Responses to Reviewer 1

Vasilakos et al. introduces an extension to the TwO-Moment Aerosol Scheme (TOMAS) that includes the effects of radioactive charging. Using the extended model, TOMASRC, the authors demonstrate that radioactive charging plays a significant role in the lifetime and transport of particles within particular size ranges. The paper is interesting and relevant to the journal, the manuscript is well written, the methods are described with sufficient detail, and the results are presented clearly. I recommend publication if the authors address the following questions and comments:

Response: We thank the reviewer for the thoughtful and constructive comments. Responses to each point raised are provided below in light blue.

A key finding of the paper is the importance of radioactive charging on particles in the coarse mode (on the order of 5 μm in diameter), but it is unclear to me that one would expect such large charged particles in the atmosphere. I suggest the authors elaborate on the extent to which charging by such large particles would be expected in the atmosphere.

Response: We thank the reviewer for the suggestion. The charging mechanisms of radioactive particles are diffusion charging and self-charging. The in-situ observation and model prediction of Renard et al. (2013) showed that below 10-km altitude and above 20-km altitude, many nonradioactive particles larger than 2 μm can be easily charged by diffusion of atmospheric ions. In particular, large particles could acquire many charges by diffusion charging (Renard et al., 2013). Self-charging results from the decay of radionuclides embedded in radioactive particles, and this charging mechanism has been verified by many laboratory scale experiments and modeling investigations [e.g., Clement and Harrison (1992), Gensdarmes et al. (2001), and Kim et al. (2016)], suggesting that large radioactive and nonradioactive particles may be easily charged in the atmosphere. The rationale behind looking at such a large range of particle sizes is to evaluate the effects of radioactive charging on the microphysical evolution of aerosol populations over a wide range covering sizes from the Aitken to the coarse mode. The presence of such large particles is also expected in dust and volcanic ash clouds (Langmann, 2013), as well as radiological debris created during nuclear events. The text below has been added/revised to explain the presence of charged large particles in the atmosphere.

Page 4, Lines 43-44 (Before revision): "These ions ejected and transferred their kinetic energy to the gas phase can combine with surrounding gas molecules or other aerosols through collision/adsorption."

Page 4, Lines 43-47 (After revision): "These ions ejected and transferred their kinetic energy to the gas phase can combine with surrounding gas molecules or other aerosols through collision/adsorption, thereby producing many ion pairs and charged aerosols. Renard et al. (2013) found that many large aerosols in the upper troposphere and stratosphere can be easily charged by diffusion of ions, and the aerosols may gain more charges as their size increases."

Page 14, Lines 359-364 (After revision): "The rationale behind looking at such a large range of particle sizes, is to map the response of aerosol populations spanning all possible sizes to radioactive charging, from the Aitken to the coarse mode. The presence of such large particles is expected in dust and volcanic ash clouds (Langmann 2013), as well as nuclear reactor debris in the case of a radiological event.

In general, I would like to see more discussion on the expected size ranges of charged particles in the discussion of the results. The authors point out that this information is not well constrained, but they also provide the example of forest fires in the introduction; one would expect charged particles from forest fires to be much smaller. What mechanisms are expected to yield charged particles? It is difficult to understand the relevance of the results without understanding anything about the size ranges.

Response: Multiple charging mechanisms exist and different particle sizes are involved in each one. Particles formed through ion-induced nucleation are a few nanometers in size (Harrison & Carslaw, 2003), while particles in volcanic ash or dust clouds that are charged through friction reside mostly on the coarse mode with diameters greater than 10 μm (Langmann 2013). For charged radioactive particles resuspended during forest fires, the bulk of radioactive material is contained in giant particles of diameters greater than 25 μm as shown for the case of forest fires conducted in controlled conditions in the Chernobyl exclusion zone (Yoshchenko et al, 2006). Note that this refers to radionuclides that were already deposited on the ground and not new particles formed through charging mechanisms. The text below has been added to the section "3. Results".

