The reviewer's comments are in black, and responses are in blue.

General comments:

Huang et al. investigated the role of secondary ice production on thunderstorm electrification under various aerosol conditions using numerical simulations. Overall, the research topic is interesting and valuable for the scientific community. However, in its current form, the manuscript needs major changes before being considered for further revision or final acceptance. I have enlisted my specific and minor comments below.

Reply: We appreciate your insightful comments. The paper has been revised accordingly and has been improved a lot. Please see our responses below.

Specific comments:

1. Model validation is done based only on radar reflectivity. Since the main objective of the paper is thunderstorm electrification it is important to validate additional variables from model simulations with observations such as flash rate, rain rate, ice number concentration, ice water path, etc. It will be interesting to see how changes in aerosol affect the flash rate in the presence of SIP mechanisms.

Reply: Thank you for your comment. Unfortunately, we don't have in-situ measurements of cloud microphysics such as ice number concentration, ice water path, etc. CloudSat data is not available for this case to retrieve the ice microphysics. The locations of lightning can be obtained from the FY4A stationary satellite measurement, we add the comparison between the modelled and observed lightning location in the revised paper (Fig. R1). The limitation of FY4A is its detection efficiency is relatively low during the daytime because of the reflectance of sunlight (Sun et al., 2022), and the flash rate cannot be obtained from FY4A. Statistically, the locations of the observed and modelled lightning are consistent.

According to your Comment 2, the 4SIP-4000 experiment has been set as a control experiment and the following analysis has been added to the revised manuscript: "*In this paper, the observed lightning data are collected by the Lightning Mapping Imager (LMI), which is mounted on the second-generation Chinese geostationary meteorological satellite FY-4A to continuously detect lightning activity in China and its neighboring areas. Additionally, the brightness temperature captured by FY4A* *satellite is used to delineate the area of the deep convective cloud. Figure R1 shows the location of the lightning events observed by LMI and simulated in 4SIP-4000 experiment. It is seen that the simulated and observed lightning locations are in good agreement with each other and are both in the low brightness temperature region, which characterizes the presence of strong convection.*"

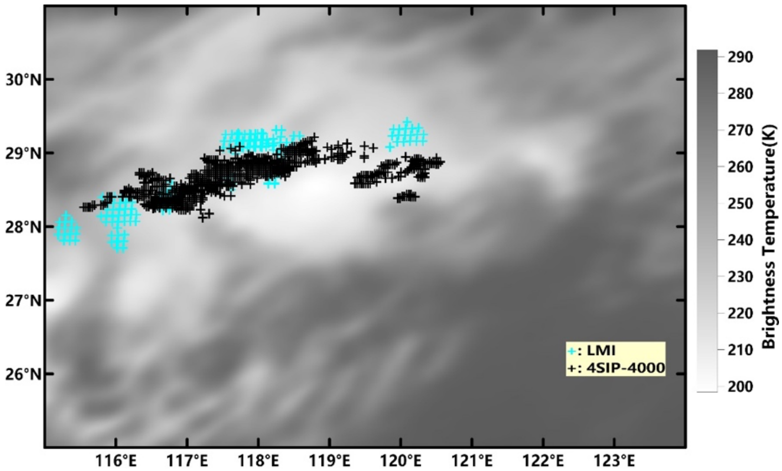

Figure R1. The simulated and observed lightning locations (sign "+"). The shaded field indicates brightness temperature.

