Reply to Referee #2 for A New Technique to Retrieve Aerosol Vertical Profiles Using Micropulse Lidar and Ground-based Aerosol Measurements

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We thank the reviewers for their time and constructive comments. Below, we reproduce each comment followed by our response. Page numbers and line numbers follow the original submitted manuscript, with new page numbers and line numbers included in parentheses.

RC2: 'Comment on egusphere-2024-3363', Anonymous Referee #2, 08 Jul 2025

Summary: This paper uses ground-based in situ observations of CCN and aerosol concentrations in combination with lidar profiles to create vertical profiles of CCN, aerosol, and INP concentrations. The focus of the analysis is correcting the lidar backscatter profiles observed at varying levels of RH to dry backscatter profiles to avoid the disconnect between optical properties and aerosol concentrations at high RH. The paper is well-written and the methods seem thorough. However, the paper is missing more discussion on the other factors that can cause optics to not be linearly correlated with aerosol concentrations, as well as a discussion of the implications of the time averaging done to retrieve aerosol/CCN concentrations from a single backscatter profile.

Authors' response: We appreciate the Reviewer's perspective. It was not our intent for the manuscript to be overly focused on RH corrections. However, that correction process became quite detailed and lengthy. Based on this comment and a similar comment from Reviewer 1, we have now expanded the description of other factors, including near-ground uncertainties in lidar signal and time averaging of lidar data. Please see our response to Reviewer 1 for additional details.

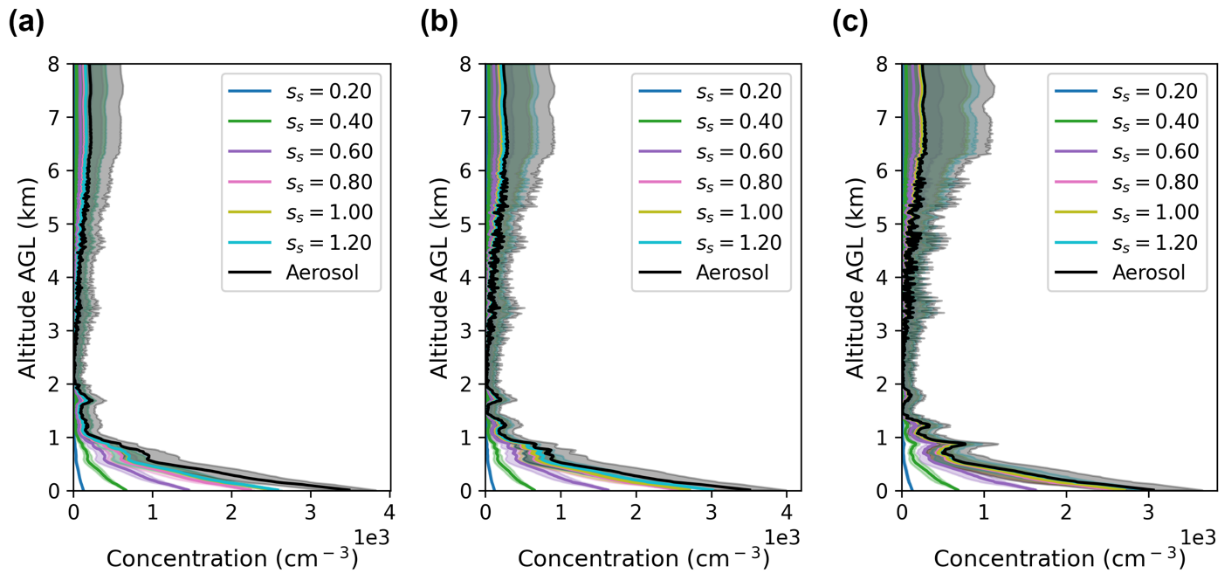
Major Comments:

Referee's comment: If I'm understanding correctly (Line 194-196), all cloud-free profiles from 2-4 hours of lidar observations are averaged into a singular backscatter profile used to retrieve the aerosol/CCN/INP profiles. More discussion of the implications of this step would strengthen the analysis, because it seems like a long time to average over.

Authors' response: Thank you for this comment. In reviewing the original text, we see that it contains an error. It should have said all cloud-free profiles were averaged for 1-3 hours rather than 2-4.