Page 13, Lines 321-330 (After revision): "Given that multiple charging mechanisms exist and different particle sizes are involved in each one, a wide range of particle size distributions was used. Particles formed through ion-induced nucleation are a few nanometers in size (Harrison & Carslaw, 2003), while particles in volcanic ash or dust clouds that are charged through friction, reside mostly on the coarse mode with diameters greater than 10 μm (Langmann 2013). For charged radionuclides resuspended during fires, the bulk of radioactive material is contained in giant particles of diameters greater than 25 μm as shown for the case of forest fires conducted in controlled conditions in the Chernobyl exclusion zone (Yoshchenko et al, 2006). Note that this refers to radionuclides that were already deposited on the ground and not new particles formed through charging mechanisms."

A key factor seems to be the charge distribution as a function of particle diameter. The authors assume a gaussian charge distribution for particles within each size bin, along with a list of citations, but it is unclear whether they are following an assumption that was made by previous authors or if this charge distribution was determined experimentally. Please state explicitly how the functional form of the charge distribution was determined in these previous studies.

Response: The Gaussian charge distribution we utilize has been used in previous studies (Clement & Harrison, 1992; Clement et al., 1995; Kim et al., 2016) as an approximation of the exact charge distribution calculated numerically. The normal distribution presents a simple, yet accurate representation of the charge distribution as shown for the case of self-charging aerosols such as ^{198}Au, as long as the ion-asymmetry ratio *X* does not significantly deviate from 1. For the internally mixed aerosol populations presented here, the gaussian is an accurate simplification of the steady-state charge distribution as shown in Kim et al. (2016). The below text has been added to the section "2.2 Coagulation of radioactive aerosols".

Page 8, Lines 175-176 and Page 9, Lines 177-182 (After revision): "The Gaussian charge distribution we utilize has been used in previous literature reports (Clement & Harrison, 1992; Clement et al., 1995; Kim et al., 2016) as an approximation of the exact charge distribution calculated numerically in Clement & Harrison (1992). The normal distribution presents a simple, yet accurate representation of the charge distribution of radioactive aerosols (e.g., Gensdarmes et al., 2001). For the internally mixed aerosol populations presented here, the Gaussian distribution constitutes an accurate simplification of the steady-state charge distribution as shown in Kim et al. (2016)."

I also think it would be helpful to show the charge distribution for an example aerosol population, perhaps as a 2D density distribution.

Response: The point is well understood. Given the accumulation of most charges occurs in the coarser aerosol, we feel that a 2-D charge distribution does not provide considerably more information to what Figures 3 and 4 already show; to avoid increasing further the size of the manuscript, we are kindly requesting that this additional figure is not included.

Would the results differ under a more realistic simulation that also includes gas condensation? It seems this analysis should be reserved for a later study, but it would be helpful to understand why this mechanism is ignored. I suggest commenting on this early on in the paper.

Response: Depletion of the gas phase would limit diffusion charging (the effects of diffusion charging from the surrounding gas to the particles are already included in the model - Equation 6 in the manuscript), while at the same time increasing the impact of self-charging due to the increase of radioactive material per particle from the condensation of radioactive gas. Condensation of gas to the aerosol phase should not impact the average charge of each size bin, but it could potentially affect the deposition rates due to the subsequent changes in particle size. This will be noted in the manuscript and left for a future study.

The authors describe the impact of particle charging on dry deposition due to enhanced or reduced coagulation rates. Would charging also impact the deposition flux in areas impacted by radioactivity? That is, could dry deposition for particles of a given size also be enhanced

or reduced due to charging?

Response: Following the resistances in series model (e.g., Seinfeld & Pandis, 2006), it is expected that areas affected by radioactivity would present different surface resistances to charged aerosol populations. Therefore, the deposition rate for particles of a given size can be enhanced/reduced close to a radioactive surface. To better explain this, the text below has been revised.

Page 3, Lines 21-22 (Before revision): "Considering charging effects of radioactive aerosols, immediately impacts their initial deposition patterns."

Page 3, Lines 22-26 (After revision): "Considering charging effects of radioactive aerosols, immediately impacts their initial deposition patterns. Following the resistances in series model described in Seinfeld & Pandis (2006), it is expected that areas affected by radioactivity would present different surface resistances to charged aerosol populations. Therefore, the deposition rate for particles of a given size can be enhanced or reduced close to a radioactive surface."

Many of the equations are difficult to read. For example, in some cases, it is difficult to distinguish between multiplication, exponents, or superscripts. I assume this will be addressed during typesetting.

Response: We thank the reviewer for the comment. This issue will be resolved during the typesetting process.

References

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