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Interactive comment on “Tunable diode laser in-situ CH₄ measurements aboard the CARIBIC passenger aircraft: instrument performance assessment” by C. Dyroff et al.

Anonymous Referee #2

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This paper by Dyroff et al. presents the performance assessment of a commercial Fast Greenhouse Gas Analyzer (FGGA, Los Gatos Res.), which was adapted for unattended air-borne CH₄ measurements within the CARIBIC program. It is focused on the characterization of the instrument in terms of stability, precision, drifts, bias, and accuracy. The calibration approach and data quality verification of the regular, in-situ and continuous CH₄ measurements in the UTLS, are important factors and their discussion as presented in this work can be of interest for the community involved in airborne measurements.

The manuscript is well written, and I recommend publication after addressing some

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minor changes suggested below.

General:

The significance of this unique data-set is briefly mentioned. However, the advantage and importance of the results are not discussed in the broader context of global source distributions, atmospheric variability (vertical and spatial), trends, and their contributions to our understanding of CH₄ cycle. Although, this is a technical paper, it will have an added value if more details and some interpretations of the 103 flight data would be presented to demonstrate the potential and relevance of such kind of data-set.

Since similar work has been done and published employing other laser spectroscopic techniques (e.g. TDLAS, CRDS, etc.), it would particularly be useful for the reader to know, what was the main factor leading to the selection of the measurement technique employed by the authors. A brief discussion on their decision would help the community to gain insight into advantages/drawbacks of existing alternatives.

According to specifications, nowadays instruments from LGR already have data-analysis software that provides correction algorithm for reporting accurate methane and carbon dioxide on dry (and wet) mol basis in real time, i.e. without additional post processing. Why is then the cross sensitivity to H₂O still subject of investigation?

During flight, pressure and temperature variations can be the limiting factors in measurement precision and long-term stability. Similarly, mechanical vibrations tend to impair the performance of the instrument. However, these issues were not included in the characterization (see also the specific comments).

Specific:

P9226/L5: Remove “The modified instrument is described.”

P9226/L6: “A laboratory characterization was performed ...” sounds like you made one single investigation. I suggest replacing with “The instrument was characterized in the laboratory with respect to...”

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P9226/L8: “For airborne operation a calibration strategy is described, that utilizes” should read “For airborne operation, a calibration strategy is described that utilizes”

P9226/L12: Replace “less accurately considered...” with “less accurately determined...”

P9227/L5: Replace “highly unknown” with “highly uncertain”

P9227/L20: Replace “for its CH4...” with “for their CH4...”

P9227/L26-27: I suggest to use the same tense (Simple Past) across the whole paragraph.

P9228/L20: “. . . was developed and is described.” I suggest “. . . is presented in detail.”

P9228/L22: “First observations are presented.” I suggest “Some illustrative examples are shown.”

P9228/L25: Give type or model number of the spectrometer

P9228/L27: “While CO₂ measurements are also obtained by the FGGA, in the present paper we focus on CH₄ measurements only, . . .” This sentence is redundant, since the information is given already in the first paragraph. Just say “Given the limited precision on H₂O and CO₂, we focus on CH₄ measurements only, . . .”

P9229/L5 Write NIR instead of N-IR.

P9229/L7 Replace “two mirrors with very high reflectivity (in our case $R \sim 0.99992$).” with “two mirrors with reflectivity of $R \sim 0.99992$.”

P9229/L14 The phrase “Due to the high mirror reflectivity, the residence time τ_{RD} of the photons traversing at the speed of light c inside the cavity, and thus the effective optical path-length l_{eff} is long compared to single or multipass absorption cells. In our particular case we obtain $\tau_{RD} \sim 10 \mu s$ on average, which corresponds to $l_{eff} = 3$ km in a $L = 25$ cm long cavity, . . .” makes the text unnecessarily wordy. Furthermore, photons

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are always traveling with speed of light in a medium like air. I recommend: “The cavity ring-down time of $\tau_{RD} \sim 10 \mu\text{s}$ corresponds to an effective optical path length l_{eff} of 3 km within a 25 cm long cell...”

P9229/L22: “laser-wavelength scanning via the laser-injection current,” should read “laser-wavelength scanning by modulating the laser-injection current,” It would be useful to know the number of data points per scan. Along the linestrength values of the species, an estimate for the absorbance (or transmission) seen by the instrument under flight conditions (in the UTLS) would be beneficial. Also the detection limit or absorbance equivalent noise should be given. Neglecting these parameters the discussion of the various linestrengths is irrelevant.