To investigate the impact of aerosol on the flash rate in the presence of SIP, we plot Fig. R2, which shows the flash rates modelled in the 4SIP experiments with different aerosol concentrations. The results indicate that a higher aerosol concentration can lead to a greater flash rate, which is consistent with previous studies (e.g., Liu et al., 2020; Shukla et al., 2022; Wang et al., 2011)

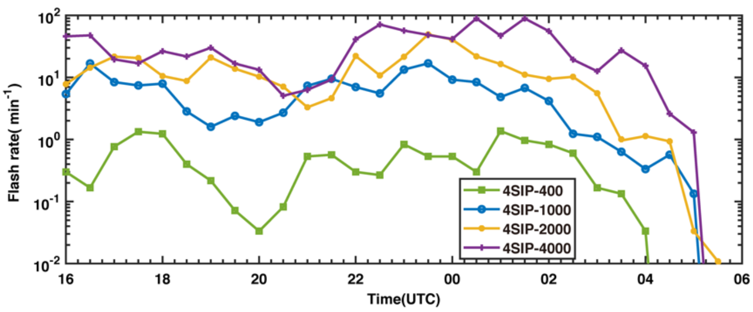

Figure R2. Flash rate modelled by the 4SIP-400, 4SIP-1000, 4SIP-2000, and 4SIP-4000 experiments.

References:

Liu, Y., Guha, A., Said, R., Williams, E., Lapierre, J., Stock, M., and Heckman, S.: Aerosol Effects on Lightning Characteristics: A Comparison of Polluted and Clean

Regimes, Geophysical Research Letters, 47, e2019GL086825, https://doi.org/10.1029/2019GL086825, 2020.

Shukla, B. P., John, J., Padmakumari, B., Das, D., Thirugnanasambantham, D., and Gairola, R. M.: Did dust intrusion and lofting escalate the catastrophic widespread lightning on 16th April 2019, India?, Atmospheric Research, 266, 105933, https://doi.org/10.1016/j.atmosres.2021.105933, 2022.

Wang, Y., Wan, Q., Meng, W., Liao, F., Tan, H., and Zhang, R.: Long-term impacts of aerosols on precipitation and lightning over the Pearl River Delta megacity area in China, Atmos. Chem. Phys., 11, 12421–12436, https://doi.org/10.5194/acp-11-12421-2011, 2011.

Sun, H., Yang, J., Zhang, Q., Song, L., Gao, H., Jing, X., Lin, G., and Yang, K.: Effects of Day/Night Factor on the Detection Performance of FY4A Lightning Mapping Imager in Hainan, China, Remote Sensing, 13, 2200, https://doi.org/10.3390/rs13112200, 2021.

2. Also, it is not a good idea to compare perturbed simulations with observations and then pick the simulations that are in better agreement with observations. Authors should set a control simulation that is more realistic considering observed conditions of aerosol/CCN, including all SIP (like in real clouds). Then compare only control simulation with observations as a part of validation. This will increase the readability of the manuscript. Also, it is important to quantify the validation results. e.g. if there is bias in observations and simulations it should be mentioned in % or actual bias.

Reply: Thank you for your comment. The aerosol concentration in mainland China is typically high, we do not have direct measurements of aerosol concentration for this case, so we refer to previous studies in the same region. Qu et al. (2017) suggested using an N_0 of 4000 cm^{-3} for southeast China. Therefore, we use the 4SIP-4000 experiment as the control experiment. In the revised paper, we now compare the control simulation with observation (Fig. R3), and the related text has been modified accordingly. The bias between observations and simulations has been quantified as follows: *"Comparison of the simulated results and observation reveals that the mean absolute errors of reflectivity at 00:00,02:00 and 04:00 are 12 dBZ, 11 dBZ and 12* *dBZ, respectively. The area where the radar reflectivity difference is less than 15 dBZ are 67%, 71%, and 67% at 00:00, 02:00 and 04:00, respectively."*

