

Response to referee #2 for the manuscript entitled '1.645 μm Differential Absorption Lidar Measurements of Atmospheric Methane Using an Er:YAG Laser'

Review for 1.654 micron differential absorption lidar measurements for atmospheric methane using an Er:YAG laser by Dimitri Edouart, Fabien Gilbert, and Claire Cenac

This paper presents initial methane mixing ratio profiles retrieved from a DIAL instrument. This paper is well written, complete, and can be published as is.

Between line 30 and 31, A new paragraph might be added that discusses the need spatial, temporal, and resolution of the methane measurements needed by the science community to help put this work in perspective.

The following paragraph will be added to the manuscript after line 34:

'The expected lidar performance in terms of spatial and temporal resolution, as well as measurement precision, depends on the intended application. From a ground-based perspective, the lidar could contribute to the validation of satellite observations, in particular those from the MERLIN lidar mission. For this purpose, high spatial and temporal resolution is not critical. An integration time of one hour combined with a vertically integrated measurement would be sufficient to validate the satellite observations. However, it will still be necessary to achieve the 1% relative statistical error on the integrated column expected for MERLIN. For applications aimed at methane city emission inventories, a spatial resolution of 1 km associated with a temporal integration time of 1 minute and a relative statistical error on methane concentration of approximately few percent are expected (Saboya et al. 2022). The requirements for methane flux measurements using the eddy covariance method are the most demanding. Indeed, this application requires, first, spatial and temporal resolutions on the order of 100 m and 10 s, respectively, to respect turbulence scales together with a precision on the order of 1% to obtain a 1-hour mean significant geophysical flux measurement (Nemitz et al. 2018).'

References:

Saboya, E., Zazzeri, G., Graven, H., Manning, A.J., and Michel, S.E., Continuous CH_4 and $\delta^{13}\text{CH}_4$ measurements in London demonstrate under-reported natural gas leakage, *Atmos. Chem. Phys.*, 22, 3595–3613, <https://doi.org/10.5194/acp-22-3595-2022>, 2022

Nemitz, E., Mannarella, I., Ibrom, A., Aurela, M., Burba, G. G., Dengel, S., Gielen, B., Grelle, A., Heinesch, B., Herbst, M., Hörtnagl, L., Klemetsson, L., Lindroth, A., Lohila, A., McDermitt, D. K., Meier, P., Merbold, L., Nelson, D., Nicolini, G., ... Zahniser, M., Standardisation of eddy-covariance flux measurements of methane and nitrous oxide. *International Agrophysics*, 32(4), 517-549. <https://doi.org/10.1515/intag-2017-0042>, 2018

Figure 2: Because you do not have temperature measurements, you would like to choose a temperature insensitive absorption line for the methane. Can you comment on the temperature sensitivity of the absorption lines chosen?

This absorption line was selected because it is the line chosen for the MERLIN space mission and, moreover, it lies within an emission band of the Er:YAG laser. The sensitivity of the methane measurement to temperature is approximately 0.4% per kelvin (see Fig. 9) for the ON wavelength used. It may be possible to identify an absorption line that is less sensitive to temperature variations around a given temperature (for example, around 300 K). However, for vertical measurements, where the temperature can vary significantly, such reduced temperature sensitivity would no longer be maintained over the entire measurement range.

Figure 6: Using a swept window in both time and range may provide a better representation of the time series rather than the "pixelate" plot shown in figure 6 (left).

We chose this representation because it faithfully reproduces the results as they were computed by the inversion algorithm. A representation that smooths the values by interpolating the results might appear more natural, but it would be less faithful to the raw data.

Line 259: Why are you comparing the CH₄ mixing ratio measure at 2.1 km when the tower observation is 5 km away?

The observation tower is located to the west and slightly north of the lidar system. A direct line of sight to the tower was not possible from our measurement site. The lidar range gate at 2.1 km is also located approximately 5 km from the tower. The preceding range gate is slightly closer to the tower, but the corresponding measurement appears to be slightly biased. At this distance, the laser beam passes over a valley, which likely induces temperature variations that could explain this bias in the retrieved methane concentration (see lines 366–369).

Line 261-262. Why is a line fit used to determine the differential absorption coefficient. You should be able to directly use the return signal and the DIAL equation to retrieve the mixing ratio. Perhaps a few sentences should be added to describe how you are completing the retrieval would be in order.

The determination of the DAOD slope is a critical step in the DIAL inversion algorithm because it is highly sensitive to the noise present in the profiles. If the noise is assumed to be Gaussian, white, and of constant variance, the use of a linear fit - and therefore the least-squares method - corresponds to the maximum-likelihood estimator and is thus statistically optimal. Applying a fit over the DIAL range gate also has the advantage of providing an uncertainty estimate for the calculated slope, thereby enabling an assessment of the statistical error.

We added the following sentence to the manuscript at line 250:

‘The use of a linear fit based on the least-squares method corresponds to the maximum-likelihood estimator when the noise is assumed to be Gaussian, white, and of constant variance. In addition, the fit provides an estimate of the uncertainty in the calculated slope and therefore allows the statistical error associated with the methane retrieval to be assessed.’

Line 364: “Figure 10 (right) ...” Figure 10 is missing from my copy of the paper.

There was an error. The correct reference is to Figure 9. This has been corrected in the manuscript.