We thank Referee #2 for the suggestions and concerns. We have addressed the comments specifically below, and have updated the manuscript accordingly. We do disagree with the perspective that this paper only presents a calibration procedure (that is not novel to this reviewer) and thus is not worthy of publication in AMT, and outline our perspective below. As some of this concern may have arisen due to a lack of clarity in our writing, we have made changes to the manuscript both to clarify that our approach is an extension of traditional calibration methods and highlight the uniqueness of the approach. We have also added figures as requested more clearly illustrated to impact of mass flow control and the appearance of our calibrations. Overall we feel these changes have improved the manuscript.

Anonymous Referee #2:

This work presents airborne data of an in-situ QCL absorption spectrometer measuring greenhouse gases N2O, CO2, CO, and H2O with a commercial Aerodyne spectrometer. Such instruments tend to show strong drifts due to changing pressure and/or temperature inside the aircraft cabin. This holds particular during ascent and descent and for unpressurized platforms.

To reduce the effect of the drift, the authors apply a calibration system, which is new according to the authors - the frequent calibration high performance airborne observation system (FCHAOS). Basically the absorption cell of the IR-spectrometer is frequently flushed with a high flow of calibration cylinders with ambient mixing ratios of the target gases tracable to the NOAA WMO scale. The authors apply a duty cycle of 120 s and purge the cell for 10 s with additional 5 s latency before measuring. The output frequency is 1 Hz. The authors show, that by applying this calibration procedure the effect of the drift is accounted for. In-flight comparisons with a PICARRO CRDS system confirms this. The correction is shown for N2O data during a research flight and demonstrates the effect of the procedure.

The paper is well written and documents the calibration procedure allowing to reduce instrumental drift particularly during ascent and descent. I fully acknowledge a clear documentation of instrumental performance and data processing. However, I can't see the novelty of the approach. Fast flow and frequent short calibration with subsequent linear drift correction is basically, what has been applied here. Note, that 1.5 slpm are not novel (e.g. Korrmann et al., 2005 used 1-1.5 slpm at 56 hPa, cell < 0.5 l) as well as linear drift correction is standard.

If the authors could show, that the regulation of mass flow (MFC) upstream the cell (and downstream the cal switch valve) is the key to guarantee short calibration times by reducing pressure pulses (as suggested in the conclusions) I would see a potential new aspect. For this they should provide e.g. comparisons between pressure and flow controlled approaches (see below). It is not shown, why a pressure controlled system should not have the same performance.

As the paper currently stands, it is a well documented calibration procedure of a commercial instrument with standard methods. Therefore I don't see the paper in AMT in its current form.

We respectfully disagree with the perspective that this paper only presents a calibration procedure (that is not novel to this reviewer) and thus is not worthy of publication in AMT. This criticism is founded on two underlying perspectives: 1) That the only new value of this paper is the presentation of a new calibration method & 2) the calibration method is not novel. We disagree with both perspectives. Firstly, the paper is not solely about a calibration method. This paper is the first presentation of this flight system and shows extensive validation with in-flight null tests and direct comparison with a Picarro. Even if the calibration approach was not novel, reporting the instrument performance and traceability with validation in such detail warrants publication on its own in AMT, and in fact is necessary for the community to have confidence in the data reported from this flight system, particularly considering the most similar flight system published cannot collect data during vertical profiles (Pitt et al. 2016). This is consistent with AMT standards and the expectation that papers "comprise the development, intercomparison, and validation of measurement instruments and techniques ..." as evidenced by similar publications focused on intercomparison and validation (for example Santoni et al. 2014 & Pitt et al. 2016).

