Response to referee #3 for the manuscript entitled '1.645 µm Differential Absorption Lidar Measurements of Atmospheric Methane Using an Er:YAG Laser'

This paper reports on an Er:YAG based methane DIAL for the retrieval of methane profiles when pointed horizontally. The paper is well structured, however, the structure causes some confusion on the logic behind selecting different resolutions and spatial averages. I recommend publication after addressing some minor comments below as well as some introductory remarks on driving requirements for the spatio-temporal resolutions and what science can be achieved with those.

- Define ICOS acronym in abstract

  The definition has been added to the abstract.

- Line 27: define TCCON

  The definition has been added

- Line 74: When introducing the fSWT, please provide attributes like switching speed, and most importantly, what the optical cross-talk is as this will drive the spectral purity of the entire transmitter. Please also discuss the switching strategy. Is the wavelength switched on every shot?

  The following sentences have been added at line 74:

  'This fast fiber switch, featuring a switching time of 100 ns, is configured in a dual-stage configuration, thereby providing a crosstalk of 35 dB between the two channels. It alternates the wavelengths on a shot-to-shot basis at the 1 kHz laser repetition rate. Consequently, DIAL measurements are repeated at a rate of 500 Hz.'

  The 35 dB crosstalk theoretically results in a spectral purity of 99.97% associated with the fSWT. Therefore, the fSWT is not the limiting factor for the measured spectral purity of 99.6%.

- Line 76: Please discuss the logic for the wavelength selections (i.e. MERLIN chooses offline wavelengths on the higher side of the online whereas HALO chose offline wavelengths lower than the online. Please also discuss the laser stabilization approach. Does the laser cavity have enough bandwidth to acquire lock on each shot, or do you operate on integers of the free spectral range to alleviate the need for high-speed cavity locking? I see this topic is touched on around lie 165, but more detail would still be good like what is the FSR, what is the metric used to ensure you are operating on cavity modes?

This line was selected because it corresponds to the line chosen for the MERLIN mission and, in addition, lies within an emission band of the Er:YAG laser. The selection of the OFF wavelength is discussed further in the manuscript (lines 160–165) and is also addressed in the following discussion regarding the bias induced by water vapor.

The seed lasers are maintained at resonance with the laser cavity using a feedback loop based on the Pound–Drever–Hall technique. The wavelength feedback is achieved by correcting the laser diode current. This control loop has a bandwidth of approximately 100 kHz. The feedback loop is activated prior to each laser shot. The wavelength drift is related to the length of the laser cavity and is therefore sensitive to ambient temperature variations. The free spectral range (FSR) is 270 MHz. The quality of the injection seeding is monitored through a beat note measurement. Further details on the laser operation can be found in the cited reference: Edouart et al. 2024.

- Line 129-131: You indicate the signals are temporally and spatially averaged to meet SNR requirements, but do not discuss what the SNR requirement is, and over what scales averaging is required to hit the SNR requirement. It's also unclear what the electrical bandwidth of the receiver is? Do you electronically band limit the receiver to match the vertical resolution after averaging/smoothing? If no, please discuss the potential sources of noise aliasing by not electronically band limiting.

  The profiles are spatially integrated in order to reach the resolution limit imposed by the laser pulse duration (see line 165). The temporal integration strategy is described in detail in Section 3.2. We rely on the Allan variance rather than on the concept of SNR. The objective is to integrate the profiles over time until the instrumental noise becomes negligible (the Allan variance then exhibits a change in slope, as explained in the manuscript). The same criterion is subsequently applied for the temporal integration of the DAOD.

  The receiver bandwidth is 40 MHz, and the acquisition is performed at 80 Msample/s (see Table 1). These values satisfy the Nyquist–Shannon criterion to prevent spectral aliasing of the noise.

