Response to RC2 of manuscript egusphere-2023-2507 submitted to Geosci. Model. Dev.

Jul, 2024

Reviewer 1:

General comments:

I am pleased with many of the authors' replies to my concerns regarding the methods used in this paper, as well as their incorporation of suggestions regarding references, notation and language. Unfortunately, some of the justification present in the authors' response was not incorporated in the manuscript. I also have further concerns regarding remaining issues in the grammar and language throughout the work.

We greatly appreciate the invaluable feedback provided by Reviewer 1. We have acted upon all points raised and check the grammar and language throughout the manuscript. We believe the current manuscript is improved through addressing the review comments.

Specific comments revisited:

2. Notations and E0 and Ees: Thank you for updating the notation. The approach that you are following (Andronache) should be cited in the paper where you are defining the coalescence efficiency.

Thank you for the comment, we cite the Andronache (2004) at the line178 and empresses the coalescence efficiency parameterization refer to the paper as follow:

'Referring to Andronache (2004), we propose a parameterization of the collision efficiency…'

3. Instantaneous charging assumption: I appreciate the thorough response justifying this assumption, but would like to see it explicitly and clearly stated in the paper as a limitation and your justification. It would likewise be nice to see the authors' expectation of what impact this assumption may have on the results discussed in the work.

Thank you for the important comments and suggestions, we agree with your point about clarifying the assumption introduction. We added the limitation of the assumption and result from that might be caused by the assumption in section 2.4 lines 221-232 as follows:

'Zhou and Tinsley (2012) observed that droplets with a 10 μm radius achieve 70% of their charge in 680 seconds. However, following Andronache (2004) simplification of the complex charging process, we assume that the charge on droplets resulting from collision-coalescence reaches equilibrium instantaneously. We also consider an extreme scenario where the charge polarity of two colliding droplets is always opposite. The assumption of instantaneous charging might lead to an overestimation of the electro-coalescence effect.'

4. DNS vs LES: Please also state in the text that this is a two-dimensional LES. (I realize it is already stated in the abstract)

Thank you for your comments! We agree that it should be stated in the text that the simulations used a 2D LES model, and we have clarified this in Section 2.7 "Numerical

Setup and Scheme", lines 361-362 as follows:
'Our simulations use two-dimensional Large Eddy Simulation (LES) methodology.'

Additional general comments:

• Can you justify or comment on the use of such complex equations for the particle charge (e.g. Eq 14, versus Eq 13) and whether this additional complexity (a) adds considerable computational burden; (b) whether it is justified in terms of the difference in results produced.

Thank you for your comments. The comparison of four electrostatic force settings is a key highlight of our work.

(a) Re the Computational Burden:

The simulation times for the Coulomb force (CB), image charge (IM), and Khain 2004 (Khain04) methods in the warm cumulus case are similar. The conductive sphere (CS) method takes about 30% longer. However, this increased computational time is justified by its higher accuracy and broader applicability across various droplet sizes.

(b) Re the Justification of Different Results:

Zhou and Tinsley (2008, doi: 10.1029/2008JD011527) demonstrate that the CS method offers superior numerical stability and accuracy, particularly for droplets of similar size. This enhanced precision is essential for accurately simulating cloud microphysical processes, making the additional computational burden worthwhile.

In conclusion, while the CS method incurs a higher computational burden, it provides superior numerical stability and accuracy. We also added a brief discussion on discussion section (lines 474-475) as follows:

'The CS method for electrostatic force should be incorporated into the cloud model, despite a 30% increase in computation time. CS method provides superior numerical stability and accuracy in simulating charge droplet interactions, particularly for charge droplets of similar size.'

• It would also be nice to see a brief discussion of any new insights that have arisen from this study in comparison to Khain04.

Thank you for your suggestion. We added a brief discussion in the discussion section to highlight the new insights gained from this study in comparison to Khain04. This will provide a clearer understanding of the advancements and differences between the two studies.

'Khain et al. (2004) evaluated electro-coalescence at a low charging rate of 5% of the maximum charge on droplets. In our simulation, we tested charging rates (α) ranging from 0.05 to 0.6, equivalent to 0.15% to 1.8% of the maximum charge. At a charging rate of 0.3, the electric force evaluated by the CS method increased domain and ensemble-averaged precipitation by approximately 5.42% compared to the Khain04 setting. The results indicate that even with weak charging, the electro-coalescence effect significantly increases precipitation.'

