Responses to Reviewer #1

We thank the reviewer for taking the time to review our paper and for the constructive comments. The page and line numbers that we quote for indicating where we changed the manuscript refer to the revised marked-up version.

General note: During preparation of the revised manuscript, we identified a technical issue that required re-running all simulations. Specifically, vertical diffusion of computational particles was not correctly accounted for in the original simulations. All simulations have been rerun with this issue corrected. The revised results remain qualitatively consistent with those originally reported, and the main conclusions of the study are unchanged, although some quantitative differences appear in detailed vertical profiles.

(1.1) One issue in the presented analysis is its connection to aerosol-cloud interactions. Defining CCN properties at low relative humidity (RH) does not account for the continued condensation of semivolatile compounds at higher RH. A portion of these compounds would condense below 100% RH, thereby altering the CCN distribution. Furthermore, most nitric acid would condense onto particles prior to activation into cloud droplets, enhancing droplet formation. Including this effect could significantly change the number of cloud droplets formed. This could potentially even reverse the observation presented in this study. Therefore, using CCN as a proxy for cloud droplet concentration in scenarios with a strong contribution form semivolatile aerosol compounds may be somewhat misleading.

> We thank the reviewer for identifying this limitation with our analysis. In order to address the effects of co-condensation, we have conducted an additional set of simulations in which the relative humidity was raised to near saturation in the upper boundary layer. Our analysis is included in Section 3.8, indicating that higher RH meaningfully impacts aerosol composition and diagnosed CCN activity by enhancing the condensation of semi-volatile species such as ammonium and nitrate. We conclude that co-condensation lowers the sensitivity of CCN activity to emissions heterogeneity, however, meaningful impacts persist at intermediate supersaturation.
>
> We have included the following statement on lines 442–454, which acknowledges the limited applicability of WRF-PartMC's approach to computing CCN activity and calls for future work to further explore the quantitative contribution of co-condensation and emissions heterogeneity to aerosol aging and CCN activity:
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> "Our study of co-condensation and its effects reveals meaningful limitations in how WRF-PartMC-LES estimates CCN activity for particles under water-subsaturated conditions. Accordingly, CCN concentrations diagnosed under subsaturated conditions should be interpreted as an indicator of activation potential rather than a direct proxy for cloud droplet number, particularly in environments with substantial semivolatile mass. While our study reveals that emissions heterogeneity may alter aerosol composition and CCN activity even when co-condensation is considered, further work is required to place quantitative bounds on the contribution of each process and the modulating role of gas and aerosol composition. Furthermore, we wish to emphasize that we do not explicitly model CCN activation and droplet growth. Although previous particle-resolved studies have included this effect (Ching et al., 2012), cloud droplet growth is excluded from the current study due to computational cost. We expect that explicitly modeling CCN activation and subsequent droplet growth would meaningfully alter aqueous-phase chemistry within cloudy cells. Past studies have shown that in-cloud aqueous chemistry can result in the spatial segregation of reactive gas phase species, reducing oxidation of volatile organics (Li et al., 2017). We therefore hypothesize that cloud droplet aging is further coupled to emissions heterogeneity through aqueous-phase chemistry, and this coupling should be investigated in future work."

(1.2) What would happen if a Lagrangian perspective were adopted, assuming air masses advect over the emission source? With the modeling framework used here, this could have been explored by halting emissions from the point source partway through the simulation and allowing the emitted compounds to disperse within

the domain. Would the effects observed in Figure 6 and beyond be averaged out due to the reversible nature of nitrate partitioning? Such a setup would more closely reflect the assumptions made in low-resolution, large-scale models.

> We thank the reviewer for this recommendation. We have conducted an additional simulation in which emissions are cut off at $t = 4$ h to investigate the reversible partitioning of nitrate. Discussion of this scenario is included in Section 3.7. As expected, the most significant impact to aerosol composition is due to reversible partitioning of nitrate. The timescale at which the aerosol state relaxes back to the aerosol composition in the no heterogeneity scenario is therefore determined by the timescale of reversible partitioning for nitrate.
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> We find that the effects of emissions heterogeneity are progressively reduced after emissions cease, but are not instantaneously averaged out; instead, the relaxation timescale is governed by the timescale of nitrate repartitioning and boundary-layer mixing.

(1.3) Line 50: The statement "yet many climate models fail to resolve this variability adequately" could be clarified. Are there actually any climate models that attempt to account for subgrid-scale heterogeneity in a proper manner for the emissions?

