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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 1 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 1

1. This paper discusses the time-dependent change in size and charge distributions of particles. The text and figures comprise 45 pages, mostly of single-spaced text. This should make a nice paper if the authors can cut several pages from it. Given that the authors acknowledge on P. 23087 that "In our previous work (Kim et al., 2014, 2015) it has been shown that Approaches 1 and 3 can reliably simulate charging of radioactive particles. . ." there seems to be sufficient overlap with previous work to warrant reducing the size of this manuscript, including the number of figures, which stands at 16. Are all these figures really necessary to draw the conclusions about this paper? Some additional comments are given below.

Response: Following the Reviewer's suggestion, we moved three figures from the manuscript to the supporting information to reduce the length and improve the readability of the manuscript. These changes do not influence the conclusions of the manuscript.

Regarding the Reviewer's comment on overlapping with our previously published work, there is no overlapping. As shown on lines 286-287 of the revised manuscript, our previous work validated Approaches 1 and 3 in terms of simulating the charging of radioactive particles. The current work has only examined the validity of Approach 2 to model the charging of the particles.

2. Introduction. "Due to atmospheric dispersion, radioactive particles (e.g. 137Cs released during the Fukushima accident were sampled in situ 150 km away…)." 137Cs from Fukushima was found worldwide rather than just 150 km away (Figure 1 of Ten Hoeve and Jacobson, 2012).

Response: We agree with the opinion of the Reviewer. We have modified lines 64 - 67 of the revised manuscript as follows:

"For instance, due to these atmospheric dispersion patterns, radioactive particles (e.g., ^{137}Cs) released during the Fukushima accident were sampled in-situ 150 km away from the emission site (Yamauchi et al., 2012), and also found in many places around the world (Hoeve and Jacobson, 2012)."

3. Introduction. "Accurate understanding of the behavior of particles is necessary to predict transport of contaminants…" Please clarify. Do you mean "evolution of contaminants" or *"removal of contaminants?" Transport of contaminants is dominated by wind speed and direction rather than the behavior of particles.*

Response: We agree with the Reviewer that wind speed and direction can significantly influence transport of contaminants. In addition to wind, we believe that the deposition rates of particles can also highly affect the transport of contaminants because particle deposition, which is largely influenced by aerosol microphysics, can determine places where contaminant-laden particles are settled. To better explain this point, lines 69 - 73 of the revised manuscript have been modified as follows:

"These examples suggest that particle deposition, which is largely affected by aerosol microphysics, can determine the fate of contaminants during atmospheric transport. Thus, accurate understanding of the microphysical behavior of atmospheric particles is necessary to more accurately predict transport of contaminants (especially long-lived ones, such as ^{137}Cs), as well as their potential environmental impacts."

4. Introduction. Please briefly explain self-charging and diffusion charging in the Introduction where it is first mentioned rather than in Section 2.2.

Response: We thank the Reviewer for the suggestion. The following sentences in the section "**2.2. Charge balance models**" have been moved to the section "**1. Introduction**" of the revised manuscript (lines 76 - 79).

"Self-charging refers to charge accumulation caused by radioactive decay which typically leads to emission of electrons from particle surfaces. Diffusion charging is attributed to diffusion of ions from the surrounding atmosphere onto the surface of particles."

5. Introduction. "Coagulation of atmospheric particles can influence their charging because the particle size distribution can highly affect the time-evolution of ion concentrations." This statement is very confusing. Do you mean, "Coagulation can affect the time evolution of the size distribution of ion concentration?"

Response: We thank the Reviewer for the comment. To better explain the meaning of the sentence, lines 86 - 88 of the revised manuscript have been modified as follows:

"Coagulation of atmospheric particles can influence their charging because the concentration of atmospheric ions is affected by the particle size distribution (Yair and Levin, 1989), thereby modifying the diffusion charging rates of the particles."

6. Introduction: "Particle charging and coagulation can mutually affect each other…" Do you mean, "particle charging can affect coagulation rate coefficients and coagulation can affect the size distribution of charged particles?" If so, please clarify this statement. Please clarify when you refer to "charge" that you are not referring to van der Waal's forces, which result in particles being polarized but with zero net charge.

Response: We thank the Reviewer for the comment. Lines 86-92 of the revised manuscript have been modified to better explain the meaning of the sentence. The revised text reads:

"Coagulation of atmospheric particles can influence their charging because the concentration of atmospheric ions is affected by the particle size distribution (Yair and Levin, 1989), thereby modifying the diffusion charging rates of the particles. Also, particle coagulation can result in charge neutralization or accumulation on atmospheric particles (Alonso et al., 1998). These effects imply that particle coagulation can influence the particle charge distribution. Thus, particle charging and coagulation can mutually affect each other and simultaneously affect both charge and size distributions in the atmosphere."

Also, the following text has been added to indicate that particle polarization is not included in this work (lines 249-250 of the revised manuscript).

"In this work, polarization of particles is not taken into account."

7. Introduction. "Previous attempts to include charging effects include…" Please include Yu and Turco (2001). This and several other papers by Yu treated charging in the coagulation kernels in a sectional coagulation model. Following Equation 10, by "collision efficiency," do you mean "coalescence efficiency?"

Response: We thank the Reviewer for the suggestion and comment. A short description of the work of Yu and Turco (2001) has been added to lines 113-114 of the revised manuscript. The coalescence efficiency is often called the collision efficiency (Jacobson, 2005; Seinfeld and Pandis, 2006). To avoid confusion, text has been added to lines of 210 - 212 of the revised manuscript.

Jacobson, M. Z.: Fundamentals of atmospheric modeling, Cambridge University Press, New York, 2005.
Seinfeld, J. H., and Pandis, S.N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley and Sons, New Jersey, 2006.
Yu, F., and Turco, R. P.: From molecular clusters to nanoparticles: Role of ambient ionization in tropospheric aerosol formation, J. Geophys. Res., 106, 4797-4814, doi: 10.1029/2000JD900539, 2001.

8. Also by "coagulation frequency," do you mean coagulation kernel or rate coefficient? Please provide units. F does not have units of frequency (s^-1) but something like cm^3 per particle per second, analogous to a chemical reaction rate coefficient. Same thing with Beta in Equation 12.

Response: We thank the Reviewer for the comments. The coagulation frequency, F and collision frequency, β are also called the coagulation rate coefficient and the collision kernel, respectively (Jacobson, 2005; Seinfeld and Pandis, 2006). Lines 191-192 and 205-206 of the revised manuscript have been modified to avoid confusion.