Responses to Reviewer 2

We thank the reviewer for his/her comments. Our responses and clarifications to each point raised are provided below in light blue.

Within their manuscript, Vasilakos et al. present an extension to the global aerosol model TOMAS (Adams and Seinfeld, 2002) to take into account the effect of particle charge to the coagulation of radioactive aerosol and its lifetime.

Response: TOMAS is not in itself a global aerosol model, but rather a microphysics module for use in a 3D model (e.g., GISS/TOMAS, GEOS-CHEM/TOMAS) for predicting aerosol size distributions, mass and number concentrations. TOMAS-RC resolves many processes pertinent to aerosol processing, such as coagulation, evaporation/condensation and diffusion, as well as aerosol charging, ionization, and ion-ion recombination. Inclusion of radioactive charging effects on coagulation can allow for its inclusion into all the relevant microphysics that govern the microphysical evolution and lifetime of radioactive aerosols.

The extension is based on previous work on the enhancement factor of charged particle coagulation in the diffusion regime by Fuchs (1964), the distribution of charge for aerosol particles of a given size, and the calculation of an average enhancement factor as a function of the distribution of charge (for the latter two, mainly Clement et al., 1995). The implementation is adapted to the requirements of computational efficiency of a global model environment and the potential effects of charged particle coagulation on aerosol lifetime is explored with a sensitivity study. It is the referee's opinion that a paper merits publication within GMD if it fulfills either one or both of the following conditions: the paper presents (1) a major extension to an existing model or an entirely new model, or (2) a scheme based on a newly developed formalism. This manuscript does not present a major extension to the TOMAS model. It is related to a single process, and even this process is only partially taken into account, as it relates exclusively to particles in the diffusion regime. Other major aspects of charged particle microphysics are not considered, such as condensation, coagulation in the molecular and transition regime, ionization, ion attachment, ion recombination, particle activation as CCN, cloud scavenging, wet removal and the influence of particle charge on dry deposition.

Response: Indeed, the basic equations have been known for years, but the same can be said for any aerosol microphysical process. Radioactive charging effects poses a difficult computational problem, which would considerably increase the CPU time of the already challenging computations of aerosol microphysics. TOMAS-RC overcomes this difficulty and allows radioactive charging effects to be implemented efficiently and accurately – which we feel is a significant accomplishment in its own. Furthermore, coagulation in various flow regimes, ionization, ion attachment and recombination are included in TOMAS-RC (Equations 3-8), as other aerosol microphysical processes (with the exclusion of wet deposition and CCN activation, which can be handled by the 3-D model hosting TOMAS-RC). For these reasons, we believe that

TOMAS-RC is appropriate for publication in GMD. The text below in section "2.1 TOMAS aerosol microphysical model" has been revised as follows:

Page 6, Lines 116-117 (Before revision): "In TOMAS, it is assumed that Brownian motion is the dominant collision mechanism (Adams & Seinfeld, 2002; Lee & Adams, 2012)."

Page 6, Lines 116-119 (After revision): "In TOMAS, it is assumed that Brownian motion is the dominant collision mechanism (Adams & Seinfeld, 2002; Lee & Adams, 2012), and the coagulation coefficient is obtained using the interpolation formula of Fuchs (1964) to consider aerosol coagulation in the continuum, transition, and free molecular regimes."

