

We thank the referee #1 for the detailed and helpful comments and would like to give our responses to each of these comments individually:

Comment: The title as well as the abstract is misleading, because in the paper the authors do not present any data from their BARCA airborne campaign. The manuscript has mainly a technical nature and should be introduced as such. Revising the title and the abstract is recommended.

Reply: The authors agree that this paper focuses on the techniques of achieving high-accuracy aircraft measurements using a commercially available analyzer, not on explanations of BARCA data. We have removed “BARCA” from the title and added “using the cavity ring-down spectroscopy (CRDS) technique”. However, the abstract just mentions BARCA as the context of these measurements, we therefore decided to leave it as is.

Comment: Unfortunately, the paper has a major shortcoming in the way the authors have discussed the pressure broadening effect. This topic needs to stand on its own. For example, the line profile definition is critical to the pressure broadening effect discussion, but one cannot understand that from reading this paper alone. The Fig 4 shown in the manuscript only represent simple simulations of the normalized Galatry profile in function of changing the  $y$  parameter by unrealistically large amounts. Except the illustrative character, this has rather little in common with real-life conditions. Moreover, the authors do not communicate the pressure values used in their experiments, although this parameter is crucial for the topic and would allow the reader to do some back-of-the-envelope calculations. This could be performed relatively easy since all the pressure broadening coefficients for  $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{H}_2\text{O}$  are known at  $1.6 \mu\text{m}$ . Actually, the authors should use these coefficient values to confine their fit, more so, because as stated “the uncertainty ... is mainly caused by the noise in the  $y$  parameter due to imperfect mathematical fit”.

Reply: To our knowledge, the parameters for Lorentzian line broadening and Galatry line narrowing are not well known for arbitrary mixtures of Ar,  $\text{N}_2$ , and  $\text{O}_2$  on this particular ro-vibrational absorption line of carbon dioxide. Furthermore, this information would not be of great utility, given that the concentrations of Ar,  $\text{N}_2$ , and  $\text{O}_2$  are not known for the filling tanks used during the campaign. We have instead opted for the empirical method of fitting the data with a variable  $y$  parameter. Rather than allow  $y$ - and  $z$ - to vary independently, we allowed them to vary in tandem with a fixed proportionality factor between them (Fitting the  $y$ - and  $z$ -parameters simultaneously is possible, but the uncertainty in these parameters from this fit is larger than is required for accurate post-correction of the data). This  $y$ -parameter was then used as a proxy for the variation of the Ar/ $\text{N}_2$ / $\text{O}_2$  mixture, and can be used to correct all synthetic gas standards. We have added explanations in the revised version (see section 6.1).

Comment: Also the whole procedure of standard gas correction for pressure broadening effect is unclear, because of the following: The CRDS analyzer uses the peak height obtained from the fit of the spectral line to determine the mixing ratio of total  $\text{CO}_2$ , but since its exact value is not known (see isotope composition issues), one ends up with two unknown parameters in the fit result: one is the real concentration or the absolute height and the other is the pressure broadening coefficient. How exactly did the authors solve this?

Reply: We would like to clarify that the CRDS analyzer only detects the absorption of  $^{12}\text{C}^{16}\text{O}_2$ , not total  $\text{CO}_2$ , and this means the peak height is directly determined by the mixing ratio of  $^{12}\text{C}^{16}\text{O}_2$ . The reported mixing ratio of total  $\text{CO}_2$  is computed using a calibration curve (correlation between mixing ratios and peak heights) obtained from laboratory measurements of ambient air standards. The corrections for synthetic air standards are performed to deal with two independent issues: one is the isotope composition ( $^{13}\text{C}^{16}\text{O}_2$  and  $^{12}\text{C}^{16}\text{O}^{18}\text{O}$ ) issue that does not directly influence the peak height. Using the estimated range for the composition of these two  $\text{CO}_2$  isotopes in the calibration gases, we could compute the concentration of total  $\text{CO}_2$  with an uncertainty of 0.06 ppm. The other issue is the pressure-broadening effect due to variations in the background gas fractions ( $\text{N}_2$ ,  $\text{O}_2$  and Ar) that directly influence the peak height. To characterize the correction for this effect, we have made a series of experiments in the laboratory. We measured three synthetic air standards (known total  $\text{CO}_2$  mixing ratios), and after correcting for the isotope effect, we attribute the difference between the isotope-corrected readings of the CRDS and the mixing ratio values of the calibration gases to the influences of pressure broadening on peak heights (the calibration curve between the mixing ratios and peak heights derived from measuring ambient air standards is used). The influences could be represented by  $d[\text{peak}]/dy$  as one value from measuring all the three synthetic air standards when the Lorentzian broadening coefficient  $y$  varies in a small range. Use of this experimental value enables us to correct the pressure-broadening effect for measuring the synthetic air standards during the BARCA campaign. We have added texts and a table to explain these in the revised version (see section 6).

