Title: Charging, Aggregation, and Electrostatic Dispersion of Radioactive and Nonradioactive Particles in the Atmosphere

Authors: Yong-Ha Kim, Sotira Yiacoumi, Costas Tsouris

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We appreciate the contributions of the Editor and the Reviewers toward improving the quality of our manuscript. Our responses follow the order of the comments provided. Changes that have been made in the manuscript are described as part of the responses.

Reviewer 1

General comments:

In this study by Kim et al. the authors investigate the time-evolution of radioactive particles, taking into account charging, aggregation, and electrostatic dispersion of the particles. A comprehensive population balance model has been developed and used in studying the effects of radioactivity on the time-evolution of both radioactive particles and non-radioactive background particles. In addition, simplified forms of the population balance model are developed and verified. The scope of the manuscript is suited for publication in Aerosol Research.

The manuscript itself has clearly been carefully prepared. It is quite well written and easy to follow. The results are presented in a logical manner.

I would suggest this manuscript to be accepted for publication after my comments, which are mainly technical corrections, have been addressed.

Response: We thank the Reviewer for the suggestion, as well as for providing constructive comments. All the comments raised by the Reviewer have been addressed in the revised manuscript. Our specific responses are given below, following the comments of the Reviewer. Revisions have been highlighted in the revised manuscript.

1. (L38) "Because such radiation sources can affect the particle charge and size distributions, understanding the influence of radioactivity is necessary to investigate many microphysical processes in atmospheric systems including radiation sources." I suggest reformulating this sentence slightly, as I found it difficult to follow whether it referred to microphysical processes, including radiation sources or to microphysical processes, which include radiation sources.

Response: We thank the Reviewer for the suggestion. As suggested, we have modified the sentence as follows:

Page 2 (Lines: 38-40) (Before revision): "Because such radiation sources can affect the particle charge and size distributions, understanding the influence of radioactivity is necessary to investigate many microphysical processes in atmospheric systems including radiation sources."

Page 2 (Lines: 38-39) (After revision): "Because such radiation sources can affect the particle charge and size distributions, understanding the influence of radioactivity is necessary to investigate microphysical processes in atmospheric systems containing radionuclides."

2. 3.4 Simulation: What was the time step used?
Response: We solved the differential equations with MATLAB ode15s which is a variable-step, variable-order solver of ordinary differential equations. To more clearly indicate the simulation procedure in the revised manuscript, we have added the following sentence:

Page 8 (Lines: 179-181) (After revision): "For the investigation, we used Matlab ode15s which is typically employed to solve discrete population balance equations [e.g., see Jama et al. (2020)]."

Jama, M.A., Zhao, W., Ahmad, W., Buffo, A. and Alopaeus, V., 2020. Analytical time-stepping solution of the discretized population balance equation. *Computers & Chemical Engineering*, *135*, p.106741.

3. L220: The word "respectively" is unnecessary.
Response: The word has been removed in the revised manuscript as follows.

Page 10 (Lines: 219-220) (Before revision): "However, discrepancies were found between the simulation results of the bivariate PBM executing the algorithm and those of Adachi et al. (1981) and Oron and Seinfeld (1989a), respectively."

Page 10 (Lines: 236-237) (After revision): "However, discrepancies were found between the simulation results of the bivariate PBM executing the algorithm and those of Adachi et al. (1981) and Oron and Seinfeld (1989a)."

4. L230: I suggest adding figures of these other comparisons mentioned as a supplementary.

Response: The figures have been added to the supplementary information of this manuscript. We have corrected the statement as follows:

Page 10 (Lines: 219-220) (Before revision): For other initially symmetric uni- and bi-modal particle charge distributions, the prediction results obtained from Eq. 4 including the algorithm were comparable to those of Adachi et al. (1981) and Oron and Seinfeld (1989a) because space charge was rarely formed during the simulations (not shown).

Page 11 (Lines: 244-247) (After revision): For other initially symmetric uni- and bi-modal particle charge distributions, the prediction results obtained from Eq. 4 including the algorithm were comparable to those of Vemury et al. (1997) and Verdoold and J. Marijnissen (2009) because space charge was rarely formed during the simulations (Fig. S1 and Fig. S2).

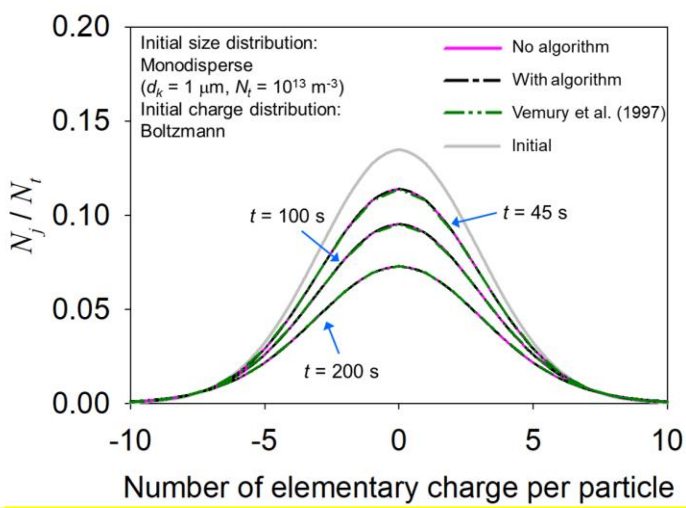

Figure S1. Time-dependent changes in the particle charge distributions predicted using the bivariate PBM including and excluding the algorithm of Fig. 2, respectively. The simulation results obtained from solving Eq. 4 were in good agreement with those of Vemury et al. (1997).

