Authors’ Response to the Anonymous Referee #2

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We are grateful to the Referee #2 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.

Comments

1. In the description of the LACIS-T facility (Figure 1 and supporting text), it would be helpful to clarify the geometry by showing axes x, y and z and indicate the reference positions for each (z = 0 being the tip of the aerosol inlet, and x = 0 and y = 0 being the centerlines of the two transverse dimensions of the duct) and define the longitudinal (z) position of the FIRH sampling and discuss why that position was chosen (I see in L142 “at the height 39 cm downstream [of] the place where the two streams merge”—meaning z = 39 cm downstream of the aerosol inlet?). It would also help to mention that the duct is oriented vertically (it is, right?) so that describing the position of the DPM sampling inlet as “below” (= displaced in z, downstream of) the FIRH beam makes sense (I was originally picturing the inlet offset in y for x “scans” and in x for y “scans”). In Figure 1, I’m not sure what information I am supposed to extract from the picture/diagram to the right of the two (x and y view) schematics of the LACIS-T measurement section, and it seems like it should be omitted or discussed.

   We agree with the suggestions and corrected the description of the facility and the measurement setup accordingly. We added the information about the vertical orientation of the measurement section of LACIS-T in sec. 2.1. We clarified the geometry by adding axes in Fig. 1. and expanding its caption.

   Following also the remarks of Referee #1, we refined our terminology to "sampling across long/short dimension" 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:

   > The sampling of the air inside LACIS-T was achieved across the glass windows at the height $z = 39$ cm, i.e. downstream of the aerosol inlet where z is the longitudinal position with $z = 0$ being the tip of the aerosol inlet, see Fig. 1. This height was selected because previous measurements related to cloud formation studies were perfomed at the same position by Niedermeier et al. (2020). The emitter and the photodetector PD2 were mounted on a rigid aluminium sleigh at the opposite sides of the wind tunnel (see Fig. 3) as close to the glass windows as it was possible (while maintaining the flexibility of easy changes of the scanning position) in

order to minimize the optical path outside the wind tunnel. Nevertheless, even despite drying the ambient air in the laboratory, parasitic absorption could not be entirely avoided (see sec. 3.3). The sleigh enables scanning the spatial variability of humidity statistics by moving the sensor horizontally along the walls of the wind tunnel. Two separate sleighs were prepared to allow measurement at both transverse orientations: across the long ($L_L = 80 \pm 0.3$ cm) and short ($L_S = 20 \pm 0.3$ cm) dimensions of the rectangular measurement section of LACIS-T, denoted hereafter with letters L and S, respectively. The sampling across the long dimension was possible at the positions $x = -3.25 \ldots 2.75$ cm due to the thickness of the window frame. In the case of the sampling across the short dimension, the positions $y = 0, -10, -20$ cm were selected in this study. The coordinates x and y denote two transverse dimensions with the origin of the coordinate system located in the center of the measurement section as shown in Fig. 3.

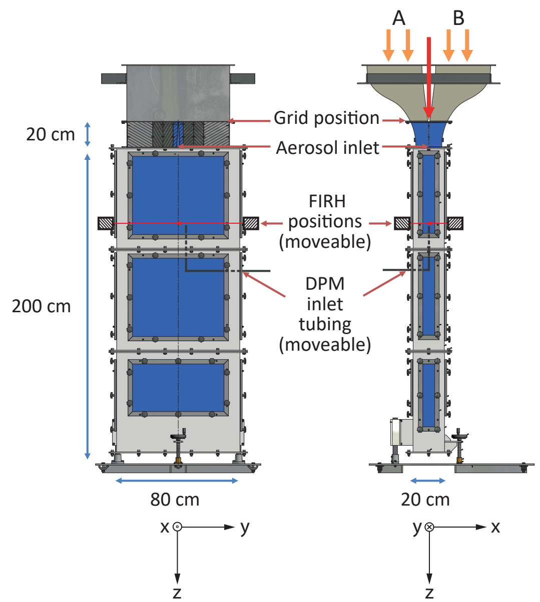

Figure 1 corrected. Schematic of the measurement section of LACIS-T. A and B mark the two air streams which are mixed in the measurement section. The red arrow marks the location where aerosol particles can be injected. Axes are included in order to display the geometry where $z = 0$ is the tip of the 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).