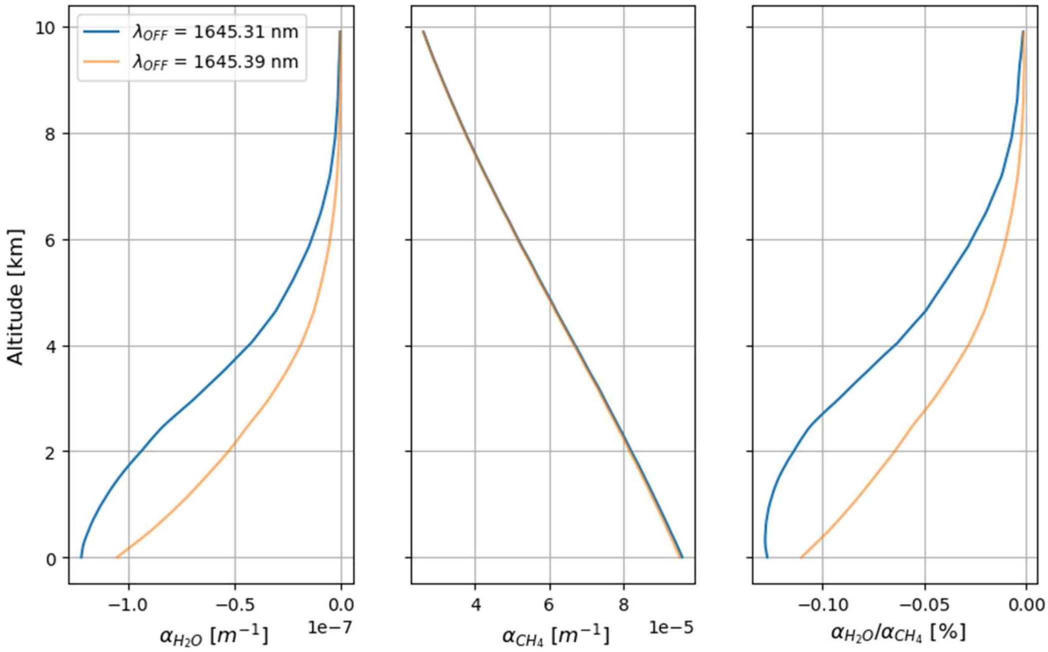
- Line 161: It may be unclear to readers why you would opt to select H_2O differentical cross section that is equal or close to zero, rather than minimizing it. Please elaborate on this topic and also include Refaat et al. 2012 where this original concept was presented. It would be good to include a vertical differential cross-section for CH_4 and H_2O to show the readers the vertical sensitivity to methane and water vapor.

  The following sentences have been added at line 161:

‘Indeed, since the residual water vapor absorption at the ON wavelength cannot be neglected without introducing a potential bias in the methane measurement, the OFF wavelength must be selected to exhibit a water vapor absorption identical to that at the ON wavelength. This ensures that the corresponding differential optical depth is zero and that the potential bias is eliminated regardless of the water vapor content (Refaat et al., 2015). The selection of the OFF wavelength must also provide a suitable differential optical depth for methane DIAL measurements.’

Refaat, T. F., Singh, U. N., Yu, J., Petros, M., Ismail, S., Kavaya, M. J., Davis, K. J., Evaluation of an airborne triple-pulsed 2 µm IPDA lidar for simultaneous and independent atmospheric water vapor and carbon dioxide measurements, Appl. Opt. 54, 1387-1398, https://doi.org/10.1364/AO.54.001387, 2015

Since the laser beam propagation is nearly horizontal (4° inclination), the elevation increase over the considered measurement range (<5 km) is less than 350 m. Nevertheless, we calculated the differential absorptions up to an altitude of 10 km for a US Standard Atmosphere and evaluated their sensitivity to altitude variations (see figure below). The relative bias induced by residual water vapor absorption remains below 0.15% and decreases with altitude.