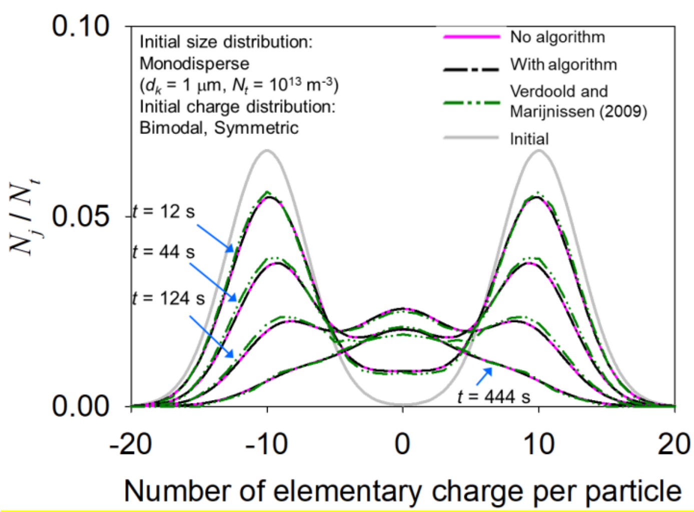

Figure S2. Time-dependent changes in the particle charge distributions predicted using the bivariate PBM including and excluding the algorithm of Fig. 2, respectively. The simulation results obtained from solving Eq. 4 were in good agreement with those of Verdoold and Marijnissen (2009).

5. L241 and L244: Incorrect use of respectively. I suggest replacing it with e.g., separately.

Response: The word has been replaced by "separately" as follows:

Page 12 (Lines: 240-241) (Before revision): "For comparison, simulations were performed with only the aggregation and electrostatic dispersion terms, respectively."

Page 12 (Lines: 257-258) (After revision): "For comparison, simulations were performed with only the aggregation and electrostatic dispersion terms, separately."

Page 12 (Lines: 242-244) (Before revision): "The results of the simulation involving both aggregation and electrostatic dispersion were closer to the measurements than those including each process, respectively."

Page 12 (Lines: 259-261) (After revision): "The results of the simulation involving both aggregation and electrostatic dispersion were closer to the measurements than those including each process, separately."

6. L285: The results not shown here could be added as a supplementary information.

Response: We have added the results to the supplementary information file and have revised the statement as follows:

Page 12 (Lines: 240-241) (Before revision): "This indirect effect was more significant for the initial distribution movement of the ^{131}I particles than ^{238}Pu particles because it took longer time to counterbalance the initial positive space-charge (not shown)."

Page 14 (Lines: 303-304) (Before revision): "The level of the indirect effect on the initial distribution movement of the ^{131}I and ^{238}Pu particles was unequal because the times needed to counterbalance the initial positive space-charge were different (Fig. S3)."

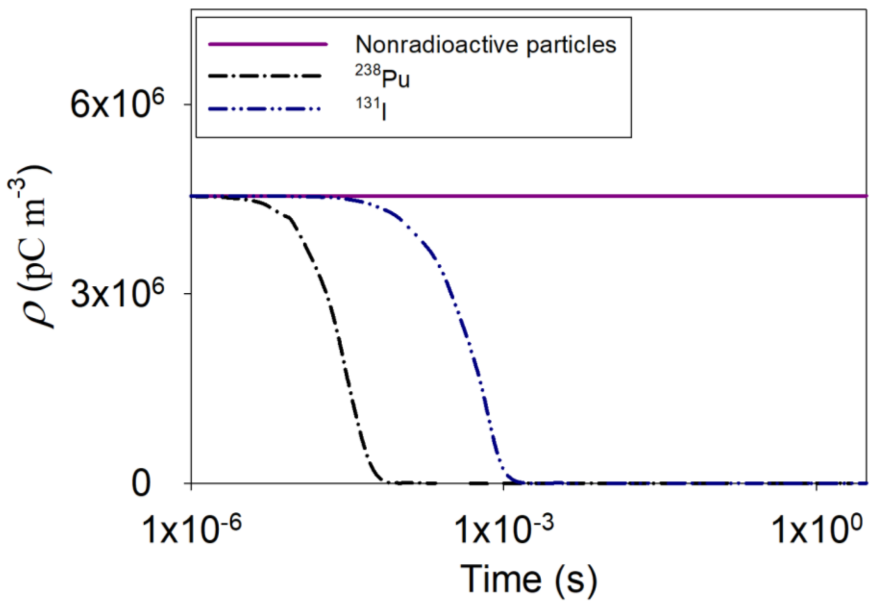

Figure S3. Changes in space charge at an early stage of the charging, aggregation, and electrostatic dispersion of radioactive and nonradioactive particles.