There are several reasons why 1-3 hours of total averaging time are used. First, the derivation of the CCN and INP vertical profiles requires ground-based measurements of aerosol size, concentration, CCN, and INP activity. During TRACER, the CCN instruments only completed a full supersaturation scan about every 30 minutes, and the INP analysis was performed on hourly impactor samples.

To demonstrate the sensitivity of our retrieval method to the time averaging window, we use the figure below to compare the aerosol and CCN concentration profile collected on 28 August using three averaging windows—2 hr 40 min, 1 hr, and 30 min—each centered on the 17:28 UTC radiosonde launch. The three averaging windows comprise 131, 55, and 29 individual lidar profiles, respectively. The overall shape and magnitude of aerosol concentration profiles remain largely consistent across all three averaging windows. However, small differences still appear, reflecting short-term variability in the aerosol field. The 30-minute average (Figure 1c) is also noticeably noisier and has more uncertainties at higher altitudes.



Comparison of aerosol concentration profile among three time-averaging periods for the example case on August 28 2022 (a) 2 hr 40 min averaging time (b) 1 hr averaging time (c) 30 minute averaging.

The figure and the following texts have been added to the supporting information.

Supporting Information, page 5, line 72 now reads: "To demonstrate the sensitivity of our retrieval method to the time averaging window, we compare the aerosol and CCN concentration profile collected on 28 August using three averaging windows 2 hr 40 min, 1 hr, and 30 min—each centered on the 17:28 UTC radiosonde launch. The three averaging windows comprise 131, 55, and 29 individual lidar profiles, respectively. The overall shape and magnitude of aerosol concentration profiles remain largely consistent across all three averaging windows. However, small differences still appear, reflecting short-term variability in the aerosol field. The 30-minute average (Figure 1c) is also noticeably noisier and has more uncertainties at higher altitudes."

The following changes have been made to the main text:

Page 9, line 195 (now page 10, line 203) now reads: "The NRB profiles of cloud-free columns, typically between about 0.5 to 1.5 hours before and after the radiosonde launch time (for a total of about one to three hours), are time-averaged."

Page 15, line 312 (now page 16, line 328) now reads: “The aerosol, CCN, and INP measurements were averaged over the same one to three hours window as the lidar data used for backscatter profile retrieval. This averaging period reflects the operational constraints of each instrument: the CCN counter requires approximately 30 minutes to complete a full scan of supersaturations, and INP samples were collected over 1-2 hour periods (Thompson, 2025).”

Referee’s comment: On average, how many profiles are included in a 2-4 hour average?

Authors’ response: The TAMU miniMPL records raw data at 1 min. However, on average, approximately one-third of the profiles were removed due to the presence of clouds. Thus, 40-120 individual profiles are included for 1-3 hours averaging period.

Referee’s comment: Is homogeneity of the aerosol profile over this time scale in this region a good assumption?

Authors’ response: Yes. In each case, we first examined the lidar attenuated backscatter time series (at 1-minute resolution) to identify time periods with relatively consistent aerosol structures. In the original text, these time series were shown in figures 8a, 9a, and 10a in the manuscript. For emphasis, we have now added the discussion below:

Page 24, line 508 (now page 24, line 502) now reads: “The NRB timeseries in **Error! Reference source not found.**a shows a persistent layer of high backscatter, visible below approximately 1 km. In addition, intermittent layers of high backscatter are observed between 1 and 3 km.”

Page 21, line 454 (now page 27, line 538) now reads: “The NRB time series in **Error! Reference source not found.**a shows limited temporal variation in attenuated backscatter profiles during the cloud-free period.”

Page 29, line 485 (now page 29, line 575) now reads: "The NRB time series in **Error! Reference source not found.**a shows some temporal variation in the attenuated backscatter profile, with a layer of high backscatter slowly decreasing from around 2 km to 1 km."

Referee's comment: How does the standard deviation associated with time averaging compare to the backscatter uncertainty already shown on the figures?

Authors' response: The uncertainty associated with time averaging is comparable to the backscatter uncertainty of the backscatter coefficient profile, as shown in the figure below, where we use the interquartile range to represent the uncertainty.