> We have included the following statement on lines 49–53 to clarify the status of sub-grid scale parameterizations in global scale models:
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> "Much of the sub-grid scale variability arises from spatially heterogeneous emissions (Qian et al., 2010), yet most climate models do not explicitly represent this variability in a general sense. Existing parameterizations in global climate models that account for sub-grid scale emissions and associated processes are typically limited to specific phenomena, such as ship tracks (Huszar et al., 2010) and contrails (Burkhardt and Kärcher, 2009), rather than providing a general treatment of emissions-driven aerosol heterogeneity."

(1.4) Line 71–72: For precision, note that SALSA by default uses a 17-bin scheme (10 + 7), similar to how M7 employs 7 modes (4 + 3) to represent externally mixed aerosol populations with high and low hygroscopicities.

> Thank you for this point of clarification. We have revised discussion of SALSA on lines 73–74 to the following: "... UCLALES-SALSA employs a 17-bin sectional scheme to represent externally mixed aerosol populations with differing hygroscopicities, ..."

(1.5) Line 85: Typo: "It is extends"

> We have fixed this typo.

(1.6) Line 94: Typo: "by by"

> We have fixed this typo.

(1.7) Line 130: In large-eddy simulation (LES) studies, heat flux is more commonly expressed in W/m^2 rather than Km/s.

> Thank you for noting this convention. We have converted the surface heat flux to non-kinematic form and note its relevance to mid-latitude settings on lines 133–135: "...295.5 $W\,m^{-2}$, which is representative of surface heat fluxes resulting from annually-averaged mid-latitude solar insolation."

(1.8) Lines 137–140: The same sentence appears to be repeated. Please remove the duplicate.

We have removed the duplicate sentence.

(1.9) Figure 4: The concentrations of nitric acid and ammonia seem quite high. Are these values realistic, or do they represent an extreme scenario? A brief discussion on this would be needed.

We have added the following statement on lines 219–221 to clarify that the concentrations modeled in this study correspond to the upper end of observed concentrations in highly-polluted conditions:

"Comparing modeled ammonium and nitric acid levels with past studies, the concentrations used here correspond to the upper end of observed values reported during brief, extremely polluted episodes in regions with substantial vehicular emissions, such as Southern California (Salmon et al., 1990; Toro et al., 2024)."

(1.10) Figure 5: If new particle formation via nucleation is not included in the study, could this omission influence the results?

We have included the following statement on lines 465–477 to clarify the omission of nucleation in WRF-PartMC-LES:

"Lastly, nucleation is not modeled in WRF-PartMC-LES due to the large computational expense associated with explicitly representing the coagulation of large numbers of ultrafine computational particles. As a result, ultrafine particle concentrations may be underestimated in the present simulations. Importantly, nucleation itself is highly sensitive to spatial heterogeneity, as nucleation rates depend nonlinearly on precursor concentrations and local thermodynamic conditions. Spatial averaging over coarse grid cells can therefore suppress the peak conditions required for nucleation, leading to an underprediction of nucleation events even in models that nominally include this process. The inclusion of nucleation could influence the results shown here, particularly through its interaction with spatially heterogeneous emissions and gas-phase precursor fields. In highly heterogeneous scenarios, enhanced nucleation rates in localized regions may be partially offset by increased coagulation with larger particles, whereas in lower-heterogeneity environments reduced coagulation could allow a more pronounced ultrafine mode to persist. The net impact of nucleation on the size distribution and CCN activity therefore depends on the relative rates of nucleation, coagulation, and mixing, as well as on the degree of emissions heterogeneity. As a result, inclusion of nucleation could either dampen or amplify differences in CCN activity between low- and high-heterogeneity cases, and explicitly resolving these interactions remains an important topic for future work."

Responses to Reviewer #2

We thank the reviewer for taking the time to review our paper and for the constructive comments. The page and line numbers that we quote for indicating where we changed the manuscript refer to the revised marked-up version.

General note: During preparation of the revised manuscript, we identified a technical issue that required re-running all simulations. Specifically, vertical diffusion of computational particles was not correctly accounted for in the original simulations. All simulations have been rerun with this issue corrected. The revised results remain qualitatively consistent with those originally reported, and the main conclusions of the study are unchanged, although some quantitative differences appear in detailed vertical profiles.

(2.1) CCN can activate below 100% RH and if so, how would this impact your results? The paper focuses on CCN activation at low supersaturation values and it would be interesting to determine if CCN activation below 100% RH results in similar trends or produces a different outcome. Perhaps, since this study is highly idealized, simulations and analysis of such a situation is not needed. However, it could be useful to comment on this in the manuscript.