7. L375: How much is "slightly"?

Response: We thank the Reviewer for pointing this out. We have removed the word, and the sentence has been modified as follows:

Page 18 (Lines: 374-375) (Before revision) Compared to the bivariate PBM, the monodisperse PBM slightly overestimated the mean particle size and space charge in absolute value.

Page 18 (Lines: 393-396) (After revision) Compared to the bivariate PBM, the particle size predicted by the monodisperse PBM was larger. After 1 hour, the mean sizes predicted by the monodisperse and bivariate PBMs were 1.04 μm and 0.89 μm, respectively, indicating a difference of 16.9%. This difference increased with time (e.g., 29.7% after 3 hours).

8. L408: "monovariate" should read "bivariate"

Response: We thank the Reviewer for the comment. We have corrected the statement as follows:

Page 21 (Lines: 408-410) (Before revision) "These results suggest that the monovariate PBM can be used to include radioactivity-induced charging and electrostatic dispersion, and the subsequent effects on particle aggregation in predictive studies of radioactivity transport."

Page 21 (Lines: 429-431) (After revision) "These results suggest that the bivariate PBM can be used to include radioactivity-induced charging and electrostatic dispersion, and the subsequent effects on particle aggregation in predictive studies of radioactivity transport."

Reviewer 2

Review of "Charging, Aggregation, and Electrostatic Dispersion of Radioactive and Nonradioactive Particles in the Atmosphere" by Kim et al. 2024

The study deals with the behaviour of radioactive particles in the atmosphere. A temporal component is taken into account and the focus is on the particle concentration and number of elementary charges per particle in the atmosphere.

The manuscript has been thoughtfully written and deals with a challenging topic which is presented in a comprehensible way.

I vote in favour of the publication of the manuscript, but I would still like to have some questions answered

Response: We thank the Reviewer for the positive recommendation and comments. All the comments raised by the Reviewer have been addressed in the revised manuscript. Our specific responses are given below, following the comments of the Reviewer. Revisions have been highlighted in the revised manuscript, and our responses to the comments of the Reviewer are shown below.

Remarks/Questions

In a previous publication, reference was also made to experiments. in this publication, experiments and data from observations are quite rare. (as in: Kim, Y.-H., Yiacoumi, S., Nenes, A., and Tsouris, C.: Charging and coagulation of radioactive and nonradioactive particles in the atmosphere, Atmos. Chem. Phys., 16, 3449–3462, https://doi.org/10.5194/acp-16-3449-2016, 2016.)

Response: Dissimilar model verification approaches led to the difference the Reviewer pointed out between the previous work and the current effort. In both studies, we developed population balance models (PBMs). While the previous publication was focused on developing PBMs involving charging and aggregation, the focus of the present work was on incorporating electrostatic dispersion into the PBMs. Measurements and theories can be used to verify microphysical processes. While our previous work used both theories and measurements for model verification, in the present work, we verified electrostatic dispersion components of the PBMs with simulation results available in the literature since this approach has been frequently used for PBM verification [e.g., see Vemury et al. (1997), Xiangrong et al. (2006), and Verdoold and Marijnissen (2009)].

Vemury, S., Janzen, C., and Pratsinis, S.E.: Coagulation of symmetric and asymmetric bipolar aerosols, J. Aerosol Sci. 28, 599-611. doi:10.1016/S0021-8502(96)00462-4, 1997.

Xiangrong, Z., Lianze, W., Cheng, W. and Fenglei, H., 2006. Effect of an external electric field on the charge distribution of electrostatic coagulation. *Journal of aerosol science*, *37*(10), pp.1370-1377.

Verdoold, S. and Marijnissen, J.C.M., 2009. A 2D fixed-sectional approach to model the coagulation of (highly) charged aerosols. *Journal of Electrostatics*, *67*(4), pp.631-639.

To more clearly indicate this model verification approach, we have revised the manuscript as follows:

Page 9 (Lines: 192-193) (Before revision) Then, the bivariate PBM was verified using Whitby et al. (1965), Kasper (1981), Adachi et al. (1981) and Oron and Seinfeld (1989a).

Page 9 (Lines: 205-208) (After revision) Then, the bivariate PBM was verified using Whitby et al. (1965), Kasper (1981), Adachi et al. (1981), Oron and Seinfeld (1989a), Vemury et al. (1997), and Verdoold and Marijnissen (2009) that have typically been used in verifying electrostatic dispersion in PBMs [e.g., see Vemury et al. (1997), Xiangrong et al. (2006), and Verdoold and Marijnissen (2009)].

Radioactive decay is often a chain reaction to other unstable nuclei, which have a different activity. has this been sufficiently taken into account?

Response: We agree that the activity of radioactive particles can increase when the decay products of radionuclides are also radioactive. This increase can affect the charging and aggregation of radioactive particles. This comment raised by the Reviewer has already been addressed by our previous work:

Kim, Y.H., Yiacoumi, S., Nenes, A. and Tsouris, C., 2017. Incorporating radioactive decay into charging and coagulation of multicomponent radioactive aerosols. Journal of Aerosol Science, 114, pp.283-300.

