Response to RC1 of manuscript egusphere-2023-2507 submitted to Geosci. Model. Dev.

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General comments:

This study investigates the impact of enhanced coalescence due to droplet charge in high-fidelity simulations using the superdroplet method. While the methodology and results are thoroughly presented, the authors' key assumption of instantaneous droplet charging is not well-founded or discussed, and likely leads to a strong overestimate of the impact of droplet charge on precipitation. Similarly, the authors do not attempt to disentangle purely microphysical effects from flow field variability that stems from microphysics-flow coupling (vs. using a method like piggybacking). These reservations about the scientific merits combined with more minor concerns about the writing itself (including typos and inappropriate references) lead me to recommend major revisions before this paper be considered for publication.

We greatly appreciate the invaluable and constructive feedback provided by Reviewer #1. We have acted upon all the points raised. We believe the current manuscript is greatly improved through addressing the review comments and further elaborating the methods and results.

Specific comments:

1. The selected references for describing the importance of droplet coalescence (e.g. line 35) are not appropriate general references for this statement. For instance, Rosenfeld 2008 specifically concerns the a controversial mechanism of aerosol convective invigoration, which has relatively little to do with coalescence. Craig 1995 discusses radiative effects which are also not inherently specific to coalescence. Forbes & Clark does not even appear in the references. More appropriate citations would include review papers, chapters from the IPCC or a classical cloud microphysics textbook, or studies which specifically investigate droplet coalescence. Likewise in line 483: the stated impact of increasing aerosol concentration is the Twomey effect, and should not be attributed to Rosenfeld 2008!

   Thank you for your important comments and suggestions, we agree that the reference to droplet coalescence is not appropriate for the point we discussed. we modified the reference for describing the importance of droplet coalescence in line 35 and line 483 as follows:

   'Droplet coalescence is one of the main processes leading to precipitation and even cloud chemistry, affecting cloud microphysics and thereby changing the global radiation budget (Pruppacher and Klett, 2010, Chapter15; Grabowski and Wang, 2013; IPCC AR6 WG1 Ch7, 2021).'

   'An increase in the aerosol concentration decreases the effective radius by increasing the concentration of small droplets, which could have a significant impact on cloud formation (Twomey, 1974)'

2. I have some issues with your notation and definitions in section 2.4. Line 144: Given that both E0 and Ees are presumably efficiencies varying between 0 and 1, they should be multiplicative rather than additive. I am also confused by the notation of Rp, rp, QR, and qr, as the text describes these radii and charges as being general to "large droplets" or "small droplets", whereas I would imagine them to be descriptive of the larger droplet R and smaller droplet r for a given pair (R, r). What does the subscript "p" refer to?

   Thanks for your comments. We will answer your comments point by point:

Re the presumed additive efficiencies: We incorporated both electro-coalescence efficiency and general coalescence efficiency using the approach outlined by Andronache 2004 (http://dx.doi.org/10.1016/j.jaerosci.2004.07.005). This method is the general way to handle electro-coalescence efficiency.

Re the notation of R_p, r_p, Q_R, and q_r: Sorry for misleading symbol, we use subscript "p" represent "particle", but as you pointed out it's confusing, we remove subscript "p" and replace "large droplets" or "small droplets" with "larger droplets of droplets pair" & "smaller droplets of droplets pair" in the text.

3. The assumption in 212-213 that droplets charge instantaneously following coalescence seems like a major flaw in this study which would lead to a strong overestimate of the effects of electro-coalescence in an LES. Given that you are using the superdroplet method for this study, you should in fact be able to model the time response of charging on a given superdroplet as an additional attribute! This would provide a much more trustworthy study of the effects of droplet charge on coalescence that could actually be used to quantify and suggest whether this effect is notable. As it stands however, this assumption undermines the findings of this study and is not adequately discussed as a limitation or confounding factor in the abstract or conclusions. Furthermore, the values chosen for alpha in the numerica l experiments are not well-justified with values or ranges measured in real clouds.