2. L111: "the exact tuning…prevents interferences" isn't quite correct. If one or both of the wavelengths were near an absorption line from another molecule, the measurement would be impacted regardless of how exact (precise) the tuning. It is the choice of the specific H2O absorption feature that is sufficiently far from interfering absorption lines that is important.

We agree. We meant that the exact tuning in addition to the proper choice of the absorption feature ensures selectivity of the measurement. We specified this in the text.

3. L148: “transverse” might be a better word than “perpendicular”, and specify relative to the direction of flow.

   We agree and changed the word.

4. L149 (and Figure 3): similar to above, it would be helpful to have the origin of the coordinates defined and the range of possible values (-x … +x, -y … +y).

   The origin is in the center of a measurement section which is indicated in Fig. 3. The ranges of the used x and y positions were added to the text. They are also listed in Table 1.

5. L151: since the two wavelengths are achieved by adjusting only the laser diode current, why are the two measurements made in separate (long) periods instead of quasi-simultaneously by rapid tuning between the two?

   Currently, our in-house developed software does not feature rapid tuning between the two wavelengths with desired accuracy and precision which are important for determination of the exact absorption cross sections. We consider wavelength modulation between the two extremes as a direction of possible improvements of our setup where the goal is to measure humidity fluctuations in the presence of cloud droplets.

6. Figure 5: I think “interferred” is meant to be “interfered”, but that is not used as an adjective. I think the appropriate term would be “convolved” as the product of the convolution of the absorption and fringe spectra.

   The label in the figure was changed into "convolved".

7. Figure 6: it would be nice to add lines indicating the locations of M and R.

   The lines indicating λ_M and λ_R were added to the figure.

8. L218 (and L265): why is the value of Tx(g)(lambda) = 0.87 so much lower than Ty = 0.98?

   The net transmission depends on the exact thickness of the glass and the exact distance between the two windows in relation to the wavelength which is of the order of 1 μm only. As discussed in lines 192-197, the range of possible values can be $0.748 \leq \mathcal{T}_2 \leq 1$. All the four derived $\mathcal{T}^{(g)}$ fit into this range. It is rather a coincidence that $\mathcal{T}_x^{(g)}(\lambda_M)$, $\mathcal{T}_y^{(g)}(\lambda_M)$, $\mathcal{T}_y^{(g)}(\lambda_R)$ are so close to 1 while $\mathcal{T}_x^{(g)}(\lambda_R)$ is not.

9. L264: the systematically high values of n from FIRH would indicate that the determinations of the contributions to the signal from the windows and ambient air were low when applied to the experimental arrangement. Or the DPM was systematically low at low H2O. Did the DPM-measured value agree with the expected based on the generated H2O in the flows?

   It is difficult to discern the true reason for this systematic difference between the instruments at low humidities. Indeed, if we consider the DPM as ground truth, then the window transmission term $\ln\left(\frac{\mathcal{T}^{(g)}(\lambda_M)}{\mathcal{T}^{(g)}(\lambda_R)}\right)$ is too high or/and ambient air absorption term $(\sigma_M - \sigma_R)n_l L_l$ is too low (see Eq. (5)) but also other factors may play a role, such as considerable uncertainties of absorption cross sections σ_M, σ_R or optical path lengths L, L_l. Following the experience gained in this study, we intend to further minimize parasitic absorption and window interference in future applications.

