

**In response to Alan Fried's comments from October the 16th, 2018.**

*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.*

- 5 *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*
- 10 *all very minor and serve to clarify some of the discussion.*

Dear Alan,

- Thank you very much for your kind and helpful comments on the procedures and analysis methods presented herein. The in-flight precisions are in fact based on ambient measurements at stable conditions. Ambient variability can thus not be completely ruled out, however in-flight precision figures could only benefit from sampling gas from pressure cylinders. We further
- 15 completely agree, that a pressure-stabilized enclosure would be the ultimate solution. This would imply heavy modifications on the instrument and render our acquired FAA certification (required for all European Research Aircraft) invalid. For this reason it has not yet been done at this point.

**1. Introduction, Line 6: change the word "remain" to "have"**

The wording has been changed to:

- 20 "[...] is a strong greenhouse gas and is expected to have the most important ozone-depleting anthropogenic impact throughout [...]"

**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).**

A brief sentence giving a reference to some of this work has been added:

- 30 "[...] Spectroscopic instruments making use of molecular ro-vibrational absorption allow for high temporal coverage through fast instrument response times (Chen et al., 2010). Some have already been used for airborne research, e.g. established IR spectrometers (O'Shea et al. (2013); Santoni et al. (2014); Cambaliza MOL (2015); Filges et al. (2015)). Significant effort led to instruments operating in the mid infrared (IR) region, e.g. liquid nitrogen cooled lead-salt diode laser based spectrometers (Fried and Richter, 2007). With the commercial availability of continuous-wave lasers emitting in the mid IR region near ambient temperature (Capasso (2010); Vurgaftman et al. (2015); Kim et al. (2015), Beck et al. (2002)) several new instrument designs have emerged (McManus et al. (2015); Zellweger et al. (2016)). [...]"

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

The relevant sentence has been changed to:

"[...] reported on a custom-built difference frequency generation (DFG) absorption spectrometer [...]"

- 40 **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"**

We are referring to established IR spectrometers in general. The wording has been changed accordingly to:

"[...] used for airborne research, e.g. 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 inrush 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?**  
Inrush currents have previously been too large, causing circuit breakers to trigger. Depending on whether computer/data analysis systems sharing the same circuit breaker, were already turned on, sudden power loss has previously implied further consequences. However, the few power loss situations we have experienced with this instrument have had no noticeable effect. The following sentence has been added for clarification:  
"[...] nominal system startup (priv. comm. Stefan Müller, MPI Mainz). Sudden power failure, due to over-current triggering aircraft circuit breakers, may lead to failures in the data analysis equipment. [...]"
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.**  
The mixing ratios will be indicated for each species in the three microwindows in a revised version of this manuscript.
7. **Page 10, Line 2: Where is the weak CH<sub>4</sub> line relative to the C<sub>2</sub>H<sub>6</sub> line which is used for spectral shifting of the C<sub>2</sub>H<sub>6</sub> feature?**  
In a revised version of this manuscript line 1/2 on Pg. 10 has been changed to: "A single adjacent CH<sub>4</sub> line, located at 2989.981 cm<sup>-1</sup> has been included in order to obtain good C<sub>2</sub>H<sub>6</sub> data even under these challenging conditions."
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<sup>-3</sup>cm<sup>-1</sup> range? The air broadening at this pressure is only ~ 0.0035cm<sup>-1</sup> 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.**  
Not including the self and water foreign broadening leads to relative errors in the range of 0-2%, depending on the species of interest. While small for C<sub>2</sub>H<sub>6</sub> and CH<sub>4</sub> with < 0.03%, the influence on retrieved CO is rather large with ~ 2%.  
"[...] Not including the self and water foreign broadening leads to relative errors in the range of 0-2% for the described setup, depending on the species of interest. While small for C<sub>2</sub>H<sub>6</sub> and CH<sub>4</sub> with < 0.03%, the influence on retrieved CO is rather large with ~ 2%. [...]"
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)?**  
Thank you very much for pointing this out. Occurrences of "Allan variance" have been changed accordingly in a revised version of this manuscript.
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.**  
The following short sentence has been added to the manuscript:  
"[...] both sampled through an upstream dryer. It should be noted, that care must be taken when interpreting the differences between slow flask samples and fast in situ measurements for high-variability flight segments. [...]"
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  $\Delta P_{\text{cabin}}/\Delta \text{time}$  comes into play between zero acquisitions, and I would expect the same thing here. Perhaps a comment on this should be mentioned.

The relevant text portion has been rephrased to:

"[...] A severe cabin pressure dependence in excess of  $0.3 \text{ ppb hPa}^{-1}$  in  $\text{CH}_4$  mixing ratio has been previously reported for airborne TILDAS instrumentation (Pitt et al., 2016). This instrumentation however physically differs from the one reported here. It is not possible to accurately compare the dependencies of one instrument relative to another since many factors/quantities involved are instrument-specific, e.g. the open-path length, the positioning and properties of optical elements, like windows and mirrors, the stiffness and thermal expansion coefficients of employed optical stands, etc.. We were nevertheless able to effectively minimize cabin pressure dependencies during operation of the QCLS instrument aboard the C130 using the calibration strategy from Sect. 2.3. [...]"