Reply to the Reviewer of Manuscript EGUSphere-2025-2600:

We would like to sincerely thank the editor and reviewer for their time, effort, and thoughtful feedback on our manuscript. The reviewer comments are shown in **blue**, with the authors' responses shown in **black** and any edited manuscript language shown in ***italicized black font***.

This manuscript deals with the relationship between aerosols, meteorological conditions and the development of cloud droplets and their size distribution. The methodology is based on both aircraft observations and numerical parcel model with bin microphysics. The overall goal is to understand the role of aerosols (number and type), constrained by meteorology, on the droplet formation mechanisms. The authors argue that aerosol entrainment is an important mechanism to explain the broadening of the droplet spectra. Overall, I think the manuscript is deserving of publication, but there are some major and minor adjustments to be made. I will point some major comments here and list a few minor comments later.

Major Comments

1. There is a disconnect between what the manuscript seems to be by reading the title, abstract and introduction and what the manuscript actually is. When I started to read the manuscript I thought it was going to be about the invigoration hypothesis, which is extensively reviewed in the introduction. However, the results section dedicates a lot of text to the description of the aerosol properties and air mass backtrajectories. While interesting, it doesn't seem to fit what I expected from the title/abstract/introduction. The same could be said about the entrainment process, which seems to be a major focus of the study and is not mentioned in the title, for instance. I will give my suggestions together with my second comment below.

2. The origin of the issue listed above is possibly related to the many foci present in the manuscript:
I) There is discussion about air masses/backtrajectories
II) There is discussion on physicochemical properties of aerosols, with vertical profiles
III) Only then the manuscript goes into the discussion that I was expecting to see from the start, which is the droplet spectra properties
IV) Compounding the many-foci issue, there is also a lot to cover from the observations side as well as from the modeling side. Therefore, there is also multiple foci on the methodology side.

With the given above, the manuscript ended up being, in my opinion, too long and without a clear message. I would suggest the authors rewrite the manuscript in a way to make their contribution more explicit. From reading the manuscript, I think the most interesting aspect was the comparison between the observations and the bin model with the entrainment discussion. Because the manuscript had so many foci, this discussion ended up being shorter and shallower than expected. For instance, the authors mention that the entrainment effect is important to reproduce the observed droplet spectra width. However, there is no figure showing this parameter explicitly. There is plenty of literature about the aerosol effect on droplet width, which could be used as context to the present study.

To summarize and provide a more objective suggestion:
I would suggest refocusing the title/abstract/introduction towards this entrainment effect and the observation-model comparison. The aerosol/backtrajectories analysis, while interesting, could be left out without hurting the overall message of the manuscript. This part could be later incorporated in a new submission focused on the aerosol discussion, in my opinion.

We sincerely thank the reviewer for these insightful points.

A primary objective of this manuscript is to provide a detailed characterization of aerosol properties and meteorological conditions during the DCMEX campaign, and how these factors influence the formation and development of cloud droplets in deep convective systems. Another publication (Daily et al., 2025) from the DCMEX project focuses on the characterization of ice-nucleating particles (INPs). The two studies establish the framework of aerosol characterization for DCMEX. In the revised manuscript, we therefore retain the discussion of aerosol physicochemical properties. Since the atmospheric sources and transport generate temporally varying aerosol properties in a given region, the source analysis using the dispersion model is needed to explain the observed temporal trends in aerosol characteristics. **To address the reviewer's question about a disconnect between the introduction and results, we have reorganized the introduction section:**
1) The introduction of invigoration hypothesis has been deleted.
2) We now emphasize that detailed characterization of aerosol amounts and properties is crucial for understanding the droplet formation and development in deep convective systems. Moreover, atmospheric sources and transport generate temporally varying aerosol types, amounts, and properties in a given region.
3) We have added the background on how entrainment processes and aerosol mixing affect the development of convective clouds.

We thank the reviewer for emphasizing the value of the comparison between the bin-microphysics parcel model and the observations, which underscores the importance of aerosol entrainment in the development of deep convective clouds. **In the revised manuscript, we have expanded the introduction and discussion of entrainment effects.** We agree that the extensive dataset obtained from individual flights offers valuable potential for developing general parameterizations that achieve improved agreement between simulations and observations. While this study focuses on case studies using a bin-microphysics parcel model to highlight the importance of incorporating entrainment, we are preparing a follow-up manuscript to conduct simulations for all flights during the DCMEX campaign. We will systematically investigate how different entrainment conditions (adiabatic, homogeneous, and inhomogeneous) affect not only cloud droplets but also secondary ice production. Regarding this, we have expanded the implications in the conclusion section.

