

Authors' Response to the Reviewer Comments

Manuscript title: Critical Role of Dust Induced Electrostatic Coagulation in the Evolution of Aerosol Size Distributions in the Atmosphere

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Response to Reviewer 2's comments:

This manuscript highlights the important role of electrostatic interactions in aerosol coagulation and their impact on the resulting PSD evolution. The study uses DMA-APS measurements to obtain joint size-charge distributions of aerosols, enabling characterization of single-particle charge properties. Based on this information, the authors combine coagulation modeling with numerical simulations, demonstrating a significant role of highly charged aerosols in the coagulation process. These results help improve understanding of atmospheric aerosol microphysical processes. Overall, the experimental design and modeling framework are generally reasonable; however, several key methodological aspects—such as size conversion, charge data processing, and model parameter assumptions—require further clarification to improve the transparency and reproducibility of the study. In addition, the applicability of some results should be more clearly defined.

1. The manuscript assumes a dynamic shape factor of $\chi=1$ in the conversion from aerodynamic diameter to electrical mobility diameter. Since mineral dust aerosols are typically irregular in shape, this assumption may introduce a systematic shift in the mapped diameter and therefore alter aerosol bin assignment in the joint size-charge distribution. Given that the reported electrostatic enhancement depends on this joint distribution, please clarify how adopting representative χ values for mineral dust ($\chi>1$) would shift the size range associated with highly charged aerosols, and whether such shifts could affect the qualitative conclusions regarding PSD evolution. Clarifying this point would help readers assess the sensitivity of the results to aerosol morphology assumptions.

Response: Thank you for raising this important point. We agree that assuming $\chi = 1$ in the conversion from aerodynamic diameter to electrical mobility diameter may affect the converted electrical mobility diameter range of highly charged dust aerosols. However, this uncertainty does not affect the qualitative comparison between the Brownian-only coagulation and electrostatic coagulation cases, because both cases start from the same initial size - charge distribution and therefore share the same underlying Brownian coagulation pathway, with the only difference being whether electrostatic interactions are included. Therefore, this assumption does not alter the qualitative conclusion of this study that electrostatic interactions substantially enhance coagulation and PSD evolution relative to the Brownian-only case.

Changes to the manuscript:

Lines 150 - 156: It should also be noted that a dynamic shape factor of $\chi = 1$ was assumed in the conversion from aerodynamic diameter to electrical mobility diameter as a simplifying assumption. Since irregular mineral dust aerosols typically have $\chi > 1$, this assumption may shift the particle size range associated with highly charged aerosols, while this assumption affects the converted particle size, but does not change the measured charge characteristics. This uncertainty does not affect the qualitative comparison between the two cases, because both cases start from the same initial PSD and therefore share the same underlying Brownian coagulation pathway, with the only difference being whether electrostatic interactions are included.

2. The manuscript uses linear interpolation along the charge axis to fill missing charge-bin concentrations. Since the high-charge tail is inherently sparsely populated, this region is typically sensitive to data-processing methods. When linear interpolation is applied, high-charge bins that would otherwise be zero or undetectable may be assigned non-zero concentrations, thereby artificially smoothing and elevating the tail of the distribution. Given that the reported electrostatic enhancement depends on the fraction and distribution of highly charged aerosols, this treatment may affect the fraction of highly charged aerosols and consequently influence the related analysis results. The authors are therefore encouraged to clarify the robustness of this interpolation step for subsequent analysis results.

Response: Thank you for raising this issue. We agree that the high-charge tail, due to its low aerosol number concentrations, can indeed be more sensitive to data-processing methods.

In this study, linear interpolation along the charge axis was applied only to a small number of missing charge bins in the size-charge joint distribution matrix. This was done mainly to preserve the continuity of the matrix for subsequent coagulation modeling. It should be noted that the prescribed Z_p range in this study is relatively broad, so the covered charge range is also broad and includes the high-charge tail. The high-charge region is therefore not entirely missing, but contains only a small number of discrete gaps. Accordingly, linear interpolation is not expected to significantly increase the proportion of highly charged aerosols.

Changes to the manuscript:

Lines 145 - 149: The interpolation uncertainty is mainly associated with the high-charge tail, where aerosol number concentrations are inherently low and therefore more sensitive to data-processing methods. However, the prescribed electrical mobility range in this study is relatively broad and still includes measured points in the high-charge tail. Therefore, the uncertainty associated with the high-charge tail is expected to have a limited influence on the reconstructed size - charge distribution.