For the experiments COMP-L and COMP-S (formerly called COMP-X and COMP-Y), both airflows A and B were conditioned identically, i.e., they had the same T_d and it was changed from measurement to measurement identically in both streams. However, there is a difference in the method of humidity control between $T_d > 0°C$ and $T_d < 0°C$ (Niedermeier et al., 2020). For $T_d > 0°C$, the airflows were led completely through the Nafion humidifiers, so the generated H2O amount is known very precisely. This is further monitored with two additional DPMs (not described in this study, one for each stream A and B). Generated H2O amount agrees well with the DPM used in this study located further downstream inside the measurement section. For $T_d < 0°C$, the desired humidity is achieved by mixing dry and humidified air. For each stream, the airflow is split into two parts. One part is left dry while the other part is led through the Nafion humidifier. Then, those two parts are mixed. The resulting water vapor amount can be calculated knowing T_d of the dry and humidified air as well as the flow rates of those two parts (being left dry and being humidified). Such calculated T_d is checked by the two monitoring DPMs. However, as the T_d of the dry part is not exactly constant, we observe a slight variability in the resulting T_d of the mixed airflow.

10. Section 5.1: I'm not sure that the arguments presented here really presents a complete argument explaining the systematically higher values measured by the DPM than FIRH (L284). The values are typically at a mean concentration (> 2e17) at which the prior experiments demonstrated good agreement, and anyway, at low values the prior experiments would predict that FIRH would be higher than the DPM measurement. Spatial differences (average vs point) would seem to require a crossover at some position since gradients along the y direction cannot explain it given the statement in L276. Given conservation of H2O in the flow, the small difference in z of the FIRH and DPM measurements shouldn't produce a significant difference (would require a source of H2O).

    In the experiments of the type SCAN (sec. 5.), the conditions were significantly different than in the earlier experiments of the type COMP (sec. 4.). During COMPs, temperature and humidity was equal in the two streams A and B. During SCANs, temperature and humidity differed between the streams which allowed for the observation of the mixing profiles presented in Fig. 8.

    We are not certain what is the main reason of the systematic discrepancy between FIRH and DPM observed in SCANs. We suppose all 3 factors originally given in the text can contribute. The second point was expanded to mention also possible angular misalignment of the FIRH optical path with respect to the desired $x = const$, $z = const$ plane. The third point was reformulated to mention the effective filtering related to the two sampling regimes relevant for FIRH and DPM. FIRH involves spatial filtering but provides high temporal resolution. DPM collects air from a relatively small, yet finite volume. The diameter of the inlet tube is 6 mm which is quite significant size in relation to the gradient of the order of 10^{17} cm^{-4} and the range of investigated x positions. Moreover, due to the particular measurement method, which requires cooling a mirror to a dew-point temperature, in the environment of rapid humidity fluctuations DPM acts as a low-pass filter with rather complex and potentially non-intuitive transfer properties. The corrected discussion reads:

> The mean n exhibits a significant systematic offset (shift) between FIRH and DPM in all four experiments. Several factors could contribute to the observed offset: (1) the limited accuracy of FIRH (see sec. 4), (2) displacements and misalignments between the FIRH optical path and the DPM inlet (i.e. inaccuracy in setting x position, angular deviation of the FIRH path from the desired direction in the plane $x = const, z = const$, deliberate shift in z between the sensors), (3) difference in sampling regime between the instruments (in fact FIRH involves spatial low-pass filtering, i.e. averaging along the optical path but provides high temporal resolution while DPM involves temporal low-pass filtering of complex characteristics but collects air from a relatively small volume). The offset is higher than observed in the comparison experiments COMP-L and COMP-S, likely due to the significant spatial gradient of humidity (up to $2 \cdot 10^{17}\ \mathrm{cm}^{-4}$). Such gradient was absent in those comparison experiments but here, due to the factors (2) and (3), it affects the outcome.

In future applications of FIRH, we intend to minimize the influence of window transmission and parasitic absorption which would reduce the number of factors involved and possibly allow to discern the main factor responsible for the discrepancies.

11. Figure 8: it would be interesting to compare the variance with dn/dx to graphically demonstrate the statement in L296/7 of the coincidence of the peak in variance with the steepness of the gradient.

    Calculated gradient dn/dx is shown in the modified Fig. 8.