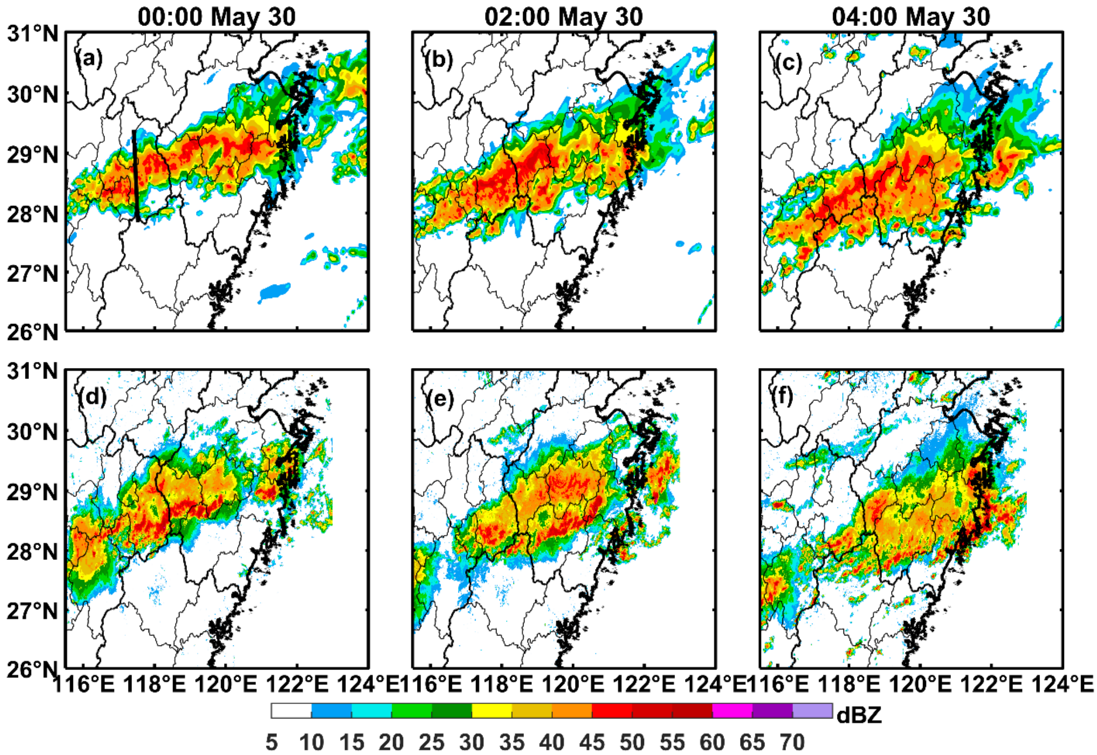

Figure R3. The (d-f) observed and (a-c) simulated radar reflectivity at 00:00, 02:00, 04:00 on 30th May. The simulated results are from 4SIP-4000 experiment.

References:

Qu, Y., Chen, B., Ming, J., Lynn, B. H., and Yang, M.-J.: Aerosol Impacts on the Structure, Intensity, and Precipitation of the Landfalling Typhoon Saomai (2006): Aerosol Impacts on Typhoon Saomai (2006), J. Geophys. Res. Atmos., 122, 11,825-11,842, https://doi.org/10.1002/2017JD027151, 2017.

3. The Author needs to justify the need for 26 simulations in the current study. Many of those simulations are not important. In their previous studies, the role of all major SIP mechanisms as well as individual SIP mechanisms on deep cloud properties including electrification has already been investigated (e.g. Yang et al. 2024). The focus of the current study should be only on how different aerosol conditions alter cloud electrification together with SIP. In my opinion, there is no need for any simulation without individual SIP mechanisms. Only the changes in the rate of SIP production can be shown under various aerosol conditions (with and without all SIP) and their

subsequent effect on cloud electrification should be discussed. In the current format, there is a lot of confusion in following the results.

Reply: Thank you for your comment. We agree, the focus of this paper is on the differences in cloud microphysics and cloud electrification under different aerosol conditions with and without SIP processes. In most of the figures, only noSIP and allSIP experiments are used. Other sensitivity experiments with individual SIP processes are only used for clarification. Based on your suggestion, we have modified Table 1 and related text in the revised manuscript to make it easier for readers to understand.

4. Line 141: Please describe briefly how the sublimation breakup was implemented in the model. Does the model have separate species of dendritic and non-dendritic crystals? If the implementation in your model has been described in the previous study give proper references.