Regarding the above discussions, we have revised the manuscript in four aspects:

1. We have re-organized the introduction to enhance the connection between the introduction and result sections. The revised introduction is:

"*The life cycle of deep convective clouds is modulated by complex microphysical processes, including droplet formation, droplet growth through condensation and coalescence, thermodynamic phase transitions between liquid droplets and ice crystals, and the development of precipitation (Arakawa, 2004). Aerosols play a key role in these processes, and aerosol-cloud interactions are considered among the largest uncertainties in estimating climate sensitivity to radiative forcing (Boucher et al., 2013; Forster et al., 2021). Aerosols can affect clouds by acting as cloud condensation nuclei (CCN) or ice nucleating particles (INP), which is termed aerosol indirect effects (Boucher et al., 2013). In the presence of aerosols or sufficient CCN, water vapor condenses onto CCN surfaces to form cloud droplets, marking the initial process in the lifecycle of convective clouds that mostly occurs at cloud base (Tao et al., 2012; Rosenfeld et al., 2014). Generally, higher CCN concentrations produce a greater number of smaller droplets and narrower droplet size distributions, which are likely to inhibit collision-coalescence and delay raindrop formation, thereby extending cloud lifetime (Rosenfeld, 2000; Tao et al., 2012). This delay can have opposing effects on convective cloud development: the increased condensational heat release*

tends to enhance cloud buoyancy and vertical development, while the resulting increase in condensate loadings partially offsets that buoyancy enhancement (Rosenfeld et al., 2008; Koren et al., 2014; Fan et al., 2018; Varble et al., 2023). However, the presence of giant CCN, such as coarse-mode sea salt aerosols, can produce initially large droplets and accelerate warm rain formation, thereby inhibiting the vertical development of convective clouds (Yin et al., 2024). These existing studies suggest that the CCN ability of aerosols determines the initial droplet number concentration and size distributions, thereby influencing subsequent cloud dynamics throughout convective cloud lifetime. Additionally, INPs can promote the heterogeneous freezing, and regulate ice crystal number concentrations during convective development (Tao et al., 2012). Detailed characterization of aerosol amounts and properties is therefore crucial for improving the representation of aerosol-cloud interactions in atmospheric models, in particular, aerosol size distribution and chemical composition which determine their CCN and INP ability (Petters and Kreidenweis, 2007). However, the representation of aerosol properties and associated indirect effects is complex and uncertain, as they are subject to atmospheric dynamic and thermodynamic conditions (Yang et al., 2020). Atmospheric transport generates temporally varying aerosol types, amounts, and properties in a given region (Yang et al., 2020). Moreover, cloud response to aerosol perturbations depends on environmental conditions, such as cloud-base temperature, updraft velocity, and humidity (Pruppacher and Klett, 2010). Previous studies have indicated that clouds with cool, high bases tend to exhibit little sensitivity in cloud-top height and precipitation to aerosol loadings, while clouds with warm, low bases display larger aerosol-induced changes (e.g. Li et al., 2011). Overall, it is critical to understand how varying aerosol properties influence convective cloud microphysics under different environmental conditions.