In this study, we focused on the charging, aggregation, and electrostatic dispersion of ^{238}Pu, ^{137}Cs, and ^{131}I particles. During simulations, the radionuclides decay as follows:

$^{238}\mathrm{Pu} \xrightarrow{\alpha} {}^{234}\mathrm{U}$

$^{137}\mathrm{Cs} \xrightarrow{\beta} {}^{137m}\mathrm{Ba} \xrightarrow{\gamma} {}^{137}\mathrm{Ba}$

$^{131}\mathrm{I} \xrightarrow{\beta} {}^{131}\mathrm{Xe}$

^{234}U is a long-lived radionuclide with half-life of 2.455×10^5 years. ^{137}Ba and ^{131}Xe are stable nuclides. These three decay products do not change the activity of the radioactive particles during the simulation time scale. ^{137m}Ba is a gamma emitter that can change ion concentrations in the surrounding environment. In the section "4.2.2 *Beta Decay of ^{137}Cs vs Beta Decay of ^{131}I*", we investigated the charging, aggregation, and electrostatic dispersion of ^{137}Cs particles, but the focus of the investigation was on assessing the effects of beta decay on the processes. Thus, the effects of gamma decay on the charging of ^{137}Cs particles were not considered here. Also, note that the charging of beta-emitting radioactive particles such as ^{137}Cs particles can be precisely predicted with considering only the effects of beta emission [see Gensdarmes et al. (2001) and Clement and Harrison (1992)].

Gensdarmes, F., Boulaud, D., and Renoux, A.: Electrical charging of radioactive aerosols–comparison of the Clement-Harrison models with new experiments, J. Aerosol Sci. 32, 1437-1458, doi:10.1016/S0021-8502(01)00065-9, 2001.
Clement, C. F., and Harrison, R. G.: The charging of radioactive aerosols, J. Aerosol Sci., 23, 481-504, doi:10.1016/0021-8502(92)90019-R, 1992.

To better clarify these points, we have modified the revised manuscript as follows:

Page 13 (Lines: 260) (Before revision) Time-dependent changes in the charge and size distributions of ^{238}Pu and ^{131}I particles were investigated.

Page 13 (Lines: 277-280) (After revision) Time-dependent changes in the charge and size distributions of ^{238}Pu and ^{131}I particles were investigated. ^{238}Pu and ^{131}I decay to ^{234}U and ^{131}Xe, respectively. Because ^{234}U is a long-lived radionuclide with half-life of 2.455×10^5 years and because ^{131}Xe is a stable nuclide, these decay products are not expected to affect the activity of the radioactive particles during the period investigated.

Page 15 (Lines: 295) (Before revision) Time-evolution of the charge and size distributions of ^{137}Cs and ^{131}I particles was investigated.

Page 15(Lines: 312-314) (After revision) Time-evolution of the charge and size distributions of ^{137}Cs and ^{131}I particles was investigated. ^{137}Cs decays to ^{137m}Ba which is subsequently transformed into ^{137}Ba by gamma decay. In our assessment of the effects of beta decay of these radionuclides on charge and size distributions, the influence of gamma decay was not considered.

General Comments

Line 77 - transport […] decrease their concentration: Please explain what the mechanism. Sedimentation/Collision and Agglomeration is important as well

Response: We have modified Lines 77-83 of the original manuscript as follows:

Page 3 (Line: 77-83) (Before revision) The transport of the charged particles and ions can decrease their concentrations (Oron and Seinfeld, 1989a&b):

$$\frac{dN_{kj}}{dt} = -\frac{B_k jeN_{kj}\rho}{\varepsilon}, \tag{2}$$

$$\frac{dn_\pm}{dt} = \mp\frac{\mu_\pm n_\pm \rho}{\varepsilon}, \tag{3}$$

where N_{kj} is the concentration of size k particles carrying elementary charge j, B is the particle mobility, $\mu_\pm$ are the mobilities of positive and negative ions, and ε is the permittivity of vacuum. This phenomenon has been observed in many laboratory-scale experiments and has been called electrostatic dispersion (Whitby et al., 1965; Adachi et al., 1981; Kasper, 1981).

Pages 3-4 (Lines: 78-86) (After revision) As shown in Fig. 1, the transport of charged particles and ions by electrostatic repulsion can decrease their concentrations in the system (Oron and Seinfeld, 1989a&b):

$$\frac{dN_{kj}}{dt} = -\frac{B_k jeN_{kj}\rho}{\varepsilon}, \tag{2}$$

$$\frac{dn_\pm}{dt} = \mp\frac{\mu_\pm n_\pm \rho}{\varepsilon}, \tag{3}$$

where N_{kj} is the concentration of size k particles carrying elementary charge j, B is the particle mobility, $\mu_{\pm}$ are the mobilities of positive and negative ions, and ε is the permittivity of vacuum. This phenomenon has been observed in many laboratory-scale experiments and has been called electrostatic dispersion (Whitby et al., 1965; Adachi et al., 1981; Kasper, 1981). Additional processes that can decrease the ion and particle concentration include recombination, attachment, aggregation, and deposition.

Line 90f - the volume size of the particle ensures a greatly increased number of atmoe and thus also a much higher activity. can you explain the size bin dependency in more detail?

