

## Authors response

Dear Authors,

Thank you very much for submitting your research to AMT. Based on the recommendations of a sufficient number of independent referees, I have decided that your article can be published subject to minor revisions.

Please take into account the recommendations and questions of the very last referee report (report #1 from referee #4) and include also your responses to report #1 from referee #2 in your authors' response. Please also verify that you have taken into consideration all suggestions of report #1 of referee #1.

In addition to these requests, I would like to ask you to carefully revise the uncertainty budget (Tables 3 and 4) and the discussion of spectroscopic uncertainties and biases, which actually seem to be too optimistic. For example, I could not find any discussion of the isotopic abundance in HITRAN which differs from the composition of the V-PDB standard as a possible bias (please also give an authoritative reference for V-PDB in line 99).

Thank you for pointing this out. There is indeed a small bias in  $\delta^{13}C$  estimate due to the isotopic abundance in spectroscopic database HITRAN. We added in the text „Note that isotopologue abundance taken in the spectroscopic database gives a  $^{13}CO_2/^{12}CO_2$  isotopic ratio of 0.011235 (<https://hitran.org/docs/iso-meta/>) which differs from the VPDB's one. This creates a bias of -0.18‰ in  $\delta^{13}C$  ». Indicated L132.

Moreover, the relative uncertainty of 0.003% in the heading of Table 1 is a far too optimistic value for a line strength measurement even of the main isotopologue. In particular it is not supported by the reference referred to in the manuscript (Benner 2016), which states that the value has been derived as statistical error obtained from a least-squares fit, thus completely ignoring the dominant systematic uncertainty sources of the study. I suggest that you either use the uncertainty estimate from HITRAN or the estimate from a suitable study.

This is right. As suggested, I decided to use the uncertainties from HITRAN2020 database. Table 3 and 4 have also been modified with HITRAN2020 spectroscopic uncertainties.

With best regards  
Christof JANSSEN

## Response to referee #4

Except for the plot in Figure 1 the impact of H<sub>2</sub>O vapor lines (although small) is not really accounted for in the calculations, in Section 3 and Table 1.

Given the lidar wavelengths used in this study no specific spectroscopic study has been made on H<sub>2</sub>O vapor lines. Water vapor absorption lines absorption cross-sections have been calculated using a Voigt line shape and HITRAN2020 database (Gordon et al. 2022), indicated in L132.

There is still no explanation why 20 mbar is used for <sup>12</sup>CO<sub>2</sub> and 5 mbar is used for <sup>13</sup>CO<sub>2</sub> for the Pound-Drever-Hall (PDH) technique (line 152)

The pressure of pure <sup>12</sup>CO<sub>2</sub> and pure <sup>13</sup>CO<sub>2</sub> absorption 80-cm path gas cell are adapted to have the narrowest line shape as possible while maintaining a rough transmission around 0.5. Indicated in the text L192.

Units are required for Wavelengths in Table 2 (all other variables have units)

Corrected in Table 2.

Line 230: "We suspect that non-random and non-correlated fluctuations between  $P$  and  $P$  remains (suspected from detection noise and signal processing) and will need further investigation in the future." Either provide some justification/evidence for the "suspected from detection noise and signal processing" claim or delete those words. It is perfectly understandable that non-random and non-correlated fluctuations will be present but any attempt to attribute them to a specific cause should have some plausible explanation or justification. There are some explanations in Line 323-326 which are useful.

Right. We have deleted the words: "We suspect that non-random and non-correlated fluctuations between  $P$  and  $P$  remains (suspected from detection noise and signal processing) and will need further investigation in the future." L234

Line 234: Please specify the long settling time of the AOM and switch

The settling time of the AOM and switch is estimated as 1- $\mu$ s. Indicated in the text L237.

Line 328: "will be needed in the future to access to absolute lidar measurements". I do not understand what the authors mean by "access to absolute lidar measurements".

L336 has been modified with "...keeping in mind that further work will be needed on lidar measurements biases."