• In general there are also several grammar mistakes that remain in this manuscript. I will address a few examples below, but I hope that the authors will undergo thorough

proofreading.
Thank you for pointing out the grammatical issues in our manuscript. We appreciate your specific examples and will undertake a thorough proofreading to address all grammatical errors.

- Section 2.5 is likely unnecessary to include, as the reader could easily look at references for the compressible nonhydrostatic equations. I would recommend instead pointing to a citation for the implementation used in SCALE.

Thank you for your comment. We deleted section 2.5, and cite Shima et al. 2020 in section numerical setup (Lines 304-307) as follow:
‘The moist air fluid dynamics in this study are computed using eqs. (71)-(81) from Shima et al. (2020). The calculations utilize SCALE’s dynamical core, employing a forward temporal integration scheme. This approach is implemented on an Arakawa-C staggered grid (Arakawa and Lamb, 1977) using a finite volume method.’

Other comments (based on the track-changes document line numbers)

- Abstract: Consider adding an introductory sentence at the start to introduce what electro-coalescence is or its impacts, something like line 520-521 in the discussion section.

Thanks for the important suggestion, we added a sentence to briefly introduce the electro-coalescence effect at the beginning of abstract as follows:
‘The phenomenon electric fields applied to droplet induce droplet coalescence was called electro-coalescence effect.’

- L17: “… particle-based microphysics method: the super-droplet method…”

Thanks for comment. We rephrase this sentence into:
‘To investigate this effect, we applied a weak electric field to a cumulus cloud using a size-resolved cloud model that employs the super-droplet method.’

- L20: “…dynamic process. We assume…”

Thanks for the comment. We rephrase the sentences to make them flow:
‘… dynamics. In the simulation, we assume…’

- L39: “rain formation” (was not corrected from first review)

Sorry for our mistake. We modified ‘cloud formation’ to ‘rain formation’ in Line30.

- L58-59: This sentence doesn’t make sense, especially the added phrase.

Thanks for the comment. we rephrase this sentence to introduce how electro-coalescence happened.
‘In cumulus clouds, vertical convection causes positive charge droplets from the upper boundary and negative charge droplets from the lower boundary to mix, leading to electro-coalescence.’

- L74: What would it mean to “eliminate” the Greenfield Gap? Do you mean that there would no longer be fewer particles in this size range?

Thank you for your valuable feedback. By ‘eliminate the Greenfield Gap’, we intended to convey that the enhanced collision efficiencies due to the electro-coalescence effect would significantly mitigate the traditionally reduced scavenging rates for particles in this size range. However, we acknowledge that ‘eliminate’ may not be the most accurate term. Therefore, we have replaced it with ‘reduce’ at L75 to better reflect the intended meaning. Thank you for bringing this to our attention.

- L291-292: Can you comment on this range (0.1-10um) and its correspondence to the Greenfield Gap?

Thank you for your comment. We have added the connection between the 0.1-10 μm range and the Greenfield Gap as follows:

‘The results indicate that the primary range for electro-coalescence is approximately 0.1 μm to 10 μm, which encompasses the Greenfield Gap.’

- L361-362 and L367-368 seem redundant
- L375 seems like an odd place for this statement. It would be better to put it in the data availability section and remove the url here.
- L450-451 is redundant with the figure caption.

Thanks for your important suggestions. We removed the redundant part and move the url to data availability section.

Reviewer 2:

General comments:

Overall, the manuscript is improved compared to the original submission, however neither all of the key points raised in the first round of review, nor the readability issues were addressed in a fully satisfactory manner.

We sincerely appreciate the invaluable and constructive feedback provided by Reviewer 2. We have addressed all the points raised and believe that the current manuscript has been significantly improved by incorporating the review comments, further elaborating on the benefits of SDM, and resolving the technical issues.

The manuscript still contains numerous technical flaws in punctuation, grammar, symbol and physical units mismatches. My first earlier comment was that "the choice of the particle-resolved method is not explained -what are the benefits, tradeoffs, limitations as compared to other modelling techniques, in the very context of modelling charged-particle interactions". The introduced change vaguely states that "particle-based microphysics method, which calculates the electro collision-coalescence kernel in real time, offers more detailed insights into droplet behavior influenced by electrostatic forces, surpassing the bin method that relies on lookup tables (Khain et al., 2004), while also demanding less computational resources". Computational demands are not explored in the present paper at all. Lookup tables are an implementation detail and can be used with both bin- and particle-resolved methods for speeding up evaluation of multi-dimensional formulae. On the other hand, particle-resolved models surpass bin-resolved models in the tractability of aerosol-cloud interactions, what is partly leveraged in the present study (while Khain et al. 2004 resorted to prescribed initial droplet size distributions). Similarly, the fidelity of representation of particle collisions is argued in literature (by the paper co-author!) to be superior in particle-resolved models (e.g., section 4.4. in Liu et al. 2023, https://doi.org/10.1007/s00376-022-2077-3). It would be worth to elaborate on it both in the Introduction as well as in the last paragraph of the Discussion section. As of now, sections Discussion and Conclusion do not refer to the particle-resolved methodology at all. It seems as all the discussion and conclusions apply equally well to bin methods - if so, worth highlighting.