Response: We agree with the Reviewer's comment. The activity of radioactive particles can increase when they aggregate. This question raised by the Reviewer has already been addressed by our previous paper:

Kim, Y.H., Yiacoumi, S., Nenes, A. and Tsouris, C., 2017. Incorporating radioactive decay into charging and coagulation of multicomponent radioactive aerosols. Journal of Aerosol Science, 114, pp.283-300.

To better clarify this point, we have modified the original manuscript as follows.

Page 4 (Lines: 99-102) (Before revision) In Eq. 4, particle charging is parameterized by s, A_k, $\beta^{\pm}_{k,j}$, and $n_{\pm}$. The self-charging coefficient, s, is characteristic of the decay modes of radionuclides [e.g., s = 9 for $^{238}PuO_2$ (Yeh et al., 1978); s = 1 for beta-emitting particles (Clement and Harrison, 1992)]. Note that for nonradioactive particles, the self-charging term disappears from Eq. 4 because the particle radioactivity is zero (i.e., $A_k = 0$).

Page 4 (Lines: 102-107) (After revision) In Eq. 4, particle charging is parameterized by s, A_k, $\beta^{\pm}_{k,j}$, and $n_{\pm}$. The self-charging coefficient, s, is characteristic of the decay modes of radionuclides [e.g., s = 9 for $^{238}PuO_2$ (Yeh et al., 1978); s = 1 for beta-emitting particles (Clement and Harrison, 1992)]. For a homogeneous radioactive particle population, the activity of single radioactive particles increases as the particle size increases because large particles contain more radionuclides. The activity of size k particles was estimated according to Clement et al. (1995) and Kim et al. (2017). Note that for nonradioactive particles, the self-charging term disappears from Eq. 4 because the particle radioactivity is zero (i.e., $A_k = 0$).

Clement, C.F., Clement, R.A. and Harrison, R.G., 1995. Charge distributions and coagulation of radioactive aerosols. *Journal of Aerosol Science*, *26*(8), pp.1207-1225.

Line 105f - the height of the atmosphere plays a major role for the ion concentration, if this has not been taken into account, please narrow down to the relevant height range (Stozhkov 2003).

Response: We thank the Reviewer for the comment. In the revised manuscript, we have added the following:

Page 9 (Lines: 180-182) (Before revision) Typical values for the properties of background air were quoted from Harrison and Carslaw (2003): (i) the air contains 5×10^8 ion pairs m^{-3}, (ii) the background ionization rate is 10^7 ion pairs m^{-3} s^{-1}, and (iii) α_{rc} is 1.6 $\times 10^{-12}$ m^{-3}.

Page 9 (Lines: 193-196) (After revision) Typical values for the properties of background air were quoted from Harrison and Carslaw (2003): (i) the air contains 5×10^8 ion pairs m^{-3}, (ii) the background ionization rate is 10^7 ion pairs m^{-3} s^{-1}, and (iii) α_{rc} is 1.6 $\times 10^{-12}$ m^{-3}. These values may represent atmospheric conditions of lower troposphere (Harrison and Carslaw, 2003; Stozhkov, 2003).

Stozhkov, Y.I., 2003. The role of cosmic rays in the atmospheric processes. *Journal of Physics G: Nuclear and Particle Physics*, *29*(5), p.913.

Line 142f – charges of particles…. : The average charges of particles is largely a multi modal distribution with a size dependency. (Wiedensohler 1987) May you explain the differences?

Response: Particle size is an important parameter of diffusion charging because large particles can capture more ions. In typical atmospheric conditions, negative ions can be more mobile than positive ions. In comparison to nano-sized aerosols, micron-size aerosols can capture more negative ions, indicating that the mean aerosol charge can be negative and the width of the aerosol charge distribution can be wider as shown in Wiedensohler (1988) and Kim et al. (2017).

Wiedensohler, A., 1988. An approximation of the bipolar charge distribution for particles in the submicron size range. Journal of aerosol science, 19(3), pp.387-389.

Kim, Y.H., Yiacoumi, S., Nenes, A. and Tsouris, C., 2017. Incorporating radioactive decay into charging and coagulation of multicomponent radioactive aerosols. Journal of Aerosol Science, 114, pp.283-300.

Equations 9-11 can predict changes in the mean charge of particles as a function of size, activity, and ion concentrations. Equation 12 can be used to approximate the charge distribution of particles with their mean charge and size. To better clarify this point, we have revised the manuscript as follows:

Page 6 (Lines: 141-144) (Before revision) The mean charge of particles predicted by solving Eqs. 9-11 can be approximately converted to the particle charge distribution using a Gaussian distribution:

$$N_{kj} = \frac{N_k}{\sqrt{2\pi}\sigma_k} \exp\left(-\frac{(j - J_k)^2}{2\sigma_k^{\ 2}} \right), \qquad (12)$$

where σ_k is the standard deviation given by Clement et al. (1995).

Page 6 (Lines: 147-152) (After revision) The mean charge of size *k* particles predicted by solving Eqs. 9-11 can be approximately converted to the particle charge distribution using a Gaussian distribution:

$$N_{kj} = \frac{N_k}{\sqrt{2\pi}\sigma_k} \exp\left(-\frac{(j - J_k)^2}{2\sigma_k^{\ 2}} \right), \tag{12}$$

where σ_k is the standard deviation given by Clement et al. (1995). The width and mean of the particle charge distribution can be influenced by the charging parameters. For example, in comparison to nanosized aerosols, the charge distribution of micron-size aerosols can be wider because they can capture more ions [e.g., see Wiedensohler (1988) and Kim et al. (2017)].

