Response to the Review Report # 2 for the manuscript, titled “δ13C carbon isotopic composition of CO2 in the atmosphere by Lidar. A preliminary study with a CDIAL system at 2-µm”

The manuscript claims the capability of measuring atmospheric carbon dioxide’s main isotopologues, 12CO2 and 13CO2, simultaneously using a coherent differential absorption lidar operating at three wavelengths within the 2-µm spectral region. While the objective of this work is to obtain the range-resolved CO2 isotopic ratio δ13C (Line 18), the conclusion clearly admits the limitation of obtaining such a ratio due to measurement precision and accuracy, as claimed (Lines 318-319). Therefore, the use of δ13C in the title is inappropriate.

From our measurements, d13C has been indeed calculated in this manuscript as claimed in the title despite a large uncertainty. Section 6 of the manuscript gives also some guidelines to measure geophysical features of d13C in the atmosphere with a lidar. We would like to keep d13C in the title as this is the topic of our work.

In fact, careful spectroscopic analysis indicates major fundamental problems in this work, other than precision and accuracy, which implies that obtaining 13CO2 measurements is impossible, and thereafter δ13C, with the described setup operating at the stated spectral range. The presented results indicate measuring 12CO2 twice using two differential settings, rather than measuring 13CO2, which leads to incorrect results interpretation by the authors. Therefore, this manuscript is rejected, and the authors should cautiously review the following issues.

The analysis and the conclusion of the reviewer is based on a misunderstanding of HITRAN2020 database. In HITRAN2020, the standard isotopic ratio 0.01118 is already included in isotopologue line intensity (https://hitran.org/docs/definitions-and-units/). Then Figure A of the reviewer proves that our spectral domain is fully relevant to measure 12CO2 and 13CO2 as claimed in the manuscript.
Please re-consider your review and conclusion with these facts.

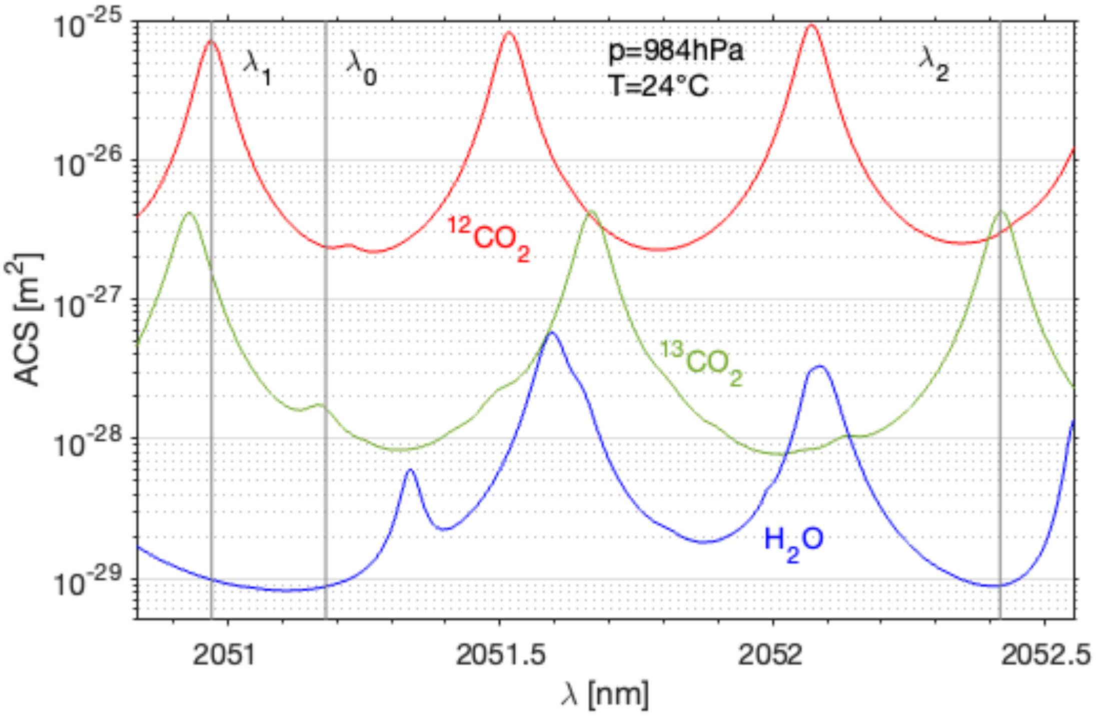
1- Insufficient Citation: A significant part of this work was previously published in the 31st International Laser Radar Conference (ILRC), held in Landshut, Germany, 2024, by some of the authors, titled “δ13C carbon isotopic composition of CO2 in the atmosphere by Lidar”. This conference paper is uncited, while it includes the same methodology, spectral analysis and instrumental setup sections presented in this manuscript. In addition, papers presenting similar work by other research teams achieving atmospheric CO2 lidar measurements were uncited, indicating insufficient literature research.

