This study addresses the uncertainty in global climate models (GCMs) arising from challenges in representing spatially heterogeneous emissions and complex aerosol processes. Using particle-resolved large-eddy simulations with the WRF-PartMC-MOSAIC-LES framework, the authors investigate how spatial variability in emissions influences aerosol aging and cloud-relevant properties such as hygroscopicity and cloud condensation nuclei (CCN) activity. The results show that CCN concentrations at low supersaturations (0.1–0.3%) can increase by up to 25% in the upper planetary boundary layer under highly heterogeneous emission scenarios, primarily due to enhanced nitrate formation. These findings underscore the importance of accounting for emission heterogeneity in climate modeling. While the use of such a detailed model provides valuable insights, the analysis and discussion should be improved in relation to topics outlined below.

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.

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.

Minor Comments:

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?

Line 70: 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.

Line 85: Typo: “It is extends”

Line 94: Typo: “by by”

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

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

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.

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