Authors’ Response to the Anonymous Referee #1

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We are grateful to the Referee #1 for the insightful comments and suggestions on our manuscript. We respond to them in detail below. The original review is given in black, our anwers in blue.

General remarks

I really enjoyed reading the first part of the paper. I had the feeling that all the questions risen during reading were answered in the subsequent sentences or paragraphs. Unfortunately, this impression was failed at Section 5 and 6. In my opinion, there was a break in the flow of the manuscript. The presentation of the results, and particularly its discussion remained non-conclusive. In the end I could not tell why the measurement was conducted for, and why was it important to carry out the measurements in a turbulent flow. How this measurement helps in such applications? I hoped that this question will be addressed in Summary and Discussion, but it was not the case. Anyhow, as I mentioned, the topic is very important, and the results are interesting and promising, but I wish a more detailed discussion with respect to the application in a turbulent flow.

We formulated the aim of the measurements in the introduction before:

> The goal of the series of experiments was two-fold: (1) to evaluate the properties of FIRH under a wide range of well-defined reproducible conditions resembling those in the real atmosphere, (2) to characterize the humidity field and turbulent fluctuations of humidity inside LACIS-T for different settings of the wind tunnel.

The key point is that previous cloud-formation studies conducted at the LACIS-T facility (Niedermeier et al., 2020) included the measurements of droplet spectra, velocity fluctuations and temperature fluctuations but did not include the measurements of humidity fluctuations. Therefore, our work complements previous efforts with an important additional piece of information.

Following the Referee’s comment, we added a paragraph at the beginning of section 5 which reminds the second goal of the study and explains the purpose of the measurements series in a more clear manner:

> In this section, we intend to reach our second goal formulated at the beginning: characterize the humidity field and turbulent fluctuations of humidity inside LACIS-T for different settings of the wind tunnel. The previous cloud formation studies conducted at this facility included the measurements of droplet spectra as well as turbulent fluctuations of velocity and temperature (Niedermeier et al., 2020) but the properties of the humidity field, specifically its turbulent fluctuations could not be evaluated so far. The knowledge about these fluctuations is of

great importance for the understanding and interpretation of past and future cloud formation studies at LACIS-T. Therefore, we performed several measurement series named scans in order to investigate the mixing of the two air streams differing in thermodynamic properties. We selected the conditions which have been already used in former studies (Niedermeier et al., 2020).

Moreover, we rearranged section 6 to underline the motivation given above.

Specific comments

1. Line 33: The authors list numerous hygrometers, but in my opinion an important type of instrument is missing, namely a photoacoustic based hygrometer. Although such a hygrometer is implicitly cited, but could also be referred here (see e.g., Szakall et al., Frontiers In Physics, 2020; or Tatrai et al., AMT, 2015). These papers address a lot of similar problems as the hygrometer of the present manuscript has, like antireflection coating, and multiple reflection from windows, for instance.

   We agree that photoacoustic spectroscopy is one of the key measurement methods in hygrometry. We supplemented the overview of current hygrometers with the suggested references (Tátrai et al., 2015; Szakáll et al., 2020).

2. Fig.1, and Fig 3: Probably that was my fault, but it was for me very difficult to figure out what is x direction and what is y direction. The caption in Fig. 3. did not help either ("x position – long path, perpendicular to what is depicted in this scheme"; does not tell for me anything). Then I found the description in line 351 which helped a lot: "across the long and short dimensions of the rectangular measurement section of LACIS-T". (Probably it was written earlier, but I have overseen it?) Please consider showing x and y directions in Figure 1. Further, in caption of Fig. 1 please indicate what DPM means.

   We refined our terminology to "sampling across long/short dimensions" and changed the acronyms denoting the experiments to COMP-L, COMP-S, SCAN-L, SCAN-S where L and S refers to long and short dimension, respectively. This convention is now explained in sec. 2.2. A "scan" is defined in sec. 5 as a series of measurements performed across long dimension at different positions x (SCAN-L) or across the short dimension at different positions y (SCAN-S). We corrected the captions of Fig. 1 and 3 accordingly and added the axes of the coordinate system in Fig. 1.

