Interactive comment on “Modification, Characterization and Evaluation of a Quantum/Interband Cascade Laser Spectrometer for simultaneous airborne in situ observation of CH₄, C₂H₆, CO₂, CO and N₂O” by Julian Kostinek et al.

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This is an excellent paper representing very careful and well thought out procedures and analysis methods. The paper is very well written and the results are very sound. I particularly like the fact that two different precision regimes have been identified (within the PBL and above the PBL) and yield different results due to differences in alignment caused by aircraft vibrations. We often see this effect in our measurements and highlighting them here is a further illustration of the care devoted to the measurements presented. The one thing that should be added is a brief section indicating how the in-flight precisions were determined. Did the authors base this on the precision of zero air measurements or the precision of ambient measurements under stable conditions? In the case of the latter, ambient variability cannot be ruled out the in-flight precisions may be even better than indicated.

I recommend final publication after the following minor points are addressed. As you can see, these are all very minor and serve to clarify some of the discussion.

1. Introduction, Line 6: change the word “remain” to “have”

2. Introduction, Line 16: Since there have been extensive measurements of atmospheric gases well before QCLs and ICLs in the mid-IR using for example, liquid nitrogen cooled lead-salt diode lasers as well as other sources, a brief sentence giving a reference to some of this work should be included. One can cite numerous sources, but one convenient way (at the risk of being self-serving) would be to cite our text book chapter which has many of these references (Chapter 2: Infrared Absorption Spectroscopy by A. Fried and D. Richter, in the book Analytical Techniques for Atmospheric Measurement, edited by D.E. Heard, Blackwell Publishing, 2006).

3. Introduction, Line 20: Please change the wording “custom-built QCL….” to “custom-built difference frequency generation (DFG) absorption spectrometer”

4. Introduction, Line 27: Are you strictly referring to established cavity ring-down instruments here or are you referring to more generally IR absorption instruments? Since you mention the “described spectrometer”, I think you should change “established cavity ring down” to “established IR spectrometers”

5. Page 4, Line 25: It would be useful to the reader to further elaborate on the meaning of “jeopardized nominal system startup”. Do you simply mean the large in rush current to get the pump going is more than the airplane circuit breakers can take, or does this
mean that this may cause damage to other parts of the instrument?

6. Figure 5: It would be very helpful to the reader to indicate the mixing ratios in the figure caption used in recording the various spectra.

7. Page 10, Line 2: Where is the weak CH4 line relative to the C2H6 line which is used for spectral shifting of the C2H6 feature?

8. Page 11: This is a very nice discussion of the various broadening parameters and how they are handled. However, this reviewer wonders how important actually including the self-broadening and water broadening are in the final fits since these are smaller by the fact the sampling pressure is 50-mb and the overall spectral stability is in the 10-3 cm-1 range? The air broadening at this pressure is only ~0.0035 cm-1 which is close to the spectral stability. Perhaps a brief mention of how the inclusion of self and water broadening changes the retrieved results should be included.

9. Figure 7 and Its Caption along with Page 13, Line 1: At the FLAIR (Field Laser Applications in Industry and Research) the Program Committee strongly recommended that references to “Allan Variances” should be denoted “Allan-Werle Variances” in honor of the late Peter Werle who adapted this concept to atmospheric measurements. Below I include a portion of the Program Committee’s Obituary for Peter Werle and its recommendation (this need not be included in the final paper but is included here for your reference). Also, what mixing ratios were used in recording Fig. 7 (zero air or calibrated standard mixing ratios)?

In 1993 Peter and colleagues published a seminal paper in Applied Physics B in which they introduced the concept of Allan Variance applied to laser diode measurements of gas concentration time series. The principle of using Von Neumann’s two-sample variance to describe the statistics of time series that exhibit power law spectra that are more dispersive than white noise (e.g., show a varying mean, or drift) was first made popular by D.W. Allan in 1966 for characterizing frequency standards. The importance of introducing this new tool to laser physicists and atmospheric scientists cannot be overstated; it provides a wealth of new information in a mathematically formal and rigorous framework that would not otherwise be easily accessible. Today virtually every group in the world carrying out ultra-sensitive laser spectroscopic measurements employs the concepts introduced by Peter and colleagues. It is indeed rare to find a high-quality paper discussing measurements without an Allan Plot. Perhaps such plots should in the future be referred to as “Allan-Werle” plots.

10. Page 15, Discussion of Fig. 10: In comparing flask and in situ measurements it should be mentioned that care must be exercised in that during times of rapidly changing ambient mixing ratios one may not get agreement between the slow flask samples and fast in situ measurements. Although this is obvious, it is worth mentioning here. I see this is discussed in the Fig. caption 13 but it is also worth mentioning here.

11. Page 18, in the discussions of cabin pressure dependence: The authors should mention that it is not possible to accurately compare the dependence of one instrument relative to another since many instrument-dependent and other factors come into play. For example, some of the dependence is due to the changing mixing ratios for the species under study in the open-air path. Additional dependencies result from movement of optical windows and other components and are instrument dependent. Also, we find that the rate of cabin pressure change is an important factor, and this is specific to the particular aircraft and the flight pattern employed. Hence, the left side of Fig. 12 may not tell the whole story. We find that the delta Pcabin/delta time comes into play between zero acquisitions, and I would expect the same thing here. Perhaps a comment on this should be mentioned.

The following does not need to be addressed in this paper but is for your informational purposes only. Given your zeroing/calibration every 5 to 10-minutes, you may find that this is too infrequent for aircraft undergoing large rates of cabin pressure changes like the NASA DC-8 where the cabin pressure can change as much as many torr/second. The ultimate solution would be to build a pressure-stabilized enclosure.