Especially, wet removal is a major process of radioactive particle microphysics and lifetime. The authors are aware of it, which is why it is mentioned as a future model development step, no global modeling results are shown, and a sensitivity study is presented instead in terms of the potential of charged particle coagulation to aerosol lifetime via dry removal only. The manuscript does not present a newly developed scheme either. The theory is based exclusively on previous work, which is then adapted to a global modeling framework. However, the adaptation is minor only: it is investigated whether it is sufficient to limit the charge distribution to the interval of twice its standard deviation, and whether accuracy is conserved as the integration step that serves to assess the average enhancement factor is increased when average particle charge exceeds 100 elementary charges. The results of the sensitivity studies are not spectacular, as the relevant influence of particle charge on particle dynamics is known and expected, which is why the authors have extended the TOMAS model to include charged particle coagulation in the diffusion regime in the first place, and their relevance is questionable, as these very preliminary results are not validated in a global modelling environment with all key processes included. For these reasons, I recommend the manuscript not to be published within GMD. The manuscript should be integrated into a forthcoming publication that includes the processes that are currently in the development stage, that fulfills the critical mass criterion for publication within GMD, and includes global modelling results with all essential processes taken into account. In doing so, the reader would also get a much clearer picture of what a more accurate representation of charged particle microphysics would imply to the simulation of radioactive particles and their lifetime.

Response: The contribution of the present work is to demonstrate that radioactive charging can have a significant impact on the atmospheric lifetime of radioactive aerosols. While processes such as wet deposition are not included in the simulations presented, the importance of radioactive charging is well supported by the sensitivity simulations. This work here is the necessary first step before TOMAS-RC is implemented in global models, which is an ongoing process (Kim et al. 2017, in preparation).

If the editor were not to follow the reviewer with their recommendation, I would like to make the following comments that in my view would help to improve the manuscript. These comments may also be helpful in case of an implementation in a forthcoming publication:

1) Equation 5 was not developed by Spellman (1970). As far as I know it dates back to the seminal work of Fuchs (1964). It is based on a number of simplifying assumptions (image forces are neglected, I think) and it applies to the diffusion regime only. The authors need to mention the underlying assumptions of this basic formula to their work. In particular, the authors need to explain, why they do not take into account the molecular and the transition regime, whilst they do take into account particles as small as 30 nm, which are well into the molecular regime.

Response: We thank the reviewer for the comment. As the reviewer pointed out, equation 5 is obtained by neglecting the effects of image forces, and it is only valid in the continuum regime. Approaches to include the effects of aerosol charging on aerosol coagulation in other regimes have been developed [e.g., Marlow (1980) and Huang et al. (1990)]. However, these approaches are computationally expensive, thereby dramatically increasing the central processing unit time of three-dimensional (3-D) radioactivity transport simulations. In contrast, equation 5 is much simpler, indicating that it is computationally more suitable for use in 3-D global transport models. Also, equation 5 is typically used in various modeling and experimental investigations into coagulation of charged particles in the molecular and transition regimes (Maisels et al., 2002a, 2002b). For example, Maisels et al. (2002b) calculated the coagulation coefficient of charged particles in the transition regime using the interpolation formula of Fuchs (1964) and equation 5, and found that the calculated coagulation coefficients were in good agreement with the measurements. Text has been added to the section “2.2 Coagulation of radioactive aerosols” to better explain these points.

Page 7, Lines 126-127 (Before revision): “$\overline{W}_{k,i}$ in TOMAS-RC is based on the “stability function” correction factor formulation (Spellman, 1970; Seinfeld & Pandis, 2006):”

Page 7, Lines 126-128 (After revision): “$\overline{W}_{k,i}$ in TOMAS-RC is based on the “stability function” correction factor formulation neglecting the effects of image forces (Fuchs, 1964; Spellman, 1970; Seinfeld & Pandis, 2006):”

Page 7, Lines 141-152 and Page 8, Lines 153-154 (After revision): Equation (5) is derived assuming coagulation of charged particles in the continuum regime. The correction factor formulations for the transition and molecular regimes are available elsewhere [e.g., Marlow (1980) and Huang et al. (1990)]. Compared to these formulations, equation (5) is less accurate [e.g., up to 10% error for the transition regime (Huang et al., 1990)]. In contrast to these formulations requiring high computational costs, however, equation (5) is much simpler and computationally more efficient, indicating that the equation may be more suitable for use in three-dimensional transport models. Also, equation (5) has been used in various modeling and experimental investigations into coagulation of charged particles in the molecular and transition regimes [e.g., Maisels et al. (2002a, 2002b)] because the equation may still provide reliable computational results. For instance, Maisels et al. (2002b) estimated the coagulation coefficient of charged particles in the transition regime using the interpolation formula of Fuchs (1964) and equation 5, and found that the calculation was in good agreement with the measurements. Thus, in this study, equation 5 was used to include the effects of particle charging on particle coagulation in all flow regimes.

