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Geoscientific Model Development Discussions

Interactive comment on "Studying the Impact of Radioactive Charging on the Microphysical Evolution and Transport of Radioactive Aerosols with the TOMAS-RC v1 framework" *by* Petros Vasilakos et al.

Anonymous Referee #2

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Within their manuscript, Vasilaskos 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. 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 chagre (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. 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 inluded.

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.

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 moelcular regime.

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.

3) Equation 9 contains an error and is not clear with respect to the distinction between mass and charge indexes.

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 con-

tained 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.

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.

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 contradition 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 inlfuence 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.

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.

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.

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