Reply: Thank you for your comment. The Fast-SBM scheme in WRF does not distinguish the shape of the ice crystals (Khain et al., 2009). It treats solid particles as ice/snow or graupel/hail. Therefore, in this study, the sublimational breakup process is applied to ice/snow crystals. This parameterization has been used in previous studies (e.g., Yang et al., 2024; Deshmukh et al., 2022; Waman et al., 2022)

Reference:

Deshmukh, A., Phillips, V. T. J., Bansemer, A., Patade, S., and Waman, D.: New Empirical Formulation for the Sublimational Breakup of Graupel and Dendritic Snow, Journal of the Atmospheric Sciences, 79, 317–336, https://doi.org/10.1175/JAS-D-20-0275.1, 2022.

Khain, Leung, L. R., Lynn, B., and Ghan, S.: Effects of aerosols on the dynamics and microphysics of squall lines simulated by spectral bin and bulk parameterization schemes, J. Geophys. Res., 114, D22203, https://doi.org/10.1029/2009JD011902, 2009.

Waman, D., Patade, S., Jadav, A., Deshmukh, A., Gupta, A. K., Phillips, V. T. J., Bansemer, A., and DeMott, P. J.: Dependencies of Four Mechanisms of Secondary Ice Production on Cloud-Top Temperature in a Continental Convective Storm, Journal of the Atmospheric Sciences, 79, 3375–3404, https://doi.org/10.1175/JAS-D-21-0278.1, 2022.

Yang, J., Huang, S., Yang, T., Zhang, Q., Deng, Y., and Liu, Y.: Impact of ice multiplication on the cloud electrification of a cold-season thunderstorm: a numerical case study, Atmos. Chem. Phys., 24, 5989–6010, https://doi.org/10.5194/acp-24-5989-2024, 2024.

5. The reasons behind the noninductive dipole structure having an upper negative region and lower positive region are not discussed properly/or are difficult to follow. Are there previous studies showing this kind of charge structure? If there is an increase/decrease in electrification due to SIP/aerosol should be quantified.

Reply: Thank you for your comment. In the SP98 noninductive charging scheme, the riming accretion rate (RAR) controls the charge sign during charge separation, and the noninductive charging occurs where RAR is greater than 0.1 $gm^{-2}s^{-1}$ (Mansell et al., 2005; Saunders and Peck, 1998). The negative region denotes that an RAR is smaller than the critical RAR and the positive region denotes an RAR greater than the critical RAR. The critical RAR is a function of temperature. The RAR is the effective water content multiplied by the graupel fall velocity (Mansell et al., 2005). The relatively low liquid water content and the small size of graupel particles in the upper region lead to a smaller RAR, which is more likely to be less than the critical RAR and result in a negative noninductive charging rate. This is also found in previous studies, for example, Fierro and Mansell (2017) simulated idealized tropical cyclones and conducted sensitive experiments to investigate the impact of the wind shear and sea surface temperature on cloud microphysics and electrification. They found that positive charging occurs between 6.5 and 8 km with negative charging above that over a deeper layer, between 8 and 11 km. This discussion is added in the revised paper in the description of Fig. 11.

According to your comment, the bias in electrification due to SIP/aerosol has been quantified as follows: "*As* N_0 *increases from 400* cm^{-3} *to 4000* cm^{-3}*, the noninductive charging strengthens, with the mean positive noninductive rate and negative rate increasing by 4.9 and 3.24 times, respectively. The inductive charging rate increases too, with positive and negative rates increasing by a factor of 8.7 and 4.9, respectively. The addition of SIP processes also has a great influence on the charging separation. On average, the rime splintering and freezing droplet shattering process has greater* *impacts than the other two. The mean positive and negative noninductive charging rates in the 4SIP experiment are 3.9 times and 46% greater than in the noSIP experiment, and the positive inductive charging rates can be enhanced by 1.2 times by implementing the four SIP processes. However, the negative inductive charging rate decreases 56.83% in 4SIP experiment than in noSIP experiment. In addition, the aerosol concentration and the SIP processes can affect the area of the positive and negative charging. As N_0 increases from 400 cm^{-3} to 4000 cm^{-3}, the areas of positive noninductive and inductive charging increase by 18% and 23%, respectively. With the addition of SIP processes, the area of positive noninductive and negative inductive rates decreases by about 3% and 23%, while the area of negative noninductive and positive inductive charging increases.*"