3. In the mixed-system simulations, the inclusion of electrostatic interactions leads to an approximately 10% difference in the peak ambient sub-500 nm aerosol number concentration compared with simulations without electrostatic effects. The authors are encouraged to clarify in the Results and Discussion section of the manuscript the significance of this difference for the

evaluation of environmental impacts such as radiative forcing and CCN number concentration, and whether it should be highlighted in the discussion of the results or regarded as only a secondary effect within the model. Providing such clarification would help readers better understand the implications of this difference for atmospheric applications.

Response: Thank you for raising this issue. We agree that, within the simulation framework of this study, the ~10% difference in the peak number concentration of ambient sub-500 nm aerosols in the mixed system should be regarded as a secondary effect. Nevertheless, it still has environmental relevance because ambient sub-500 nm aerosols contribute substantially to CCN number concentrations and aerosol radiative effects.

Changes to the manuscript:

Lines 359 - 364: Although the magnitude of this difference is modest compared with the much larger the PSD changes of highly charged dust aerosols, the ~10% difference indicates that electrostatic interactions affect the evolution of ambient sub-500 nm aerosols. This effect is environmentally relevant because these aerosols are closely related to aerosol radiative effects and CCN number concentrations. At the same time, it should still be regarded as a secondary effect, as it is much smaller than the PSD changes observed for highly charged dust aerosols themselves.

4. The study adopts a closed-box framework, in which no new aerosol input is considered. Under this assumption, aerosol charges gradually neutralize and decay through coagulation. However, during real atmospheric transport, dust plumes may continuously introduce newly emitted or mixed-in aerosols that can retain relatively high initial charge. Given that the reported electrostatic enhancement depends on the presence of highly charged aerosols, the authors are encouraged to clarify whether such continuous input of highly charged aerosols could delay charge decay and sustain stronger electrostatic coagulation, thereby affecting the reported PSD evolution.

Response: Thank you for this important comment. We agree that during real atmospheric transport, if a dust plume continuously mixes with newly emitted or entrained highly charged dust aerosols, this sustained input may indeed slow the charge decay of the system to some extent. It may also allow strong electrostatic coagulation to persist for a longer time, thereby affecting the PSD evolution. In contrast, this study adopts a closed-box framework and does not include any replenishment by newly emitted or entrained highly charged dust aerosols. As a result, the charge level of the system decreases more rapidly than in real atmospheric conditions with continuous input, and the persistence of electrostatic coagulation may therefore be underestimated. Newly emitted or entrained dust aerosols may retain higher charge levels than dust aerosols after long-range transport. However, this does not alter the main conclusion of this study that electrostatic interactions associated with highly charged dust aerosols substantially enhance coagulation relative to the Brownian-only case and thereby accelerate PSD evolution.

Changes to the manuscript:

Lines 334 - 342: The dust aerosols used in this study are more representative of freshly

generated dust aerosols. As coagulation and related processes proceed during atmospheric transport, the charge states of dust aerosols are generally expected to decrease. Therefore, the PSD evolution trend shown here is more representative of dust source regions and near-source conditions. However, in real atmospheric transport, dust plumes may continuously mix with newly emitted or entrained aerosols that retain relatively high initial charges. Such continuous input could delay charge decay and sustain stronger electrostatic coagulation over a longer timescale, thereby affecting PSD evolution during transport. The above discussion suggests that the transition from freshly generated, highly charged dust to more aged and weakly charged dust during atmospheric transport deserves more explicit consideration in future modeling studies.

5. In Fig. 3, the caption refers to aerodynamic diameter, whereas the main text indicates that aerosol sizes were converted to mobility diameter. To avoid potential confusion, the authors are encouraged to clarify in the caption which diameter is shown on the x-axis and to ensure consistent terminology between the text and the figure.

Response: Thank you for pointing out this issue. The aerosol diameter shown on the x-axis in Fig. 3 is actually the electrical mobility diameter. The original caption incorrectly referred to it as “aerodynamic diameter,” which may have caused confusion. Following the reviewer’s suggestion, we have corrected the caption of Fig. 3 in the revised manuscript and ensured consistent terminology between the main text and the figure caption.