Line 171f: the selected radioactive elements themselves decay into radioactive atoms and can change their type of radiation. has this been taken into account?

Response: The decay products of the selected radionuclides are ^{234}U, ^{137m}Ba, ^{137}Ba, and ^{131}Xe. ^{234}U is a long-lived radionuclide with half-life of 2.455×10^5 years. ^{137}Ba and ^{131}Xe are stable nuclides. These three decay products do not change the activity of the radioactive particles during the simulation time scale. ^{137m}Ba is a gamma emitter that can change ion concentrations in the surrounding environment. In the section "4.2.2 *Beta Decay of ^{137}Cs vs Beta Decay of ^{131}I*", we investigated the charging, aggregation, and electrostatic dispersion of ^{137}Cs particles, but the focus of the investigation was on assessing the effects of beta decay on the processes. Thus, the effects of gamma decay on the charging of ^{137}Cs particles were not considered here. Nevertheless, note that the charging of beta-emitting radioactive particles such as ^{137}Cs particles can be precisely predicted with considering only the effects of beta emission [see Gensdarmes et al. (2001) and Clement and Harrison (1992)].

Gensdarmes, F., Boulaud, D., and Renoux, A.: Electrical charging of radioactive aerosols–comparison of the Clement-Harrison models with new experiments, J. Aerosol Sci. 32, 1437-1458, doi:10.1016/S0021-8502(01)00065-9, 2001.
Clement, C. F., and Harrison, R. G.: The charging of radioactive aerosols, J. Aerosol Sci., 23, 481-504, doi:10.1016/0021-8502(92)90019-R, 1992.

To more clearly indicate these points, we have modified the revised manuscript as follows:

Page 13 (Lines: 260) (Before revision) Time-dependent changes in the charge and size distributions of ^{238}Pu and ^{131}I particles were investigated.

Page 13 (Lines: 277-280) (After revision) Time-dependent changes in the charge and size distributions of ^{238}Pu and ^{131}I particles were investigated. ^{238}Pu and ^{131}I decay to ^{234}U and ^{131}Xe, respectively. Because ^{234}U is a long-lived radionuclide with half-life of 2.455×10^5 years and because ^{131}Xe is a stable nuclide, these decay products are not expected to affect the activity of the radioactive particles during the period investigated.

Page 15 (Lines: 295) (Before revision) Time-evolution of the charge and size distributions of ^{137}Cs and ^{131}I particles was investigated.

Page 15(Lines: 312-314) (After revision) Time-evolution of the charge and size distributions of ^{137}Cs and ^{131}I particles was investigated. ^{137}Cs decays to ^{137m}Ba which is subsequently transformed into ^{137}Ba by gamma decay. In our assessment of the effects of beta decay of these radionuclides on charge and size distributions, the influence of gamma decay was not considered.

Line 176 – "we assumed" : how was this justified, also with regard to the source

Response: The values for the ionization rate coefficient of ^{238}Pu, ^{131}I, and ^{137}Cs were obtained from Clement and Harrison (1992) and Kim et al. (2015). Predictions using these values agreed well with charge measurements. To more clearly justify the assumption, we have revised the manuscript and corrected a typo for the ionization rate coefficient of ^{238}Pu, as follows:

Page 8 (Lines: 175-177) (Before revision) To involve alpha- and beta-radiation leading to ionization of air molecules during simulation, we assumed that ^{238}Pu can produce 15,000 ion pairs per alpha decay (Clement and Harrison, 1992), while ^{137}Cs and ^{131}I can generate 2,067 and 1,945 ion pairs per beta decay, respectively (Kim et al., 2015).

Page 8 (Lines: 185-190) (After revision) To involve alpha- and beta-radiations leading to ionization of air molecules during simulation, we assumed that ^{238}Pu can produce 150,000 ion pairs per alpha decay, while ^{137}Cs and ^{131}I can generate 2,067 and 1,945 ion pairs per beta decay, respectively. These values are the ionization rate coefficients of the radionuclides; the value for ^{238}Pu was obtained from Clement and Harrison (1992), while the values for ^{137}Cs and ^{131}I were given by Kim et al. (2015). Predictions using these values agreed well with charge measurements of the radioactive particles (Clement and Harrison, 1992; Kim et al., 2015).

Line 230f; Figure 4: Why was a particle size of one micrometre chosen? the accumulation of particles in the atmosphere is around 200 nm (sedimentation-driven). the calculations can be redone/added with this relevant variable (Rose et al. 2021)

Response: The goal of Figure 4 is to verify the bivariate PBM coupling electrostatic dispersion with aggregation. We employed a conventional approach used to verify bivariate PBMs tracking the rates of aggregation and electrostatic dispersion. The approach is to compare simulation results of models developed in this study with those of Oron and Seinfeld (1989) and Adachi et al. (1981) who assumed 1-µm aerosols with uni- and bi-modal charge distributions [e.g., see Vemury et al. (1997), Xiangrong et al. (2006), and Verdoold and Marijnissen 2009)]. For the comparison, all simulation conditions should be identical. Thus, we chose 1-µm as the particle size. To more clearly justify the particle size selected for the simulations, we have revised the manuscript as follows.

