

This manuscript addresses complex aerosol processes involving emission spatial heterogeneity using an idealized WRF-PartMC-MOSAIC-LES framework, the first of its kind, to address a source of uncertainty of such factors in global climate models (GCMs). In particular, the authors study how spatial variability in particle emissions influences aerosol aging and subsequently, cloud condensation nuclei (CCN) activity and hygroscopicity. The main finding of the manuscript appears to be that under low supersaturation conditions, CCN concentrations increase by 25% atop the boundary layer, with high spatial heterogeneity, given a six-hour period of surface warming, drying, and boundary layer deepening with no wind. This work, while highly idealized, clearly shows the importance of capturing emission heterogeneity in cloud-aerosol interactions which is not represented well in the GCMs.

Overall, the paper is well written, the scientific objectives are expressed clearly, the limitations are described, and figures are well made. This work contributes to the scientific understanding of this complex problem by forwarding the scientific understanding of the complex aerosol-cloud interactions while accurately describing the limitations of such an idealized study. However, I do have some further considerations for improving the manuscript and would like some clarifications. There are some questions/comments to address and some very minor grammatical issues to fix. See below:

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.
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.
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.

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.
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?
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.

Other very minor things:

Typo line 85: "it is extends

Typo line 94: "by by"

Repeated sentences around line 138