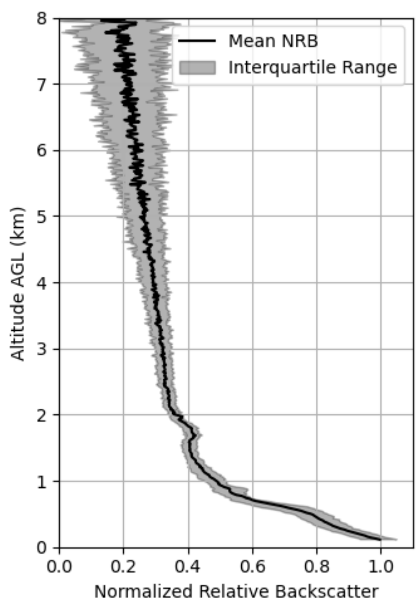


Figure S3 Mean (solid black line) and interquartile range (gray area) of NRB profiles for the example case on August 28, 2022.

The figure and discussion below are included in Section 4 of the Supporting Information.

Supporting Information, page 4, line 61 now reads: "We assess the uncertainty associated with temporal variability of attenuated backscatter by calculating the interquartile range (25th–75th percentile) of the 1-minute NRB profiles within the averaging window. For the example case (in the Method section) on August 28th shown in the figure above, the relative magnitude of temporal variability in NRB is comparable to the retrieval uncertainty presented in Figure 4b of the main manuscript. Because range-dependent noise grows with height, the temporal average of NRB shows greater variability aloft; the backscatter coefficient profile retrieval steps apply smoothing to reduce this noise, so the retrieved backscatter profile has lower uncertainty at those heights.

Reviewer comment:
In lines 569-574, there is discussion of the spatial heterogeneity of aerosol vertical profiles between Galveston and LaPorte, which are only 46km apart. Given the potential for air mass transport over tens of kilometers during a 2-4 hour time frame, the assumption of temporal heterogeneity may lead to some vertical features being washed out. A quantitative assessment of the temporal variability would enhance confidence in the resultant aerosol profile retrievals.

Authors' response: Some features may indeed vary on the time scale of our analysis, but a more detailed quantitative assessment of variability is difficult given the time resolution of the ground-based measurements. Nevertheless, aerosol vertical profiles remained distinctly different between Galveston and La Porte, even after 2 h 40 min of temporal averaging, as shown in Figure 12 in the main text.

Referee's comment: I would recommend replacing "accurate" with "realistic" in Line 585 and elsewhere in the paper, because "accurate" implies validation against independent observations, which is not presented here. "Realistic" better reflects that the profiles follow expected physical behavior. Additionally, additional discussion of the scenarios in which this method will not work would strengthen the final section. It is alluded to earlier in the paper (Line 409) that the approach only holds for well-mixed aerosol layers, but I think this needs to be reiterated here. As soon as the aerosol chemical composition or size distribution differs from what is measured at the surface, there will be errors introduced as the exact relationship between aerosol/CCN concentration and optical properties changes. It may also be beneficial (if you have a number) to discuss how often such well-mixed aerosol layer cases for which this method holds were observed in the TRACER campaign.

Authors' response: We thank the reviewer for the thoughtful suggestions. We agree that "realistic" is a more appropriate term than "accurate" and have changed the wording everywhere in the text. We note that the discussions of the method's limitations due to the assumption of the well-mixed aerosol column are included in the Method section 2.5 and the fourth paragraph of the Conclusions.

Reply to specific comments:

Reviewer's comment 1: Line 30: Can you clarify why the focus here is on convective processes? Assuming because it's mostly what is observed in the Houston area, but this makes it sound like aerosols don't also impact stratocumulus cloud processes.

Authors' response: The referee is right to point out that the non-convective clouds could be included in the introduction. We initially focused on the convective clouds since it is the focus of the TRACER project. However, aerosols affect both convective and non-convective clouds.

On Page 2, line 29 (now Page 2, line 28), the text now reads: "Consequently, changes in aerosol concentrations could influence many properties and processes of both convective and non-convective clouds (Tao et al., 2012; Fan et al., 2016; Twohy et al., 2005)."

Reviewer's comment 2: Line 81: "multiwavelength" not capitalized

Authors' response: Thank you. We have corrected this.

Reviewer's comment 3: Line 85: "relationship between"

Authors' response: Corrected.