Thank you for your important comment. The droplets get charged instantaneously is a main assumption of this work, we make this assumption base on following two reasons:

- Based on the findings of Zhou et al. 2012, the charging time for 10-micrometer droplets is approximately ten minutes, which is relatively short compared to the overall development and precipitation duration of clouds. Moreover, for the droplets in our simulations, we have set a maximum limit of 50 elementary charges for droplets of any radius to constrain the impact of electro-coalescence effects from exceeding our estimates. Notably, 50 elementary charges are significantly less than the maximum charge that droplets can carry; observational data from Beard et al. 2004 reveal that real stratocumulus droplets of 10 micrometers carry about 100 elementary charges. Therefore, our assumption of instantaneous charging is based on realistic observations and numerical simulation results.
- As the reviewer suggested, we could simulate the charge amount and charging time dynamically as attributes of a super-droplet. However, because the electric potential gradient within clouds and the charge carried by droplets interact, the droplet's charge is not solely a function of time. As the charge attribute of a super-droplet changes at each time step, the cloud's electric potential gradient would also change, thereby affecting the charging efficiency. Addressing this would require significantly more computational resources. Currently, our parametrization is an initial attempt, and we aim to use simpler assumptions to model the electro-coalescence effect and obtain preliminary results. In future work, we also plan to parameterize the prediction of droplet charge.

4. I also take issue with the comparisons made between different simulations given that the flow-field and microphysics are fully coupled. A more appropriate way to analyze the purely microphysical effects of electro-coalescence would be through the common technique of piggybacking, as other studies have shown that differences due to small perturbations to the flow field often outpace differences related to microphysics effects. I do like the approach of using 50 ensemble members to analyze statistics of the superdroplet simulations, but I'm not convinced that this would help isolate

microphysics from flow-field variability. Furthermore, in section 2.5 and 2.6, is it not clear whether the simulations performed are DNS or LES as there is no sub-grid scale turbulence model (line 340), nor is there any mention of what impact neglecting SGS turbulence would have on the results.

Thank you for your insightful comments. Regarding the disentanglement of purely microphysical effects from the flow field, we recognize the efficiency of the piggybacking technique in isolating microphysical impacts from dynamic interactions, as demonstrated in other studies. In this study, we constructed idealized simulations specifically designed to highlight the electro-coalescence effects on warm cumulus clouds. The results indeed showed a significant difference between the scenarios with and without electro-coalescence under identical flow conditions, emphasizing the distinct influence of electro-coalescence. For future work, we plan to incorporate the piggybacking technique to further isolate and verify the microphysical effects from flow-field variability. This will enhance our understanding and ensure a more robust analysis of the interplay between microphysics and atmospheric dynamics.

Re the simulations performed as DNS or LES, we are using SGS for turbulence model for dynamics, not using any SGS turbulence model for cloud microphysics, such as collision-coalescence enhancement, velocity fluctuation, and supersaturation fluctuation, the simulations performed is LES. We clarify the simulations performed is LES and the impact of neglecting sub-grid scale (SGS) turbulence as follows in line 360:

'We are employing a subgrid-scale (SGS) turbulence model for dynamic processes, but not for cloud microphysics processes such as collision-coalescence enhancement, velocity fluctuations, and supersaturation fluctuations. This approach may lead to an underestimation of the collision rate of charged droplets (Lu & Shaw 2015). The simulations conducted are based on the Large Eddy Simulation (LES) methodology.'

5. In general when discussing figures and results, details from the figure caption which describe the various lines are repeated in the full text unnecessarily (e.g. lines 240-245, line 356-364, and elsewhere throughout). This repetition should be removed and avoided.

