Author Response to Anonymous Referee #1

We thank the reviewers for their useful and constructive comments. We shall address each of the reviewer's comments in turn. Reviewers comments are coloured red and the responses are coloured black.

All page/line numbers refer to the position in the amended manuscript, available on the interactive discussion page.

The manuscript by Pitt et al. describes the deployment of a commercial quantum cascade laser absorption spectrometer on an atmospheric research aircraft. Measurements of nitrous oxide and methane are reported and both laboratory and in-flight data are used to evaluate the total uncertainty of the data. Deployment of QLAS on an aircraft in general suffers from changing environmental parameters. Here the dependencies on changing ambient water vapor concentration and cabin pressure, are investigated in detail. In general the paper is suitable for publication in AMT. It is well written and should be published after some minor modifications.

From my point of view it seems that the strong dependency of the cell pressure on changing ambient pressure could be avoided. Why don't you use a forward facing inlet? The additional ram pressure would result in a wider dynamic range over which the cell pressure could be held constant. Also, the cell pressure of ca. 70 hPa could be further reduced, which would also enhance the dynamical range and reduce the pressure broadening of the absorption line, without affecting the sensitivity strongly.

We agree that a forward facing inlet would allow for the cell pressure to be controlled down to lower ambient pressures. However, the rear facing inlet provides better protection from liquid water ingress, which would otherwise cause problems associated with flooding of the mass flow controller and particulate filter (note that all chemistry instruments on the FAAM aircraft are sampled from rear facing inlets). Considering the results of the laboratory tests discussed in P8/L10 – L18, we feel that the practical benefits of a rear facing inlet outweigh the negatives.

In the chapter dealing with the water vapor correction the authors use two different methods to correct ambient data in order to achieve dry mole fractions of the trace gases under investigation: laboratory measurements with humidified standard air and simultaneous QLAS based H2O measurements. I would assume that the laboratory results should be representative of all processes affecting the measured volume mixing ratio (density and pressure broadening effect). Nevertheless it seems that the "calculated" correction is superior. Why is that?

This is an insightful and useful question. Essentially we believe that the spectroscopic correction in the TDLWintel software performs better because it includes the water vapour pressure broadening effects in the mole fraction retrieval itself, calculating and accounting for the physical line parameter changes directly, rather than assuming an empirical relationship of the form of Eq. (2). This has the advantages of: a) correctly accounting for changes in

water vapour pressure broadening associated with measured cell pressure change; b) reducing the sensitivity of the correction to drift in the measurement of the water vapour mole fraction. A discussion of these effects has now been added to the amended text (P12/L3 – L21 in amended manuscript) in line with the above.

It should also be stressed that both the scale factor method and the spectroscopic method involve using laboratory (empirical) results to determine the corresponding coefficients. The text has also been amended (P10/L5 – L18) to better clarify this point.

I am quite surprised that the dependency of the measurements on changing cabin pressure is so strong. Although we also observe some dependency of the optical alignment on pressure changes, this effect is much smaller in our set-up. I wonder whether there are other factors that affect the measurements here. Unfortunately, detailed information on the pressure regulation ahead of the cell and the pressure measurement in the cell itself are not provided. Pressure measurements in general depend on the kind of sensor used and in our experience often suffer from changing environmental conditions (T,P). Is the pressure reading of the sensors used in your set-up absolute or relative to ambient pressure? In the later case, the strong dependency on cabin pressure might be due to pressure measurement principle.

We are very interested to learn that the reviewer has observed a pressure dependency of smaller magnitude with their set-up. It is our hypothesis that the effect may, at least in part, be associated with fringes caused by an etalon effect between plane-parallel surfaces in the optical path (although the optical bench has been designed to avoid this, it is unlikely to have eliminated it completely). Small changes in optical alignment, or slight expansion/contraction of the optical pathlength, would then have the effect of moving these fringes across the spectrum, thus subtly altering the shape of the baseline in the region of the absorption lines. In such a situation one would expect the magnitude of this effect to depend on the precise set-up of the optical bench. Further work is required to establish if this effect is indeed responsible for the pressure dependency observed. An amendment to the text has been made (P15/L30 – L31 & P16/L8) to clarify that changes in the spacing, as well as the alignment, of the optics may be responsible.

Regarding the question on the pressure measurement, an MKS electronic pressure controller was used to regulate the pressure ahead of the cell, as described in P7/L20 – L30. This reads absolute pressure, and is designed to operate across a wide range of ambient pressures and temperatures, and so should not exhibit sensitivity to cabin pressure or temperature over the ranges experienced during flight.