- Section 3.1: Please elaborate on why you are only using a 25 point moving average (resulting in 47 m resolution on the DIAL retrieval). The differential cross-section over this delta R is almost negligible and therefore makes it difficult to achieve high precision.   Furthermore, increasing the range over which the signals are average increases SNR by deltaR^3/2 power. Please discuss why larger range averages are not utilized and what science is driving to higher spatial resolutions.

The 47 m resolution corresponds to the resolution limit imposed by the laser pulse duration (see line 165), but it is not the resolution used to calculate the differential optical depth. To calculate the differential optical depth and, consequently, the DAOD slope, we perform linear fits over 10 points, which reduces the spatial resolution of the XCH4 measurement to 470 m (see lines 248–250). We agree that, in order to improve the XCH4 precision, increasing the range gate length is more advantageous than increasing the temporal integration time (see Eq. 12 and line 385). To further clarify the requirements in terms of spatio-temporal resolution for the different scientific applications, the following paragraph has been added at line 35.

'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 $\delta^{13}CH_4$ 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., Klemedtsson, 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

- Lines 220-230: as with the previous comment, please discuss why you opt to significantly increase the temporal averaging at the expense of retaining higher vertical resolution? Why not utilize a range dependent delta R for the DIAL retrieval to better leverage the 3/2 power scaling for the vertical averaging. A discussion on the error propagation as to how you arrived at these average schemes would be good for the reader.

As explained previously, the strategy for temporal integration is based on the Allan variance rather than on the SNR. Increasing the vertical resolution is highly effective for improving the precision (with a dependence of ∆R^(-3/2)), and using a range gate size that increases with distance is an excellent approach to maintain the precision over increasing ranges. The present work is a demonstration of measurement capability, and the targeted spatio-temporal resolutions and precision levels can be adapted depending on the scientific applications. An analysis of the achieved precision and of the error propagation as a function of SNR is provided in Section 4.1.2. It shows that the obtained precision is sufficient to observe the standard deviation of atmospheric methane fluctuations. This analysis can be used to adjust the precision and spatio-temporal resolutions according to the targeted applications.

- Section 3.2 – it is now noted that the resolution is reduced to 470 meters, but it is framed with respect to the cross sections. Please discuss why the differentical cross-sections are computed over a 470 meter window and also discuss why the lidar signals are not smoothed to this same spatial resolution?

We use a linear fit over 10 points to determine the DAOD slope and, consequently, the differential cross section. The spatial resolution is therefore reduced to 470 m.

The determination of the DAOD slope is a critical step in the DIAL inversion algorithm, as it is highly sensitive to the noise present in the profiles. Assuming Gaussian, white noise with constant variance, the use of a linear fit, and therefore of the least-squares method, corresponds to the maximum likelihood estimator and is thus statistically optimal. Applying a linear fit over the DIAL range gate also provides an uncertainty estimate on the calculated slope, thereby enabling the evaluation of the statistical error.

We have 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 340: The laser spectral purity may be 99.6, however, the system spectral purity may be different owing to the performance of the fast optical switch. Please report what the optical cross-talk is of the switch and how that affects the spectral purity of the pulsed laser.

Please refer to the response above regarding the comments on line 74. The switch crosstalk is 35 dB, which would limit the spectral purity associated with the switch to 99.96%. Furthermore, in the present configuration, each seeder is only frequency-locked immediately before its corresponding laser shot. For example, the ON seeder is locked just before the emission of the ON pulse, while during this time the OFF seeder is free-running and may slightly drift away from the cavity resonance. Consequently, its injection seeding efficiency into the laser cavity is lower than that of the ON seeder, and its contribution to the spectral impurity is likely lower than that estimated from the switch crosstalk alone.

In this laser configuration, the spectral purity is limited by the spectral linewidth of the seeders. We use DFB laser diodes with a spectral linewidth on the order of MHz. Since the acquisition of a spectrally narrower ECDL, we have observed an improvement in the laser spectral purity.