Page 10 (Lines: 213-216) (Before revision) This algorithm was verified by comparing simulation results of Eq 4 with those of Adachi et al. (1981) and Oron and Seinfeld (1989a) who predicted time-dependent changes in the concentrations of charged particles by aggregation and electrostatic dispersion.

Page 10 (Lines: 230-232) (After revision) This algorithm was verified by comparing simulation results of Eq 4 with those of Adachi et al. (1981), Oron and Seinfeld (1989a), Vemury et al. (1997), and Verdoold and J. Marijnissen (2009) who predicted time-dependent changes in the concentrations of charged 1-µm particles by aggregation and electrostatic dispersion.

Line 272f: the changed behaviour with regard to the charge distribution may also have something to do with the electronegativity/affinity of the elements?

Response: We thank the Reviewer for the comment. Because all simulation results were obtained using the developed PBM, the temporal evolution of the particle charge distributions was driven by the microphysical and radiological processes included in the model. Because the model didn't include specific parameters to implement the electronegativity and affinity of the elements involved, the effects of the electronegativity and affinity of these elements were not considered in this study. To better clarify this point, we have added the following sentence to the manuscript.

Page 8 (Line: 170) (Before revision) The PBM was used to investigate the dynamics of charge and size distributions of radioactive and nonradioactive particles.

Page 8 (Lines: 178-179) (After revision) The PBM was used to investigate the dynamics of charge and size distributions of radioactive and nonradioactive particles. The models did not include parameters to consider the effects of electronegativity and affinity of the elements involved.

Line 294 137Cs vs 131I - the electron affinity differs greatly for these elements and is also reflected in the charge distribution as the corresponding ions are strongly favoured (positive charged alkali metals and negatively halogens). can this be the explanation?

Response: Because all simulation results were obtained using the developed PBM, the temporal evolution of the particle charge distributions was driven by the microphysical and radiological processes included in the model. Because the model didn't include specific parameters to implement the electronegativity and affinity of the elements involved, the effects of the electronegativity and affinity of these elements were not considered in this study. To better clarify this point, we have added the following sentence to the manuscript.

Page 8 (Line: 170) (Before revision) The PBM was used to investigate the dynamics of charge and size distributions of radioactive and nonradioactive particles.

Page 8 (Lines: 178-179) (After revision) The PBM was used to investigate the dynamics of charge and size distributions of radioactive and nonradioactive particles. The models did not include parameters to consider the effects of electronegativity and affinity of the elements involved.

Line 375f: can you please add numbers and percentages to your statements?

<u>Response</u>: We have added the particle size and percentage differences to the manuscript as follows.

Page 18 (Lines: 374-375) (Before revision) Compared to the bivariate PBM, the monodisperse PBM slightly overestimated the mean particle size and space charge in absolute value.

Page 18 (Lines: 393-396) (After revision) Compared to the bivariate PBM, the particle size predicted by the monodisperse PBM was larger. After 1 hour, the mean size predicted by the monodisperse and bivariate PBMs was 1.04 µm and 0.89 µm, respectively, indicating a difference of 16.9%. The difference increased with time (e.g., 29.7% after 3 hours).

Line 379/Figure9: please add a legend in all your graph for the colored lines (the arrows do not point unambiguously to the lines)

<u>Response</u>: We have added legends to Figure 9.

(Before revision)