Another key uncertainty in understanding the development of convective clouds is the entrainment and mixing process. Theoretically, cloud-droplet growth in a closed (adiabatic) parcel leads to narrower size distributions as vertical development progresses, tending to suppress the onset of coalescence through differential gravitational sedimentation (Pruppacher and Klett, 2010). However, observations have revealed that cloud-droplet size distributions are relatively broader than those in ideal adiabatic parcels, and this result is usually attributed to a consequence of entrainment and mixing and secondary activation (Blyth, 1993; Chandrakar et al., 2016). Despite its recognized importance, the representation of the entrainment and mixing process in models remains uncertain. It is suggested that inhomogeneous mixing typically dominates when cloud droplets are small, as their evaporation rates significantly exceed the mixing rate of clouds with surrounding subsaturated air (Pruppacher and Klett, 2010). In this way, a subset of cloud droplets evaporates completely, leaving the others in the volume unchanged. When cloud droplets are larger and their evaporation rates are comparable to the mixing rate, homogeneous mixing dominates the system and the influence of inhomogeneous mixing weakens (Pruppacher and Klett, 2010). With homogeneous mixing, droplets evaporate by uniformly reducing their size across the population, leaving droplet number density largely unchanged except through simple dilution. While some studies suggest that entrainment and mixing in convective clouds are almost completely inhomogeneous (e.g. Burnet and Brenguier, 2007; Braga et al., 2017a), some other studies propose that inhomogeneous mixing may dominate early cloud development when droplets are small, and then homogeneous mixing may become more prevalent as convective clouds evolve (e.g. Lehmann et al., 2009). It is unclear whether the entrainment is predominantly homogeneous mixing, inhomogeneous mixing, or a combination of both. An improved understanding of how these mixing mechanisms dominate throughout the development of convective clouds is essential for improving the representation of their microphysical processes. Moreover, the consequence of entrainment processes on cloud microphysical evolution can be regulated by surrounding environmental conditions, such as relative humidity (RH) and aerosol characteristics (Koren et al., 2010). In particular, aerosols entrained with surrounding air can act as additional CCN and promote secondary activation of droplets above cloud base. Such additional activation has been identified as an important factor contributing to the broadening of droplet size distributions, toward small droplet diameters (Lehmann et al., 2009; Chandrakar et al., 2016). An

improved understanding of aerosol entrainment will reduce the current uncertainty in predicting droplet number concentrations and size distributions.

Overall, existing studies show that the impacts of aerosols, meteorological conditions, and entrainment processes on the formation and development of deep convective clouds are highly complex, which introduces large uncertainties in understanding the climate effects associated with deep convective systems. Advancing understanding in this area requires coordinated observational and modeling efforts across diverse convective regimes with different aerosol and meteorological conditions. To improve the representation of aerosol-cloud interactions and cloud microphysical processes in deep convective systems, the Deep Convective Microphysics Experiment (DCMEX, July to August 2022) was conducted over the Magdalena Mountains, New Mexico (local time = UTC – 6 hours). Isolated convective clouds can frequently form and develop over these mountains during the summer season in North America, providing ideal laboratory-like conditions for investigating deep convective processes. Earlier studies primarily characterized convective clouds and precipitation development in this area (i.e. Blyth and Latham, 1993). The DCMEX campaign provided coordinated and comprehensive observations of aerosols, cloud microphysics, and thermodynamics of the deep convective system over the Magdalena Mountains (Finney et al., 2024). The campaign benefits from significant advances in measurement techniques, including high-resolution cloud probes with improved particle sizing and phase discrimination, and state-of-the-art aerosol instruments capable of resolving physicochemical properties (Finney et al., 2024). In this study, we focus on the influence of aerosol properties, meteorological conditions and entrainment processes on the formation and development of cloud droplets during the campaign, combining observations with bin-microphysics parcel model simulations. This study aims to improve the representation of microphysical processes in this region and similar deep convective systems in future model studies."

2. We have also expanded the discussion of entrainment processes in the development of convective clouds. In the revised manuscript, we examined three entrainment scenarios, including purely homogeneous mixing, purely inhomogeneous mixing, and a hybrid approach of early-stage inhomogeneous mixing (from the cloud base up to ~ 1 km above) followed by homogeneous mixing. We also examined the effects of aerosol entrainment under the three entrainment scenarios. By comparing different scenarios, a combination of early-stage inhomogeneous mixing and following homogeneous mixing as well as the inclusion of aerosol entrainment would provide the best match with observations. The revised manuscript in Sect. 3.2 is:

