confirm an accurate release rate? Clarify.

We did not have a mass flow controller in this study. The manifold used in this study used a feedback loop to keep a constant gas delivery pressure during the controlled release. However, the apparatus in this study did not compensate for small temperature variabilities during the release campaigns. So, a gravimetric measurement would provide a more accurate measurement of the mass of gas released. Line 210 now reads:

We used the mass given by the scale to determine the amount of gas released in each release campaign since it provides a more direct estimate of the release rate than the one obtained using the gas manifold. Previous gas release study has successfully used scale data to verify the flow rate of mass flow controller (Coates et al., 2017).

11. Ln 220: “*The WindTrax input data consisted of ... and appropriate wind statistics.*” Describe the wind statistics used.

In lines 225 and 226: We have replaced “appropriate statistics” by adding mean, variance and covariances of wind velocity and temperature, obtained using the sonic anemometer data

12. Ln 225: “*The source area (Fig. 4) used by WindTrax to infer fluxes was set to match the 12.5 m<sup>2</sup> area of the CH<sub>4</sub> point sources.*” Was the area source at ground level?

The source was set to 0.7m above the ground. Lines 231 now reads:

and the source level was set to 0.7 m above the ground which is the same height as the manifold outlets.

13. Ln 233: “*... where  $F$  is the flux,  $\sigma_F$  is flux error,  $\sigma_{2rd}$  is downwind (background) mixing ratio error,  $\sigma_{2ru}$  is upwind mixing ratio error,  $cov(d,u)$  is the covariance of the downwind and upwind errors.*” Are the  $\sigma_2$  values variances (of what variable)? Reference a good description of Eq. (2).

A reference for Eq 2 is the work of Herman et al., 2021, where this equation is based on sensitivity analysis found in Coddington et al. 2016. The errors  $\sigma_2$  are concentration variances, summed in quadrature to give the total error. The text has been altered to fully explain the variables with proper references.

14. Ln 241: “*A CH<sub>4</sub> release at a rate of 3078 g day<sup>-1</sup> is shown in Fig. 5.*” It would be good to remind the reader of the significance of this release rate (e.g., it corresponds to X number of cattle)?

In line 247 we added:

A CH<sub>4</sub> release at a rate of 3078 g day<sup>-1</sup> equivalent to 15 head of grazing cattle is shown in Fig. 5.

15. Ln 244: “*This measurement demonstrates that the DCS system can detect small CH<sub>4</sub> enhancements equivalent to the ones caused by a small herd of cattle located at approximately 50 m from the downwind laser beamline.*” A) Can this conclusion be justified statistically? B) From Fig. 5 it appears there is no difference between  $r_d$  and  $r_u$  for some periods. The magnitude of  $(r_d - r_u)$  will depend on windspeed (i.e.,  $u^*$ ), and plotting  $(r_d - r_u)$  vs  $u^*$  would presumably show the DCS system is not detecting downwind enhancement for higher winds (when there is more dilution of the tracer). This is not a unique problem for DCS as any sensor would show similar trends with  $u^*$ .

We agree with Reviewer 1 that the statement in line 244 is not clear and the conclusion cannot be justified statistically. We have revised the statement to better describe the results in the Figure 4 and the impact of windspeed on the enhancement. Lines 251 to 255 now read:

However, the wind speed affected the DCS ability to measure these small concentration enhancements by diluting the methane plume as can be seen when the wind speed values were high during the afternoon of 04 Feb 2023 (Fig. 4a). The two-path DCS measurement was also capable of capturing the temporal dynamics of the CH<sub>4</sub> background driven by changes in atmospheric boundary layer conditions.

16. Ln 246: “*The two-DCS measurement path geometry is also capable of capturing and rejecting the temporal dynamics of the CH<sub>4</sub> background driven by changes in atmospheric boundary layer conditions.*” What does “rejecting the temporal dynamics” mean?

We agree with Reviewer 1 that the phrase ‘rejected’ is unclear and it has been removed from the text.

17. Ln 252. “*To determine if any mixing ratio biases exist between the North and South beamlines that may lead to incorrect flux values... The average CH<sub>4</sub> flux computed using WindTrax was  $\pm 974$  g day<sup>-1</sup> equivalent to approximately 5 head of cattle ...*”. This paragraph is confusing and needs more explanation. A) Remind the reader of what you are doing, e.g., using path concentrations during a period with no gas release in order to determine precision of the DCS+IDM calculation? B) What is the difference between the given uncertainty in the average CH<sub>4</sub> flux (+/- 217) and uncertainty using WindTrax (+/- 974)?

The purpose of this measurement is twofold: 1) to determine if there is any statistically significant bias between the north and south concentration measurements, and 2) use the no-release concentration result to provide minimum measurement sensitivity. Additionally, the statement is unclear and incomplete since the value of +/- 974 is the average flux error from Eq. 2. We have rewritten the statement to clarify the important results.

