

Reviewer's comments are in black, and responses are in blue.

General comments:

In fact, I also reviewed another paper by the author (Yang et al. 2024; <https://doi.org/10.5194/acp-24-5989-2024>), which can be considered a sister paper in some respects. In comparison to these two papers, the first one (Yang et al. 2024) seems more innovative because it appears to have established a new model. This paper only conducted sensitivity experiments with different aerosol concentrations using the model from the previous paper, which provided useful references but seemed to have lessened in terms of innovation.

Reply: We appreciate your comment. We agree that this study is based on the model in previous studies, although the technique is not new, the question we address is important and not well understood. To our knowledge, no study has investigated the role of different SIP processes in cloud electrification under different aerosol conditions. Our results show the RS process is the most important SIP process in a polluted environment, while the SD process is more important in a clean environment. We believe this can be a useful reference for future studies.

Specific comments:

1. First, we acknowledge that different secondary ice production (SIP) processes or different aerosol concentrations play an important role in cloud electrification, which has been previously studied in many related studies (Mansell and Ziegler 2013; Tan et al. 2015; Phillips and Patade 2022; Yang et al. 2024). At this stage, it seems less urgent to continue discussing the impact of different SIP processes on electrification under different aerosol conditions. Because cloud microphysical characteristics change successively after the SIP process and aerosol concentration changes, the part related to electricity will also naturally change. When the charging rate changes, the evolution of the charge structure is only a more superficial feature. We know that there are many experiments or hypotheses related to the SIP processes, but which process of SIP actually plays a role in clouds and whether they all play a role at the same time is still a question worth exploring. Therefore, we are still unsure whether 4SIP will play a role at the same time.

Reply: Thank you for your comment. We acknowledge that previous studies have revealed the effects of aerosol concentration or SIP processes, but to our knowledge, no

study has explored in detail the role of different SIP processes under different aerosol conditions. Mansell and Ziegler (2013) tested 13 different aerosol concentrations but only considered the rime-splintering process. Tan et al. (2015) investigated the effect of different aerosol concentrations on electrification but not in-depth on the SIP processes. Phillips and Patade (2022) investigated the impact of aerosol concentration and three SIP processes, respectively, but they did not report which SIP dominates under different aerosol conditions. These studies are good references, but we think this is worth conducting a study focusing on the role of different SIPs under various aerosol conditions because the results will tell us which SIP process is more important in a polluted and clean environment, and can deepen our understanding of the difference between maritime and continental thunderstorms.

We agree it is still a question that which process of SIP actually plays a role in clouds and whether they all play a role at the same time. In fact, this is one of the purposes of this study. Our results suggest the RS process is the most important one in an environment with high aerosol concentration, and the SD process is more important when the aerosol concentration is low. This conclusion is the same no matter whether all the four SIP processes are considered or individual SIP process is implemented in the model. However, the charging rate is the greatest when all the four SIP processes are turned on.

We acknowledge that the conclusions are obtained only from a case study, and some of the results are different from the other cases in previous studies. To illustrate the similarity and difference between this study and previous ones, we add the following discussions in the revised paper:

In this paper, aerosol concentrations from 400 to 4000 cm⁻³ are considered, the results suggest generally a higher aerosol concentration leads to stronger charge separation, but the aerosol impact on cloud microphysics and electrification is not linear. This is also found in some previous studies, for example, Mansell and Ziegler (2013) tested 13 different aerosol concentrations from 50 to 8000 cm⁻³ to investigate the effect of aerosols on storm electrification and precipitation. They found that the graupel concentration increases as the CCN concentration increases from 50 to 2000 cm⁻³, but slowly decreases as the CCN concentration increases from 2000 cm⁻³. Tan et al. (2015)

designed simulation experiments with CCN concentrations from 50 to 10000 cm⁻³. They found that more cloud droplets, graupel, and ice crystal production lead to a stronger charge separation as aerosol concentration increases from 50 to 1000 cm⁻³. In contrast, as the aerosol concentration increases from 1000 to 3000 cm⁻³, the mixing ratio of ice crystals decreases, the noninductive charging is weakened, while the inductive charging rate has no significant change.