Thank you for your thoughtful explanation and attached reference.

- Re the technical issues: We addressed and corrected them in the text and the Detailed comments section.
- Re the elaborate of SDM: We agree your point about elaboration the reason of choose particle-resolved method worth to mention in the Introduction, Discussion and Conclusion section. Sorry didn't make sense on last modification.

  In Introduction section, we elaborate SDM as follows:

  l.91-93: 'Lagrangian particle-based approaches accurate solutions for the collision-coalescence process compared to bin microphysics schemes, as they overcome the limitations imposed by the assumptions of bin schemes (Grabowski et al., 2019; Liu et al., 2023).'

  We rephrase the sentence in Description of the cloud model to clarify the benefit, the limitation and trade off of SDM compare to bin scheme, as follows:

l.101-107: 'In this study, we assume that the charged droplets are well-mixed in the warm cumulus cloud and focus on the electro-coalescence effect. A Lagrangian particle-based cloud model is used with the particle size resolved treatment following the SDM by Shima et al. (2009, 2020). Compared to bin microphysics schemes, the Super-Droplet Method (SDM) eliminates numerical diffusion and provides more accurate solutions for well-mixed volumes (Grabowski et al., 2019). Despite its sensitivity to super-droplet initialization and a higher variance than observed in reality (Liu et al., 2023), SDM is well-suited for this study.'

We added the superior of SDM in Discussion as follows:

l.467-469: 'The particle-based approach SDM provides explicitly cloud-aerosol interaction simulation, such as the role of CCN in rain formation (Grabowski et al., 2019). According to our simulation results, the electro-coalescence effect on precipitation is sensitive to the aerosol concentration.'

We also mention the importance of SDM in Conclusion as follows:

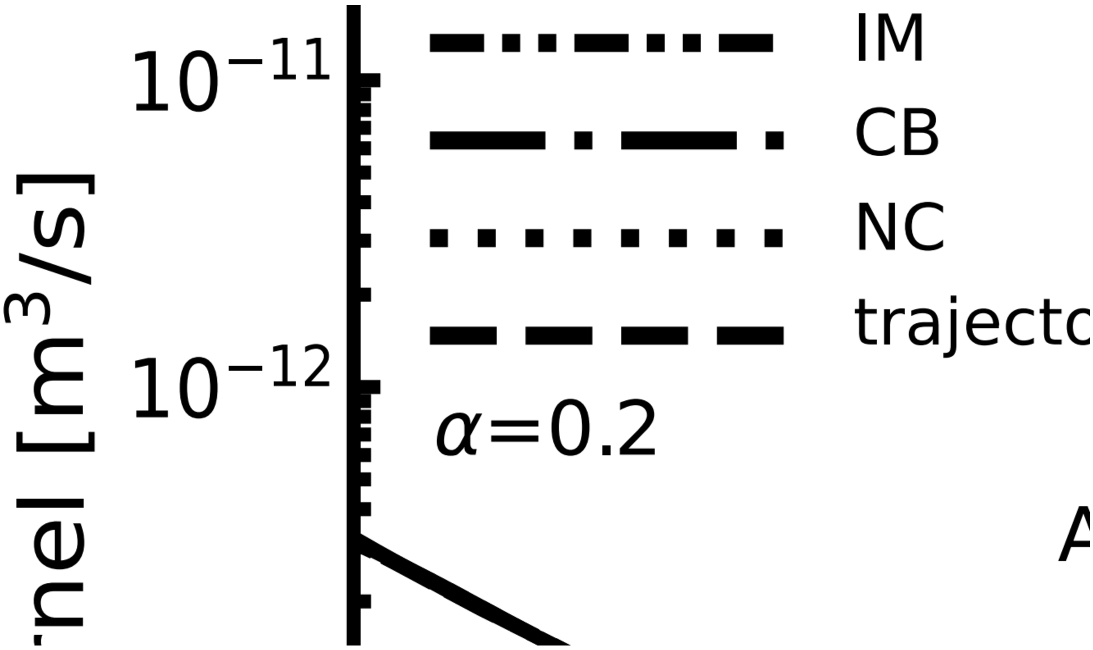
l. 479-481: 'A new simulation with the exact treatment of the electrostatic force for opposite sign charge case based on the particle-based approach SDM provides a good estimation of the effect of electro-coalescence in the Greenfield gap region.'