P9230/L4 The sentence “The nearest strong H₂O absorption line is located well outside the scan window . . .” is obsolete. Remove it.

P9230/L8 Replace “performing a fit to obtain the CH₄ mixing ratio” with “performing a spectral fit to obtain the CH₄ mixing ratio”

P9230/L9 For a better reading, avoid using the same expression within one sentence. “Fit results along with some housekeeping parameters are transferred via RS232 serial connection to the housekeeping computer for storage.” What are these housekeeping parameters? Just name it.

It is rather unexpected that the most important data such as the measured spectra are not saved. The authors do not even try to give an explanation, which is even more annoying. Nowadays, data-storage solutions should make easily possible to save (and archive) such precious and unique data. This would allow traceability, transparency and, if required, re-processing of the measured values. A brief explanation is required at this point.

P9230/L15 Figure 1 shows a rather general scheme, which can also well be a laboratory set-up or an unmodified commercial instrument. A photograph of the 19” rack

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system would be more adequate. Again, it is not clear whether the power supply unit was custom developed or is a commercial device. What is the noise (level, spectrum) specification of the unit? What about its thermal stability?

P9231/L3 “a proportional valve is used. . .” Add manufacturer and type.

P9231/L3 “a proportional valve is used to establish a constant pressure of 180 ± 0.05 hPa within the cavity. This pressure was chosen in order to reduce the width of the molecular absorption lines, and thus to minimize potential cross-interference”

This pressure value is the standard setting for the FGGAs and the authors can shorten this paragraph. On the other hand, in a recent publication of O’Shea et al. using similar instrumentation, it is argued that 50 Torr (~ 67 hPa) is a good compromise between increased absorption and line-feature selectivity. How this statement fits to your finding?

The authors should also discuss the response (or lag) time of the system including the whole sampling line.

P9231/L10: The authors state that the instrument is stabilized to 40°C , but it would be useful to add some stability range to this value. Furthermore, the thermal stability of the instrument should be discussed. The temperature sensitivity test (between 15 and 40°C) of the spectrometer is missing from the assessment, although it may explain some of the drifts observed by the authors. In general, a brief discussion of environmental impacts on instrument performance and potential sources causing the instrumental drifts would increase the scientific value of the manuscript. Later on (P9237/L17) the authors even mention that the in-strument performance may suffer, but they fail to quantify this effect.

P9232/L8: Remove indent, otherwise the next phrase becomes a general statement, which is not necessarily true. Depending on the time-scales of the drift, the Allan variance can be even higher when the integration time is increased.

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P9232/L16: Remove indent

P9232/L11: Replace “departs slightly” with “deviates”

P9232/L26: A brief explanation is required, why the authors used a second laser for the CO₂ that has anyway low precision and thus give up the 100% duty cycle. Also, what was the noise level of the original instrument prior modification? What is the detector dark-noise level?

P9233/L13: The units for CH₄ should be ppb, not ppbv. The WMO scale for CH₄ is defined nmol/mol and ‘ppb’ is used as an abbreviation. (see e.g. Dlugokencky et al., JGR, 110, 2005, doi: 10.1029/2005JD006035).

P9234/L8: Instead of “Next the cross sensitivity to water vapor (H₂O) is analyzed” I suggest “In the following, the cross sensitivity to water vapor (H₂O) is investigated”

P9236/L21: replace “indivdial” with “individual”

P9236/L28: replace “sparticular” with “particular”

P9237/L1 and L5: instead of “ordinate (bias)” just write “offset” It would be interesting to know, whether the instrument exhibits hysteresis during repeated shut-down/start-up cycles. Is the observed offset part of this phenomenon? Moreover, it would also be interesting to discuss whether a frequent zero-gas addition approach would also be appropriate. A converter could be potentially small and flight compatible.

P9237/L1: replace “red+” with “red cross”

P9238/L10: Avoid starting a phrase with numbers. I suggest writing “About 90% ...”

P9238/L16: “FLASK” is not an acronym, so just write “flask”.

P9244/L 3: add comma after furthermore.

P9244/L4 For better reading replace “obtain” by “achieved” in the sentence: “With this calibration method we obtain a total uncertainty of 3.85 ppb ...”

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