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Interactive comment on "Modification, Characterization and Evaluation of a Quantum/Interband Cascade Laser Spectrometer for simultaneous airborne in situ observation of CH_4, C_2H_6, CO_2, CO and N_2O" *by* Julian Kostinek et al.

Anonymous Referee #4

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The paper by Kostinek et al. presents ground-based and in-flight performance assessments of a commercial trace gas analyzer (TILDAS, Aerodyne Research Inc.) after its adaptation for airborne operation. The subject is highly topical and targets a key issue that every scientist is facing when taking decision on analyzer selection. This can even be critical considering the stringent place and measurement-time limitations of flight campaigns. Here, the author's choice is on a dual-laser direct absorption spectrometer with multi-species (i.e., five compounds) detection capabilities deploying

state-of-the-art mid-IR laser sources (both QCL and ICL). The in-flight intercomparison with CRDS-based instruments and flask samples is an important element of the manuscript that can be of interest for the community involved in airborne measurements. The manuscript is well written, and I recommend publication after addressing some comments and changes listed below.

General:

The title should better reflect the content of the manuscript. Given that for the measurements the spectrometer is equally using both QCL and ICL devices, this should be weighted the same. Furthermore, the instrument was mainly adopted and not modified for airborne operation. Thus, my suggestion is to write: "Adaptation and performance assessment of a dual-laser mid-IR direct absorption spectrometer for ..."

The abstract should focus on summarizing briefly the highlights and findings of the presented work. Thus, I suggest starting directly with L7, "Here we demonstrate. . ."

Although the introduction contains a brief hint about the large number of available meas-urement techniques for airborne atmospheric measurements, it is unfortunate that the authors completely refrain to motivate their choice for a particular analyzer. Clearly there are some benefits of having multi-species capabilities at slightly higher sampling rate, but how this compensates for the obvious limitations, such as cabin pressure dependence, frequent high-flow calibration requirements, tedious post-processing of the raw spectral data, high power and calibration gas consumption, etc.? A more elaborate discussion on ad-vantages/disadvantages of the chosen approach would significantly improve the manuscript.

In the same context, it is also not obvious in the present form, why the authors decided for an extensive calibration scheme and additional data post-processing instead of developing a purged and sealed enclosure around the instrument. Apparently, most of the relevant drifts or biases are due to ambient air (H2O mainly) absorptions outside the multi-pass cell.

Considering that the spectral retrieval software of the manufacturer has been around for more than 20 years, supporting a large number of custom-built instruments applied in a wide range of applications, one would assume that the software experienced a continuous development and incorporates many fine-tuning/customizing features in order to optimize also the fitting process. Therefore, it is highly interesting and valuable if the custom retrieval software (JFIT) significantly improves the performance. This must, however, be more clearly documented by a side-by-side comparison of the results of both software packages. Especially, the additional shift parameters introduced by JFIT and its co-allocation to various segments of the spectral window seems rather subjective and should be quantified in terms of performance improvements. The authors refer to Fig.8 (p13) to show that the tuning rate of the lasers is stable, which seems to be in contradiction with the many shift parameters introduced in JFIT.

Similarly, the discussion of the water vapor correction should contain further details about the observed biases and drifts in the whole range of water concentrations experienced during flights. Measuring water concentration at absolute level is challenging, so the authors should show the observed correlation between generated humidity and spectroscopically measured water mixing ratios. Also, some discussion is required to make clear how the additionally in-troduced broadening effect improves the measurements, and compare this to the impact of the significant spectral instability (10-3 cm-1) and potential temperature fluctuations of the sampled gas and of the cabin during flights.

Since the instrument is a unique platform, where two different mid-IR laser sources are oper-ated side-by-side, a more detailed comparison of performances and noise characteristics of QCL/ICL would certainly be an added value to the manuscript.

Specific:

Pg2, L27: need more clarification what is meant by sequential and truly concurrent sensing. Otherwise, there should be a short note about the importance/benefit of mea-

suring at 0.5 Hz instead of 2 Hz.

Pg2, L25: a reference to the paper at this stage is enough; especially, that Section 2 starts with the same information.

Pg2, L29-31: How to interpret these cited works? In the present context, they give the impres-sion that there is no open question regarding the suitability of QCLS for airborne measure-ments.

