

## Review of the submitted article:

# **Modification, Characterization and Evaluation of a Quantum/Interband Cascade Laser Spectrometer for simultaneous airborne in situ observation of CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, CO<sub>2</sub>, CO and N<sub>2</sub>O**

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### General comments:

This paper describes the performances of an analyser of the major greenhouse gases in air on board of an aircraft. Details are provided on the analyser hardware, the analytical software and the calibration method, followed by an evaluation of the performances by comparison with other analysers present during the same flight. While the analyser itself is not new and was already described in a previous paper (McManus 2011), in this work the number of analysed compounds was extended, the in situ performances were looked at more deeply, and the calibration method was improved. The paper is generally well written, well-structured, clear, and provides lots of details on the instruments and methods. However the section on performance evaluation needs some more work, both in its content and format. I therefore recommend a minor revision before the paper can be published in AMT.

### Comments on the terminology

- Units to be written in plain (not italic) format
- The format to display a value with its unit is “value–space–unit” for example “204*m*” on page 4 should be written “204 m”
- “mixing ratio” to be replaced by “amount fraction”, expressed in mol mol<sup>-1</sup> (nmol mol<sup>-1</sup> for ppb, μmol mol<sup>-1</sup> for ppm).
- Names of molecules to be written in plain (not italic) format.
- Allan deviation seems to be confused with Allan variance. When values are reported in the same unit as the concentrations, this should be a deviation. Please check the correct usage over the document.

### Specific comments by section:

Section 2.1: the text describes two sealed cells containing CH<sub>4</sub> and N<sub>2</sub>O. Where are they on Figure 1? Please indicate the purity of the gas and its pressure.

Section 2.3: please provide more information on the calibration mixtures. In particular NOAA standards are all identified within NOAA database and you could just provide their reference to allow the users looking at all values measured by NOAA. At least please indicate the nominal amount fractions, their uncertainties, and the isotopic composition for CO<sub>2</sub>. This last value is of importance as you noticed a bias between the CO<sub>2</sub> amount fractions measured with your instrument and those measured by the PICARRO.

Section 4, ground–based performance: the reported Allan deviations seem a bit large. Compared to McManus 2011 on CO<sub>2</sub> for example, a factor 10 is noted. Please consider revising the statement that “values are in good agreement with the values reported by Aerodyne” and/or provides further support. Is there an effect of the calibration system described in 2.3, which is said to be used to check the stability and the linearity?

Section 5: while the traceability of measurements with the QCL is clear (calibration with NOAA standards), nothing is indicated regarding the PICARRO. This is needed to fully understand the origin of biases. It seems that an anchored to NOAA is assumed, but this deserves further details (which standards? How many calibration steps? Isotopic composition?).

When both instruments are compared, it would be more useful to express the difference in amount fraction, both in the text and in the graphs. This should then be compared with their uncertainties, not taking into account common sources of uncertainties such as NOAA uncertainty if all amount fractions are expressed on the same scale. Going beyond this, some consideration on how this compares with the Data Quality Objectives set by WMO would be of interest.

The treatment of the constant bias found between the AERODYNE and the PICARRO analysers needs to be improved. Are both analysers calibrated directly with NOAA standards? How different are the calibration gases? It would be valuable to estimate the bias one could expect from the isotopic difference, as done for example in the paper of Chen et al. (*Atmos. Meas. Tech.*, 3, 375–386, 2010), and compare with the observed bias. Indeed, an observed bias of  $10 \mu\text{mol mol}^{-1}$  seems very large.

Section 5, uncertainties: it is not so common to see combined uncertainties considered in such measurements, and the effort of the authors is certainly valuable. However some consideration on how these values compare with other instruments would be required. Is the calibration procedure specific to this instrument? Does this imply a larger uncertainty than for others? Would you say this instrument has comparable precisions than others?

Section 5, discussion on instruments precisions: Allan deviations (not variance) were measured before the flight and during the flight. It is not very clear how those values compare. One would expect the lowest values during ground-based measurements, presumably recorded on gas mixtures with constant flow rate and pressure. During the flight, other sources of instabilities can increase the noise of the instrument. However some of the values appear to be lower during the flight (above the planetary boundary layer only). This would need some further explanation.

Conclusions: the advantages and drawbacks of the aerodyne instrument could be better highlighted. The large number of species analysed together is certainly an interesting feature, but it seems to come with increased noise compared to CRDS analysers. Is that really the case or is this a wrong impression coming from an increased in-flight noise which could impact other analysers as well?

#### Line-by-line comments:

#### **Page 3**

Line 5: you may clarify that “DLR” in the title is the name of the laboratory owning the spectrometer.

#### **Page 4**

Line 25: you may keep SLPM for the flow rate, but indicate the value in  $\text{mL min}^{-1}$  as well

#### **Page 5**

Line 19: why the use of “cross-calibrated” rather than “calibrated”? Does it involve a particular method?

Line 32: the entire sentence may be rewritten to express more clearly that no dilution was introduced at this stage, which is why you do not need to take into account an uncertainty on the flow rate measurements.

#### **Page 10**

Line 14–15: what is meant by “not accurately constrained”? There is certainly an issue with the difference in isotopic composition between the sample and the calibration gas, and this aspect deserves a better treatment in the paper. However at this point you are describing the fit of the spectra, and the statement about constraining the isotopic composition of the sample is unclear. Does this mean constraining the fit? The fit window?

#### **Page 12**

Line 14 “excluding absolute error”. Do you mean uncalibrated or expressing the precision only?

Line 15: “values reported by Aerodyne”. Which paper? McManus 2011?

Line 2–3: “we were not able to reproduce...” seems a rather negative introduction for a positive result, as everything was made to be insensitive to the cabin pressure. Consider rephrasing.

Comments on figures:

Figure 7 it is not clear if the amount fractions are provided after calibration or not. The legend seems to indicate calibrated values, but the y-axis in the right plot indicates “Methane RAW [ppt]” which would mean raw values before calibration. Please clarify.

Figures 10 and 11: it is too uneasy to compare both analysers on the plots. Differences would be more interesting, as the paper does not include any consideration on the amount fractions of the gases.

Figure 12: y-axis of the right plot is the methane amount fraction. Use a symbol and unit such as “ $x_{\text{CH}_4} / (\text{nmol mol}^{-1})$ ” and indicate in the legend “ $x_{\text{CH}_4}$  is the methane amount fraction”.