References:

Fierro, A. O. and Mansell, E. R.: Electrification and Lightning in Idealized Simulations of a Hurricane-Like Vortex Subject to Wind Shear and Sea Surface Temperature Cooling, Journal of the Atmospheric Sciences, 74, 2023–2041, https://doi.org/10.1175/JAS-D-16-0270.1, 2017.

Mansell, E. R., MacGorman, D. R., Ziegler, C. L., and Straka, J. M.: Charge structure and lightning sensitivity in a simulated multicell thunderstorm, J. Geophys. Res., 110, D12101, https://doi.org/10.1029/2004JD005287, 2005.

Saunders, C. P. R. and Peck, S. L.: Laboratory studies of the influence of the rime accretion rate on charge transfer during crystal/graupel collisions, J. Geophys. Res., 103, 13949–13956, https://doi.org/10.1029/97JD02644, 1998.

6. The discussion section needs improvement. Authors should discuss and compare their results in comparison with previous studies showing aerosol/SIP effect on electrification. If the results are not in agreement with the previous findings, please include some discussion on the factors associated with it.

Reply: Thank you for your comment. The discussion section has been modified in the revised manuscript:

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 ℃ 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:

Huang, S., Jing, X., Yang, J., Zhang, Q., Guo, F., Wang, Z., and Chen, B.: Modeling the Impact of Secondary Ice Production on the Charge Structure of a Mesoscale Convective System, JGR Atmospheres, 129, e2023JD039303, https://doi.org/10.1029/2023JD039303, 2024.

Huang, Y., Wu, W., McFarquhar, G. M., Xue, M., Morrison, H., Mil- brandt, J., Korolev, A. V., Hu, Y., Qu, Z., Wolde, M., Nguyen, C., Schwarzenboeck, A., and Heckman, I.: Microphysical pro- cesses producing high ice water contents (HIWCs) in tropical convective clouds during the HAIC-HIWC field campaign: dom- inant role of secondary ice production, Atmos. Chem. Phys., 22, 2365-2384, https://doi.org/10.5194/acp-22-2365-2022, 2022.

Mansell, E. R. and Ziegler, C. L.: Aerosol Effects on Simulated Storm Electrification and Precipitation in a Two-Moment Bulk Microphysics Model, Journal of the

Atmospheric Sciences, 70, 2032–2050, https://doi.org/10.1175/JAS-D-12-0264.1, 2013.

Phillips, V. T. J. and Patade, S.: Multiple Environmental Influences on the Lightning of Cold-Based Continental Convection. Part II: Sensitivity Tests for Its Charge Structure and Land–Ocean Contrast, Journal of the Atmospheric Sciences, 79, 263–300, https://doi.org/10.1175/JAS-D-20-0234.1, 2022.

Field, P. R., et al.: Ice formation and evolution in clouds and precipitation: measurement and modeling challenges: secondary ice production: current state of the science and recommendations for the future. Meteorological Monographs, AMSMONOGRAPHS-D-16-0014.1, https://doi.org/10.1175/AMSMONOGRAPHS-D-16-0014.1, 2016.

Shi, Z., Li, L., Tan, Y., Wang, H., and Li, C.: A Numerical Study of Aerosol Effects on Electrification with Different Intensity Thunderclouds, Atmosphere, 10, 508, https://doi.org/10.3390/atmos10090508, 2019.