2) It should also be noted that the size range of the molecular regime increases with height in the atmosphere. In this study, the considered height is limited to 1000 m. But this is unrealistic for particles as small as 30 nm, which are well mixed within the entire height of the troposphere. Please explain.
Response: The chosen height is based on previous work from Chesser et al. (2004), which can encompass a plume from a nuclear incident. TOMAS explicitly calculates the Knudsen number in the subroutine that does coagulation, and uses the beta correction factor for the non-continuum regime from Seinfeld and Pandis (2006). Therefore, for the atmospheric column used in this study, the expanding size of the molecular regime throughout the column is taken into account.

3) Equation 9 contains an error and is not clear with respect to the distinction between mass and charge indexes.
Response: To improve the equation's readability, the relevant text has been revised as follows:

Page 9, Lines 206-210 (Before revision): To overcome this limitation, the average correction factor $\bar{W}_{k,i}$ proposed by Clement et al. (1995) and validated by Kim et al. (2016), which can consider the interaction of all charged aerosols, was employed.
Page 9, Lines 206-208 (After revision): To overcome this limitation, the average correction factor between particles of size i, k $\bar{W}_{k,i}$ proposed by Clement et al. (1995) and validated by Kim et al. (2016), which can consider the interaction of all charged aerosols, was employed.

$$\bar{W}_{k,i} = 1 + \frac{\sum_{j\neq 0}^{\infty} N_{k,j} N_{i,j} \left(W_{k,i}^{-1} - 1\right)}{\sum_{j}^{\infty} N_{k,j} \sum_{j}^{\infty} N_{i,j}}$$

4) The authors do not explain their choice to not represent charge distribution explicitly, and why they would rather use a parameterized version of charge distribution developed by Clement et al. (1995). The purpose of the scheme is to the simulate the transport of radioactive particles globally. The bulk of radioactive contamination is contained within the larger particles that present a large number of elementary charges. For these particles it may probably be assumed that their charge distribution is known, as shown by observations. However, this circumstance, if given, needs to be mentioned and explained in the manuscript for reasons of clarity and readability. Furthermore, as the authors' scheme performs quite a fastidiuous calculation for the assessment of the average enhancement factor, which is almost tantamount to an explicit representation of charge distribution with respect to

coagulation, I would ask myself whether it would not be preferable to represent charge distribution explicitly with respect to all particle processes via particle charge bins, similarly to particle size and mass. An explicit representation would allow simulating the interaction of radioactive and non-radioactive aerosol more accurately. The authors need to explain their choice.

Response: The calculation of the charge distribution is carried out bin by bin, similar to the particle number and mass (Equation 6 in the manuscript). Kim et al. (2016) has conducted extensive analyses on the validity of using a Gaussian distribution to approximate the charge distribution and found that the errors associated with such an assumption only become significant in particles with diameters smaller than 40 nm. An explicit representation of the charge distribution would be computationally demanding when compared to a Gaussian distribution, as demonstrated in Clement et al. (1995). Furthermore, the average charge and deviation values used in the Gaussian distribution are derived from the exact distributions (Clement & Harrison 1992), which even further reduce errors and computational burden. Text has been added to the section "2.2 Coagulation of radioactive aerosols".

