

We thank the referee #2 for the detailed and helpful comments and would like to give our responses to each of these comments individually:

Comment: The authors repeatedly refer to the 'measurement of CO₂' in a casual way, rather than to the 'measurement of CO₂ mixing ratios' or 'CO₂ concentrations'. Even though the context usually provides enough information to avoid confusion the measured quantity should always be stated in my opinion.

Reply: We have paid more attention to this issue in the revised version. Measurements of CO₂ mixing ratios' or 'CO₂ concentrations' are used in the revised version to avoid possible confusion.

Comment: Generally more attention to detail would have been desirable. Should mixing ratios not be given in ppmv (by volume) rather than ppm?

Reply: Mixing ratios of CO₂ should be given in μmolmol^{-1} (ppm), not ppmv. Because the CRDS analyzer was calibrated against air standards with known mixing ratios in dry mole fractions (ppm) that are traceable to the WMO scale, therefore, the reported CO₂ mixing ratios in this paper should be given in ppm. Please note that CO₂ is a non-ideal gas, the mole fractions of CO₂ in ppm will not be the same as volume mixing ratios of ppmv.

Comment: The title of the paper does not appear to summarize the content of the paper in an appropriate way, since mixing ratios of CO₂ and CH₄, as they were acquired during the BARCA campaign are not discussed. This, however, is the expectation generated using the present title. I recommend that the authors consider a new title.

Reply: we have changed the title, as also suggested by the first referee.

Comment: Section 1 P3129, L1: A reference(s) should be given after "...since the 1930s."

Reply: A reference has been given in the revised version.

Comment: Section 2 P3130, L22-24: The sentence should be rephrased. "... thus providing parts-per-billion concentrations ..." -> "... thus typically providing parts-per-billion mixing ratio ..." "... which is unaffected by the initial strength of the light source." -> "... which are in good approximation independent of the intensity fluctuations of the excitation light source."

Reply: We accept the recommendation and have made changes in the revised version.

Comment: P3130/31: Details of the ring-down setup have been published previously (Crosson, 2008). In my opinion the main technical features and specifications of the setup, that are of interest to the reader in the context of the present study, are inadequately summarized here.

Reply: More descriptions have been added in the revised version.

Comment: Also no spectroscopic information is given in section 2. Which crosssections were used for gas concentration retrieval - (refs?). How are cross-sections affected by the temperature - was that taken into account? Only in section 6 it is stated that, after fitting a profile to a line feature, the peak height was used to determine the mixing ratio of CO₂ in air. Why were concentrations not obtained from integrated spectra?

Reply: We do not use absorption cross sections to retrieve the gas concentrations. Instead, we calibrate the analyzer with air standards. The line-broadening effects do not affect the total area of the absorption line, but they do affect the peak height. The CRDS analyzer uses the absorption peak heights to derive concentrations of the three species; the peak height is used because of the higher noise present in the peak area measurement, due to systematic noise in the absorption baseline as well as noise in the measurement of the wavelength. Because the peak height is not constant for a given mixing ratio, variability in the line-broadening thus leads to systematic errors in the reported mixing ratios. For the carbon dioxide and methane lines employed in the CRDS analyzer, the line-broadening effects, if not corrected, would lead to systematic errors of about 40% of the dilution effects. We have added these in the revised version.

Comment: The choice of the term "partially reflecting mirror" (in two places) is somewhat unlucky considering the fact that the cavity mirrors' reflectivity was $R > 0.99995$.

Reply: The use of the term 'partially reflecting mirror' follows standard nomenclature in the optical community --- the term is used in situations in which the both the transmission and reflection of the mirror are used in the optical train, as is the case here. The instrument would not work if $R=1.00000$ (or, if the transmission $T=0.000$), because no light would enter the cavity, and no light would reach the detector.

Comment: The wavelength ranges and line assignments for the retrieval of CO₂ and CH₄ mixing ratios (~1603 nm for CO₂, and ~1615 nm for CH₄/H₂O according to (Crosson, 2008)) are not stated. It is of interest to have more spectroscopic information in this paper, because the effective independence of the approach from potential H₂O interferences is claimed to be one of the major achievements here. Since some of the improvements listed at the end of section 2 can only be fully appreciated if sufficient detail is given on the CRD analyzer - more information should be provided; perhaps in Figure 1. Please note that AMT is a journal that reaches an audience with significant technical interest.

Reply: More descriptions have been added to the texts and Figure 1 in the revised version.

Comment: Section 3 Please explain to the reader what is meant by the "dilution effect"?

Reply: We have added some sentences to explain the dilution effect in the revised version.