Despite introducing changes to the model code and providing new source archive at Zenodo, the title and text still refers to the version number from the original submission - a change in version number is needed.

Thank you for your comment. The changes made to the model code are optimizations in code writing and do not affect the microphysics processes or simulation results. Therefore, we believe we are still working with SDM version 2.3.0 and did not alter the version number in the text and title.

Despite authors' statement on provision of vector graphics in figures, provided pdf evidently contains raster graphics.

Thanks for the comment. We checked the graphics in manuscript and replaced the raster graphics to vector graphics. (e.g., zoom of fig.1:)



Detailed comments:

- l. 16: remove size-resolved (it is unclear what size refers to)
  Thanks for your comment. Removed 'size-resolved' in the sentence.
- l. 17: add "probabilistic" before "particle-based"
  Thanks for your comment. Added 'probabilistic' before 'particle-based'.
- l. 17-20: split into multiple sentences, suggest adding information on the processes represented in the microphysics model.
  Thanks for the suggestion. Split into three sentences and added microphysics processes in the model.
- l. 27: rephrase "droplet charge is lower charge limit"
  Thanks for the comment. rephrase 'droplet charge is lower charge limit' to 'droplet charge is at the lower charge limit'
- l. 38: do these references support "and even cloud chemistry"?
  Thank you for comment. These references do not support 'and even cloud chemistry', we removed this part in the text.
- l. 39: missing space in "Chapter15"
  Thanks for comment. Added space in 'Chapter 15'.
- l. 42: is the non-chronological order of references intentional?
  Thanks for your comment. Not intention, sorry about that…We modified these references in chronological order.
- l. 55: why is the µm unit typeset in different font?
  Thanks for your comment. It is modified to the same font.
- l. 57: rephrase "opposite sign charged affect by"
  Thanks for the comment. rephrase this sentence to:
  'In cumulus clouds, vertical convection causes positive charge droplets from the upper boundary and negative charge droplets from the lower boundary to mix, leading to electro-coalescence.'

- l. 65: rephrase “series of trajectory simulation work by”
  Thanks for the comment. rephrase this sentence to:
  ‘The trajectory simulation studies by’
- l. 65: is the non-chronological reference order intentional?
  Thank you for the comment. Rephrase the references in chronological order.
- l. 71: “micrometer” used here, but “micron” elsewhere
  Thanks for comment. Replace ‘micron’ and ‘micrometer’ by ‘μm’.
- l. 85: “5% of maximum charge amounts of natural droplets” seems unclear, also perhaps better not to start a sentence with a digit
  Thanks for the comment. Rephrase the sentence to:
  ‘Khain04 set a charge rate equal to 5% of the maximum charge of natural droplets, which is 2.5 times larger than the values reported by Zhou et al. (2007), to study electro-coalescence impact on rain enhancement and fog elimination.’
- l. 90: avoid using surnames as person indications, these should be used only as reference labels (also, plural “simulations”?)
  Thanks for your comment. Modified surnames by references and plural:
  ‘…was used in Andronache (2004) and Wang et al. (2015) simulations.’
- l. 92: what “real-time” refers to? is it different than in other cited studies? (it is elaborated on in l. 101, but still unclear why so central - super-particle method could also use a lookup table, it is just a way of speeding up evaluation of a multi-argument function...)
  Thank you for your comment. We agree with your point and rephrase this sentence as follows:
  l.91-96: ‘Lagrangian particle-based approaches accurate solutions for the collision-coalescence process compared to bin microphysics schemes, as they overcome the limitations imposed by the assumptions of bin schemes (Grabowski et al., 2019; Liu et al., 2023). In this study, we estimate the effect of electro-coalescence from J_z on warm cumulus clouds by an exact treatment of electric forces using the conducting sphere (CS) method, using the Super-Droplet Method (SDM), a Lagrangian particle-based cloud microphysics scheme.’
- l. 95-97 the “will be addressed in future work” statement seems awkward for an introductory section
  Thanks for the comment. We removed this pert from the sentence.
- l. 103: super-droplet method was already mentioned, but the acronym is only defined here - move the definition to first occurrence
  Thanks for the comment. We moved the acronym to the introduction section (Line 95).
- l. 104: -droplets vs. -droplet, also since SDM was just defined, why not start using the acronym?
  Thanks for the suggestion. We replaced all the ‘super-droplet method’ by the acronym after first defined.
- l. 109: multiplicity was never mentioned before
  Thanks for the comment. the ‘multiplicity’ indicate the ‘super-droplet represents multiple droplets with the same attributes and position’ at the begging of the

sentence.