"*Many observations and modeling studies have suggested that entrainment and mixing processes are important for the evolution of cloud microphysics (Burnet and Brenguier 2007; Lehmann et al., 2009; Braga et al., 2017a). In this study, three entrainment scenarios were examined, including purely homogeneous mixing (denoted as "Hom"), purely inhomogeneous mixing ("Inhom"), and a hybrid approach of early-stage inhomogeneous mixing (from the cloud base up to ~ 1 km above) followed by homogeneous mixing ("Inhom+Hom"). In the simulations, the entrainment rate of surrounding air was constrained by thermodynamic profiles derived from dropsonde measurements. The simulated entrainment rates under different entrainment scenarios are shown in Fig. S8. With the inclusion of entrainment processes, which considered the dilution of cloud layers by entrained subsaturated air and the associated evaporation of cloud droplets, the simulated N_d and LWC were substantially reduced throughout the cloud depth compared to adiabatic simulations. As seen in Figs. 7a and 7b, the inclusion of entrainment improved the agreement between simulated and observed LWC profiles (red, blue, and green solid lines). Simulated cloud-base N_d values were more consistent with observations under the "Inhom" and "Inhom+Hom" scenarios (blue and green solid lines in Figs. 7c and 7d), whereas they were overestimated under the "Hom" scenario (red solid lines in Figs. 7c and 7d). However, all three scenarios exhibited a pronounced decrease in N_d with vertical development, in contrast to the relatively constant N_d in observations. Simulated R_e values were substantially overestimated under the "Inhom" and "Inhom+Hom"*

scenarios (blue and green solid lines in Figs. 7e and 7f), due to the underestimated N_d, particularly at higher cloud levels. The "Hom" scenario produced R_e profiles closer to observations (red solid lines in Figs. 7e and 7f) but continued to predict excessively narrow droplet size distributions (red solid lines in Fig. 8). All three entrainment scenarios failed to reproduce small cloud droplets as vertical development progressed (red, blue, and green solid lines in Fig. 8).

Previous studies suggest that aerosols entrained with surrounding subsaturated air can be activated in the rising air parcel, broadening droplet size distributions toward small droplet diameters (Lehmann et al., 2009; Chandrakar et al., 2016). To account for this effect, aerosol entrainment (EA) was further incorporated into the simulations under three entrainment scenarios, denoted as "Hom+EA," "Inhom+EA," and "Inhom+Hom+EA" (red, blue, and green dashed lines respectively in Figs. 7 and 8). The entrained aerosols were assumed to follow the same size distributions as those measured near the cloud base (Fig. S7). For both cases, the further inclusion of aerosol entrainment made a negligible difference to the simulated LWC under three entrainment scenarios. In the Period 1 case (C300), incorporating aerosol entrainment improved the consistency of simulated N_d profiles with observations, capturing the slight decrease at low cloud layers followed by relatively constant values aloft. In particular, the "Inhom+EA" and "Inhom+Hom+EA" scenarios produced N_d values close to observations, whereas the "Hom+EA" scenario still overestimated N_d. Aerosol entrainment also broadened the droplet size distributions under three entrainment scenarios, effectively filling the deficit in small-droplet populations. Under the "Hom+EA" scenario, the overestimated N_d resulted in fewer large cloud droplets and slightly underestimated R_e, particularly at higher cloud levels. In contrast, the "Inhom+EA" scenario produced a higher concentration of large droplets relative to observations, as vertical development progressed. This is likely due to excessive collision-coalescence in the simulations, as inhomogeneous mixing promotes the evaporation of some droplets while leaving others unaffected, thereby enhancing droplet growth and shifting the droplet size distribution toward larger diameters (Pruppacher and Klett, 2010). These results suggest that homogeneous mixing should be incorporated as vertical development progresses. Overall, the "Inhom+Hom+EA" scenario provided the best agreement with observed N_d and cloud droplet size. In the Period 2 case (C303), aerosol entrainment presented the same effects on simulated LWC, N_d, and cloud droplet sizes as in the Period 1 case. However, the two cases exhibited slightly different sensitivities to aerosol entrainment under the "Inhom+Hom+EA" scenario. While the simulated N_d, R_e and droplet size distributions showed good agreement with observations in the Period 1 case, the R_e and large-size droplets were slightly underestimated in the Period 2 case due to the overestimated N_d as vertical development progressed. This discrepancy is likely due to different vertical profiles of aerosol concentrations and size distributions between the two cases. In the Period 1 case, N_a and aerosol size distributions remained relatively constant from the cloud base to above (Figs. S9a and b). In the Period 2 case, N_a decreased monotonically with height, and larger particles were less abundant above cloud base compared with measurements near cloud base (Figs. S9c and d). The assumption of constant aerosol size distributions during the entrainment process aligned well with the Period 1 case. However, it likely resulted in overestimated N_d in the Period 2 case, due to excessive entrained aerosols in simulations, which in turn led to underestimated R_e. Employing vertically resolved aerosol size distributions in the model would likely have little impact under Period 1 conditions but could improve the representation of droplet number and size evolution under Period 2 conditions. Additionally, this discrepancy is likely attributed to the parameterization of the initial radius of ascending cloud parcels in simulations (Table S3). The simulated entrainment rate is sensitive to the parcel radius, with smaller radius resulting in stronger entrainment and more rapid dilution. However, since the initial radius cannot be directly constrained by measurements, this also introduces uncertainty in simulations. In this study, simulations of cloud droplet formation and development based on different assumptions highlight the importance of incorporating the entrainment of surrounding subsaturated air, particularly aerosol entrainment. The incorporation of aerosol entrainment is crucial in reproducing the width of droplet size distributions. Although