Comment: Section 3.1 Figure 1 does not contain enough details and the corresponding caption should be improved.

Reply: We have added more descriptions in the revised version.

Comment: P3132, L8: Ambient air in a tank was supplied to the humidifier. Where was the ambient air taken from - laboratory air, outside air?

Reply: This ambient air tank was prepared in our laboratory using dried and compressed outside air. We have specified this in the revised version.

Comment: P3132, L18/19: Experiments were performed above room temperature to avoid water vapour condensation in the inlet line. How does that compare with the in-flight conditions? What inlet losses could be expected during the airborne mission.

Reply: The objective for performing the experiments under above room temperature is to supply water vapor of high mixing ratios to the CRDS analyzer so that we could better describe the nonlinearity of water vapor correction curves. If inlet losses of water vapor occurred under flight conditions, these would not have caused a problem for water corrections, because the CRDS analyzer measures the water vapor mixing ratios of the air in the cavity, which are the exact concentrations that should be used for correcting water vapor effects for CO₂ and CH₄. However, condensation is not expected under flight conditions, since due to the rearward facing inlet, the pressure in the inlet tube is lower than ambient, which reduces the dew point of the sample air. We have added these in the revised version (see section 3.1).

Comment: P3132, L22/23: A linear or exponential drift of CO₂ mixing ratios as determined downstream from the humidifier is reported due to the 'interaction' between CO₂ and H₂O. A significant drift is not apparent in Fig. 2a,b; The blue line appears constant over the time interval shown. Do Figs. 2a,b show already corrected mixing ratios? Can the drift be quantified? Can the nature of the interaction be specified - chemical reactions?

Reply: Figs 2 a-b show examples of the CO₂ and CH₄ concentrations when the H₂O switches between ~ 0 % to ~ 2.9 %. The drift was caused by solubility of CO₂ in water contained in the humidifier. The magnitude of the drift for CO₂ is about several tenths of a ppm within an hour period.

Comment: P3133, L6/7: The quadratic dependence of the concentration ratio [CO₂wet]/[CO₂dry] on the water concentration [H₂O] is empirically described by virtue of parameters denoted a, b (for CO₂), c and d (for CH₄). The values given should carry a unit.

Reply: The unit for the parameters of "a" and "c" is %⁻¹, and the unit for the parameters of "b" and "d" is %⁻². We have added the units in the revised version.

Comment: Section 3.2 From the first sentence in this section the reader gets the impression that the temporal constancy of the parameters a to d was to be investigated, and not the transferability of the parameters from one analyzer to another. If the temporal constancy of a,b,c and d is the main aspect in this section, then the following assumption of the authors is not clear to me: "The assumption is that if the coefficients are transferable between instruments, they are also likely to not change over time." How can the long term time stability of an instrument be compared with the performance of two setups at the same time? This requires further justification.

Reply: Transferability between two individual analyzers suggests stability. However, we realize that the statistics from testing two analyzers is still weak. Further water tests are ongoing with various analyzers. We have added the explanations in the revised version (see section 3.2).

Comment: P3133, L20: 'Similar experiments' are not specified sufficiently.

Reply: We have specified this in the revised version.

Comment: P3133, L23/24: The way how two analyzers are connected to the humidifier is not shown, a separate part in Fig. 1 for instance would be appropriate.

Reply: This is shown in the modified Fig.1 in the revised version.

Comment: P3134, L5-8: The authors state that, "Because the water vapor measurement by the analyzer is based on a single stable H₂O spectroscopic feature which is spectrally close to the CH₄ spectral feature, we expect the measurement of the water vapor to exhibit the same highly stable performance over time that has been demonstrated on both CO₂ and CH₄." Since there are two lasers in the device the long term stability for the detection of CO₂ and CH₄ / H₂O may be different. Furthermore, in the last part of the section the performance on CO₂ detection is used to estimate the expectation of instrument stability for H₂O. Is it possible to apply some Alan variance type of analysis to data to quantify the long term stability of the instrument in this context?

Reply: The difficulty in assessing the stability of the water calibration with Alan variance analysis is that it is impossible to provide sufficiently constant water vapor mixing ratios. Note that the required precision for water vapor to achieve 0.1 ppm accuracy in CO₂ is 250 ppm.

Comment: P3134, L13: "In CO₂, these analyzers appear to drift less than 0.5 ppm (Richardson et al., 2009) over two years of operating time,..." Under what operational conditions of the analyzer? What does "appear to drift" mean in this context. The reference is practically not traceable and hence not helpful in this context.

Reply: We have cited another paper in the revised version.