3. Line 97: Please revise: "one can calculate water vapor concentration" – I found the word "easily" superfluous.

   We removed the word "easily".

4. Line 132: Why did you use an electrooptic amplitude modulator? Semiconductor lasers can be easily modulated with their currents. Was that because of the disturbing effect of a residual wavelength modulation? Furthermore, in the Summary you mention the difficulties with measuring at two wavelengths with this setup. Would that be possible to apply

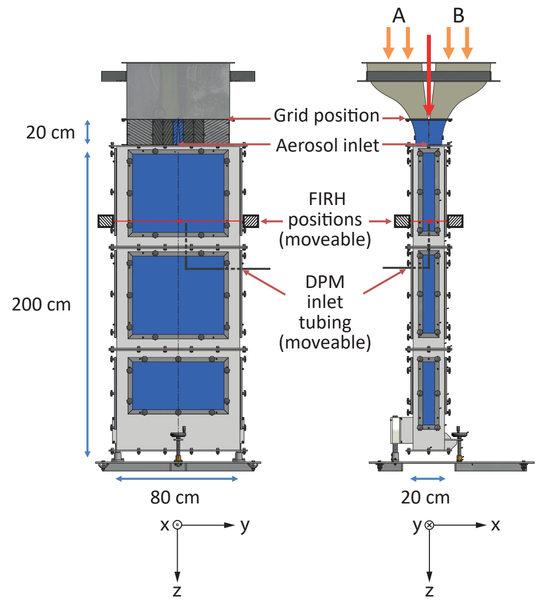

Figure 1 corrected. Schematic of the measurement section of LACIS-T. A and B mark the two air streams which are mixed in the measurement section. The red arrow marks the location where aerosol particles can be injected. Axes are included in order to display the geometry where $z = 0$ is the tip of the aerosol inlet, and $x = 0$ and $y = 0$ are the centerlines of the two transverse dimensions of the measurement section. The red lines denote the position of the Fast InfraRed Hygrometer (FIRH) optical paths. The thick grey lines denote the inlet tubing of the dew point mirror (DPM) hygrometer. Adapted from Niedermeier et al. (2020).

wavelength-modulation instead of amplitude modulation, and to apply 1f or 2f detection? That would also eliminate the problem with the window signal, I suppose.

We did not modulate the laser light intensity with the laser current because manipulating the current introduces changes in wavelength and, in consequence, in absorption cross section. Our measurement strategy accounts for the dependence of absorption cross section on water vapor concentration (sec. 3.1.) due to self-broadening which would be more challenging to achieve with wavelength modulation applied.

We applied slow wavelength variation when analysing the influence of the windows (sec. 3.2.). The amplitude modulation applied in actual humidity measurements served for the purpose of reducing signal noise. We consider 1f or 2f detection as a direction of possible improvements of our setup where the goal is to measure humidity fluctuations in the presence of cloud droplets.

5. Line 192: Are the two windows here the two opposite windows in the setup, i.e. in LACIS-T?

   Yes. We clarified this sentence.

6. Caption Figure 5: The assumed concentration given here is the water vapor concentration in LACIS-T?

It is one example value of water vapor concentration in LACIS-T selected from the range of n considered in this study (see Table 1). We used such water vapor concentration in experiments COMP-L and COMP-S.

7. Line 200: I understand that the windows were large, so any antireflection coating or tilting would not work. But the laser spot is small, so not the whole window should be tilted or coated.

   We agree that a fixed coating of certain spots on the glass windows would work. However, it would reduce the scanning flexibility to these spots and thus the universal purpose of the wind tunnel. Furthermore, it should be mentioned that FIRH is not a fixed instrument at LACIS-T, i.e., the wind tunnel windows are not customized for this particular instrument and the associated wavelengths. FIRH was used during a measurement campaign to evaluate the properties of FIRH and to characterize the humidity field and turbulent fluctuations of humidity inside LACIS-T.