Reviewer's comment 4: Line 105: Clarify what exactly "dry" ambient conditions mean for this case

Authors' response: The literature we cited here assumes ambient conditions of less than 50% relative humidity are dry. For clarity, this has been added to the text.

Page 4, line 105 (now page 4, line 105) now reads: "Their results show that CCN concentration at 0.3% supersaturation in dry ambient conditions (where RH ≤ 50%) strongly correlates with the HSRL-2 measured extinction and backscatter."

Reviewer's comment 5: Line 145: "first" not capitalized

Authors' response: Corrected.

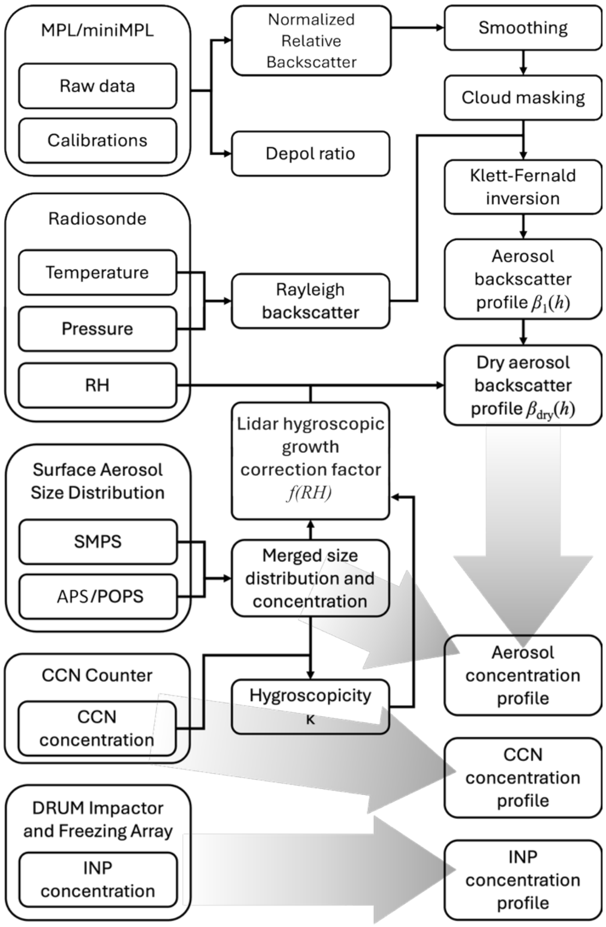
Reviewer's comment 6: Line 145: What defines an enhanced operation day? Is it dependent on atmospheric conditions or just a pre-determined day to deploy additional platforms?

Authors' response: Enhanced operation days were defined from field-campaign forecasts indicating a strong sea breeze and atmospheric conditions favorable for isolated deep convection. On these days, the TRACER team launched additional radiosondes. In hindsight, this terminology was unclear to readers, so we have removed the term "enhanced operation day" and instead describe the forecast criteria explicitly.

Page 6, line 145 (now page 6, line 145) now reads: "During the IOP, when forecasts indicated a strong sea breeze and conditions favorable for isolated deep convection, the TAMU ROAM-V was deployed at Seawolf Park in Galveston, Texas, and at several inland sites (Rapp et al., 2024)."

Reviewer's comment 7: Fig 2: Can you write out what NRB stands for? Since I don't think it's a super common acronym, this could be confusing for someone scrolling through the figures first before reading.

Authors' response: Good point. NRB stands for Normalized Relative Backscatter. Figure 2 in the manuscript has been updated as shown below:



Overview of the aerosol, CCN, and INP profile retrieval routine.

Reviewer's comment 8: Line 245-249: I am assuming this is expected in this region due to a primarily polluted urban boundary layer but maybe state it a little more explicitly if this is what you're implying.

Authors' response: This section of the manuscript aims to convey that the total backscatter coefficient above 2 km closely matches the Rayleigh backscatter from air molecules, indicating that the atmosphere at these altitudes is relatively clean with minimal aerosol presence. This agreement also serves as a quality check for the Fernald retrieval, as the Rayleigh backscatter is calculated independently and not retrieved. Matching profiles suggest that the retrieval is well constrained and performed correctly.

Page 12, line 244 (now page 13, line 255) now adds: "The total backscatter coefficient profile closely follows the molecular (Rayleigh) backscatter profile above 2 km AGL, indicating that aerosol contributions are minimal at these altitudes and that the backscatter is dominated by scattering from air molecules. This consistency also suggests that the Fernald inversion is performing well, since the molecular backscatter is independently calculated and provides a reference baseline."