Regarding the calibration approach, it certainly can be considered a natural extension of previous calibration approaches. In spite of the apparent triviality of the approach, the combination of high flow rate, mass flow

control and high frequency, short duration calibrations with near-ambient concentration cal gas has never been applied to other modern GHG systems. The most recent, state-of-the-science instrument paper on a flight QCLS N₂O system concluded they couldn't use data during vertical profiles (Pitt et al. 2016)—an important limitation for a flight instrument. While our approach may seem simple, we are able to achieve unprecedented in-flight performance in the face of dynamic environmental variables (cabin pressure, etc.), thus achieving a better duty cycle and more robust performance than any published flight N₂O system. We feel the overall approach is novel (and we note the other reviewers do as well), but even if the calibration method is felt to be simple, the extensive validation presented in the manuscript is new and necessary to document this systems performance.

Some of this concern may have arisen from lack of clarity in our writing. In response, we have made some changes to clarify that this is an extension of traditional methods and pinpoint the uniqueness of this approach (see abstract, intro, conclusion).

Also as requested, we have added a figure and discussion on the importance of the mass flow control approach (contrasting with the p-control setup) to more clearly illustrate some of the different elements/novelty of the setup. This is outlined more below.

Main point:

If the use of an MFC is the key novelty this should be clearly documented in the analysis. The current Fig.1. and the text states, that three-way solenoid valves are used (p.4, 1.16/17). In case of a calibration I expect a direct connection between the pressure transducer (calibration tank) and the MFC controlling the cell flow and thus a pressure pulse. The inlet line is probably closed during calibration. In case of switching from ambient to calibration I still expect a short pressure pulse perturbing MFC and cell pressure. This will probably stabilize after a few seconds since the MFC limits the flow, but I do not see the advantage or novelty over a calibration using overflow of calibration gas by flushing the inlet at ambient pressure, which has been applied since years to GHG measurements by TDLAS (or QCLAS).

Note, that many QCLAS or TDLAS systems often are calibrated by flushing the inlet line with higher flow rates than the cell flow. The calibration gas tube is directly connected to the inlet and thus ambient pressure solely via a t-connector in the inlet line. Calibration gas is just switched via an open/close valve. This ensures a minimum pressure perturbation of the cell due to the open connection of the calibration line to the inlet.

This has been established over a long time (e.g. Wienhold et al., 1998) and a potential advantage - if existing - via the proposed procedure in Gvakharia et al., should be documented.

Thank you for bringing this issue up. We are familiar with the excess flow, pressure control setup, and have flown such a system many times. We note one disadvantage of the excess flow setup is that it can lead to contamination of other instrument sampling for nearby inlets and thus is often not preferred (with a second weakness being larger cal gas consumption). Even with the excess flow setup, there will invariably be some pressure fluctuations in the cell when switching to calibration gas. Depending on the specifics of the pressure control setup, to achieve stable pressure control over the entire dynamic range sampled by the aircraft makes it a challenge to prevent any pressure blips when switching to calibration gas, though it may be possible. We show below here an example of a calibration with a pressure control setup (including excess flow) compared with the mass flow control. For our system, we have peak-to-peak fluctuations on the order of 0.8 Torr occur with the pressure control setup and transient pressure fluctuations that do not stabilize within 10s. With mass flow control, they are reduced to ~0.04 Torr, an order of magnitude smaller, and stabilize in much shorter times. We have added this figure and some related discussion to the manuscript in Section 2.2.





The plot below shows the calibration signal in flight, with ambient pressure around 730 mb. Similar to what was seen in the pressure plot above, the fluctuation in pressure is minimal when the valve changes. We have also added this figure to the manuscript in Section 3.2.



p.13, 1.5: How do the respective Allan variance plots look like for the Nulltest? How do they compare to a lab test?

The top set of plots shows Allan variance for the 04/26 null test. The bottom set shows Allan variance plots for when gas was sampled on the ground (note tanks are dry so there is no H₂O). As is illustrated, the in-flight performance closely matches the ground performance, except noise is increased by a factor of 2. We have added text accordingly to make this point in Section 4.1.



Fig.4: y-Axis: mixing ratio instead of concentration (also check the main text).

Thank you for the suggestion, we have updated Figure 4 as well as Figure 2 and checked the consistency of the main text.