Referee #1 (Alan Fried)

This is an impressive study which describes a newly developed QCL-based absorption spectrometer for highly accurate balloon-borne water vapor measurements in the UTLS. The instrument was validated in the laboratory employing SI-traceable reference gas standards from: a dynamically diluted gravimetric permeation system; and from gravimetrically prepared compressed gas water vapor mixtures. Various absolute water vapor mixing ratios from 2.5 ppm up to 180 ppm were generated and measured over the range of temperature and pressure conditions representative of the UTLS.

Using these standards, improved spectroscopic broadening and pressure shift parameters were determined employing a quadratic-speed dependent Voigt fitting profile, which accurately reproduced the measured line shapes without systematic residuals over the entire pressure range. The resulting fitting approach and empirically determined parameters improved the resulting accuracy over the more traditional Voigt line profile using HITRAN 2020 parameters. The authors clearly demonstrated that their balloon-borne instrument is capable of retrieving UTLS water vapor mixing ratios with a calibration-free instrument with an absolute accuracy to better than ±1.5 % with respect to the SI-standards.

The results of this study and the future deployment of this instrument will represent a significant advancement in UTLS measurements of the most important Greenhouse gas. This paper is Excellent in its: Scientific Significance, Scientific Quality, and the Quality of Presentation. This reviewer highly recommends publication with only minor revisions, as detailed below, for improved clarity.

We are very grateful for your nice words and supportive feedback.

1. In the Abstract, it would be desirable to define the acronym ALBATROSS

The acronym ALBATROSS is a somewhat freely chosen name resulting from a random pick of letters of a descriptive phrase: "Balloon-borne laser spectrometer for UTLS water research". To avoid repetitive phrases in our abstract, we decided to leave the definition in the Acknowledgement section.

2. Line 18, page 2: It would be useful to the reader to indicate right up front to write: "The aim of this work is to validate the accuracy and precision of a newly developed **open path** mid-IR quantum-cascade …..", even though this is indicated in the next section.

We agree, implemented as suggested.

3. Page 3, Starting on Line 13 in the discussion of "Rapid spectral sweeping …", although this approach is discussed in Graf et al. (2021), it would be useful for the reader to add another sentence right after "….Liu et al., 2018) further explaining this approach here. For example, you could add "

In this approach the laser driving current is applied in pulses, followed by a moment of complete shutdown of the laser to re-establish its initial temperature".

We have slightly modified the respective paragraph to include more details about the laser driving as suggested:

"Rapid spectral sweeping of the QCL is achieved by periodic modulation of the laser driving current, following the intermittent continuous wave (ICW) modulation approach (Fischer et al., 2014). In this approach, the laser driving current is applied in pulses (typically 100 μs long), followed by a short period of complete shutdown of the QCL. The ICW driving is obtained using custom developed analogue electronics (Liu et al., 2018)."

4. Regarding Fig. 1, I am a little confused by the conceptual diagram of the solenoid valve following the diluted permeation source. I would expect the common port on the MFC3 end and not on the Zero air input end. Also, I was expecting a multi-port valve whereby the diluted permeation flow is directed out to a vent port without flow disruption when the Zero air is switched in. I realize this is a conceptual diagram, but it would be important for clarity to represent this more accurately.

Indeed, the common port of the solenoid valve is connected to MFC3, in order to switch between the diluted permeation source and zero air. The orientation of the solenoid valve is now indicated by the labels "COM" (MFC 3), "NO" (normally open - zero air) and "NC" (normally closed - permeation source) in the schematics. Also, there was an overflow between the solenoid valve and MFC3, which was not indicated in the previous schematics. This is now included in the revised version of Figure 1.

Also, for completeness the authors should mention whether or not they observe any hysteresis in the time response passing through MFC3 into the MP cell upon switching in and out the diluted permeation flow and the Zero air. Is the 3 hour equilibrium time dictated by hysteresis in this MFC, by the MP cell, by the connecting lines, or all of the above?

