Review Report for the manuscript, titled "δ^{13}C carbon isotopic composition of CO_2 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, $^{12}CO_2$ and $^{13}CO_2$, 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 CO_2 isotopic ratio δ^{13}C (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 δ^{13}C in the title is inappropriate.

In fact, careful spectroscopic analysis indicates major fundamental problems in this work, other than precision and accuracy, which implies that obtaining $^{13}CO_2$ measurements is impossible, and thereafter δ^{13}C, with the described setup operating at the stated spectral range. The presented results indicate measuring $^{12}CO_2$ twice using two differential settings, rather than measuring $^{13}CO_2$, which leads to incorrect results interpretation by the authors. Therefore, this manuscript is rejected, and the authors should cautiously review the following issues.

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 "δ^{13}C carbon isotopic composition of CO_2 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 CO_2 lidar measurements were uncited, indicating insufficient literature research.

2- Incorrect Spectral Analysis: Figure 1 presents the absorption cross-section spectra for $^{12}CO_2$, $^{13}CO_2$ and H_2O, with unclear scaling parameters. It is unclear how the curve marked "Total" was obtained. The $^{13}CO_2$ 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.

First, Figure A presents the absorption cross-section spectra for $^{12}CO_2$, $^{13}CO_2$ and H_2O 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 H_2O magnitude.

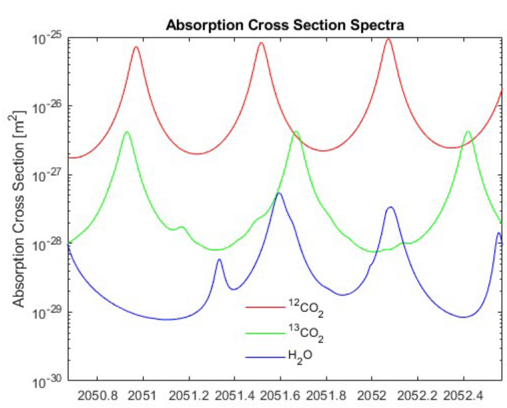

Figure A Absorption Cross Section Calculations using HITRAN 2020.

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 H_2O and 0.01118 for $^{13}CO_2$. Figure B clearly indicates that the 0.01118 factor was not included in the $^{13}CO_2$ profile of Figure 1 in the submitted manuscript (nor the ILRC paper), whereas the H_2O scaling factor of 25 was included. This leads to the wrong conclusion of the ability to measure $^{13}CO_2$ using on-line (ON13) wavelength shown in Figure 1.

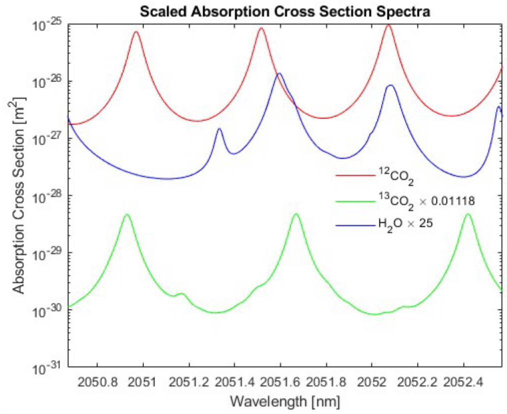

Figure B Absorption Cross Sections of Figure A scaled according to paper.

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 $^{12}CO_2$ with some influence from H_2O but without any key contribution from $^{13}CO_2$. The $^{13}CO_2$ absorption coefficient is about 3 orders of magnitude lower than $^{12}CO_2$ and therefore does not contribute to the total absorption. This is due to lower $^{13}CO_2$ absorption strength within this 2-µm region, as presented in Table 1, and lower $^{13}CO_2$ abundance of 0.01118, as presented in the caption of Figure 1.

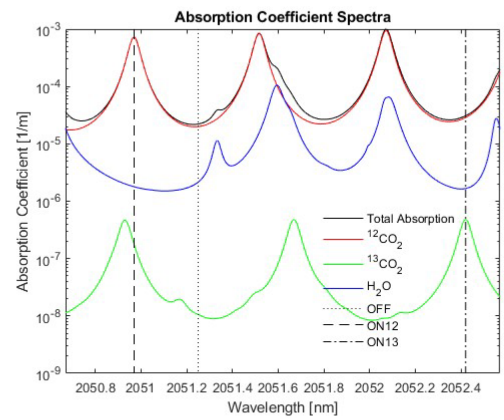

Figure C Absorption coefficient spectra using US standard model for water vapor and 727.00 and 4.72 ppm for $^{12}CO_2$ and $^{13}CO_2$, repsctively.

This indicates that $^{13}CO_2$ is unmeasurable at all using the claimed settings, while $^{12}CO_2$ is measured twice, using two on-line and single off-line wavelengths.

<u>Improper Interpretation for the Lidar Measurements:</u> The lidar results presented in Figures 8 (c) and (d) show a high correlation between C_{12} and C_{13} measurements, which is unrealistic. Generally, $^{13}CO_2$ abundance is either uncorrelated or anticorrelated to $^{12}CO_2$, due to the Suess effect. High correlation between C_{12} and C_{13} measurements confirms measuring $^{12}CO_2$ twice using two different spectral settings. C_{12} in Figure 8(c) represents $^{12}CO_2$ measurement with high sensitivity, due to high differential absorption coefficient between ON12 and OFF, of Figure C. C_{13} in Figure 8(d) again represents $^{12}CO_2$ measurement with lower sensitivity, due to lower differential absorption coefficient between ON13 and OFF, of Figure C, but not $^{13}CO_2$ 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.

<u>Incorrect Dry Air Terms in Equations:</u>

In Equations 2 and 3 the term for dry air is given by $(1 – C_{H2O})n_{air}$, which is wrong. The correct term for dry air is $n_{air} / (1 + C_{H2O})$.