

## Authors' Response to Referee Comments #1

We would like to thank the Referee for the constructive comments and helpful suggestions on the manuscript, which helped us to further improve the clarity of the paper. Below, we give detailed responses (in blue) where appropriate.

**General comments:** There are only two minor remarks that apply to the paper in its current form, which should be published after minor corrections. First, the capability of measuring at high temporal resolution is pointed out at several instances all over the paper and in the abstract. However, apart from the better than 1 Hz requirement and the capability to measure at 1 s time resolution, no clear number is given with respect to the time resolution reached by the final instrument. I would suggest putting such a number (or a qualifier such as sub-Hz) in the abstract and the conclusion to make things clear.

In the context of balloon-borne soundings, a few meters vertical distance per measurement are considered as high resolution. There are two requirements for such a rate to be reached: Not only the capability of a fast sampling guarantees high effective resolution; also, the air exchange rate in the sampling zone must be adequate. Therefore, we realized an open path sampling system that allows a high throughput of air. In addition, the recording of individual spectra (full scans) takes place at a rate of 3 kHz. These single spectra are then co-averaged 3000 times to improve the signal-to-noise ratio (SNR) and give roughly one humidity value per second. In principle, though, the number of co-averages can be reduced, leading to higher temporal resolution, but on the expense of precision. Thus, technically, the effective acquisition rate may be increased up to 3000 individual humidity measurements per second.

We agree that this should be mentioned more clearly in the manuscript. Therefore, we added additional information to the Section 2.3.4 *Laser driving and data acquisition (P8, L205)*: *"An individual spectrum is recorded during the applied current ramp that lasts for 200 us and consists of 25'000 measured intensity points. A dedicated FPGA functionality sums up in real-time a predefined amount of individual spectra -- usually a few thousand spectra per second -- in order to improve the SNR. The number of averaged spectra and the duty cycle can be individually selected, taking into account the trade-off between precision, temporal resolution, and covered wavelength range."*

In addition, we add the following sentence to the methods description subsection of the balloon-borne experiment:

*"The spectra were recorded at a repetition period of 300 us, which is a good trade-off between broad spectral coverage and speed, as it enables the co-averaging of 3000 spectra to reach 1 Hz temporal and 5 m vertical resolution."*

Second, an uncertainty budget (L301+) for the water vapour measurements is provided, but the discussion does not include any of the systematic biases that were discussed in the previous paragraphs. While degassing from the balloon seems to be an unwanted effect that can be overcome using a longer rope or by measuring in descent, the problem of degassing into the 9 mm interior optical path has not yet been overcome by the present design. According to Fig. 7, this problem is apparent in the >13km altitude range, where a correction is applied that brings the water mixing ratio to constant values. What is the uncertainty of the measurements in this altitude range? Then, there is (another?) apparent offset (>10%) between QCLAS and CFH in the 11 to 13.5 km range. Where does this come from? I could not find an adequate discussion, but obviously, the proposed degassing correction does not account for the discrepancy. This systematic bias also warrants discussion in the text and a corresponding number should appear in the uncertainty budget.

We agree that the discussion of uncertainty contributions is somewhat confusing. Therefore, we modified the Section 3.2.1 to make it clearer, discussing the reasons for the recorded undesired water absorption (1) within the instrument enclosure, (2) within the laser housing, and (3) due to balloon-related desorption in three different paragraphs. In this section, we also mention the attempt of correcting for contribution (1) and (2).

Concerning (1), the outgassing of humidity within the instrument, the actual design does not yet offer a solution to the problem, indeed. There are ongoing efforts for providing a technical solution, though. During the reported measurements, the only option to correct for this offset was to subtract the estimated internal humidity contribution, based on the measurement of the embedded capacitance RH-sensor. However, the conditions in question ( $<200$  hPa,  $<8\%$  RH) were far outside the specifications of this device, even yielding negative values for a relevant fraction of the measurement period. We have tried several strategies to calibrate this sensor against the QCLAS data, but it is very likely that this sensor exhibits non-linear behavior, especially at very low RH. This would largely explain the offset that already appears at about 11 km, meaning that the capacitance sensor underestimates low RH. At the moment, this is the only way to correct for the signal due to internal humidity, although with little information about the quality of this correction. We state this fact more clearly in the text by reformulating the corresponding sentences: *"While this correction partly removes the trend, the sensor fails to deliver plausible absolute values in the stratosphere, where the environmental conditions ( $p < 200$  hPa,  $RH < 8\%$ ) are far beyond the specifications for this capacitive RH-sensor. Therefore, it is likely that the remaining bias between CFH and the corrected QCLAS data is caused by a significant error of the internal humidity measurement."*

Having discussed these unwanted, avoidable, or external contributions to the uncertainty we have identified in the experiment, Section 3.2.2 focuses on intrinsic method- or instrument-related sources of uncertainty that remain even under optimized conditions. For the demanded completeness, we add an additional category (0) *spurious water vapor (external and internal)*, although this is not the focus of this section.