   Following the experience related to the influence of interference in the windows gained in the course of this study, an alternative method of reducing interference fringes by wavelength modulation was designed by Winkowski and Stacewicz (2021). We intend to apply this method in future experiments with FIRH.

8. Line 215: What does "perpendicular orientation" here mean?

   Perpendicular to the one which is discussed in the previous sentences and shown in Fig. 6, i.e. across the short dimension of the wind tunnel. We clarified this in the text.

9. Line 220: The effects of reflection are discussed. Would such a reflection not worsen the laser efficiency when coming back to the active material of the laser? Or is this somehow avoided?

   This effect is not relevant for our setup because there is no coupling between the reflected beam and the fiber. We used single mode fibers and the lens couplers with a very small angle of acceptance. Then the coupling is not possible without special adjustments.

10. Line 230: Why is the parasitic absorption so different for the x and y directions?

    Absorption $\mathcal{A} = 1 - \mathcal{T}$ depends on cross section σ, concentration n and optical path L. Parasitic absorption is the same for the two directions in terms of absolute values because optical path outside the wind tunnel is $L_l = 5.0 \pm 0.3$ cm for both directions. However, in line 230 we consider parasitic absorption in relation to the absorption inside the wind tunnel which is larger for longer optical path inside (80 cm vs 20 cm). We rephrased this sentence to avoid confusing the value of parasitic absorption with the ratio of parasitic absorption to the absorption in the wind tunnel.

11. Line 242. Please consider providing the formula (maybe in the Appendix). It could be interesting for the readers or other researchers with similar applications.

    As explained in sec. 4, we neglected the dependence $\sigma(n)$. Then Eq. (5) provides a direct formula for n:

    $$n = \frac{1}{(\sigma_M - \sigma_R)\,L}\left[\ln\left(\frac{I_1(\lambda_M)\mathcal{T}^{(g)}(\lambda_M)}{I_2(\lambda_M)}\frac{I_2(\lambda_R)}{\mathcal{T}^{(g)}(\lambda_R)I_1(\lambda_R)}\right) - (\sigma_M - \sigma_R)\,n_l L_l\right].$$

We used a common linearized approximation based on total derivative for a function of many variables $n = n(x_1, \ldots, x_i, \ldots, x_m)$

$$\Delta n \approx \sum_i \left| \frac{\partial n}{\partial x_i} \right| \Delta x_i$$

which applied to the above formula and assuming

$$\begin{aligned}
\frac{\Delta I}{I} &= \frac{\Delta I_1(\lambda_M)}{I_1(\lambda_M)} = \frac{\Delta I_2(\lambda_M)}{I_2(\lambda_M)} = \frac{\Delta I_1(\lambda_R)}{I_1(\lambda_R)} = \frac{\Delta I_2(\lambda_R)}{I_2(\lambda_R)} \\
\frac{\Delta \sigma}{\sigma} &= \frac{\Delta \sigma_M}{\sigma_M} = \frac{\Delta \sigma_R}{\sigma_M} \\
\sigma_M &\gg \sigma_R
\end{aligned}$$

can be simplified to a form

$$\Delta n = \left(2\frac{\Delta \sigma}{\sigma} + \frac{\Delta L}{L} \right) n + \frac{1}{\sigma_M L} \left(4\frac{\Delta I}{I} + \frac{\Delta \mathcal{T}^{(g)}(\lambda_M)}{\mathcal{T}^{(g)}(\lambda_M)} + \frac{\Delta \mathcal{T}^{(g)}(\lambda_R)}{\mathcal{T}^{(g)}(\lambda_R)} \right) + n_l \frac{L_l}{L} \left(\frac{\Delta n_l}{n_l} + \frac{\Delta L_l}{L_l} \right)$$

where as σ_M we plugged a fixed value corresponding to a typical water vapor concentration in LACIS-T ($5 \cdot 10^{17}\ \mathrm{cm}^{-3}$). The term proportional to n in the r.h.s. is what we called relative error, the remaining part of the r.h.s. was called absolute error.

