Exploring the Aerosol Activation Properties in a Coastal Area Using Cloud and Particle-resolving Models

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Summary

Yu and coauthors applied cloud parcel model and particle-resolved aerosol model to investigate under shallow convection conditions how aerosol evolution affect cloud-formation properties of the aerosol populations. They found that significant different between aerosols in boundary layer and high altitude in terms of CCN properties, also discrepancies in CCN activation ratio due to internal mixing assumption and that discrepancy increases with environmental supersaturation.

This study enhances our understanding of cloud formation properties of aerosol particles undergoing aging process under actual meteorological conditions when the air parcels leave the boundary layer. I would suggest the editor to consider accepting the manuscript for publication after emphasizing the novelty of this study and addressing the following questions or comments.

<u>General and major comments</u>

1. The unique contribution of this study is to incorporate shallow cumulus convention information to aerosol (particle-resolved) modeling in order to simulate the aerosol aging process (under shallow cumulus convection condition) and the associated CCN properties.

However, a lot of details about the methodology is missing. This part is important in that it is closely related to the conclusion of this study and provides unique contribution apart from existing studies of aerosol mixing state and CCN properties. Besides, lack of methodological details weakened the reproducibility and quality of this work, however this could be improved by providing more simulations details.

1.1 How is the Cloud Model 1 (CM1) configured to drive large-eddy simulations of ideal shallow cumulus convection? What are the specific model input and related parameters?

1.2 What do you mean by 'ideal shallow cumulus convection conditions'? Are these conditions based on any previous studies, any citations?

1.3 The authors tracked 484 parcels from the large eddy simulations, how to devise the four scenarios based on the 484 parcels? I don't see the technical details here in the manuscript. How do we know these 4 scenarios are representative? Besides, a table show the model input of these 4 scenarios is recommended for convenience.

1.4 Line 121-122, the authors extracted the temperature, pressure, kinetic diffusion coefficients for scenarios setup. Are these parameters the environmental input parameters to PartMC-MOSAIC simulations? How are relative humidity (or any other humidity measures) input to PartMC-MOSAIC?

1.5 Line 127-130 listed the four scenarios. What is meant by 'high altitude'?

1.6 Besides, it is not clear about the time dimension of the two set of model simulations, the CM1 and PartMC-MOSAIC. How long does the CM1 simulation last? 6 hours or 9 hours,10 hours or longer? It says PartMC-MOSAIC run for 30 hours, however, some features are shown e.g. concentration of Cl and nitrate increase drastically from the 2^{nd}, 6^{th} and 10^{th} hour for scenarios B, C, and D respectively in Figure 1 (left panel). I wonder what is the relationship between the two 'time axes' of CM1 and PartMC-MOSAIC.

1.6 Also, it is not clear what is the relationship between the CM1 model output and PartMC-MOSAIC model input.

1.7 How do you deal with exchange of gases and aerosol particles (dilution) between the Lagrangian parcel of PartMC-MOSAIC and the surrounding environment? Is there any model output from CM1 that can guide PartMC-MOSAIC simulations in this regard?

1.8 Line 134, it says that horizontal eddy diffusivity at the high altitude was about one-fifth of the value near the surface. Is there any previous study supporting this assumption?

1.9 Line 140 and Table 1, how did authors use MERRA-2 reanalysis to derive concentration of aerosol at ground level and at high-altitude? A table showing the aerosol properties derived from MERRA-2 would be helpful, for example, this study is about coastal area, what is the concentration of sea salt aerosol and organic? How high is the 'high altitude'? Since authors also mentioned that vertical variability of aerosol is important, this vertical information is required to be clearly described.

2. About the quantification of the aerosol mixing state impact on cloud droplet formation property, presented in section 4 and figure 5, there is no details about how the composition averaging was performed. This is important because the conclusion about impact of mixing state and the error on CCN properties rely on this calculation.

3. Besides, there are already many studies about the relationship between aerosol mixing state and CCN properties of aerosol (the authored also cited some of these studies). What is the uniqueness and novelty of this model study? The authors emphasized their study incorporated shallow cumulus convention model, which is great. However, the authors are expected to explain the importance of such advancement and how does it compare to existing observational studies (even though there is no modeling study of similar kind).

4. Line 264, why scenarios B and C differ in aging process? More explanation is expected.

5. Line 286, why some hydrophilic species tend to form on smaller particles? What are those species? More explanation is expected.

6. Line 360, why at high supersaturation, CCN activation error becomes larger for some scenarios? More explanation is expected.

7. In figure 4b, why is there a peak for scenarios B, C and D at 2nd and 6th hour?

Specific comments

Apart from the general comments above, there are some specific questions or items to be clarified throughout the manuscript.

1. Section 4, (p.14) There is no details about the calculation of ‘chi’, the mixing state index. In the figure 4a, the D_alpha and D_gamma lie between 1.4 to 2.0, why is the range so small? There are so many chemical species listed in lines 160-164.

2. What is the definition of error in Figure 5? Does positive sign mean overestimation or underestimation of CCN by composition averaging? How about negative sign?