Lines 263 to 265 now read:

CH<sub>4</sub> dry mole fraction and WindTrax were used to compute an average CH<sub>4</sub> flux of 1.3 g day<sup>-1</sup> and standard deviation of  $\pm 217.5$  g day<sup>-1</sup>. This standard deviation value is equivalent to approximately one head of cattle, assuming an emission rate of 200 g head<sup>-1</sup> day<sup>-1</sup>.

18. Ln 258. Figure 6 caption (and Fig. 7, 8). Explain what the error-bars represent ... flux uncertainties due to DCS measurement uncertainty (and this does not include IDM uncertainty).

The error bars are only uncertainties due to the DCS measurement since the IDM uncertainties are much smaller. This has been clarified in the caption of Figure 6.

Figure 6: Time series of (a) wind friction velocity and direction, (b) dry CH<sub>4</sub> mole fraction ( $r_{\text{CH}_4}$ ), and (c) release and IDM computed CH<sub>4</sub> flux a case of no released gas. The error bars are uncertainties due to the DCS measured concentrations calculated using Eq. 2. Wind arrows point in the direction from which the wind is blowing.

19. Ln 266: “WindTrax computed average CH<sub>4</sub> flux was (4002 ± 1498) g day<sup>-1</sup>, showing a good agreement to the actual release CH<sub>4</sub> flux of 3970 g day<sup>-1</sup>.” A) What does the ± value represent here? B) The emission “recovery” fraction (4002/3970 = 1.008) is phenomenally good. This is worth some commentary and context given other IDM tracer release studies (e.g., a compilation of verification studies is given in the appendix of Harper et al., 2010: The effect of biofuel production on swine farm methane and ammonia emissions. J. Environ. Quality).

The flux value includes the average and the error computed by Equation 2. We have removed the bracket notation and written the values explicitly out in the text.

Lines 275 to 277 now read:

WindTrax computed average CH<sub>4</sub> flux was 4002 g day<sup>-1</sup> and the flux uncertainty due to DCS concentration errors (Eq. 2) ± 1498 g day<sup>-1</sup>, showing a good agreement to the actual release CH<sub>4</sub> flux of 3970 g day<sup>-1</sup>.

We thank the reviewer for bringing attention to the ‘recovery factor’ and results published in Harper et al. To address the comment we have added the following statement to the text.

As a point of comparison, Harper *et al.*, 2010 summarized the accuracy of IDM in 13 controlled release studies. They expressed the IDM accuracy by a recovered rate, given by  $(F_{\text{IDM}}/F_{\text{release}}) \times 100$ , finding an average recovery rate of 95% for all the studies. We estimated our recovery rate to be 100.8 (4002/3970



× 100) using the data shown in Fig. 7a. This is a noteworthy result indicating that the combination of DCS with IDM can produce flux estimates with high accuracy.

20. Ln 290: “*The agreement between the computed and actual CH<sub>4</sub> fluxes in this study shows that DCS can precisely measure the small concentration enhancements due to a herd of beef cattle in the field at distances up to 100 m from the source area.*” A) It would be good to add the caveat about the effect of windspeed on detectability (see comment 15 above). B) I would like the authors to comment on the base level uncertainty (~ 5 cows), e.g., what does this imply about the minimum number of cattle that could be measured?

We have addressed this concern with the text added with respect to comment 15.

21. Ln 302: “*For example, prior calibrations are often necessary when using multiple FTIR systems to perform multi-path gas concentration measurements.*” What is meant by “prior calibrations”? Cross-calibration of different instruments? This is not just an FTIR problem, but a problem with all types of concentration sensors.

Cross calibrations will be necessary when using multiple instruments. We have clarified this point in lines 313 and 314, which now read:

For example, cross calibrations are often necessary when using multiple line average sensors to perform multi-path gas concentration measurements.

22. Line 305: “*Expected CH<sub>4</sub> horizontal gradients in grazing systems are often small, as demonstrated in this study, so small instrument biases can lead to large errors when inferring fluxes.*” Good point.

Thank you for the comment.

23. Ln 310: “*The driving rationale of this work is to quantify the net CH<sub>4</sub> fluxes produced by cattle grazing system ...*” Are the authors suggesting a role for the DCS system in measuring soil fluxes (by either micrometeorological approaches or chambers)? Is this realistic given the generally small magnitude of the soil fluxes or the logistics of pairing DCS with a chamber.



No, we are not suggesting using the DCS system to measure small soil fluxes. The DCS measurements could be combined with other measurements, such as eddy covariance or soil chambers, to determine fluxes of soil and cattle components. We wrote in line 324:

Separating animal and soil contributions to the net CH<sub>4</sub> fluxes will require a combination of measuring approaches, such as chamber and micrometeorological measurements (e.g. eddy covariance measurements).