Line 377: "better space resolution" should be better spatial resolution

Corrected L386

## Response to Referee# 1

The paper describes the use of a 3-wavelength coherent differential absorption lidar to make measurements of CO<sub>2</sub> isotopic ratios in order to improve our understanding of sources and sinks. Data was collected using the lidar and compared to an in-situ co-located gas analyzer. The paper correctly identifies the importance of isotopic ratio measurements and provides a general description of DIAL theory, the spectroscopy and the experimental setup. The data is analyzed and presented in a reasonable fashion.

However, the manuscript needs some revisions before publication. The language and nomenclature are at times confusing and difficult to understand.

Corrected. The nomenclature has been significantly improved both in the text and in the figure legends and captions when it was judged as confusing.

L69 Equation (1)  $\alpha_{i,exp}$  and  $\tau_{i,exp}$  have been introduced and are for absorption and optical depth measured by the lidar,  $\alpha_i$  is now the corrected absorption coefficient from water vapor absorption

L82 Equation (4) has been added

Figure 1: wavelengths have been modified in agreement to the equations notation

Figure 3 :  $\tau_{i,exp}$  has replaced  $\tau_i$

Figure 4 : Time has replaced  $\tau$  for x-axis to avoid confusion with DAOD notation

Figure 6 : a  $\sim$  has been added to  $\Delta\sigma$  in agreement with equation notation. Legend has also been modified to be in agreement with equation notation.

The impact of water vapor interference in the absorption measurements is not adequately discussed.

L250-270. The impact of H<sub>2</sub>O interference both in the absorption and in the air density has been quantified in the new 5.3 section detailing the precision budget and in the new Table 3.

A reference or brief explanation to “Squarer and Levin estimators” would be useful.

Reference Gibert et al. 2006 has been added where the referee could find more details and related publications.

There are several places where the precision is discussed (based on statistical error) but the accuracy (or bias) is not adequately explained. However, the authors do state the biases “will need further investigation”.

A detailed review of precision and accuracy is now provided in two new sections 5.3 and 5.4 and in the new Table 3 and Table 4 with references (L250-300)

This is a difficult measurement to make with a lidar and the authors, much to their credit, conclude that these measurements may seem “to be out of range for a coherent DIAL system with reasonable range and time resolution”.

## Response to Referee #2

The influence of atmospheric parameters should be included to justify more than 1 nm wavelength separation between off-line and on-line, including error budget estimates.

The error budget is now provided in two new sections 5.3 and 5.4 and Table 3 and 4 (L250-300). In 5.4, the spectral variation of aerosol extinction and backscatter has been addressed and the induced potential biases on absorption coefficients quantified (L286-299).

Detailed technical discussions are missing in some sections.

Several technical features have been added especially when it was linked to the precision and accuracy budgets.

L 138-142 and Table 2 – specification has been added concerning the spectral stability.

L145 the role of the EOM and an additional double stage optical switch is detailed

L154-155 and Table 2- specification has been added concerning the spectral purity

Additional references for similar work should be included and addressed.

Similar work related to  $^{13}\text{CO}_2$  measurements has been presented in the introduction.

Other work known by the authors concerns water vapor isotopologue Lidar studies. A reference has been added in the introduction (L54) and is discussed in the text (L232 and L269).

### Response to reviewer #3

First, I would like to thank Reviewer #3 for his constructive review of my manuscript. I answered to the different points below and modification in the new manuscript are indicated with line number.

I have reviewed the manuscript below, which was submitted to the journal *Atmospheric Measurement Techniques*:

“Delta 13C carbon isotopic composition of CO<sub>2</sub> in the atmosphere by Lidar. A preliminary study with a CDIAL system at 2- $\mu$ m,” by authors Fabien Gibert, Dimitri Edouart, Didier Mondelain, Claire Cénac, and Camille Yver.