> We thank the reviewer for this question. In classical Köhler theory, CCN activation requires supersaturation with respect to water (RH $>$ 100%) and therefore does not occur below saturation. However, growth of particles under subsaturated conditions through hygroscopic water uptake and co-condensation of semi-volatile species can substantially modify particle size and composition prior to activation. This effect is explicitly examined in the additional high-RH simulations introduced in Section 3.8, which were added in response to Reviewer 1. We find that such pre-activation growth shifts CCN activity toward lower supersaturations and moderates, but does not eliminate, the influence of emissions spatial heterogeneity.
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> We have included the following statement in Section 4, lines 447–454 acknowledging our limited treatment of CCN activity and anticipated outcomes if explicit modeling of CCN activation at subsaturated conditions were incorporated:
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> "...Furthermore, we wish to emphasize that we do not explicitly model CCN activation and droplet growth. Although previous particle-resolved studies have included this effect (Ching et al., 2012), cloud droplet growth is excluded from the current study due to computational cost. We expect that inclusion of CCN which activate at subsaturated conditions would meaningfully alter the aqueous phase chemistry within cloudy cells. Past studies have shown that in-cloud aqueous chemistry can result in the spatial segregation of reactive gas phase species, reducing oxidation of volatile organics (Li et al., 2017). We therefore hypothesize that cloud droplet aging is further coupled to emissions heterogeneity through aqueous-phase chemistry, and this coupling should be investigated in future work."

(2.2) Could you use a profile where there's a more humid boundary layer? I think a boundary that would support cloud formation would be useful in that the results would show aerosol-cloud interactions in an environment that actually forms clouds. This would help support your explanations of what should occur given supersaturation conditions being realized in your environment, which don't actually happen based on the boundary layer profile used in the experiment.

> We thank the reviewer for this recommendation. Although our model is not able to explicitly represent CCN activation and cloud microphysics, we have conducted an additional set of simulations in which the relative humidity was raised to near saturation in the upper boundary layer. Our analysis is included in Section 3.8, indicating that higher RH meaningfully impacts aerosol composition and computed CCN activity by enhancing the condensation of semi-volatile species such as ammonium and nitrate. We conclude that co-condensation lowers the sensitivity of CCN activity to emissions heterogeneity, however, meaningful impacts are still found at intermediate

supersaturation. We have included an additional statement in Section 4, lines 442–447, which acknowledges the limited applicability of WRF-PartMC's approach to computing CCN activity and calls for future work to further explore the quantitative contribution of co-condensation and emissions heterogeneity to aerosol aging and CCN activity:

"Our study of co-condensation and its effects reveals meaningful limitations in how WRF-PartMC-LES estimates CCN activity for particles under water-subsaturated conditions. Accordingly, CCN concentrations diagnosed under subsaturated conditions should be interpreted as an indicator of activation potential rather than a direct proxy for cloud droplet number, particularly in environments with substantial semivolatile mass. While our study reveals that emissions heterogeneity may alter aerosol composition and CCN activity even when co-condensation is considered, further work is required to place quantitative bounds on the contribution of each process and the modulating role of gas and aerosol composition."

(2.3) For the high heterogeneity emission scenario, is there any value in having multiple point sources (plumes) and testing sensitivity to that configuration vs. one point source? I'm thinking that industrial regions often have more than just one smoke stack or concentrated emission point. Maybe this is better suited for future work, but worth considering.

We agree that more realistic emission patterns should be explored, and have included the following statement in Section 4, lines 424–430:

"Additionally, all emissions—both gas-phase and particulate—are temporally constant after spin-up and spatially collocated, reflecting a deliberate idealization used here to isolate the effects of emissions spatial heterogeneity. This configuration may not fully capture the complexity of real urban emission patterns where different sources (e.g., traffic, industry, biomass burning) are spatially and temporally decoupled. Future studies should investigate the impact of emissions heterogeneity on the aerosol state in response to realistic emission patterns such as numerous point sources with spatially segregated reactive species."

(2.4) Is there really a need to run the model at 100 m grid spacing for what this work is trying to demonstrate? There wasn't much of a discussion on the role of turbulence (only briefly mentioned in the introduction) which I would assume is important to address when running LES scale simulations. Another way of thinking of this is, what do we learn from the LES scale simulation that is not well represented or cannot be produced in a meso-scale simulation for this work? This is not clearly articulated in the explanation of the model setup or in the results.

We have included the following statement in Section 4, lines 455–464 to clarify that LES is required here to resolve emission heterogeneity at sub-kilometer scales, rather than to study turbulence itself.

"In this study, use of LES is primarily motivated by the length scale of heterogeneous emission patterns. The current work resolves emissions heterogeneity down to 100 m, which is well below the grid spacing of typical meso-scale and global models and allows explicit representation of sharp spatial gradients in gas and aerosol concentrations that would otherwise be averaged out. Given the idealized configuration employed here, the resolved turbulence primarily serves to provide realistic boundary-layer mixing under laterally homogeneous conditions, and its detailed structure is not a central focus of the present analysis. Nevertheless, turbulence is expected to play an important role in modulating the interaction between spatially segregated emissions, gas-phase chemistry, and aerosol aging in more realistic settings. Past studies have shown that turbulence facilitates the mixing of spatially segregated reactive gas species, and future applications of WRF-PartMC-LES should therefore investigate how turbulence modifies the rate and spatial structure of aerosol aging in domains with topography, heterogeneous surface forcing, and complex emission patterns."