Pg2, L32: the main objective of the paper is missing. What is the final goal of this investigation? Which measurement data and for what purpose are they going to be used? Is the data quality adequate to answer the research questions?

Pg3, Sect 2.1: this section needs some re-work, e.g. statement like “optics compartment con-tains all optical elements” is redundant, while Laser#1 and #2 without clear definition has no sense. I suggest giving the driving specifications of the lasers (current and temperature) as well as their optical power output. Specify the exact detector type.

Pg4, L1: obviously there are many hundreds of reflections within the multi-pass cell, which leads to significant decrease of optical power of the laser beam. What is the reflectivity of the mirrors and how much is the out-coupled ICL intensity?

Pg4, L3: I doubt that the laser devices are directly coupled to the Peltiers. There should be a buffer heat-sink between.

Pg8, L15: the relative frequency changes seem to be the same, which is also illustrated by Fig8. So what is the real benefit for using five different shift parameters?

Pg9, L1: how large were the temperature fluctuations within the optical compartment? What was their effect on the spectral retrieval?

Pg9, Fig 9: specify the averaging time of the spectra.

Pg9, Fig 9: where is the CH4 line in the C2H6 fit-window? What is causing the strong bias in the residual in the CH4 fit-window?

Pg10, L7: a short clarification should be added why the authors chose this difficult spectral window? The range around 2224.5 cm-1 would, e.g. contain all the species with significantly less spectral interference. The ambition of getting the CO_2 along with CO and N_2O introduces severe compromises in the achievable spectral sensitivity and selectivity. Adding the fact that the selected CO_2 line is not even the main isotopologue and seems to have large systematic bias, I seriously doubt whether this compromise is worthwhile.

Pg11, L9: indicate the precision and accuracy of the generated water vapor mixing ratios. What about hysteresis effects, i.e. humidifying vs. drying cycle?

Pg12, L4: How well can the results obtained at 1 SLPM transferred to the 23 SLPM operation regim? What about simply using an empirical correction factor on the retrieved mixing ratios instead of introducing the broadening coefficient in the fitting procedure?

Pg12, Fig.7: it seems that the plot shows the deviation instead of variance. The Allan deviation plot indicates that the instrument drifts already after 30 s even though operated under ground-based conditions. During flight, pressure and temperature variations, as well as mechanical vibrations tend to impair the performance of the instrument at even shorter time-scales. Considering the long-path of the optical cell, I wonder whether the authors did observe any correlated noise behavior when changing gas flow through the cell from 1 to 23 SLPM? As such, it would be useful to see the distribution diagram (or at least to give quantitative estimates of their spread) of the calibration gas measurements during flights.

Pg13, L9: what is meant by software based frequency lock mechanism?

Pg13, Fig8: what is the influence of the sudden frequency shift discontinuities on the retrieved mixing ratios?

Pg17, L4: what would be the required isotopic composition of such a CO_2?

Pg17, L8: give an estimate of the overall calibration gas consumption for the 18 flights

and shortly discuss options for optimization.

Pg19, L10: as mentioned earlier, it would be useful in this context to show the distribution di-agram of the calibration gas measurements performed at every 10 min interval and repre-senting about 10% of the measurement time.

Pg19, L17: it is somehow unclear what applies: in the previous section (pg18, L2) the authors claim that they were unable to reproduce the cabin pressure dependence, but in the conclusion is argued that the known cabin-pressure dependencies are effectively minimized by frequent two-point calibration.

Pg19, L24: was the frequency lock mechanism active during flight operation only? Do the fre-quency-"jumps" correlate with laser heat-sink temperature changes?

Pg20, L2: Having an uncertainty of 1 ppm and systematic bias of 10 ppm on the CO_2 retrieval, projecting towards isotope ratio measurement is quite steep.

Technical corrections:

Pg1, L12: "truly" is not a proper attribute for simultaneous. Remove it.

Pg2, L15: check reference, because Santoni et al. used QCLS instead of CRDS

Pg2, L20: as above, Richter et al. used DFG instead of QCL

Pg3, L16: here and across the manuscript add space between value and unit. Also the chemical formula should be always printed in Roman (upright) type (see e.g. IUPAC Green Book).

Pg4, L3: avoid using laser diode when referring to ICL/QCL devices.

Pg8, L4: replace "micro" by "fit" window.

Pg21, references: check for typos and completeness, e.g. at L10, L13, L22, etc.

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