The term related to water vapor concentration in the lab can be analogously obtained from Eq. (1) and Clausius-Clapeyron equation:

$$\frac{\Delta n_l}{n_l} = \frac{\Delta T_l}{T_l} + \frac{M_v L_v}{R T_l} \frac{\Delta T_{dl}}{T_{dl}}.$$

where M_v is molar mass of water, L_v is latent heat of vaporization.

We shortly explained the above method in sec. 4., however refrained from presenting the entire derivation as it is pretty straightforward once the method is known.

12. Line 254: Here the measurement was conducted with two air streams. If I understood correctly, the former measurements were carried out without flow. The measurement conditions should be described correctly and at the beginning of the paragraphs. Here it is also not clear how the sampling for the dew point mirror was done. Or was the inlet permanently in LACIS-T, as shown in Figure 1?

    The measurements of window transmission described in sec. 3.2 were carried out without the flow. It is specified in line 203. The subsequent measurement series were COMP-L (formerly called COMP-X, see point 2 in this response) and COMP-S (formerly called COMP-Y) in which indeed the two air streams were used, however with the same velocity, temperature and humidity in both. The dew point mirror inlet was permanently inside LACIS-T as in Fig. 1, however its horizontal position was changed so that it is always beneath (i.e. downstream of) the optical path of FIRH. This was specified in lines 93 and 270. We clarified the description of DPM sampling in sec. 2.1 and 4 as well as of the measurement conditions in sec. 4.

13. End of Section 4: For me the explicit determination of the detection limit or the minimum detectable concentration of FIRH is missing. From the calibration it could be determined, right? Something like 1.5 E17 cm-3.

    We preferred not to specify the exact detection limit as it depends on the optical path length and such estimation would need to rely on the dew-point mirror measurements. Instead, we provide a range of water vapor concentration $1.0\ldots6.1\cdot10^{17}\ \mathrm{cm}^{-3}$ for which we verified the agreement between FIRH and DPM.

14. Line 283: Again the question: was the DPM inlet permanently mounted? Or was that movable? One could perform a scan with DPM if its inlet is movable.

    The DPM inlet was permanently inside LACIS-T but its position was changed in accordance with FIRH so that the inlet was always beneath (i.e. downstream of) the optical path. This was specified in line 93. The scans were performed simultaneously with the two instruments FIRH and DPM. The positions of both were adjusted manually which might have caused some inaccuracies.

15. Line 286: That FIRH measurements represent an average along the optical path is not a new information, it is mentioned a few lines earlier.

    We removed the repetition.

16. Line 295: "The profiles of n ..." – was already mentioned.

    We removed the repetition.

17. Line 317: Is it possible to measure the air flow and get information about the velocity profile? Applying an LDV, for instance?

    The air-flow was measured independently with a Hot-Wire anemometer and the results are given in Niedermeier et al. (2020) as already stated in the text. LDV would also be possible, however, it requires the insertion of seed particles as tracers. Currently, particle insertion is only possible via the aerosol inlet. This would mean that these velocity measurements would be limited to the locations where the particles are. Those particles might also influence the humidity field (through water adsorption/absorption) which we want to avoid here.

18. Line 333, 335: Vibration and oscillation of the window are the same thing, if I understand correctly. Why were the windows vibrating? Some mechanical vibration from the whole facility?

    Yes, we refined to one term: vibrations. The windows vibrate to a minor extent due to the mechanical vibrations of the whole facility. For example, thermostats are used for the adjustment of the air-flow temperature. These thermostats cause vibrations that are damped by the design of the wind tunnel, but are still transferred to the measurement section and thus to the glass windows.