The response time of the instrument after switching between the diluted permeation flow and zero air varied in function of two factors: the gas pressure and the water vapor content. A lower gas pressure resulted in systematically longer response times and higher zero levels that suggests enhanced desorption effects of H_2O from surfaces. Furthermore, an elevated water vapor content involved a longer response time, a clear indication for memory (surface) effects. The complex interplay of these two mechanisms hinders us to make any quantitative statement of such hysteresis effects, also because of the lack of repeated measurements at the same pressure conditions.

Nevertheless, the contribution of the MFC to the response time of the instrument was evaluated at the end of the campaign, by connecting the synthetic air source directly to the multipass cell (i.e., bypassing MFC3 and the solenoid valve). This showed no significant variation in the zero level reached by the instrument, indicating that the role played by the MFC is negligible. Hence, the effects discussed above are mainly due to the multipass cell and the sampling line, although, their relative contributions were not assessed in details.

We added a more detailed description of the observed effects in Section 2.4 of the revised manuscript:

"This behavior is mainly due to the strong surface adsorption/desorption properties of H_2O, causing

a memory effect to the system. Throughout the validation experiments, we observed a tendency towards slightly elevated zero levels whenever the gas pressure in the sampling line was lowered. Furthermore, the response time of the instrument showed a clear correlation with the humidity content of the measured gas. While these effects may, if not properly taken into account, e.g. by the empty-cell spectrum normalization, affect the accuracy of the measurements, they are largely absent during flight conditions, where the instrument is operated in open-path configuration. In this case, there is no sampling line, the gas flow is much larger, and the surfaces of the SC-MPC are drastically reduced, as the lids are removed and the gas-surface interaction is limited to the narrow inner circumference of the cell."

Also, a brief discussion of the actual liquid water source used in the permeation device and its purity is warranted

The water within the permeation device was ultrapure water. This information is now included in Section 2.2 of the revised manuscript.

"The permeator […] was filled with ultrapure water (resistivity 18.2 MΩ cm at 25 °C, corresponding to a purity > 99.999 %)."

5.   Table 1 headings and descriptor at the bottom, needs further clarification/modification. The labels "Nominal value" and "Actual value" need to be defined more clearly in the descriptor below. These terms refer to the generated water vapor mixing ratios from the dynamic-gravimetric method and the spectroscopic measured values, respectively. The descriptor "expanded measurement uncertainty" is confusing here, as I believe you are combining two terms: the expansion in the number of significant figures in the determined spectroscopic values from the nominal values expected from the generation system and the combined uncertainty from all the parameters employed in the spectroscopic measurement? Is this correct. Are these uncertainty limits at the 1σ levels? Do the nominal values from the standards generation system have additional significant figures than those shown in Table 1? I ask this because aren't you ultimately relying on agreement between your qSDVP fitting procedure and the standards generation system output (as implied on page 10, line 3)? If your absolute water mixing ratios from the standards generation system does not have more significant figures than those indicated in Table 1, aren't you reporting too many significant figures in your expanded actual values? Perhaps some clarification here?

The label "Nominal value" in Table 1 does not refer to the actual H_2O amount fractions generated by the dynamic-gravimetric method, but rather to the *target* H_2O amount fractions that we aimed to generate by this method. Conversely, the "Actual value" refers to the *actual* generated H_2O amount fractions (and their expanded uncertainties), determined upon calibration of the permeator and the dynamic dilution unit before and after the measurement campaign. The spectroscopically measured H_2O amount fractions are not given in Table 1, but only in Table 3 (expressed as differences with respect to the "Actual values" generated by the dynamic-gravimetric method). Hence, the number of reported significant figures is consistent with the accuracy of the reference gas generation method (< 1.5 % at all conditions).

For the sake of clarity, we have now replaced "Nominal value" with "Target value" in Table 1, and keep "Actual value" to indicate the H_2O amount fractions generated by the permeator. The same applies to the pressure values given in the same table.

6.   The comparison between the polynomial baseline fitting and the empty cell approach is impressive and is important since only the former can be used in real atmospheric measurements.

Good remark. We added a sentence in the Conclusions to highlight this point.

"Furthermore, the comparison between the normalization methods, i.e., empty-cell and polynomial-baseline (shown in the Supplementary material) demonstrates the applicability of these results to the analysis of real atmospheric (open-path) data as well."