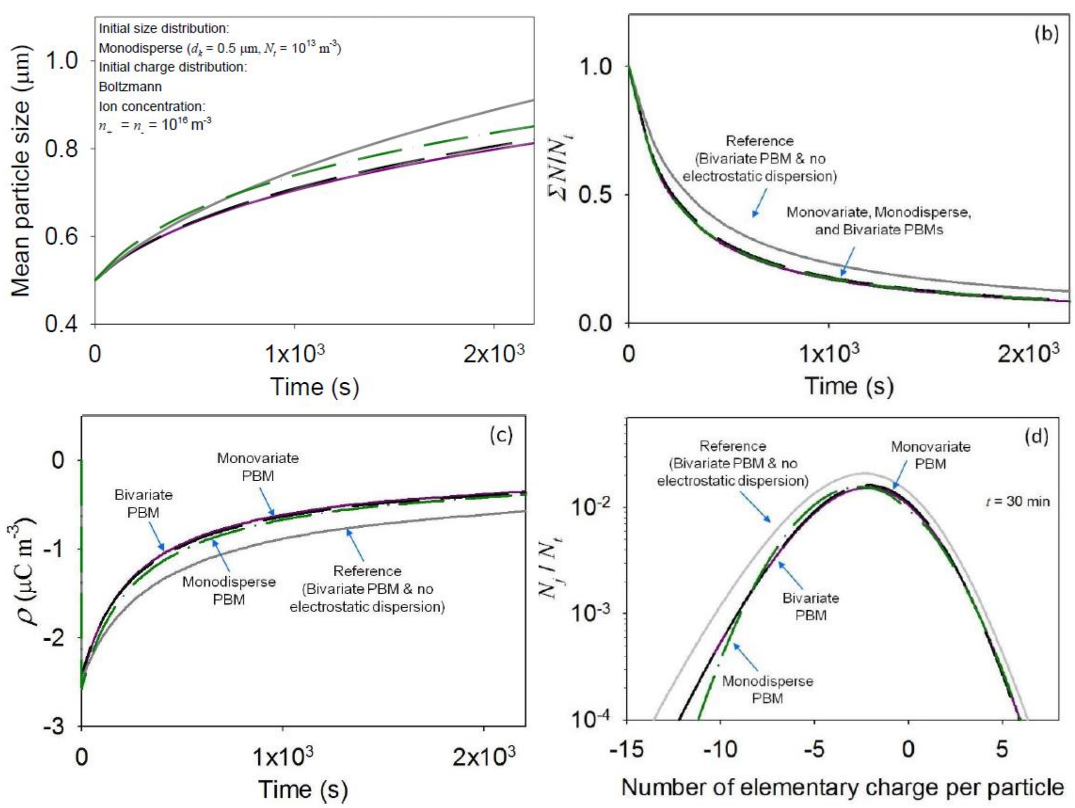

Figure 9: Time-dependent changes in the mean size (a) and total concentration (b) of particles, the space charge density (c), and particle charge distribution (d) in a spatially homogeneous atmospheric system. Reference refers to simulation results obtained using Eq. 4 in the absence of the electrostatic dispersion term.

(After revision)

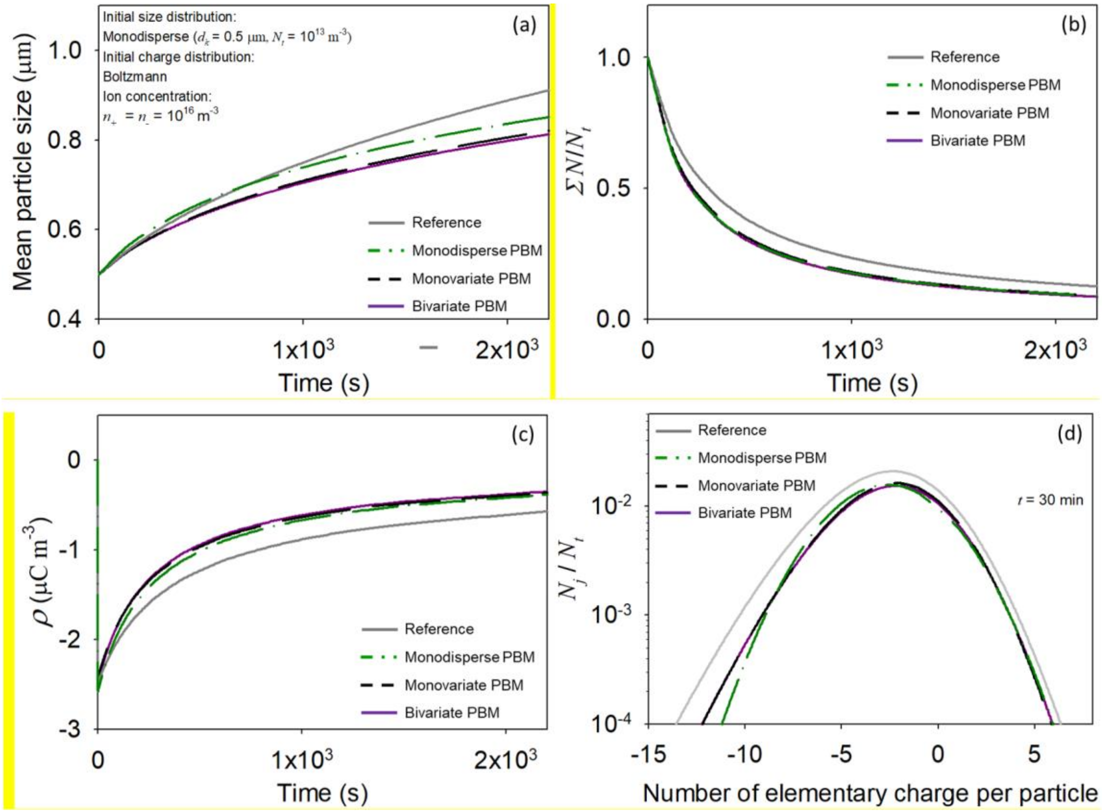

Figure 9: Time-dependent changes in the mean size (a) and total concentration (b) of particles, the space charge density (c), and particle charge distribution (d) in a spatially homogeneous atmospheric system. Reference refers to simulation results obtained using Eq. 4 in the absence of the electrostatic dispersion term.

Minor Comments

Line 67 – can easily change to favored

Response: We have revised the sentence as follows.

Page 3 (Lines: 66-68) (Before revision) In a spatially homogeneous atmospheric system containing charged particles and ions, positively charged particles can easily collide with particles and ions with negative charge because of electrostatic attractive forces generated between positive and negative charges.

Page 3 (Lines: 65-67) (After revision) In a spatially homogeneous atmospheric system containing charged particles and ions, positively charged particles can easily collide with negatively charged particles and negative ions because of electrostatic attractive forces generated between positive and negative charges.