12. L315: the hypothesis here could have been tested by comparing with an experiment including a flow (aerosol-free) from the aerosol inlet with n = (nA + nB)/2 that would be more representative of the typical aerosol-inclusive studies at LACIS-T.

    We agree with this idea. Unfortunately, at the time of the experiments we were not aware of such a strong importance of aerosol flow on humidity field but quantified this issue in the course of later data analysis.

13. Data availability: per the AMT data policy, it is (at the least) encouraged that authors make the supporting data publicly available via some repository.

    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.

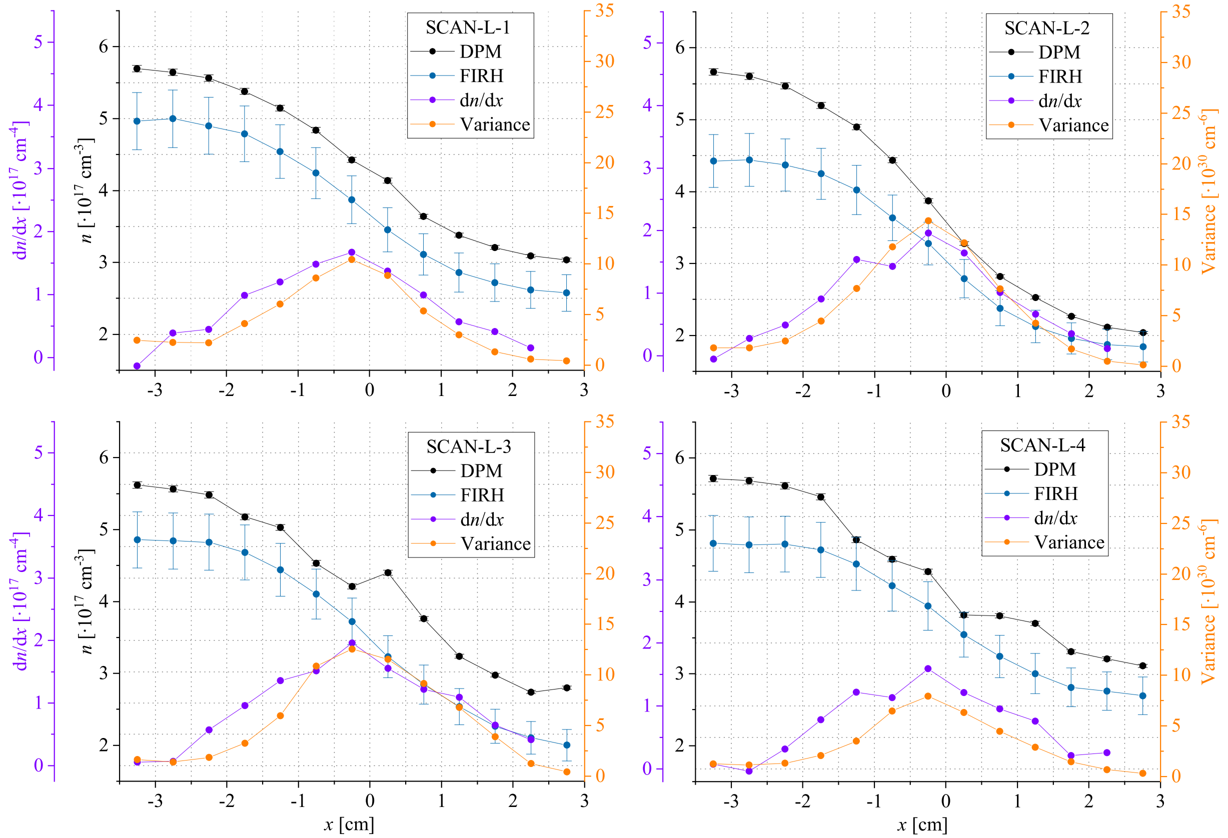

Figure 8 corrected. Turbulent mixing of the two streams differing in thermodynamic properties (given in Table 1) observed in the course of the four experiments: the profiles of mean n, its variance and gradient dn/dx with respect to the position x.