Comment: Section 4 P3134, L24: The analyzer was placed in an environmental chamber to simulate flight conditions of a non-pressurized aircraft cabin. No details are given on the "environmental chamber".

Reply: We have added information about the environmental chamber in the revised version.

Comment: P3135, L8: What is meant by 'undisturbed stability'? In Figure 3 there are mixing ratio spikes apparent at ca. 51500, 54000, 55900, 57000 s for CH₄ and at ca. 54200 and 55900 s for CO₂. Is there an explanation for these?

Reply: 'Undisturbed stability' means "the difference in the mean values is insignificant", and we have rephrased this in the revised version. The mixing ratio spikes happen when the environmental chamber was increasing the pressure. The pressure increase rates at these moments are much larger than those expected to have during flight. We have explained this in the revised version.

P3135, L24/25: "The time delay between the time air enters the inlet until it reaches the sample cell varies according to the bypass flow and relevant volumes." What is the 'bypass flow' and what are the 'relevant volumes' - no experimental details are given here?

Reply: We have modified this in the revised version.

Comment: Section 5 This section compares the mixing ratios of CO₂ obtained with the CRD analyzer and an NDIR CO₂ analyzer aboard the aircraft during the BARCA campaign. At first arguments on the timing of the measurements are made and finally a mean difference (0.22±0.09 ppm) and standard deviation (0.23±0.05 ppm) are stated. The order in which arguments and data are presented here was difficult to follow, since the purpose of initially comparing the timings of the duty cycles in the CRD and NDIR analyzer only becomes obvious in the last sentence of the section. The correlation optimization should be included much earlier in the text or the order in which various aspects are presented should be rearranged altogether. The caption of table 1 is too short. It does not even mention that the data refer to CO₂. Details on the timing are also not included in the table. Column 2 or the caption should contain a year as well.

Reply: We have reorganized the paragraphs and added more descriptions to the Table 1 in the revised version.

Comment: Why was flight 001 removed from the average (not stated in the main text)?

Reply: Flight no. 001 has been used to calculate the average difference from the direct comparison. However, comparison after cross-calibration was not available because the reference gas for this flight was filled with one air standard that was not measured by the CRDS analyzer during the campaign. We have specified this in the note of Table 1 in the revised version.

Comment: Section 6 Was there a specific reason to use exactly four tanks of synthetic air? What determined that number?

Reply: Three tanks were used as calibration gases and the other one was used as a target gas for long term surveillance. The CO₂ mixing ratios of these tanks were determined by a modified Li-Cor, Inc. LI-6251 infrared gas analyzer before the shipment of these tanks to Brazil about one year before the BARCA campaign. Detailed descriptions about the calibrations can be found in Daube (2002). We have specified this in the revised version.

Comment: P3137, L24: Why were absorption cross-sections not obtained from integrated spectra. The corresponding information would be better placed in the experimental section.

Reply: Because the absorption cross-sections were not used. Also see the responses to the referee #1's comment.

Comment: P3138, L14: Why was the z parameter not independently established during the measurements in Brazil - was there a specific reason?

Reply: Ideally, changes in both y and z should be used to correct the pressure-broadening effect for measurements of synthetic air. However, the z parameter was not independently fitted during those measurements in Brazil, because the line-narrowing effect could not be clearly distinguished from noise in these data. This is due to the fact that the inert gas composition varied over a very small range of values in the filling tanks, and because the line narrowing effect is of much smaller magnitude than the Lorentzian line broadening effect. Therefore, we only discuss correcting the pressure-broadening effect based on the variation in the y parameter, assuming that the z parameter is linearly correlated to the y parameter. We have added the explanations in the revised version (see section 6.1).

Comment: Fig. 4 shows modeled profiles according to the Galatry model (Varghese and Hanson, 1984). The authors should consider showing experimental results here. Especially since on P3139 it stated that the uncertainty in the correction is mainly caused by the uncertainty in the y parameter owing to the fit error.

Reply: The uncertainty of this correction is mainly caused by the noise in the y parameter due to noise in the loss and wavelength values of the individual data points that make up the complete spectrogram. It is important to note that this noise is the reason that we use the height rather than the area of the absorption profile to quantify the gas concentration. We have shown experimental data (CRDS readings, mean and standard deviation values of Δy , known concentrations (for the synthetic air standards in the laboratory), and the predicted $d[\text{peak}]/dy$ for each tank) in a table (see Table 2) in the revised version.

