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Comment

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

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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.

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Of course I can agree with the reviewers' opinion. The reason why we did not go into any deeper interpretation of our measurements is that we clearly wanted to focus on the technical aspects and data quality of our data set. Interpretation of the data will be done in future work in dedicated papers/journals.

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.

A statement was added to Par. 1 of Sec. 2.

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 than the cross sensitivity to H₂O still subject of investigation?

A comment was added at the end of Par. 1 of Sec. 3.3 that states the reason for our decision to investigate H₂O cross sensitivity.

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).

There are some occasions where the instrument was not yet properly warmed up when we started the measurement in flight. Then we sometimes observed what might be a

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direct correlation of temperature and calibration offset. However, it is difficult to fully disentangle the processes that may lead to changes in calibration offset. Admittedly, we have not performed dedicated laboratory measurements in this respect. A new paragraph has been added as Par. 6 of Sec. 4.2.

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

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. . .”

Changed.

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

Changed.

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

Replaced.

P9227/L5: Replace “highly unknown” with “highly uncertain” P9227/L20: Replace “for its CH4. . .” with “for their CH4. . .”

Replaced.

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

Tense changed.

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

Changed.

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

Changed.

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

Done.

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, . . .”

The original sentence is preferred. No changes.

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P9229/L5 Write NIR instead of N-IR.

Done.

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

Replaced.

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 $\tau_{RD} \approx 10 \mu s$ on average, which corresponds to $l_{eff} = 3 \text{ km}$ in a $L = 25 \text{ cm}$ long cavity, . . .” makes the text unnecessarily wordy. Furthermore, photons are always traveling with speed of light in a medium like air. I recommend: “The cavity ring-down time of $\tau_{RD} \approx 10 \mu s$ corresponds to an effective optical path length l_{eff} of 3 km within a 25 cm long cell...”

I agree that the wording may be slightly lengthy. It is done as a means to introduce all variables of Eq. 1 prior to Eq. 1. I find this enhances readability overall.

P9229/L22: “laser-wavelength scanning via the laser-injection current,” should read “laser-wavelength scanning by modulating the laser-injection current,”

Changed.

It would be useful to know the number of data points per scan.

The number of data points per scanned spectrum is not accessible information for the

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user of the FGGA. The spectrum in Fig. 2 was recorded with an oscilloscope connected to the FGGA photodetector.

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.

The absorbance for the 1987 ppb is now stated in Par.1 of Sec. 3.1.

Also the detection limit or absorbance equivalent noise should be given. Neglecting these parameters the discussion of the various linestrengths is irrelevant.

The noise equivalent fractional absorbance is now stated together with the precision results at the end of Par. 2 of Sec. 3.1.

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

Removed.

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

Replaced.

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

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connection to the housekeeping computer for storage.” What are these housekeeping parameters? Just name it.

Done.

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.

I agree with the concern about not being able to store raw spectra. This feature is unfortunately not available for this early FGGA model. According to the LGR website, recent models allow for spectra storage. A comment was added.

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 system would be more adequate.

A photo of the modified FGGA was added to Fig. 1.

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?

A comment was added that we use standard Vicor DC/DC converters (full specifications are to be found on the Vicor website). Detector noise is now discussed in Par. 6 of Sec. 2.

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

Done.

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 FGAs 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 a compromise between increased absorption and line-feature selectivity. How this statement fits to your finding?

I agree. We simply use the standard pressure. Paragraph shortened.

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

The response of the instrument is now discussed in the new Sec. 4.5. A new Figure shows a comparison to fast (5 Hz) O₃ measurements. No response slower than the 1 Hz measurement frequency of the FGGA can be observed.

P9231/L10: The authors state that the instrument is stabilized to $40 \pm 0.5^\circ\text{C}$, 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

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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 instrument performance may suffer, but they fail to quantify this effect.

See reply to the last general comment. A new paragraph Par. 6 has been added in Sec. 4.2.

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.

Indent removed.

P9232/L16: Remove indent

Done.

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

Replaced. ***

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?

A sentence is included in Par. 1 of Sec. 2 that the FGGA has two separate laser channels for CH₄ and CO₂ measurements, which are operated in an interleaved fashion. We as user have no control as to switch off one of these channels. A modification by

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LGR in the operative phase of the instrument is not possible due to aircraft certification restrictions. The precision of the original instrument was very similar to the one achieved with the modified one, as is now stated in Par. 3 of Sec. 3.1

The detector dark noise prior and after instrument modification are now discussed in Par. 6 of Sec. 2. Based on the reviewers comment we noticed that we drew a wrong conclusion regarding the electronic noise of our modified instrument. Unlike originally stated in Sec. 3.1, the detector dark noise of the modified instrument is identical to the original instrument. Therefore a noisier detector signal cannot explain the differences to other researchers results with similar instruments. This statement was thus removed from the manuscript. Importantly, the performance of our FGGA has not suffered dramatically by our modifications.

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).

Of course. Changed to ppb.

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"

Changed.

P9236/L21: replace "indivdual" with "individual"

Replaced.

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

Replaced.

P9237/L1 and L5: instead of “ordinate (bias)” just write “offset”.

Done.

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?

During the flight sequences, the flight-to-flight scatter of the offset was around 5–10 ppb, and occasionally up to 15 ppb. A sentence was added in Par. 5 of Sec. 3.4.

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.

A sentence was added in Par. 5 of Sec. 3.4 which states that we do not consider zero measurements because the natural range of observed CH₄ is large, and thus provides us with a well defined calibration function.

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

Replaced.

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

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Done.

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

Done.

P9244/L 3: add comma after furthermore.

Done.

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 ...” Done.

Please also note the supplement to this comment:

<http://www.atmos-meas-tech-discuss.net/6/C4082/2014/amtd-6-C4082-2014-supplement.pdf>

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 9225, 2013.

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