Comment: On the other hand, the section about isotope correction is presented in a too detailed manner, although, this issue is well known in the laser spectroscopy community and not only, see e.g., Bowling et al., *Agricultural and Forest Meteorology* 118 (2003) 1–19 and Griffis et al., *Agricultural and Forest Meteorology* 135 (2005) 44–60; It is also needless to make a lengthy discussion on this issue, as the absolute values regarding the isotope composition of the  $\text{CO}_2$  are not known in the present study and only a good guess about their expected value is used.

Reply: The impact of  $\text{CO}_2$  carbon isotopes ( $^{13}\text{CO}_2$  and  $^{12}\text{C}^{16}\text{O}^{18}\text{O}$ ) on total  $\text{CO}_2$  concentrations is well known; however, use of synthetic air standards for calibrating an analyzer based on the laser spectroscopy technique is rather new to the  $\text{CO}_2$  concentration measurement community. The suggested papers mention isotope effects on spectroscopic measurements of  $\text{CO}_2$ , but do not discuss fully the implications for high precision calibrations needed to obtain inter laboratory comparability. An estimated range for the isotope values enables us to estimate the corrections within an acceptable uncertainty.

Comment: I would suggest that prior to any important flight campaign the standard gases that are going to be employed for calibration should be analyzed and characterized by mass spectrometry for their composition, including not only the species in interest, but also the background gas fractions ( $\text{N}_2$ ,  $\text{O}_2$ ) and, when necessary, then also for isotopic species. The logistic and analytical expenses are negligible in comparison to a flight campaign organized abroad. This will assure the highest accuracy of the data, make any inter-comparison straightforward and avoid comments as above.

Reply: The isotope and pressure-broadening effects are emerging problems for high-precision measurements of CO<sub>2</sub>. Prior to the CRDS technique, synthetic air standards are widely used and are sufficient for calibrating analyzers for accurate CO<sub>2</sub> concentration measurements using the NDIR technique. We also recommend not using synthetic air standards to calibrate a CRDS analyzer. On the other hand, ambient air standards could be used without the need to characterize the compositions of N<sub>2</sub>, O<sub>2</sub> and Ar because these compositions do not vary large enough to have a significant pressure-broadening effect on atmospheric CO<sub>2</sub> concentration measurements (see section 3 in the revised version). As far as the isotope effect is concerned, it depends on the origin of the CO<sub>2</sub> in the standard gases. If it is from ambient air, the variations in carbon isotope will have an insignificant impact on the measurement of total CO<sub>2</sub> concentration. However, if it is from burning fossil fuel, the isotopes need to be characterized. We have added a recommendation to use ambient air calibration gases in the summary.

Comment: P3130 Line 1: "... highly accurately" is not a common term, I would recommend the use "of high accuracy" instead.

Reply: We accept the recommendation.

Comment: P3130 Line 1 and 2: "... without the need to dry the sample air or to employ inflight calibrations. It was necessary to perform in-flight calibrations and careful air drying techniques ...". The logical connection between the two sentences is missing and the two statements are actually in contradiction. Please rephrase it.

Reply: We have added "previous" before "all in situ" to avoid the confusion.

Comment: P3130 Line 20: "CRDS is a technique which introduces a gas sample into a high-finesse optical cavity ...". There are many techniques which introduce the gas sample into a high-finesse optical cavity. I recommend rewriting the sentence as follows: "In the CRDS technique the gas sample is analyzed in a high-finesse optical cavity ...".

Reply: We accept the recommendation.

Comment: P3131 Line 1: "light at a specific wavelength". The value is known, so why not use it? Near-infrared region at 1.6 μm

Reply: Yes, the wavelengths are known, and different for the three species of CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O. We have added them in the revised version.

Comment: P3131 Line 8: write "are" instead "is determined".

Reply: We accept the correction.

P3131 Line 19: "solid state memory" I would rather call it as solid state drive (SSD) or RAM drive. These devices are of course based on solid state memory, but in the computer you install the drive which has to emulate a hard disk drive interface.

Reply: Yes, we have changed it to “solid-state drive”

Comment: P3132 Line 8: replace “from an ambient air tank” with “from a pressurized ambient air tank”

Reply: We have provided more detailed description about the water experiment, including the tank information.

Comment: P3133 - P3134: In my opinion, the accuracy of the water dilution experiment significantly suffered by the fact that the humidity content of the gas entering into the CRDS analyzer is not known in absolute values. The authors also do not specify the humidifier type and its accuracy. The water signal of the CRDS should be calibrated for exact dilution determination, which is rather complicated since one has the significant pressure self-broadening effect also for the water. Nevertheless, the authors should inform the reader about the quantitative values of the dilution effect and the pressure broadening effect due to the humidity. This would be very useful in judging the magnitude of such corrections and eventually comparing them with other existing analyzers.