our SMPS measurements did not provide continuous vertical profiles of aerosol size distributions due to limited time resolution, future studies should consider an approach of parameterizing fractional entrainment constrained by observed vertical profiles of aerosol number concentrations, to further improve model performance."

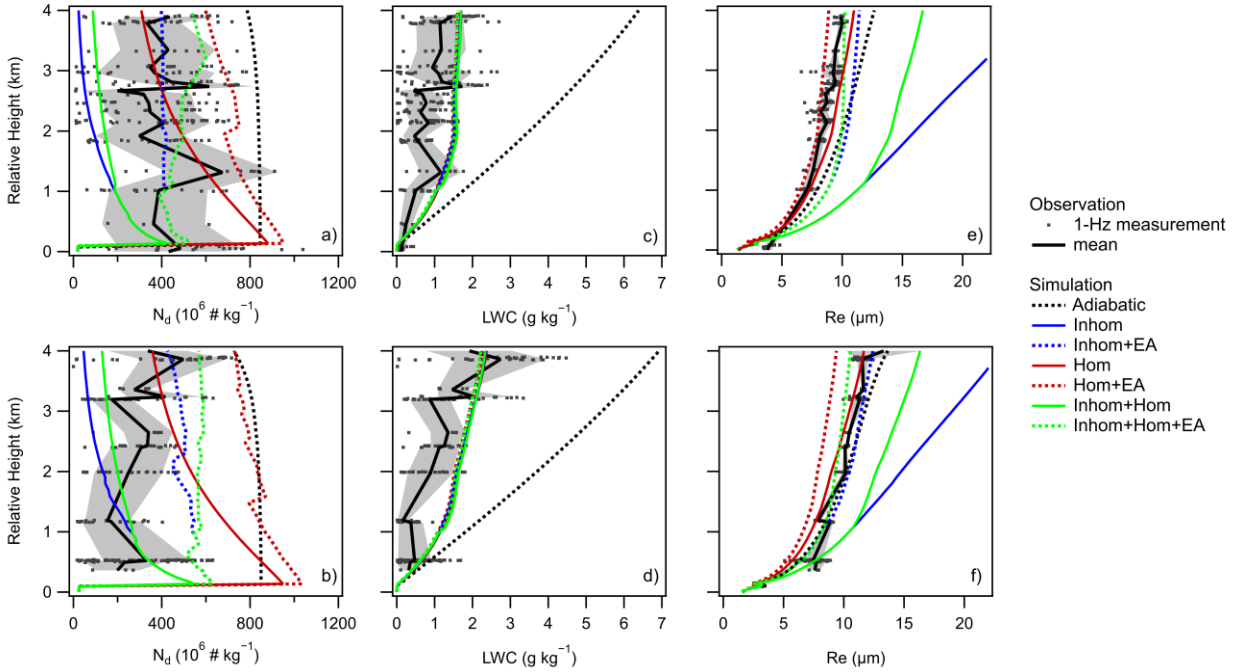

Figure 7. Vertical profiles of observed and simulated cloud droplet number concentration (N_d, kg^{-1}), LWC (g kg^{-1}) and effective radius (R_e, μm) for the Period 1-C300 case (upper plots) and the Period 2-C303 case (bottom plots). It is noted that, for consistency with the model, the units of N_d and LWC are expressed in "kg^{-1}", as opposed to "m^{-3}" in Sect. 3.3.1. The dots represent 1-hz measurements from the CDP. The solid black lines and shades represent average and standard deviation of observed values. The dashed black, pink and green lines represent simulations under different scenarios respectively. The y-axis is the relative height with respect to the LCL height. It is noted that the pink and green dashed lines overlap in the plots of LWC.