- l. 113: specifying particular chemical composition seems misleading at the level of method description -there is nothing in the method that constraints it to ammonium sulphate!
  Thanks for the comment. We removed ‘ammonium sulphate’ and rephrase the sentence.
- l. 126: remove “and”
  Thanks for comment. Removed ‘and’ by ‘,’.
- l. 135: non-chronological order of references; also: should be Rogers & Yau instead of Yau & Rogers
  Thanks for the comment. Rephrase the references in chronological order and replaced ‘Yau & Rogers’ by ‘Rogers & Yau’. Sorry for didn't correct this issue in the first round referee.
- l. 147: worth mentioning here that charge effects on the equilibrium saturation vapour pressure are neglected (see, e.g., Weon & Je 2010, https://doi.org/10.1063/1.3430007) - then at least this section would be somewhat justified in the paper
  Thanks for the comment and important suggestion. We mention charge effect on the equilibrium saturation vapour pressure as follows:
  ‘Note the charge-induced reduction in surface tension decreases the equilibrium vapor pressure (Weon & Je 2010).’
- l. 178: “viscosity rate” ~ “dynamic viscosity”

  Thanks for the comment. Replaced ‘viscosity rate’ by ‘dynamic viscosity’ at line 179.
- l. 187: is this what is meant: “droplet accepts less than 1 elemental charge”?
  Thanks for the comment. We rephrase this sentence to make it more clarify and readable as follows:
  ‘The rear collision range is relevant for droplets smaller than 0.1 μm, the droplets typically accept fewer than 1 elemental charge, meaning the electric force does not significantly impact the collision process.’
- l. 194: is μ_a the same as μ defined in line 177?
  Thanks for the comment. Yes, they are same, deleted repeat definition.
- l. 204: r_{nt} is defined as dimensionless ratio in (12) but eq. (13) suggests it should have length dimensionality
- Thanks for your important comment. We mixed up two symbols for the distance of droplets. The symbols of eq (12) and (13) are corrected.
- l. 209: ditto - r2nt is added to dimensional r2
  Thanks for the comment. Ditto.
- l. 210: period at line beginning, unopened parenthesis...
  Thank you for your review. These technical issues have been corrected.
- l. 233: ϵ_0 was already defined in l. 205
  Thanks for the comment. Deleted repeat definition.
- l. 233-235: two consecutive sentences begin with almost the same phrase

Thanks for the comment. Rephrase two sentences as follows:

'The air breakdown voltage: $U_b \sim 3\times10^6\ \mathrm{Vm^{-1}}$, determines the maximum charge that cloud droplets can carry (Meek and Craggs, 1953). Consequently, the maximum charge that droplets can carry…'

- l. 237: grammar: "we following Andronache (2004)"

  Thanks for the comment. Rephrased this sentence as follows:

  'To simulate droplets in a weak electric field, we followed Andronache (2004) and described the mean charges on the larger and smaller droplets in a pair as a function of their radii as follows'

- l. 281: R symbol mismatch - previously used for particle radius
- l. 295: "and the time derivatives for condensation/evaporation" - predicate missing?

  Thanks for the comments. We deleted the initial section 2.5 followed the suggestion of reviewer 1.

- l. 313-314: "size distribution were adjusted to 3, 6 or 9 times" sounds as if size parameters were adjusted

  Thanks for the comment. We rephrase this sentence, split the data source and aerosol background factor into two sentences as follows:

  'The aerosol number concentration and size distribution were based on the data provided by Van Zanten et al. (2011) for the RICO intercomparison case. Note that aerosol concentrations are multiplied by factors of 3, 6, or 9, depending on the aerosol background conditions.'

- l. 321: shouldn't this section go before 2.6?

  Thanks for the comment. The 'Numerical setup and schemes' section is moved before 'Design of our numerical experiment' section.