Thank you for your comment, we removed the repeat caption of plots in the text and rephase this sentence for each figure such as:

'Figure 1 displays a comparison of the collision-coalescence kernel for droplet radii of 40 μm (black lines), 20 μm (green lines), and 10 μm (red lines) across different calculation methods. The plots vary by line style to represent different analytical treatments and the inclusion or absence of static electric forces, with specific settings for the droplet charging rate shown in Figures 1(a) and 1(b).'

6. Lines 401-406 suggest that the trend going from LA to MA conditions is opposite from the trend going from MA to HA conditions, when the figure in fact indicates that the trend is consistent. Increasing the aerosol concentration appears to uniformly delay and reduce the precipitation quantity.

Sorry for the inconsistent description and figures, we rephase the conclusion about precipitation on different aerosol background in lines 409-416 as follow:

'Under NC settings, the Twomey effect demonstrates that higher aerosol concentrations lead to smaller particle radii in clouds, reducing precipitation efficiency. Conversely, when electrostatic forces are introduced, these higher aerosol concentrations substantially enhance precipitation across different scenarios. Specifically, in high aerosol (HA) conditions, the precipitation enhancement reaches 782% over the no charge (NC) setting; for medium aerosol (MA) conditions, it's 467%

higher; and for low aerosol (LA) conditions, the increase is 110%. This illustrates the significant role electrostatic forces play in modulating cloud dynamics and precipitation responses to aerosol variations.'

Technical comments

- Line 33: "play a key role in cloud formation" should be "rain formation"

  Thanks for the comment, we replaced "cloud formation" with "rain formation" in line 27.
- Line 62: the Greenfield Gap should be concisely defined

  Thanks for your suggestion, we added a concisely definition of Greenfield Gap in lines 69-71:

  'The so-called Greenfield gap, identified by Greenfield (1957), describes the reduced concentrations of particles in the 0.1 to 1 micrometer size range. Greenfield gap could be eliminated with sufficient charging of the droplets.'
- Line 200: there is an extra close-parentheses ")" after Khain04

  Thank you for your comment, removed extra ")" in line200.
- Line 203: there is an extraneous comma after "the IM treatment"

  Thank you for your comment, removed extra "," in line203.
- Line 228: if alpha is a ratio, it should be unitless and have a maximum value of 1. Can you clarify the definition here?

  Thank you for your comment, the alpha is an empirical parameter, when alpha=2 for average conditions of strongly electrified clouds and has an upper limit of alpha=7 that can occur by conduction charging (Pruppacher & Klett, 1997, Chapter 18, based on available observations). we rephase the alpha definition part(Lines 242-244) as follow:

  'The charging rate α is an empirical parameter (α is referred to herein as the droplet charging rate) that varies between 0, which represents neutral particles, and 7, which represents highly electrified clouds associated with thunderstorms (Andronache, 2004).'
- Line 305-306: Why are the number concentrations written this way, as "3 x" something?

  Thanks for your comment, we agree that this statement is unreadable, and we have rephrased the sentence defining aerosol number concentration in lines 312-315 as follows:

  'The aerosol number concentration and size distribution were adjusted to 3, 6 or 9 times from that given in Van Zanten et al. (2011) for RICO intercomparison case. The aerosol number concentration and size distribution is given by a bimodal log-normal distribution: The particle number concentrations of the two modes are $N_1 = 90$ cm^{-3} and $N_2 = 15$ cm^{-3}, respectively.'
- Line 312-313: "SDM requires less computational cost..." compared to what? Be specific; it requires more computational cost than a bulk method!

  Thanks for comment, we clarified SDM requires less computational cost than a bin model in line 325-326 as follow:

  'SDM requires less computational cost to accurately simulate clouds and precipitation compare to bin scheme (Shima et al., 2009)'
- Equation 27: what is p?

  Thanks for the comment, we remove the useless subscript 'p' in the formula.
- Lines 433-435 are repeated from earlier in the paragraph.

  Thanks for comment, removed repeat sentence in line436.
- Lines 485-490 are a very good summary of key findings and insights from this study

Thank you so much for your encouragement!