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Title: Charging and Coagulation of Radioactive and Nonradioactive Particles in the Atmosphere
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We appreciate the contribution of the Reviewer 2 to improving the quality of our manuscript. Our responses follow the order of the comments provided by the Reviewers. Changes that have been made in the manuscript are described as part of the responses.

Reviewer 2

1. In this work authors proposed three approaches for the description of charging and coagulation of radioactive and nonradioactive particles in atmosphere. The work is based on recent theoretical developments by Kim et al. (2014, 2015) in which, it was found that, electrostatic interactions caused by radioactivity can significantly modify the particle charge distribution. In the present work, mutual effects of charging and coagulation were examined. The applicability and accuracy of the new approaches depends on several factors, such as the size of particles and the initial conditions, while the computational cost may differ by ~ 2 - 3 orders of magnitude from each other. The proposed approaches can be employed in various studies, such as, the influence of radionuclides on electrification phenomena in atmosphere.

Overall, I believe that the present work is of a good quality and it is suitable for publication. Apart few minor points described below, the manuscript is well written and reasoned. The analysis of the results is very detailed, while the limitations, accuracy, and the range of applicability of each approach has been carefully examined. Comparison of theoretical predictions with experimental results seems to validate the presented approaches.

Response: We thank the Reviewer for the overall positive evaluation of our manuscript, as well as for providing useful comments to improve the quality of this manuscript. Our specific responses to the concern raised by the Reviewer are given below, following the Reviewers' comments.

2. As a general comment, throughout text and in figures, particle sizes are expressed sometimes in micrometers (µm) and some other times in nanometers (nm). For the readability of the paper, I believe that authors should decide to use consistently only one of the two units.

Response: We thank the Reviewer for the general comment. As suggested by the Reviewer, the manuscript has been revised with consistency in the units, to improve its readability. Specifically, we decided to use µm for particle size.

3. Symbol n_k first appearing in Eq.-7 has not been defined. Is it obtained from the decay rates $n_{k,j}$ which was defined before? The same for symbols $\beta^{+}_{k,j}$ and $\beta^{-}_{k,j}$ in the same equation, which are different from $\beta^{+}_{k,j}$ and $\beta^{-}_{k,j}$ appearing before.

Response: We thank the Reviewer for pointing out it. The symbols, mentioned by the Reviewer as n_k and $n_{k,j}$, are actually the Greek letter η_k and $\eta_{k,j}$, respectively. Both symbols represent the radioactivity of particles. As written in lines 133-134 of the revised manuscript, *k* and *j* are the indices of the size and number of elementary charges of particles, respectively. Thus, η_k represents the radioactivity of size *k* particles while $\eta_{k,j}$ denotes that of size *k* particles with elementary charge *j*. To avoid confusion, we replaced the symbol η by *A*. Similar explanations are applicable to the symbols representing the ion-particle attachment coefficients, $\beta^+_{k,j}$, $\beta^-_{k,j}$, $\beta^+_{k,J}$, and $\beta^-_{k,J}$. The first 2 of these terms have low *j* as subscript, which is the number of elementary charges, while the last 2 terms have capital *J* as subscript which is the mean charge of particles. This distinction is made in the text on line 161 of the revised manuscript.

4. In Eq.-10 in the definition of the time-evolution of particles, the symbol n(x, j) was picked to denote the number densities of particles. This may be confusing since the same symbol n has been used before to denote number concentration of charges. Using instead symbol N similar to Eq.-11 may be more clear.

Response: We thank the Reviewer for the comment and suggestion. In equations 1-9, "*n*" is only used to represent the concentrations of ions. We admit that $n(x, j)$ is similar to some symbols to denote the ion concentrations such as n_+. Literature reports [e.g., Oron and Seinfield (1989)], however, show that *n(x, j)* typically represents the number density of particles while *N* normally denotes the concentration of particles in size bins. We believe that $n(x,j)$ should not change. Alternatively, the symbols of the ion concentrations have been modified from n_+, n_-, and n_0 to n^+_{ion}, n^-_{ion}, and n^0_{ion} in the revised manuscript to avoid confusion, respectively.

Oron, A., Seinfeld, J. H.: The dynamic behavior of charged aerosols: III. Simultaneous charging and coagulation, J. Colloid Interface Sci., 133, 80-90, doi:10.1016/0021-9797(89)90283-X, 1989.

5. In Eq.-11, the property distribution factor $\eta_{l,m}$ has not been described. Also, it may be confusing that the same symbol was used before for the decay rate of the radioactive particles.

Response: Lines 202-203 of the revised manuscript has been modified to describe the property distribution factor. We agree with the Reviewer's opinion on the symbol of the particle decay rate. To avoid confusion, the radioactivity symbol, η has been changed to *A* in the revised manuscript.

6. Page 23805, Line 22-23: Equation numbers (in parentheses) used for each method seems to be redundant at this place since they have given in the previous paragraph just above.

Response: We thank the Reviewer for the comment. The equation numbers have been removed from the revised manuscript.

7. Page 23808, Line 22: No units are given for the diffusion charging rate.

Response: As pointed out by the Reviewer, the units of the diffusion charging rate have been added to line 327 of the revised manuscript.

8. For the case of charging and coagulation kinetics of nonradiative particles, approach 1 was evaluated using the approximate analytical solutions suggested by Alonso (1999) (paragraph 3.2.1). I believe that the authors should shortly describe the previous approach and how it differs from the present one. At this end, it should become transparent the novelty of the present approach. Moreover, it should be clarified, why the results of the present approach are compared to the approximate analytical solution and not to the corresponding numerical solution of the rigorous population balance equations given by Alonso.

Response: We thank the Reviewer for the comments. Short descriptions of the work of Alonso et al. (1998) and Alonso (1999) are given in the section "**1. Introduction**." (Lines 115-118 of the revised manuscript). To additionally explain the work of Alonso and his coworkers, text has been added to the section "**3.2.1. Bivariate Population Balance Model for Approach 1**(lines 344 - 352 of the revised manuscript)."

Alonso (1999) found that results of the approximate analytical solutions are in good agreement with those of the numerical solution of the population balance equation of Alonso et al. (1998). This finding suggests that the analytical approach of Alonso (1999) may be useful to assess the validity of new numerical solutions. Thus, the comparison of results of Approach 1 with those of the numerical solution was omitted. Lines 344-352 of the revised manuscript have been modified to better explain this point. The revised text reads:

"Based on the numerical approach of Alonso et al.(1998), Alonso (1999) suggested an analytical approach to simultaneously investigate charging and coagulation kinetics of nonradioactive particles, smaller than 0.02 µm in diameter. Results of the analytical approach agreed well with those of the numerical approach, but the applicability of both analytical approaches may be limited as discussed in section Introduction. The analytical approach, however, was found to be useful to validate numerical solutions of population balance equations including diffusion charging and coagulation (Alonso, 1999). In this study, the analytical approach of Alonso (1999) was used to evaluate Approach 1 because Approaches 2 and 3 are not applicable to particles smaller than 0.02 µm, as shown in Figure 2a."

Alonso, M., Hashimoto, T, Kousaka, Y., Higuchi, M., and Nomura, T.: Transient bipolar charging of a coagulating nanometer aerosol. J. Aerosol Sci., 29, 263-270, doi:10.1016/S0021-8502(97)10007-6, 1998.

Alonso, M.: Simultaneous charging and Brownian coagulation of nanometre aerosol particles, J. Phys. A: Math. Gen., 32, 1313-1327, doi:10.1088/0305-4470/32/8/003, 1999.