The stronger charge separation induced by higher aerosol concentration may modify the structure of total charge density. For example, Shi et al. (2019) found that the charge structure at different convective intensities (by controlling the environmental humidity and temperature stratification at an initial time) became more complex as the aerosol increased. Sun et al., (2024) showed that compared to the low aerosol concentration case, a notable inverted dipole charge structure was simulated in the high aerosol environment. The modelled charge structure in different cases may be different, depending on multiple factors such as the thermodynamic properties and liquid water content (Phillips and Patade, 2022; Zhao et al., 2020), and is possibly related to the different parameterizations of electrification used in various studies (Phillips and Patade, 2022). Nevertheless, all these studies, including the present paper, demonstrate the flash rate can be enhanced by higher aerosol concentration, which is regarded as a key explanation for the higher flash rate over continents than over ocean.

Previous studies have pointed out that the SIP processes strongly affect cloud microphysics and electrification (Waman et al., 2022; Huang et al., 2022; Phillips and Patade, 2022). In this study, we further show that the RS process is the most important one in an environment with high aerosol concentration, and the SD process is more important when the aerosol concentration is low. This conclusion is consistent with previous studies which suggest the RS process can strongly affect the charge separation in continental thunderstorms (Huang et al. 2024; Yang et al. 2024), and the SD process may be a more efficient SIP mechanism in maritime convection, in which more supercooled rain drops are observed (Field et al., 2017). Phillips and Patade (2022) investigated a convective cloud with a cold base, they suggested the IC process is more active than the RS and SD process as the droplets are too small. In our case, the IC process is only efficient at temperatures colder than -10 °C in the mature stage. The

sublimational breakup process has the least impact, which is also found in mature convective clouds simulated by Waman et al. (2022).

Regardless of the differences in various studies, it is commonly found that the aerosol concentration and SIP processes both have great impacts on cloud microphysics and electrification. An increase in aerosol concentration leads to a nonlinear enhancement of the charging rate. The RS process is the vital SIP process in a polluted environment, and the SD process is more important in a clean environment. Therefore, accurate representations of the various SIP processes under different aerosol conditions are important for the model simulation of lightning activities.

References:

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2. Regarding the structure of the paper, in the description of the model in section 2.2.2, the content of this section is almost a duplicate of Appendix B in Yang et al. (2024). Is there a need for duplicate descriptions here?

Reply: Thank you for your comment. According to your suggestion, the equations in Section 2.2.2 have been removed, and the related text has been modified.

3. From the results in Fig. 10, compared to the experiments with $N_0=400$, the experiments with $N_0=4000$ show a stronger charging rate, which also corresponds to a stronger electric field intensity in these two groups of experiments (Fig. 16). However, in Fig. 10, compared with noSIP-4000, the non-inductive charging rate of 4SIP-4000 is stronger, but the polarity and height of the charging rate change very little, which may not cause a significant change in the polarity of the charge structure. On the contrary, the inductive charging rate has undergone significant changes. This may be the reason for the change in the polarity of the charge structure of 4SIP-4000 in Fig. 13.

Reply: Thank you for your comment, and sorry for the confusing statement in the original manuscript. As shown in Fig. 11 in the revised manuscript, the charge structure, especially the reversal temperature, can be significantly altered by increasing CCN concentration. Therefore, the modified charge structure in 4SIP-4000 experiment is not only a result of SIP but also the increased CCN concentration. In fact, the reversal temperature is less affected by SIP because the polarity and height of the charging rate change very little by comparing the noSIP-4000 and 4SIP-4000 experiments. Therefore, it is incorrect to state the SIP processes can change the structure, it is the combined effect of increased CCN concentration and SIP processes that modify the charge structure.

The magnitude of noninductive charging rate is much larger than that of inductive charging, so we believe it is still the noninductive charging dominates the changes in total charge structure. To confirm this, we made a sensitivity test using noninductive charging only (Fig. R1), it is seen from the figure that the modelled charge structure is similar compared to that in the original paper, which included both charging mechanisms.