Comment: P3138, L19: ... normalized peak height and the width of the spectral profile... -> ... height and the width of the spectral profile ... The discussion on the pressure broadening appears too short. The conclusions in the last paragraph of the section seems not well supported by the information provided in section 6.1

Reply: We have corrected the sentence, added more discussions on the pressure broadening in the revised version. We have modified the conclusions about the accuracy as well.

Section 6.2 When referring to the isotope ratio in the sample or reference mixture parenthesis should be used around the symbols. It should be made clear that mixing ratios are meant here throughout. Hence ... $^{13}\text{C}/^{12}\text{C}$ _sample ... -> ...($^{13}\text{C}/^{12}\text{C}$)_sample ... and ... $^{13}\text{C}/^{12}\text{C}$ _reference ... -> ...($^{13}\text{C}/^{12}\text{C}$)_reference ... and ... $^{18}\text{O}/^{16}\text{O}$ _sample ... -> ...($^{18}\text{O}/^{16}\text{O}$)_sample ... and ... $^{18}\text{O}/^{16}\text{O}$ _reference ... -> ...($^{18}\text{O}/^{16}\text{O}$)_reference ...

Reply: These have been modified according to the recommendations in the revised version.

Comment: P3140, L18: Please explain GLOBALVIEW-CO₂C13.

Reply: We have added a citation for this.

Comment: P3140, L20: What logistic difficulties are referred to here?

Reply: These tanks were in Brazil and it took very long time to get them back to the laboratory in the US mainly due to the problems associated with shipping hazardous materials out of Brazil.

Comment: P3141, L3: Corrections due to variation of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of 0.14 to 0.16 ppm are claimed to be small in comparison to the pressure-broadening impact. This does not seem to be the case. They seem to be within the same order of magnitude (-0.22 ppm to 1.68 ppm on Page 3139).

Reply: One order of magnitude smaller when considering the range of the corrections.

Table 2 should be explained better. The caption is cryptic and I recommend to improve the column titles.

Reply: We have modified the caption and the column titles in Table 2 (now Table 3) in the revised version.

Comment: Section 7 P3142, L6: ... Of green house gases during ... -> ... Of the green house gases CO_2 and CH_4 ... ->

Reply: We have modified this in the revised version.

Comment: SUGGESTED TECHNICAL CORRECTIONS

P3129, L7: ...aircraft -> ...aircrafts

Reply: The plural form for aircraft is aircraft

P3129, L10: ...on board aircraft ... -> ... on board an aircraft ...

Reply: We think both are ok.

P3129, L28: ... and CH_4 with ... -> ... and CH_4 concentrations with ...

Reply: Yes

P3130, L3: ...in all in situ ... -> ... in all previous in situ ...

Reply: Yes

P3131, L4: ...off the laser ... -> ... off the lasers ... (probably plural since there are two lasers).

Reply: The two lasers do not work at the same time. At one time, light from one of the two lasers enters the cavity.

P3131, L7: ... spectrum ... -> ... spectra ...

Reply: Yes

P3131, L26: ... CO_2 better ... -> ... CO_2 to be better ...

Reply: Yes

P3134, L17: ... to just drift of ... -> ...to a drift of ...

Reply: We have changed to "A drift of ... translates to a drift of only ... "

P3134, L27: The analyzer measured a standard gas during ... -> The analyzer measured mixing ratios of standard gases CO₂ and CH₄ during ...

Reply: Yes

P3135, L9: ... compared to under normal ... -> ... compared to normal ...

Reply: Yes

P3135, L12: ...aboard research aircraft or aboard commercial ... -> ...aboard a research aircraft or aboard a commercial ...

Reply: Yes

P3137, L10: ... very linear ... -> ... linear ...

Reply: Yes

P3137, L17: ... one year's storage ... -> ... a one year storage period ...

Reply: Yes

P3137, L19: ... we try ... -> ...we tried...

Reply: Yes

P3137, L20: ... we use the ... -> ... we used the...

Reply: Yes

P3138, L22: Delta Peak is not defined in the text.

Reply: We have defined the definition in the revised version.

P3139, L11: ... total CO₂ by ... -> total CO₂ concentration by ...

Reply: Yes

P3141, L7: ...as well since ... -> ... as well, since ...

Reply: Yes

P3143, L10: Remove line break.

Reply: Yes

References

P3144, L32: ...filed... -> ...field...

Reply: Yes

Figures

The time scales in Figs. 2a,b and 3 seem arbitrary - what is the relevance of the times on the axes?

Reply: The time scales are according to the UTC time of the computer clock of the CRDS analyzer. The numbers are the seconds that have lapsed since the beginning of the day when the experiment was carried out. We have added explanations in the caption of this figure.