Page 8, Lines 157-165 (After revision): Kim et al. (2016) has evaluated an approach assuming a Gaussian distribution to approximate the charge distribution and found that the errors associated with such an assumption only become significant for particles with diameters smaller than 40 nm. An explicit representation of the charge distribution would be extremely computationally demanding when compared to a Gaussian distribution, as demonstrated in Clement et al. (1995) and Kim et al. (2016). Furthermore, the average charge and deviation values used in the Gaussian distribution are approximated from the exact distributions (Clement & Harrison, 1992), which further reduce the error while achieving desirable computational efficiency.

5) The authors need to show much more clearly what they are up to with the model extension that they present, and in this respect, it would be nice to see a few global modelling results. It is not at all clear what the potential of their scheme really is. In a complex and non-linear system, such as particle dynamics, the effects shown by authors under limited process conditions could all but vanish, thus underlining that publication of this manuscript was premature. Also, the effects will strongly depend on an accurate representation of charge distribution. However, this quantity is parameterized and not simulated explicitly. For these reasons, the physical validation of the present model extension will require a global modelling component, a sensitivity study is not sufficient.

Response: The purpose of the paper is to develop and test an extension to a microphysics model. While it is true that the system is highly complex and the interactions are non-linear, TOMAS accounts for all the pertinent aerosol processes, and therefore is able to resolve them. Preliminary global transport modeling results have shown effects of the microphysical behavior of radioactive particles on the transport of radioactivity, indicating that publication of this manuscript is not premature; however, more work is needed to include all the particle processes in the global transport model before we can publish the results.

6) In their sensitivity study, the authors state several times that the smaller particles are almost neutral on average, and that for this reason, their particular charge is less important to their evolution within a plume of radioactive particles. In my opinion, this finding is in contradiction with previous results in the field of the atmospheric aerosol that were obtained within studies on the growth dynamics of charged secondary particles (see, e.g., Yu and Turco, 2001). These studies indicate an essential role of particle charge within the entire size spectrum. They might be worth considering in the context of global modelling of radioactive particles. The smaller particles carry less radioactive matter but might still be interesting in terms of their much larger lifetime and expected range of transport. Particles considered in this study are as small as 30 nm. I would expect these particles to be strongly influenced by the atmospheric aerosol. My impression that the authors underestimate the influence of the atmospheric aerosol on the evolution of the radioactive particles might be wrong. But it would certainly be related to a lack of discussion of the modelling context. The authors need to discuss if their finding of a marginal influence of small particle charge to their growth dynamics are expected to hold in a global modelling study with interacting atmospheric aerosol.

Response: Our finding is not in contradiction with previous literature reports, since it pertains to primary-released particles that are charged only through the mechanisms described in the paper. Even though small particles carry minute charges, the impact of this charge is profound, since their coagulation rates with larger particles are significantly enhanced (Figure 2), leading to the rapid removal of these smaller particles by coagulation (Figures 3 and 5), something described in section 3.2 Dry deposition fluxes of radionuclides.

7) Global modeling schemes encounter regularly unanticipated stability and computational expense issues, once they are actually used in a global modeling environment. The inclusion of global modelling results is an essential numerical validation step of the scheme that is presented, and a section on the computational expense of the scheme should also be included. The verbal finding that it is efficient simply is not enough.

Response: This is a valid concern and addressed in a future manuscript by Kim et al., where TOMAS-RC is implemented in a Global Climate Model (GCM). However, the sensitivity tests we conducted span many orders of magnitude for both particle numbers and sizes (Figures 6 through 8 in the manuscript), which are the main input for the TOMAS module to include radioactive charging effects; no instabilities were observed and the model performed skillfully and efficiently in every scenario. Text has been added to the “3. Results” section.

Page 13, Lines 317-320 (After revision): "During these investigations using the TOMAS-RC model under various initial conditions, computational issues (e.g., computational instability which can suddenly increase computational costs) were not observed.."

8) The text contains a number of errors, in particular words are missing in several instances. Please correct and consider revising your text more thoroughly before submission.
We thank the reviewer for bringing this to our attention. We have reviewed the whole text carefully to address this point in the revised manuscript.

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