Reviewer's comment 9: Fig 6: Can you make panel (a) wider? The numbers on the top really run together and are difficult to read

Authors' response: Thank you for this comment. We have edited the figure and updated the figure description to clarify the constant *κ* lines.

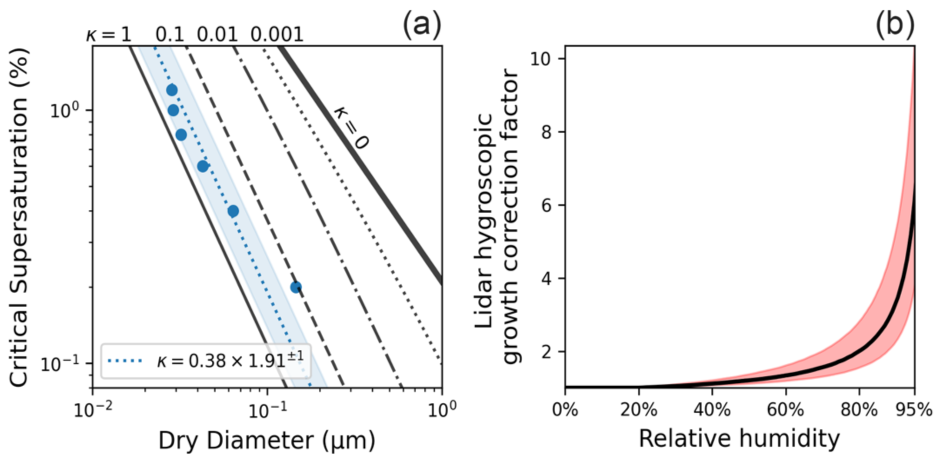


Figure 1 (a) Blue scattered points represent pairs of critical supersaturation and corresponding critical dry diameter derived from aerosol size distribution and CCN measurements. The blue dotted line represents the derived geometric mean of aerosol hygroscopicity κ, and the shaded region represents the one geometric standard deviation of κ. κ = 1 line is shown in solid black line; κ = 0.1 is shown in dashed line; κ = 0.01 line is shown in dash-dotted line; κ = 0.001 line is shown in dotted line; and κ = 0 is shown in thick solid black line. (b) Lidar hygroscopic growth correction factor as a function of relative humidity. The shaded area represents the uncertainties of derived κ.

Reviewer's comment 10: Line 379: How does ammonium sulfate compare to/represent the aerosol types typically observed during TRACER? Somewhere when you're describing the campaign can you discuss the typical/dominant aerosol types observed during the campaign?

Authors' response: While the exact refractive index of ambient aerosol during TRACER is not fully characterized in this study, we assume that of ammonium sulfate and compute a volume-weighted average with water at each RH. This choice has limited influence on the hygroscopic scattering enhancement correction, which depends on the relative increase in scattering with RH rather than the absolute backscatter coefficient.

Page 18, line 379 (now page 19, line 405) now reads: "In the absence of detailed aerosol composition data, the refractive index of ammonium sulfate is frequently adopted as a representative value in aerosol optical calculations, as it provides a reasonable approximation for non-absorbing, hygroscopic particles (Zieger et al., 2013; Ghan and Collins, 2004). In reality, sulfate, nitrate, organic aerosols, soot, and soil dust aerosols are all frequently observed in Houston in varying proportions depending on air mass origin (Thompson, 2025; Lei et al., 2025)."

Reviewer's comment 11: Should there be a section in here somewhere (even if brief) about the radiosonde observations and the uncertainty of their temperature/RH profiles?

Authors' response: We agree. Radiosonde information is shown in table 1 in the manuscript. The iMet-4 radiosonde reports temperature uncertainty of ±1.0 °C, and relative humidity uncertainty of ±5%.