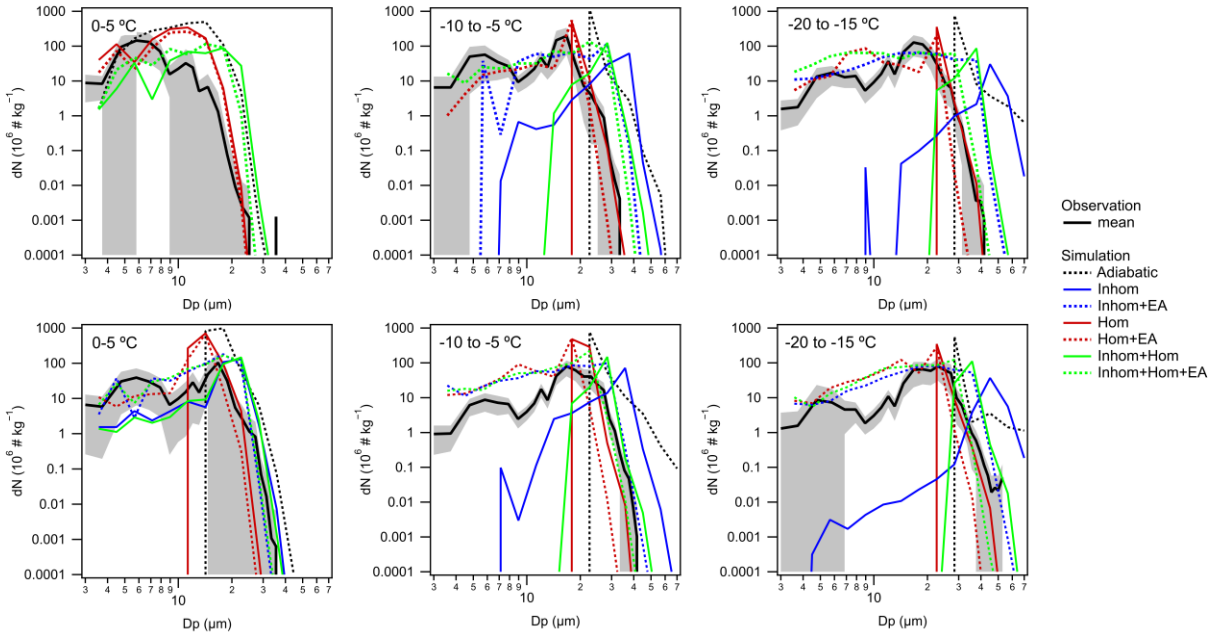

Figure 8. Observed and simulated cloud droplet size distributions for the Period 1-C300 case (upper plots) and the Period 2-C303 case (bottom plots), as a function of T ranging from near cloud base to high level. The solid black lines and shades represent the average and standard deviation of observed values within specific T ranges. The dashed black, pink, and green lines represent simulations under different scenarios, respectively.

3. We have added the entrainment rate (μ_j) to address the reviewer's comment of "no figure showing this entrainment parameter explicitly". We have added the exact handling and formulation of mixing with ambient air and secondary aerosol activation in the supplementary. The entrainment rate (μ_j) is the key parameter for describing the entrainment process. Vertical profiles of simulated μ_j are now shown in Fig. S8.