Sun, M., Li, Z., Wang, T., Mansell, E. R., Qie, X., Shan, S., Liu, D., and Cribb, M.: Understanding the Effects of Aerosols on Electrification and Lightning Polarity in an Idealized Supercell Thunderstorm via Model Emulation, JGR Atmospheres, 129, e2023JD039251, https://doi.org/10.1029/2023JD039251, 2024.

Tan, Y. B., Shi, Z., Chen, Z. L., Peng, L., Yang, Y., Guo, X. F., and Chen, H. R.: A numerical study of aerosol effects on electrification of thunderstorms, Journal of Atmospheric and Solar-Terrestrial Physics, 154, 236–247, https://doi.org/10.1016/j.jastp.2015.11.006, 2015.

Waman, D., Patade, S., Jadav, A., Deshmukh, A., Gupta, A. K., Phillips, V. T. J., Bansemer, A., and DeMott, P. J.: Dependencies of Four Mechanisms of Secondary Ice Production on Cloud-Top Temperature in a Continental Convective Storm, Journal of the Atmospheric Sciences, 79, 3375–3404, https://doi.org/10.1175/JAS-D-21-0278.1, 2022.

Yang, J., Huang, S., Yang, T., Zhang, Q., Deng, Y., and Liu, Y.: Impact of ice multiplication on the cloud electrification of a cold-season thunderstorm: a numerical case study, Atmos. Chem. Phys., 24, 5989–6010, https://doi.org/10.5194/acp-24-5989-2024, 2024.

Zhao, P., Li, Z., Xiao, H., Wu, F., Zheng, Y., Cribb, M. C., Jin, X., and Zhou, Y.: Distinct aerosol effects on cloud-to-ground lightning in the plateau and basin regions of Sichuan, Southwest China, Atmos. Chem. Phys., 20, 13379–13397, https://doi.org/10.5194/acp-20-13379-2020, 2020.

Minor comments:

1. Line 17: delete up

Reply: Thank you for your comment. The “up” has been deleted in the revised manuscript.

2. Line 82: Mention WRF model version

Reply: Thank you for your comment. The WRF model version is v3.9.1 and has been added to the revised manuscript.

3. Line 97: Occurred in

Reply: Thank you for your comment. The “occurred at” has been changed as “occurred in” in the revised manuscript.

4. Line 133: change depended to depending, filed to field

Reply: Thank you for your comment. These two have been revised in the manuscript.

5. Line 134: change could to can

Reply: Thank you for your comment. The “could” has been changed to “can” in the revised manuscript.

6. Line 148: SP98 should be defined here

Reply: Thank you for your comment. The SP98 is the name of the noninductive charging scheme proposed by Saunders and Peck (1998) and its definition is added in the revised paper.

7. Line 152: signs

Reply: Thank you for your comment. The “sign” has been changed to “signs” in the revised manuscript.

8. Line 185: Describe boundary conditions in this section

Reply: Thank you for your comment. The FNL reanalysis data with 0.25° × 0.25°

resolution are used to provide the boundary conditions.

9. Line 198: What was the slope value in the Twomey equation

Reply: Thank you for your comment. The slope value in the Twomey equation is 0.4.

10. Line 225: change that are to that is

Reply: Thank you for your comment. The “that are” has been changed to “that is” in the revised manuscript.

11. Line 227: mention bias in %

Reply: Thank you for your comment. The bias has been quantitatively described there.

12. Line 228: delete of

Reply: Thank you for your comment. The “of” has been deleted in the revised manuscript.

13. Line 230: microphysics and electrification processes

Reply: Thank you for your comment. The “process” has been changed to “processes” in the revised manuscript.

14. Line 287: change than that in to than in

Reply: Thank you for your comment. The “that” has been deleted in the revised manuscript.

15. Figure 15: define legends scg, scsi etc

Reply: Thank you for your comment. The “scg”, “scsi” and “sctot” have been defined in the revised manuscript.