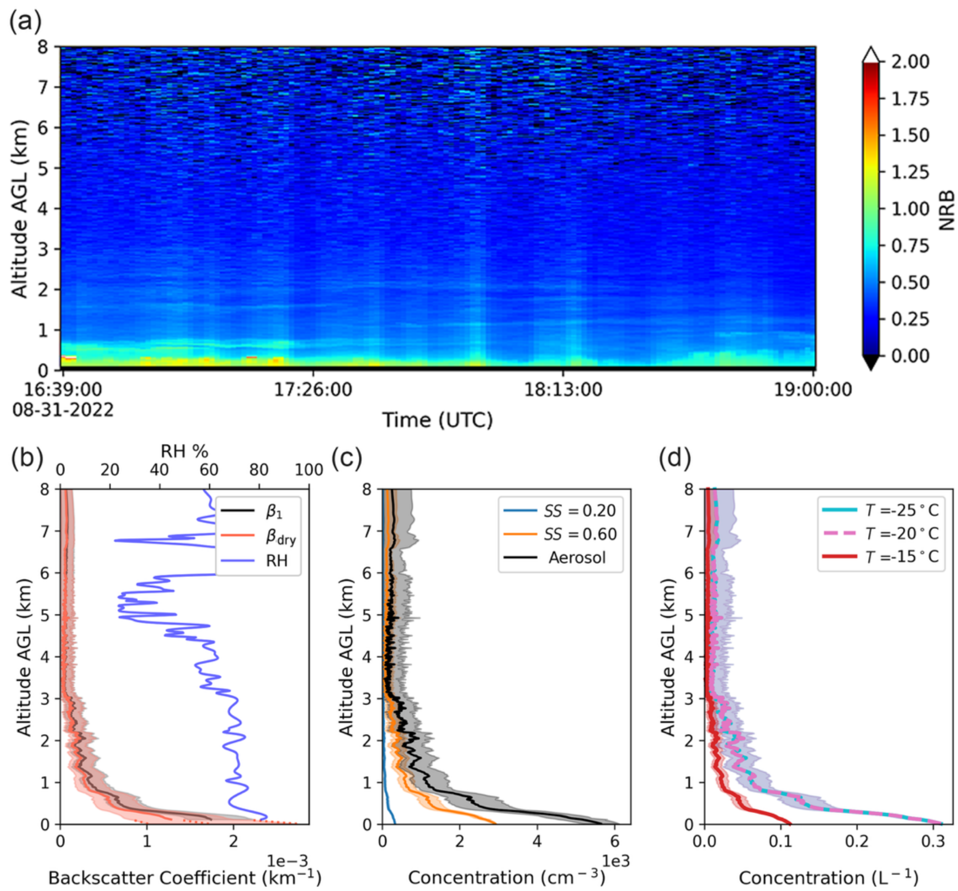
Page 19, line 417 (now page 21, line 450) now reads: "The radiosonde has a relatively small uncertainty in relative humidity measurements, specified as ± 5%."

Reviewer's comment 12: Fig 7: Would a log-scale for the x-axis be helpful? It's hard to see what's going on near the y-axis with all the lines really close together.