19. Figure 12: The inlet figure has no scale, so it is difficult to understand it.

    We added the scale to the insert in Fig. 12.

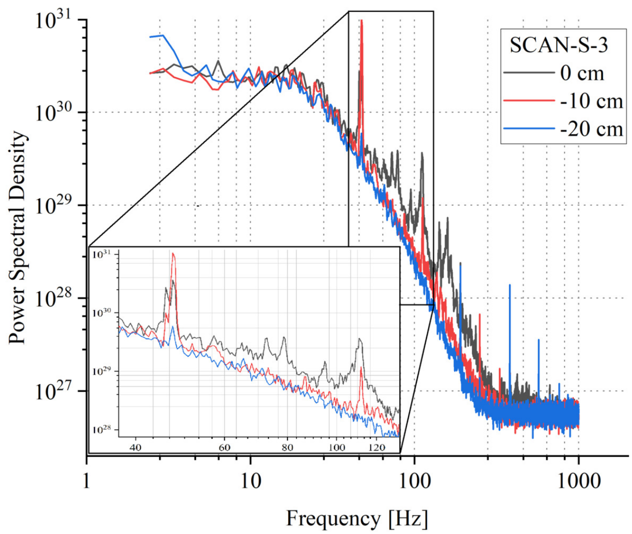

Figure 12 corrected. Power spectral densities of the timeseries $n(t)$ recorded at various positions y during SCAN-S-3. The insert shows the peaks in the spectrum described in the text.

20. Lines 372-375: I did not understand the motivation of this discussion. Are the results meaningful in this aspect or not?

    We are confident with the conclusions given in lines 372-375. We suppose the Reviewer might have meant lines 375-379 which indeed contained some discussion that, we agree, is unnecessary in this section. We removed those sentences.

21. Line 380: It is claimed here that the measurements "provided new insights into the properties of turbulence and turbulent mixing in LACIS-T". This is not obvious for me and that is what I meant in my General remarks.

    We removed this sentence as superfluous. The next one explains what we meant by new insights, namely that the results on humidity fluctuations complement the previous characterizations of turbulent velocity and temperature fields from Niedermeier et al. (2020). Following the general remarks given by the Reviewer, we included a new paragraph at the beginning of sec. 5. to clarify the usage of these investigations and also rearranged sec. 6.

22. Data availability: I suggest the authors using a data repository for publishing the data, at least the ones corresponding to the figures.

    We prepared a dataset corresponding to the figures and will reference it in the final version of the manuscript.

References

Niedermeier, D., Voigtländer, J., Schmalfuß, S., Busch, D., Schumacher, J., Shaw, R. A., and Stratmann, F.: Characterization and first results from LACIS-T: A moist-air wind tunnel to study aerosol-cloud-turbulence interactions, Atmospheric Measurement Techniques, 13, 2015–2033, https://doi.org/10.5194/AMT-13-2015-2020, 2020.

Szakáll, M., Mohácsi, Á., Tátrai, D., Szabó, A., Huszár, H., Ajtai, T., Szabó, G., and Bozóki, Z.: Twenty Years of Airborne Water Vapor and Total Water Measurements of a Diode Laser Based Photoacoustic Instruments, Frontiers in Physics, 8, 384, https://doi.org/10.3389/FPHY.2020.00384/BIBTEX, 2020.

Tátrai, D., Bozóki, Z., Smit, H., Rolf, C., Spelten, N., Krämer, M., Filges, A., Gerbig, C., Gulyás, G., and Szabó, G.: Dual-channel photoacoustic hygrometer for airborne measurements: Background, calibration, laboratory and in-flight intercomparison tests, Atmospheric Measurement Techniques, 8, 33–42, https://doi.org/10.5194/AMT-8-33-2015, 2015.

Winkowski, M. and Stacewicz, T.: Optical interference suppression using wavelength modulation, Optics Communications, 480, 126 464, https://doi.org/10.1016/J.OPTCOM.2020.126464, 2021.