- l. 324: "code is not accessible through this site" should better go into the preceding parenthesis

  Thanks for the comment. The url of SCALE and the sentence about accessible were moved to the 'Code and data availability' section to keep text coherence.

- l. 343: "common" ~ "coupling"?

  Thanks for comment. The 'common time step' refers to the uniform time interval at which the primary calculations of the SDM are updated, followed the jargon of section 5.4 'Operator splitting of the time integration' of Shima et al. (2020).

- l. 341: worth rephrasing "processes for aerosol/cloud/precipitation particles are integrated separately" as it seems misleading - aerosol, cloud and precipitation particles are not treated separately

  Thanks for comment. We removed 'for aerosol/cloud/precipitation particles' for disambiguation.

- l. 350 (again): please elaborate what are "Lagging processes" and "overall system dynamics"

  Thanks for the comment.

- Re the 'Lagging processes':

  Lagging processes refer to those processes that occur on shorter timescales within the simulation, such as condensation and evaporation. These processes can change rapidly and thus require more frequent computational updates to ensure accurate representation. Prioritizing these processes in the computation helps to capture their dynamics effectively and prevents numerical instability.

- Re the 'overall system dynamics':

  Overall system dynamics encompass the behavior and evolution of the entire system over time, considering the interactions and feedbacks between various components and processes. In our simulation, this includes the interactions between fluid dynamics, cloud microphysics.

  To enhance readability and comprehension, we have rephrased the sentence as follows:

  'Processes with shorter timescales are prioritized in the computation to ensure they accurately capture their subsequent impacts.'

- l. 350: "prioritized in computational priorities" – pleonasm

  Thanks for the comment. Removed 'priorities'.

- l. 398: rephrase "The results of the domain and ..." (domain- and ensemble-averaged?)

  Thanks for the comment. Rephrase the sentence beginning to :'The results of the domain water path, averaged over 50 ensembles…'

- l. 401: "cloud" ~ "clouds" (or otherwise "produces")

  Thanks for the comment. Modified 'cloud' to 'clouds'.

- l. 454: suggest removing "will be evaluated in our next paper"

  Thanks for the comment. Removed 'and will be evaluated in our next paper'.

- l. 459: "103" ~ "on the order of 100" ?

  Thanks for the suggestion. replaced '103' by 'on the order of 100'.

- l. 468: rephrase "two factors larger"

  Thanks for the comment. Rephrased to 'is twice as large as'

- l. 472: what "the cloud model" refers to?

  Thanks for the comment. Replaced 'cloud model' to 'cloud microphysics scheme'.

- l. 489: "the effective radius" was never introduced, in general this sentence appears quite abruptly here (perhaps introduce subsections in section 4?)

  Thank you for the comment. We agree that this sentence is abrupt and have removed it. We have retained the discussion of the simulation results.

- l. 505: acknowledge that alpha was arbitrarily prescribed here

  Thanks for the comment. We have added the acknowledgment of alpha as follows

  '…as a function of the arbitrarily prescribed charging rate α...'

- l. 506: remove "we leave them for the future work"

  Thanks for the comment. We have removed 'and we leave them for the future work'.

- l. 508: please elaborate on how it can be done and how this works brings us closer?

  Thanks for the comment. We elaborate on the outlook of this study in section conclusion as follows:

‘Cloud radiation feedback is one of the sources of uncertainty in the climate model (Zelinka et al. 2017). The electrostatic force effect parameterization for different cloud types should be indicated to improve climate model accuracy. This study reveals the electrostatic force effect on warm cumulus clouds, contributing to the parameterization of electrostatic microphysical processes.’

- l. 550: Davis 1964a - is it different from Davis 1964b, if so, add needed information
  Thanks for the comment. We rechecked they are the same, deleted repeat references.
- l. 552: Davis 1964b - add DOI: https://doi.org/10.1093/qjmam/17.4.499
  Thanks for the comment. We have added the doi to reference of Davis 1964b.
- l. 601: add permanent URL: https://www.jstor.org/stable/113853
  Thanks for the comment and suggestion. Added url to reference of Rayleigh 1878.
- l. 608: “eulerian” ~ “Eulerian”; “lagrangian” ~ “Lagrangian”

  Thanks for the comment. We corrected the spell mistake at the reference of Sato et al. (2018).

l. 657: it is Rogers & Yau, not Yau & Rogers (already pointed out in the first round of review)
Thanks for the comment. We apologize for the oversight. We have corrected the reference to "Rogers & Yau" as suggested.