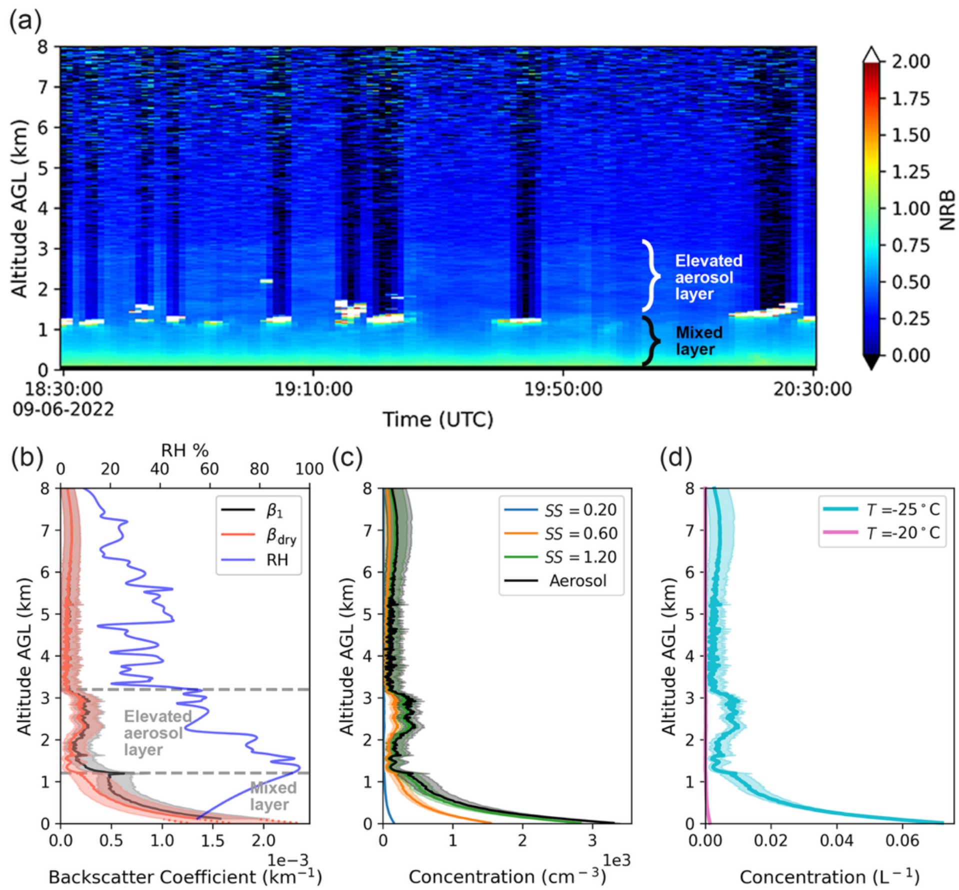
Authors' response: We appreciate the reviewer's suggestion to use a log-scale for the x-axis in Figure 7. However, we chose a linear scale to maintain consistency and interpretability across the different panels, which include both backscatter coefficient and aerosol concentration. A log-scale would compress higher values while overemphasizing minor differences at lower values.

Reviewer's comment 13: Fig 8: Could panels b, c, and d be put below panel a so all are larger and easier to read?

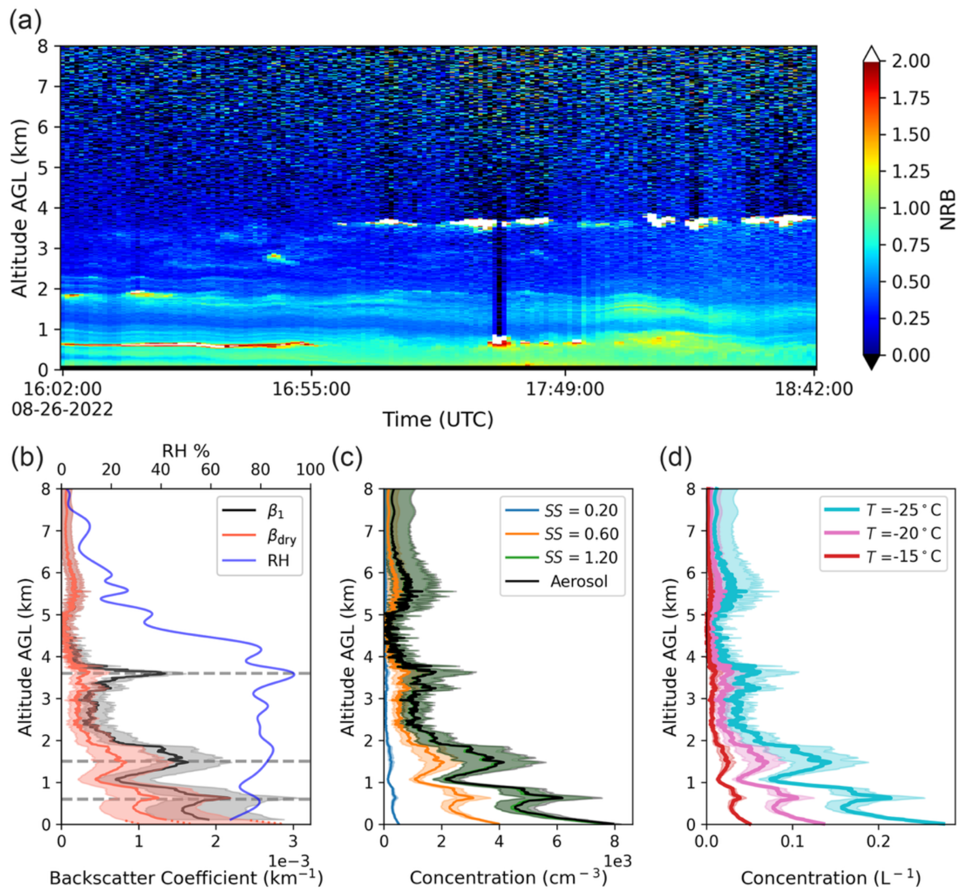
Authors' response: We appreciate the feedback. the figure 8, 9 and 10 are edited and shown below. The original section 3.3 has moved to 3.1 based on later comments.