#### Overview:

The manuscript describes initial measurements of atmospheric 12CO<sub>2</sub> and 13CO<sub>2</sub> with a lidar and discusses the results in context of carbon source analysis. The measurements were made from the ground in a nearly horizontal path by using 3 wavelengths coherent DIAL lidar that operates in the spectral region between 2050 and 2053 nm. The lidar uses an off-line wavelength, one at the absorption peak of a 12CO<sub>2</sub> line, and the other on the absorption peak of a 13CO<sub>2</sub> line. The lidar used was a previous coherent DIAL lidar that had been updated with a higher power laser and the new capability to tune to the targeted 13CO<sub>2</sub> line. The theory of the CDIAL lidar measurements are reviewed and measurements of 12CO<sub>2</sub> and 13CO<sub>2</sub> made over a several km long atmospheric path are shown and compared to those from an in-situ sensor. The measurement precisions and accuracies are discussed in the context of those needed for determination of atmospheric CO<sub>2</sub> fluxes. The primary measurement challenge is the limited capability to measure 13CO<sub>2</sub>. This is mainly caused by its weak line absorption due to its small atmospheric concentration of ~ 4 ppm, which is roughly 1% of 12CO<sub>2</sub>. The manuscript also discusses possible approaches to improve the 13CO<sub>2</sub> measurement precision and accuracy.

#### Findings and Recommendation:

The manuscript addresses work to address an important area to better remotely sense and understand fluxes of carbon between the Earth's surface and atmosphere by measuring the isotopic ratio of atmospheric CO<sub>2</sub>. It gives a detailed review of the theory of the coherent DIAL lidar measurements. It reports important lidar measurements including those extended in time, comparisons to the in-situ sensors and evaluation of its measurement stability via Allan variance. The manuscript is well written and cites a large number of relevant references. Although found that some updates needed, I recommend accepting an updated version of this manuscript after the mandatory changes are incorporated.

#### Mandatory changes:

1. The lidar's wind speed measurements are mentioned a few times, but there is little discussion of them in the manuscript and the wind measurement results aren't shown. If the focus is on CO<sub>2</sub> measurements, then I recommend just mentioning the lidar is also capable of wind measurements and give a reference.

Yes. We added the following sentence:

L194. Doppler frequency shift is used to infer the radial wind speed at each wavelength (Gibert et al. 2015).

2. The lidar's demonstration measurements are made over a few km long nearly horizontal path. They are compared to in situ measurements, which are much more accurate, especially for 13CO<sub>2</sub>. Given the in-situ sensor's higher accuracy and the lidar's relatively short range, the potential benefits of using a lidar for these types of atmospheric measurements (ie in the

atmosphere near the in-situ sensor) is unclear. This needs to be clarified in the introduction and conclusion.

Note that in the introduction of the reviewed version of the manuscript we added some information about the interest to make  $\delta^{13}\text{C}$  measurement by a range-resolved DIAL system. Note that the lidar short range  $\sim 10$  km could be significantly improved (10 km) with an off-center  $^{13}\text{CO}_2$  R30 absorption line locking of lidar wavelength 1.

L54. “the horizontal profiling and 2-D mapping of  $\delta^{13}\text{C}$  field above the surface by Lidar will bring outstanding information on sources/sinks pattern and origin.”

L 55. “Even the vertical profiling will help to characterize the local/ long distance transport of  $\text{CO}_2$  in a similar way as for stable water vapor isotopologue Lidar measurements (Hamperl et al. 2022).”

We also added the interest for a future  $\text{CO}_2$  space lidar mission:

L58. “Ultimately, the capability of  $^{13}\text{CO}_2$  lidar measurements opens the way to a global monitoring of  $^{13}\text{CO}_2$  from space using the IPDA technique to improve global carbon inversion systems (Chen et al. 2017).”

3. The manuscript’s introduction needs to be clearer about what accuracies and resolutions are needed for this type of lidar to be useful in determining flux signatures, especially given the small change their fluxes make in  $\Delta \text{CO}_2$ .

Searched precision and resolution for  $\delta^{13}\text{C}$  DIAL have been added in the introduction from Widory and Javoy, 2003.

L48 “However, hundreds of meters from the source, these anomalies are reduced to sub 1 % variations due to efficient mixing of the atmosphere (Widory and Javoy, 2003). “

L57. “However, 1 % precision with hundreds of meters range resolution has not yet been reached for  $\text{CO}_2$  DIAL system. “

L63 “more recent works (Gibert et al. 2015) reaching a precision of 0.5% with 150-m and 15-min range and time resolution, respectively, close to what is needed for  $\delta^{13}\text{C}$  observations.”