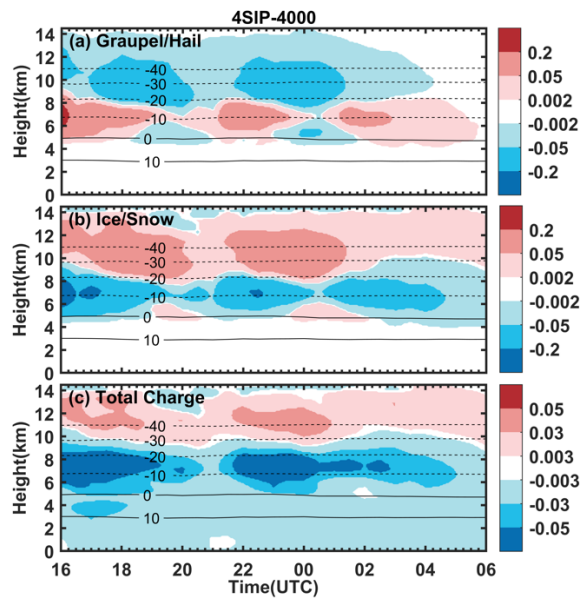


Figure R1. Time-height evolution of mean charge carried by (a) graupel/hail (a-d) and (b) ice/snow particles as well as (c) mean total space charge (unit is nC m^{-3}) from the simulation 4SIP-4000 with noninductive charging only.

Many previous studies concluded that noninductive charging process is the main charging process of thunderstorms and the inductive charging alone would be insufficient to strongly electrify a storm (Brooks and Saunders, 1994; Jayaratne et al., 1983; Mansell et al., 2005; Saunders and Peck, 1998; Takahashi, 1978). This paper also provides the same conclusion that the noninductive charging rate is much greater than the inductive charging rate (shown in Fig. 11 in the revised manuscript). In addition, the Fig. 15 in Yang et al. (2024) shows the graupel charge density and noninductive charging rate in noSIP experiment and RS experiment with only noninductive charging considered. In Fig. 15 in Yang et al. (2024), the distribution of graupel charge density still produces a significant variation. We agree that the inductive charging rate has undergone significant changes, so the effect of induced charging rates is also of concern. Based on the above discussion, it can be concluded that both noninductive charging process and inductive charging process have impact on charge structure, and the former has a greater impact.

References:

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4. The paper mentions that without SIP, the aerosol does not change its charge structure. However, as shown in Fig. 11a, even without the SIP process, the charging rate within the cloud significantly changes with the change in aerosol. The height-time variation diagram may not reflect the actual charge structure, and a cross-section diagram of the charge structure should be provided.

Reply: Thank you for your comment, and we are sorry for the incorrect statement. We agree that the aerosol concentration strongly affects the charge structure, and in fact, it has a stronger impact than the SIP on the reversal temperature and height. This can be readily seen from the time-height diagram in the original manuscript. In the revised paper, we have modified the statement and showed that the charge structure can be significantly affected by increasing aerosol concentration. According to your suggestion, the cross-sections of total charge density from the noSIP experiment and the 4SIP experiment are shown below. The cross-sections show a more complicated charge structure, but in general, it is consistent with the time-height diagram and demonstrates that the increase in aerosol concentration can strongly affect the charge structure.

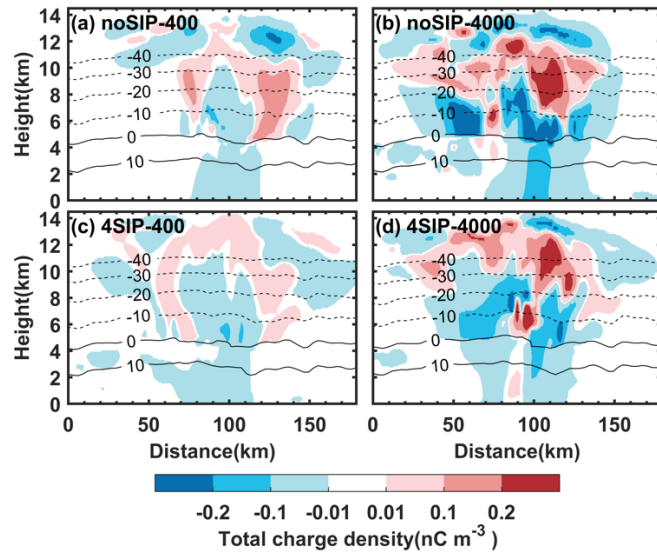


Figure R1. The cross-section diagrams of the charge structure from (a-b) noSIP experiment and (c-d) 4SIP experiment.