Updated Figure 8 in the manuscript.



Updated Figure 9 in the manuscript.



Updated Figure 10 in the manuscript.

Reviewer's comment 14: Lines 454, 469-471: Previously, you mention this approach only holds for well-mixed layers, but here you are using an example with an elevated aerosol layer. I would mention that this does not impact the main point of this section, which is demonstrating the hygroscopic growth correction. Either that, or maybe don't even address the elevated aerosol layer or only do it at the end? Right now, it feels like you're picking a bad case right out of the gate immediately after finishing the methodology.

Authors' response: We thank the referee for this suggestion. We agree that it is clearer to show a baseline case first with a simpler atmospheric vertical structure. We have moved the 3.3 case study section to the 3.1 case study section and added further discussion of the results as comment 16 suggests.

Page 21, line 454 (now page 26, line 536) now adds: “Although an elevated aerosol layer exists, it does not affect the correction for the enhanced scattering from hygroscopic growth in the mixed layer.”

Reviewer’s comment 15: Fig 9: Is it worth addressing the multiple aerosol layers here? This case also doesn’t appear super well-mixed to me.

Authors’ response: We appreciate the reviewer’s observation regarding the presence of multiple aerosol layers and the degree of vertical mixing in Figure 9. We have added clarifying statements in the revised manuscript to address these points. Specifically, we now note that enhanced backscatter near cloud levels may indicate distinct aerosol layers or a high humidity layer, and that the dry backscatter profile suggests a possible elevated aerosol layer around 1.5 km. We also acknowledge that this feature may reduce the accuracy of retrieved aerosol properties at that level, while the removal of the humidity-induced peak at 3.6 km supports the robustness of our correction method.

Page 22, line 485 (now page 28, line 574) now adds: “The high attenuated backscatter signal near the cloud levels may reflect the presence of distinct aerosol layers or result from higher humidity enhancing aerosol scattering.”

Page 23, line 491 (now page 29, line 584) now adds: “The dry backscatter profile suggests that an elevated aerosol layer may be present near 1.5 km, while the uncorrected peak at 3.6 km is likely dominated by humidity-enhanced scattering rather than a distinct aerosol layer.”

Page 23, line 497 (now page 29, line 594) now adds: “The retrieval of aerosol, CCN, and INP concentrations may be less reliable around 1.5 km due to the possible presence of an elevated aerosol layer. However, the successful removal of the humidity-enhanced scattering peak near 3.6 km is encouraging, suggesting that the applied κ value may be reasonable throughout the column.”

Reviewer's comment 16: Line 498: Could this be the first case study you show? It might be good to show the most straightforward application first to give a reader confidence in your method before you introduce complications of multiple cloud layers or more pronounced hygroscopic growth.

Authors' response: Yes, we agree, and we have moved case study 3 to case study 1. Previous lines 499-517 are now moved to current lines 492-521. Previous lines 472-497 are moved to current lines 522-560. Previous lines 472-497 are now moved to current lines 561-607.

Reviewer's comment 17: Line 518: The placement of this section between your aerosol profile results felt a little confusing – would it make more sense in the methods section maybe?

Authors' response: We appreciate the reviewer's suggestion regarding the placement of this section. However, we chose to keep it in the current location because the Methods section is reserved for describing the procedural details of the aerosol, CCN, and INP retrieval methods. This section serves as a necessary transition that sets up the subsequent comparison between the ARM and TAMU results.

References:

Fan, J., Wang, Y., Rosenfeld, D., and Liu, X.: Review of aerosol–cloud interactions: Mechanisms, significance, and challenges, Journal of the Atmospheric Sciences, 73, 4221-4252, https://doi.org/10.1175/JAS-D-16-0037.1, 2016.

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Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, Atmospheric Chemistry and Physics, 13, 10609-10631, https://doi.org/10.5194/acp-13-10609-2013, 2013.