Corrected. Citations related to CO2 absorption measurements by lidar have been added in the introduction as suggested by the reviewer :

“Several lidar teams have been interested in measuring CO_2 absorption with DIAL systems since almost twenty years with precursor work using DIAL systems in the 2 µm spectral band and coherent detection (Koch et al. 2004, Gibert et al. 2006) and more recent works (Gibert et al. 2015). The spectral band of 1.6 µm has also been considered with coherent DIAL (Yu et al. 2024) or direct detection DIAL, using the advantage of low noise internally amplified photodetector (Shibata et al. 2017; Yue et al. 2022; Stroud et al. 2023) although the obtained precision was limited by the ten times lower CO_2 absorption optical depth at such wavelength. “

2- Incorrect Spectral Analysis: Figure 1 presents the absorption cross-section spectra for 12CO2, 13CO2 and H2O, with unclear scaling parameters. It is unclear how the curve marked “Total” was obtained. The 13CO2 spectral profile is not multiplied by the VPDB isotopic ratio of 0.01118, as stated in the figure caption. Proper spectral analysis is required to justify the lidar measurement presented later. Therefore, the spectral analysis of Figure 1 was reproduced to fully understand the problem.

We agree that Figure 1 and associated caption is unclear and could lead to a somewhat misunderstanding. To clarify the picture, we remove the total ACS curve and we modify the caption :
« Absorption cross-section (ACS) for 12CO2, 13CO2 and H2O for pressure 984 hPa and temperature 24°C (Voigt profile). Isotopic ratio is taken into account. The DIAL wavelengths chosen in this work are indicated. »
New Figure 1 is below:



First, Figure A presents the absorption cross-section spectra for 12CO2,13CO2 and H2O without any scaling factors. The profiles in Figure A were obtained from HITRAN 2020 database using Voigt line model and closely match the profiles presented in Figure 1 except for H2O magnitude.

Figure A corresponds to former Figure 1 except for H2O where ACS has been multiplied by a factor 25 assuming mean H2O mixing ratio is 1% and CO2 mixing ratio is 0.04%. To clarify Figure 1, standard ACS from the database (without factor 25 due to relative CO2/H2O abundance in the atmosphere) has been displayed.

Second, Figure B presents the same profiles of Figure A but with amplitude scaling using the same factors claimed in the caption of Figure 1, which include 25 for H2O and 0.01118 for 13CO2. Figure B clearly indicates that the 0.01118 factor was not included in the 13CO2 profile of Figure 1 in the submitted manuscript (nor the ILRC paper), whereas the H2O scaling factor of 25 was included.This leads to the wrong conclusion of the ability to measure 13CO2 using on-line (ON13) wavelength shown in Figure 1.

Figure B is a nonsense. Isotopic ratio is already included in HITRAN ACS (https://hitran.org/docs/definitions-and-units/).

Third, following proper analysis, figure C presents the absorption coefficient spectra for the same molecules, after including the number density and atmospheric abundance of each molecule, as well as the total absorption. From Figure C, it is evident that the total absorption is dominated by 12CO2 with some influence from H2O but without any key contribution from 13CO2. The 13CO2 absorption coefficient is about 3 orders of magnitude lower than 12CO2 and therefore does not contribute to the total absorption. This is due to lower 13CO2 absorption strength within this 2-μm region, as presented in Table 1, and lower 13CO2 abundance of 0.01118, as presented in the caption of Figure 1.
This indicates that 13CO2 is unmeasurable at all using the claimed settings, while 12CO2 is measured twice, using two on-line and single off-line wavelengths.

Figure C is also a nonsense. When isotope ratio is included in the line intensity, the absorption of the isotopologue (i.e. here 13CO2) is calculated with the abundance of the main isotopologue (i.e. here 12CO2 ~400 ppm) and not with the abundance of 13CO2 ~4 ppm.
The analysis/conclusion of the reviewer is obviously wrong when you know that isotopic ratio is included in HITRAN ACS.

Improper Interpretation for the Lidar Measurements:
The lidar results presented in Figures 8(c) and (d) show a high correlation between C12 and C13 measurements, which is unrealistic.

Both in situ and lidar measurements are displayed in Figure 8. The reviewer comment concerns 12CO2 and 13CO2 measurements provided by the in situ PICARRO gas analyzer which is a reference sensor for such measurements. I don't understand what « unrealistic » means here.

Generally, 13CO2 abundance is either uncorrelated or anticorrelated to 12CO2, due to the Suess effect. High correlation between C12 and C13 measurements confirms measuring 12CO2 twice using two different spectral settings.

No, this is definitively wrong as shown by the in situ PICARRO measurements in Figure 8 and Figure 9a. Despite a strong correlation in PICARRO C12 and C13 measurements, there is still some information in d13C represented in the Keeling plot. These d13C measurement have to be extrapolated to the origin (1/CO2→ 0) to get some information of sources and sinks. Please see Widory, D. and Javoy, M.: The carbon isotope composition of atmospheric CO2 in Paris, Earth and Planetary Science Letters, 215, 289-298, doi: 10.1016/S0012-821X(03)00397-2, 2003

C12 in Figure 8(c) represents 12CO2 measurement with high sensitivity, due to high differential absorption coefficient between ON12 and OFF, of Figure C. C13 in Figure 8(d) again represents 12CO2 measurement with lower sensitivity, due to lower differential absorption coefficient between ON13 and OFF, of Figure C, but not 13CO2 as claimed. That explains the failure to obtain the δ13C ratio by applying Equation 4 to the results of Figure 8 for the same molecule.

Wrong remark due to wrong understanding of HITRAN database.

Incorrect Dry Air Terms in Equations:
In Equations 2 and 3 the term for dry air is given by (1 – CH2O)nair, which is wrong. The correct term for dry air is nair / (1 + CH2O).

Correct. It was a typo that has been corrected in the preprint version of the paper. In the calculations we used for the dry air: nair / (1 + CH2O). C_{H2O} and C_{12} being dry air mixing ratio. Equations 2 and 3 have already been corrected in the preprint version.