

Editor review: Using Open-Path Dual-Comb Spectroscopy to Monitor Methane Emissions from Simulated Grazing Cattle

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The paper is appropriate for AMT but requires some minor and semi-major revisions before publication. The comments from 2 anonymous referees are valid and should be addressed. In addition I would like to add my editor's technical comments and corrections below:

L16, also L219 (Eq 1) and many other instances: the usage of "mixing ratio" and "mole fraction" throughout the manuscript is not correct usage. For a mixture of A and B, eg A=CH₄ and B=CH₄-free air, the mixing ratio is defined as A:B, and the mole fraction is A/(A+B). At 2 ppm levels the difference between the two is small and the names are often interchanged, if incorrectly so. But at higher levels it is significant. Eg the mixing ratio of O₂ : air is 21:79 =0.26, the mole fraction of O₂ in air is 21/100 = 0.21. In eq 1, if X_{ch4} is the mole fraction in whole (wet) air, X/(X-X_{h2o}) is corrected for the variable water content and referred to as the dry air mole fraction. All usage of these terms should be searched, reviewed and corrected throughout the manuscript.

We thank the editor for clarifying the terminology. This discrepancy is due to poor or confusing definitions in the literature. We have replaced the term 'mixing ratio' with the term 'dry mole fraction'

L62. I believe McGinn used TDL instruments not FTIR, please also note anonymous reviewers comment on FTIR vs laser instruments. I agree to use open path as the descriptor, not FTIR.

We have corrected this mistake in Reviewer 1, Comment 4.

L76: replace "ideal" with "potentially valuable" – this paper is trying to show this to be true – "ideal" assumes that it is (and you do not need to write this paper...)

This phrase has been replaced.

L82: reference to Newville et al is not sufficient, the reader should be able to find the reference through a doi or similar.

The zotero bibliography editor dropped the doi, this has been added by hand to the references.

L96: IGMs (plural) not IGM's (possessive). Please check for other cases.

The possessives have been replaced.

Table 1: this would be easier to read with a vertical line after the 1st and 4th columns. Also if CH₄ were given as enhancements, at 2000 nmol/mol, not as mole fractions.

The columns have been added to the table. Please see the response to Reviewer 1, Comment 9.

L140 - 145 . I have trouble to follow this calculation of SNR and detection limits on several levels – I request that it be completely rewritten.

This concern was addressed in response to a comment from Reviewer 2.

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.

L141, what is meant by “normalised” here? Concentration/amount (and to what level), pathlength? What are the units?

See above comment. This originally was written as ‘optical depth’ . It has been removed.

L142 should read $(1 - \exp(-\alpha.L))$ for absorption, the 1 is missing. This equals $\sim \alpha.L$ if $\alpha.L$ is small

Optical depth is $\alpha.L$.concentration and dimensionless.

(eg $\text{cm}^2 \text{ molec}^{-1}$). $\text{cm}.$ molec cm^{-3}). What units have you used here?

SNR is calculated in measured intensity or transmission spectra, not in optical depth, which is not linearly related except for weak absorption. They are only the same for weak lines. A given noise level corresponds to a much larger increment of concentration for a line that is already strongly absorbed in the background. It is linedepth:noise that matters for detection limit, not signal:noise

So I have trouble to interpret the calculation of 5 nmol/mol uncertainty or detection limit, especially in view of the L141 comment above – I don’t know the pathlength or concentration which lead to the 0.03 “normalised” optical depths, and noise should be applied to the transmission spectrum, not optical depth. It makes a big difference if the 0.03 od is for 1 nmol/mol or for 2000 nmol/mol.

Finally, please state how you define detection limit – commonly this an amount equivalent to 3 x noise in the spectrum.

See above comment. The estimation has been removed.

L147: 12/18/2022 - please avoid this date format, it is ambiguous in an international journal. Although unambiguous in this instance, it is safer to use 18-Dec-2022 or 2022-12-18 format.

The date formatting has been fixed throughout the text.

L150: Figure 1 should be Figure 3, and 2=> 4. Please check all figure captions, numbers and cross references in the text.

All figure caption numbering has been checked in the text.

L173-174 ... that were used ... (not was used)

The tense has been fixed.

L183, 185, 189 : (PT100, FLIR etc) is not sufficient to identify the supplier. Normal usage is model number, manufacturer and location, so they can be followed up.

The manufacturer location has been added where missing.

L219 see L16 comment

This has been fixed.

243: Would be better expressed as “Data from a CH₄ release”. The figure referenced is also incorrect on this line.

The phrase has been added and the figure reference number has been checked.

L254-259. I cannot see how the last sentence in this paragraph relates to what comes before it. If the measured bias from the up-down measurement is 1 +/- 217 g /day using “the IDM”, where does the 974 g/day using Windtrax come from?

This concern has been addressed in response to Reviewer 1, Comment 17.