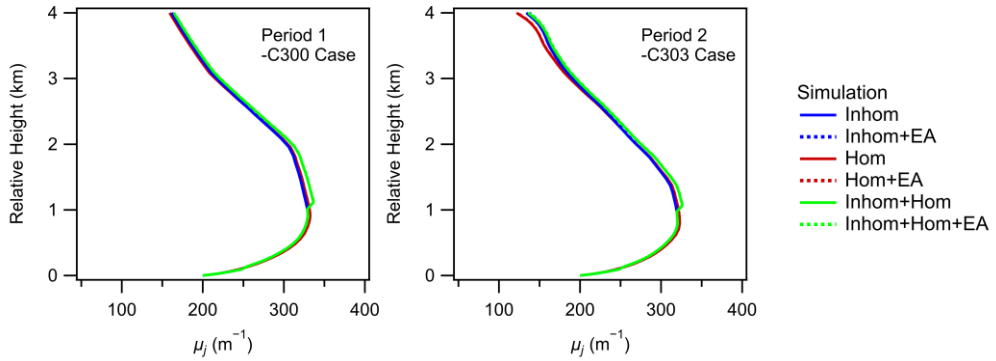

Figure S8. Figure S8. Vertical profiles of simulated μ_j (m^{-1}) for the Period 1-C300 case (left) and the Period 2-C303 case (right). The y-axis is the relative height with respect to the LCL height.

4. To expand the discussion of implications, the revised conclusion is:

"*This study provides a valuable dataset for improving parameterizations of aerosol-cloud interactions and offers critical constraints for the accurate representation of microphysical processes in this area and similar deep convective systems for future model studies. The comparison between observations and bin-microphysics parcel model simulations also highlights the importance of incorporating entrainment processes from surrounding environments, particularly aerosol mixing, in future simulations of cloud droplets in deep convective systems.* ***Future simulation studies should be conducted for all flight cases, to develop general parameterizations that achieve improved agreement between models and observations. In addition, Future studies should investigate how different entrainment conditions (adiabatic, homogeneous, and inhomogeneous) affect not only the development of cloud droplets but also ice production***."

Minor Comments

Lines 40-43: no references are given. Please provide appropriate references.

In the re-organized introduction, this sentence has been deleted.

Lines 44-45: there are more references to add here, most notably the IPCC reports

References have been added.

Aerosols play a key role in these processes, and aerosol-cloud interactions are considered among the largest uncertainties in estimating climate sensitivity to radiative forcing (Boucher et al., 2013; Forster et al., 2021).

Lines 54-55: again, no references given about the aerosol effect on cloud cover, lifetime, etc

The introduction has been rephrased, and references have been added.

Generally, higher CCN concentrations produce a greater number of smaller droplets and narrower droplet size distributions, which are likely to inhibit collision-coalescence and delay raindrop formation, thereby extending cloud lifetime (Rosenfeld, 2000; Tao et al., 2012).

Line 89: would it be nice to mention which advances in measurement techniques?

The added manuscript is:

The campaign benefits from significant advances in measurement techniques, including high-resolution cloud probes with improved particle sizing and phase discrimination, and state-of-the-art aerosol instruments capable of resolving physicochemical properties (Finney et al., 2024).

Line 111 and Line 114: please avoid using "etc"

The "etc" has been deleted. The revised manuscript is:

The FAAM Bae-146 was equipped with a suite of instruments to measure atmospheric dynamics and thermodynamics, including such as wind speed and direction, temperature (T), and RH.

Ground-based meteorological stations provided such as surface T, RH, pressure, and precipitation intensity.

Line 178: cloud development → cloud parcel development? (since it is a cloud parcel model)

Accepted

Line 210: remains → retains?

Accepted

Lines 292-294: no need to repeat the "(0.1 to 3 micron)" parenthesis, since N_a and N_{sa} were already defined earlier

The "(0.1 to 3 micron)" has been deleted.

Line 310: please consider citing "Isoprene nitrates drive new particle formation in Amazon's upper troposphere" by Curtius et al. (2024). It is a recent and relevant paper on the subject, available in Nature: https://www.nature.com/articles/s41586-024-08192-4. Also note that the aerosol-rich layer in the Amazonian upper troposphere is mostly above 8 km, which seems to be the upper limit in your study.

The revised manuscript is:

Recent studies over the Amazon have highlighted the important role of extremely low-volatility organic compounds (e.g. Isoprene-derived organonitrates) formed from the oxidation of biogenic volatile organic compounds (VOCs) in driving this NPF process in upper troposphere (Zhao et al., 2020; Curtius et al., 2024). The observed ultrafine-particle-rich layer in Amazon's upper troposphere is mostly above 8 km, which is near the upper limit of the flight altitude range in this study.

Figure 7 (and others): there should be legends in the figures themselves to explain what the different lines are. The figures should be as self-sufficient as possible, without needed to read the caption.

The legends have been added into Figures 7 and 8.

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