#### Technical:

L45 "these are yet itself ..." → "these are yet themselves ..."

Done

L46 Mention the Kigali (2016) amendment to the Montreal Protocol.

Done

L66 - 67 "The tunability of the laser allows to record entire transmission spectra at fast scanning rates, from which the number of molecules can be deduced." An entire rovibrational spectrum of  $\sim 100$   $\text{cm}^{-1}$  is hardly accessible for a conventional high-resolution laser. The sentence should be rephrased accordingly.

The term 'spectrum' was used here to distinguish from single wavelength measurements. For clarity, the sentence is rephrased: *"In this application, we exploit the fast tunability of a quantum cascade laser to record transmission spectra at kHz repetition rate that cover a few wavenumbers. From these spectra, the number density of target molecules is deduced using the Beer-Lambert-law, which describes the attenuation of radiation in absorbing media."*

L109 "while the absorption signal can be optimized by the selection a wavelength range with strong" → "while the absorption signal can be optimized by the selection of a wavelength range with strong" or "while the absorption signal can be optimized by selecting a wavelength range with strong"

Done

L112 "due to the (a) symmetric stretching mode ( $\nu_1$  and  $\nu_3$ ) ... ". Use of singular is confusing here. Better write, "due to the symmetric ( $\nu_1$ ) and asymmetric ( $\nu_3$ ) stretching modes ..."

Done

L119 "According to the HITRAN-database, the absorption parameters for this line, especially the intensity, are known with an accuracy better than 2 %." While the reference to HITRAN is correct, the authors should also cite the original work that has led to the entry in HITRAN. Otherwise, the people who did the (spectroscopy) work don't get the credit.

Done, we now cite the original sources of the used parameters, i.e. self- and air-broadening, air-broadening temperature dependence, the spectral line intensity, and the line position.

L211 The SI recommended  $\mu\text{mol/mol}$  is to be preferred over ppmv, especially because SI recommended abundance (mixing) ratios are used elsewhere in the article (e.g. Figure 6).

In our view, the correct terminology is "amount of substance fraction", given in mol/mol units. However, the scientific community in this field most frequently uses the term "(volume) mixing ratio" in units of ppmv (e.g. [Vömel et al. \(2016\)](#), [Lossow et al. \(2019\)](#), etc.). To avoid confusion we included a clear definition on L95 and we now consistently use the unit (u)mol/mol throughout the manuscript.

L218 "mixing ratios is" → "mixing ratio is"

Done

L233 - 235 "Numerous additional tests ... A detailed discussion of these experiments can be found in Graf (2020)" It is not quite clear how relevant the information is. Either the phrases should be omitted or the authors should give a very short summary/conclusion of the relevance of these tests for the current paper.

Rephrased: *"Additional tests assessing the mechanical and thermal sensitivity have been performed, elaborating the performance of the instrument under field conditions. This includes climate chamber experiments at stratospheric pressure, temperature, and humidity; however, due to technical limitations, without the possibility of controlling more than one of these parameters simultaneously. A detailed discussion..."*

L256 - 261 "In the right panel of this figure, the relative deviation is plotted, showing a standard deviation of 13 % between 2 and 10 km altitude." The way the paragraph is organised is confusing, because the authors mention the standard deviation first and the mean deviation of 3 % only at the end. The order in which variables are presented should be reversed.

The two sentences are reorganized. The 3 % mean deviation is now stated first in connection to the left panel of 7(b), mentioning the standard deviation of 13% later.

L327 "A standard deviation of 1.1 % relative to the finally chosen configuration ...". This sentence needs to be rephrased. It is not clear what "relative to" means in this context.

Revised sentence to: *"The evaluated mixing ratios  $\chi$  reach a stable plateau in the region  $\Delta\nu = 0.4 \text{ cm}^{-1}$  for baseline polynomials of the order 3 to 6. Within this region (red area), the calculated  $\chi$  show a standard deviation of only 1.1%. Therefore, the central values  $\Delta\nu = 0.4 \text{ cm}^{-1}$  and  $P_4(\nu)$  are the selected evaluation settings to analyze the presented experimental data. In Fig. 9(b), the mixing ratio retrieved with this configuration is set to unity. Other combinations within the red area show maximal deviations of +3.2 % and -1.2 %.*

L335 "which are currently not available for the herein used molecular transition" → "which are currently not available for the molecular transition used here"

Done

L336 "herein selected absorption line" → "selected absorption line" L382 H<sub>2</sub>O → "H<sub>2</sub>O" L442 H<sub>2</sub>O - → "H<sub>2</sub>O"

Done

L444 Camy-peyret → Camy-Peyret

Done

Figure 6 labels are too small and hardly legible

Done

Figure 8 Label identifies water vapour in "atmospheric." Should be "in atmosphere."

Done

Figure 9 Graphs are too small. It is difficult to identify the different retrieved mixing ratios

Done; plots enlarged.