4. Line 29 the phrase: “devastating consequences for ...” please reword using phrasing from a relevant review paper.

The sentence has been modified:

L28 “ $\text{CO}_2$  is the main anthropogenic greenhouse gas responsible for the current global warming. In 2024, its global annual average concentration in the atmosphere has reached more than 420 ppm and the global mean near surface temperature is 1.5 °C above the 1850-1900 average, with significant consequences for present and future life on planet Earth (WMO 2025, IPCC 2023).”

5. In Figure 2 the components used to measure the transmitted pulse energies or powers (ie  $P_0$ ) need to be shown. Also it needs to show the key blocks for the signal processing after the detector

$P_0$  is not the transmitted power but the backscattered power at reference non-absorbed wavelength (indicated L82). For a DIAL measurement, an accurate measurement of the transmitted power is not necessary.

Signal processing scheme has been included in Figure 2 and Figure 2 caption has been modified.

6. In Figure 1 the lines for the 3 wavelengths appear faint and need to be darker or clearer  
Corrected. Lines are black now.

7. For the lidar measurement comparisons to in situ, please give the azimuthal angle of the wind vector (if measured) relative to the lidar's azimuthal pointing angle.

Figure 8 has been modified. Horizontal wind direction at 10 m has been added and is compared to lidar line-of-sight azimuth.

8. In Table 2 the signal processing is described as spectral accumulation, while on Line 177 it references 2000 shot averaging for each wavelength. Please clarify what is meant by spectral accumulation, how it is performed and what is being averaged (the signal or its spectrum)?

Range-gate DFTs at a given distance R are averaged over a number N of shots. In coherent detection, only spectra are used and accumulated.

Table 2 has been clarified: "Real time 38.4-m range gate Discrete Fourier Transform (DFT) spectrum accumulated over 1-s (2000 shots) "

9. In Figure 3, (a)-(d) are plotted to 10 km, but the later measurements primarily go to 3 km, while Fig. 3e only to 2 km. Since the most useful range is limited to 3-4 km please replot (a)-(d) accordingly.

Figure 3 has been modified.

10. In Figure 5, the data points are very hard to see. They need to be replotted with symbols that are larger or have more contrast.

Figure 5 data points have been magnified and contrast has been improved.

11. Both plots in Figure 5 show that both tau values increase linearly with range, Please comment on what that implies for the spatial variability of 12CO<sub>2</sub> & 13CO<sub>2</sub> in the path. If there is high spatial uniformity of 12CO<sub>2</sub> and 13CO<sub>2</sub> in the path, then what does that imply about benefits of lidar vs in-situ measurements?

Although Figure 5 shows that tau values increase linearly with range, this does just mean that 12CO<sub>2</sub> and 13CO<sub>2</sub> concentrations are almost constant over the lidar LOS. But this actually does not mean anything because CO<sub>2</sub> concentration fluctuations are in the order of the ppm range which is lower than 1%. 1% variation of the slope (concentration is proportional to the slope and not tau in DIAL measurement) cannot be seen in Figure 5. Therefore, Figure 5 cannot be used to discuss the benefits of lidar vs in situ measurements. For those who are interested in, Gibert et al. (2015) range-resolved measurements answer to this question.

12. In Table 4, it is unclear whether the values are for before or after bias correction. Please clarify.

Table 4 gives just an assessment of potential systematic errors on 12CO<sub>2</sub> (C12) and 13CO<sub>2</sub> (C13) mixing ratios. The only bias that is corrected is the one due to Eq. (9) (indicated at L295). Table 4 caption has been clarified.

13. It seems other possible sources of systematic measurement error could be caused by the lidar hardware. Possibilities might include a wavelength dependent response of the optics or detector that measures the P<sub>0</sub> values for each wavelength or slight changes in the beam pattern from the transmitter for the 3 wavelengths. Please briefly address the possibilities of

the lidar hardware being a source of systematic error.

Concerning P0, please see the answer of point 5. There was some misunderstanding here.