Reply: In the water dilution experiment, the water vapor readings from the humidifier were not used for correcting the water vapor dilution effect. Instead, the highly precise readings of water vapor provided by the CRDS analyzer were used. We have added several paragraphs in section 3 in the revised version. To summarize, if considered on an individual basis, each of these uncertainties (dilution, line-broadening, water vapor nonlinearity, and absolute water vapor calibration) could result in a substantial overall uncertainty in the dry gas mixing ratios for carbon dioxide and methane. However, we can take advantage of the fact that the CRDS analyzer provides highly precise readings of all three gases. By performing careful experiments, we can derive empirical expressions that directly relate the actual dry gas mixing ratios for carbon dioxide and methane to the measured water vapor readings. All the effects discussed are then combined into a single expression which does not rely on careful understanding of each effect considered individually. The detailed description of the water dilution experiment and the quantitative values of the dilution and pressure-broadening effects are given in the revised version (see section 3).

Comment: A linear correlation between two instruments obtained at a given time is not a sufficient prove for the transferability of the water correction functions. Such statements may lead to confusions and probably to false assumptions in follow-up water correction/calibrations attempts. Referencing an instrument to another without any real calibration is giving cause for concern.

Reply: We have changed the wording in the revised version. The purpose of correlating water measurements is to check the transferability of water correction functions. We do not claim the accuracy of water vapor concentration measurements here.

Comment: P3133 Line 19: “subsequent text” should not read as “subsequent test”?

Reply: We accept the correction.

Comment: P3134 Line 23: “The water vapour measurements of the two analyzers are linearly correlated”. This clearly indicates that there should be something more than just dilution and pressure broadening effect, otherwise one would expect that the analyzers measuring the same gas at the same conditions should not only linearly correlate, but rather give the same result. Therefore the word “water calibration” should be replaced by correction, correlation or scaling factor in this section.

Reply: We have changed “water calibration” by “water correction” and clarify that this effort is to make the two analyzers report same water concentrations for the same input, although the reported values may not be accurate.

Comment: P3134 Line 5: replace “single stable H<sub>2</sub>O spectroscopic feature” with “single H<sub>2</sub>O absorption line”

Reply: We accept the recommendation.

P3134 Line 22: “we applied temperature and pressure variations that typically occurred during flight.” The authors highlight the main characteristics of the analyzer which are important for measurements on board aircraft. However, there is still another factor which would be interesting to investigate. This is the “mechanical stress due to shock and vibration” as stated in the introduction. Any thoughts here?

Reply: we did observe a slight increase in the variations of the cavity pressure for several short periods during BARCA flight. We believe that an imperfect control of the sampling gas flow has been caused by mechanical vibrations, which leads to the slightly large variations in the cavity pressure. Two modifications have been made: one is to change the orientation of solenoid valves in the flow control unit from vertical to horizontal, because larger vertical vibrations are expected than horizontal vibrations during flight; the other is to modify the flow control frequency so that it is less influenced by the vibrations during flight. After these two modifications, the slightly larger variations in the cavity pressure for short periods have diminished to an insignificant level during later flights. We have added these in the revised version (see section 4).

Comment: P3138 Line18: “the CRDS analyzer might have drifted since the calibrations were made 4 months before the campaign.” This assumption disagrees with the statement on P3134 Line 12, where the authors expect a drift of 0.5 ppm over two years, which is clearly much less than the 0.2 ppm in four months.

Reply: The drift of 0.5 ppm over two years was observed in one CRDS analyzer of the same type. However, we do not expect every analyzer behaves exactly the same. The number of 0.5 ppm was only used to do some back-of-the-envelope calculations.

Comment: P3138 Line 2: “the Lorentzian broadening parameter was measured as part of the field campaign, and that data, along with a laboratory investigation of the dependence of the peak height of the absorption lines on Lorentzian broadening, were used to correct the calibration

tank data reported by the CRDS analyzer.” I cannot see any link between the “Lorentzian broadening parameter measured during the field campaign and the laboratory investigation of the dependence of the peak height of the absorption lines on Lorentzian broadening”. The effective pressure broadening is a sum of individual pressure broadening coefficients weighted by the partial pressure of each gas component. Therefore, the pressure broadening of humid ambient air will be different from the pressure broadening caused by a dry and synthetic air.

Reply: The link was explained in the texts that followed. As explained above, a series of laboratory experiments were carried out to derive the value of  $d[\text{peak}]/dy$ . This value was then used to correct the pressure-broadening effect. We have added explanations in the revised version and a new table (Table 2).

Comment: P3142 Line 12: replace “The CRDS analyzer performed highly stably under” with “The CRDS analyzer demonstrated high stability under”

Reply: We have modified this in the revised version.

Comment: P3142 Line 22: replace “The CRDS analyzer performed highly stably without” with “Highly stable operation of the CRDS analyzer was found even without”.

Reply: We have modified this in the revised version.