(2.5) Following from question 4 above, emission flux data observations are not at 100 m resolution, correct? If so, how feasible is this framework for testing against observations for future implementation?

We have added the following discussion on lines 430–436 to clarify that although gridded emission inventories are often too coarse for LES, key source types such as point and line sources can already be represented accurately at sub-kilometer scales, while diffuse area sources require additional preprocessing and downscaling.

"Emission flux observations and inventories are not uniformly available at 100 m resolution. While many gridded inventories used in regional and global-scale modeling are too coarse for direct use in LES, several important source types can be represented accurately at sub-kilometer scales. Point sources (e.g., power plants, industrial stacks) and line sources (e.g., road networks) are often well constrained spatially and can be implemented directly in high-resolution simulations. In contrast, diffuse area sources that are typically reported at county or regional scales require additional emission preprocessing, downscaling, or data-fusion approaches to distribute fluxes at finer resolution. Developing such preprocessing workflows represents an important step toward applying WRF-PartMC-LES in observationally constrained, realistic settings."

(2.6) For some of the figures, for example Figure 7, it would be nice to give the readers a sense of the temporal variability of aerosol species mixing ratios instead of just at t=6 h. I wouldn't say this is a must, but it would be helpful to see.

We thank the reviewer for this recommendation. In response to this suggestion and another reviewer's recommendation to include simulations where emissions are turned off after a certain period of time, we have included Figure 13 (included here for convenience). This figure compares the evolution of aerosol composition for both the high heterogeneity scenario in which emissions continue unabated and a similar scenario but with emissions turned off at $t = 4$ h. Vertical profiles are plotted in hourly increments to illustrate the temporal evolution of the aerosol state.

(2.7) Typo line 85: "it is extends"

We have fixed this typo.

(2.8) Typo line 94: "by by"

We have fixed this typo.

(2.9) Repeated sentences around line 138

We have removed the duplicate sentence.

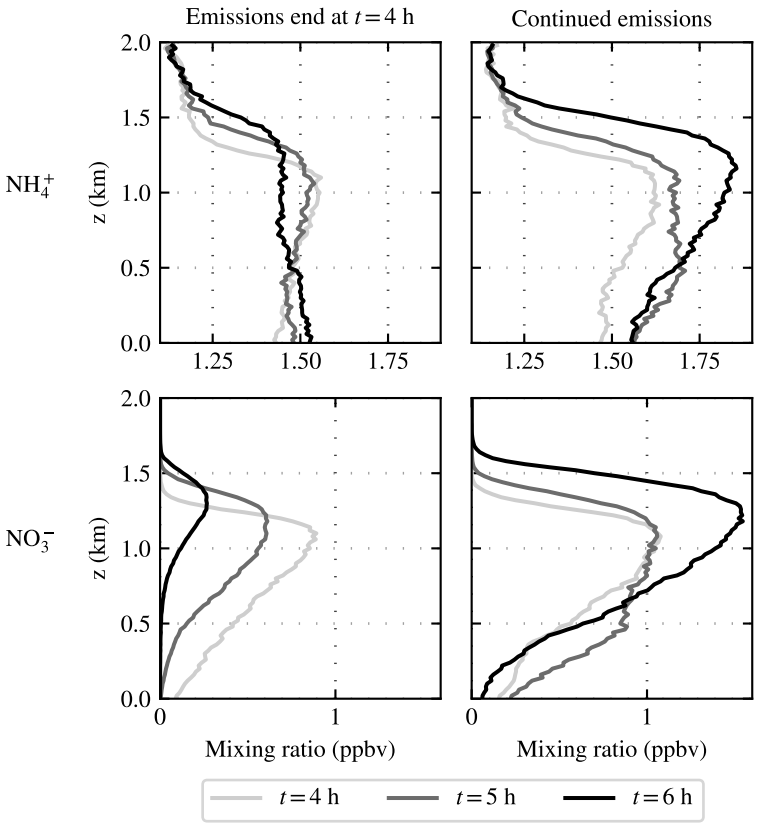

Figure 1: Figure 13 in Frederick et al., 2026: "Vertical profiles of ammonium (top row) and nitrate (bottom row) for two versions of the high heterogeneity scenario in which emissions are turned off at $t = 4$ h (left column) and emissions continue through the remainder of the simulation (right column)."