Concerning the source of systematic error this has been added L323:

“These biases are likely to be due to the long settling time of the emitted pulse shaped by the AOM (Fig. 2). As seen in Figure 3a, a part of the emitted pulse (and the remaining power after it) is reflected by the optics after the polarizer (Fig. 2) which may create a different bias on atmospheric backscattered power at each wavelength.”

14. In Figure 7 the better known (in situ) measurements are plotted on the y-axis with the lidar values on the x-axis. Since here the primary question here is about the lidar measurements it seems their values should be plotted on the y-axis. Also, if possible, please plot the measurements made on the different days with different colors or symbols.

Figure 7 has been modified with lidar measurements in the y-axis and in situ in the x-axis. Bi-square regression has been made again.

I tried to put the measurements of different days with different markers and colors but I gave up because there was a significant overlap between points that prevents the reader to distinguish efficiently the different points of the different days. Then, I decided to keep the same markers and colors for the whole dataset.

15. In Figure 9 the dots for the lidar measurements are difficult to see. Please replot in a darker color or larger symbol size.

Figure 9 has been modified and the size of the markers has been magnified.

#### Recommended changes

1. The manuscript doesn't clearly address the targeted application scenario of this type of lidar. For example, it solely intended for research, or for deployment for single units or a network? Is the targeted use primarily intended for horizontal, slant or vertical path measurements?

The different targets of such instrumental development have been clarified (see mandatory point 1.) with additional references.

2. Line 46, specify what type of gas emissions

Natural gas emission has been specified L47

3. Line 49 “interesting ways”. Do the authors mean simpler or more affordable?

I meant “simpler”. Corrected L51

4. Line 53 “outstanding information.” Do the authors mean new information?

Yes “new information”. Corrected L55

5. Line 62 “confronted” Do the authors mean compared to?

Yes “compared to”, Corrected L74

6. Line 78, 1st two equations, the equal signs did not get type set correctly

Corrected. See new equation (3) L92. These equations have been significantly modified after reviewer 1 comments.

7. Line 113. “the ACS are larger by one order of magnitude” Do the authors mean compared to those in the 1.6 um band?

No. The  $^{13}\text{CO}_2$  ACS are larger by order of magnitude than expected by the isotope ratio (two orders of magnitude). The sentence has been modified by: “ACS are larger by one order of

magnitude than expected by the isotope ratio, that mitigates, for one part, their non-optimal DAOD. “ L125

8. On page 5 please comment on why the wavelengths are switched at 60 Hz.

As written L160, “The wavelength switch is fixed at 60 Hz (switch every 100 pulses at fixed wavelength) as a compromise to limit switch disturbance on the measurements (switch cross-talk is limited to 30 dB) and keep identical atmospheric aerosol backscatter signal for the three wavelengths.”

Experiments have been achieved where the optical switch was used in different configurations and we noted an impact (bias on the slope) on DIAL measurement when the switch was used at high frequency (On-Off wavelength switch at 2 kHz). To do that we operate the system in (Off-Off) configuration to see if we could measure a 0 optical depth. This issue disappears when we operated the switch at lower frequency, i.e. 60 Hz.

At such frequency, we checked that the atmospheric backscatter remained the same.

9. On page 5 the diameter of the lens used for the common transmit receive path is given as 50 mm. Diffraction limited lens are commercially available with much larger diameters and it seems that using one would improve the lidar’s CNRs and the measurement precisions.

Please comment on why this lens diameter was chosen.

Numerical simulations were made to calculate heterodyne efficiency with different lens diameter 50 – 100 mm. 100 mm diameter lens only improves the heterodyne efficiency for a distance longer than 2 km. In addition, coherent detection with larger aperture is more sensitive to the turbulent conditions close to the surface ( $C_n^2$ ) (large  $C_n^2$  entails a decrease of coherent radius and then increases the number of speckles for a given aperture which decreases the heterodyne efficiency). In our conditions, i.e. horizontal measurement over a distance of 2 km, 50 mm diameter lens offers the best results. This was confirmed by experimental tests.

We added “ (optimal heterodyne efficiency for horizontal measurement close to the surface over a distance of 2 km)” L170