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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A semi-analytical method for calculating rates of new sulfate aerosol formation from the gas phase" *by* J. Kazil and E. R. Lovejoy

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Reply to Referees

We would like to thank the referees for their insightful comments which provided excellent guidance in the revision of the manuscript and led to substantial improvements.

The individual points brought up by the referees are commented in the following.

Reply to Referee #1

Referee: *The authors should present a more general motivation for their work in the introduction. They could mention the importance of nucleation as a source of new aerosol particles in the atmosphere, the need to include effective ways of dealing with new-particle formation in atmospheric models, and the current situation with regard of existing parameterizations on different nucleation mechanisms. It is also important to point out that the role of chemical compounds other than sulfuric acid and water nucleation in atmospheric nucleation is likely to be very important, at least in the boundary layer of the lower troposphere.*

Authors: We have revised the ”Introduction” section of the manuscript and added a Section ”Representing secondary aerosol formation in atmospheric models”, where we give an in-depth motivation of our work, and where we discuss

- the importance of aerosol nucleation from the gas phase for the atmosphere
- the (known) involved compounds
- the need for the effective representation of aerosol formation from the gas phase in atmospheric models
- and the current state thereof.

Referee: *The interpretation of results presented in Figures 5 and 6 (pages 2185 and 2186) should be enhanced a little bit. Most importantly, the authors have not really discussed the totally different roles of self-coagulation (coagulation of nucleated clusters with themselves) and inter-modal coagulation (coagulation of nucleated clusters with larger pre-existing particles). For example, inter-modal coagulation always reduces the formation rate of 2.5 nm particles compared with the nucleation rate (because it is always a sink of nucleated clusters). The role of self-coagulation is more complicated*

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because it acts as a sink of clusters but at the same time enhances the growth rate of nucleated clusters. Furthermore, while inter-modal coagulation is active practically always, self-coagulation is important only at very high nucleation rates. The different roles of these two coagulation mechanism are clearly visible in different regions of Figures 5 and 6.

Authors: We address this point in the revised manuscript by a more detailed discussion and distinction of intermodal and self-coagulation.

Referee: *The authors give the wrong impression that the method by [Kerminen and Kulmala(2002)] does not take into account coagulation at all. In reality, the method includes inter-modal coagulation but not self-coagulation (which is important only at high nucleation rates).*

Authors: We have corrected the corresponding passage, and state in the revised manuscript that

[Kerminen and Kulmala(2002)] have developed an analytical method to calculate the formation rate of particles of a given size from the formation rate of particles of a smaller size. The method accounts for coagulation with preexisting aerosol, but neglects self-coagulation both as a sink as well as a source of particles.

Referee: *The level of agreement (given by percentages of values within a certain limit from a numerically accurate value) in sections 8.2, 8.3 and 8.4 depend very much on the chosen value range of different parameters and their statistical distribution. This should be brought up explicitly in the manuscript.*

Authors: We address this point by the following in the revised manuscript:

While the parameter grid on which the particle formation rates are compared covers typical tropospheric conditions, the resulting samples will produce an incomplete

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picture of the differences between the particle formation rates: The extent and resolution of the grid introduce a sampling uncertainty. Moreover, the presented deviations between the different particle formation rates are not a representative measure of their performance when used in an atmospheric model, as the joint probability distribution of the parameters controlling aerosol formation needs not to be uniform in the atmosphere.

Referee: *Finally, the authors should help the readers a little bit in the conclusions section by writing a short paragraph that summarizes the "good" and "bad" features of their parameterization compared with existing parameterizations. Also, it would be nice to see some recommendations by the authors for further work in this field.*

Authors: We address this point by summarizing the "good" and "bad" features of our method in the revised manuscript.

Reply to Referee #2

Referee: *Part one of the manuscript gives a very detailed description of the theoretical model construction and does not need any further corrections. However, the second part in detail chapter 8 (Error analysis) could be improved in several ways. Hereby I will not force the authors to include these advices but more encourage them to think about the possibility if the readers could benefit from it.*

First I believe a table explaining the different computer simulations performed for chapter 8 would be helpful to get an easier overlook. Although the authors try to describe all runs and the outcome in detail a more clear way at least for some readers is a simple table including the model set up and the results.

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Authors: We have added a table in the revised manuscript detailing the methods and assumptions used in the calculation of the different particle formation rates. We have also added figures showing the cumulative error occurrence of our semi-analytical particle formation rates compared to reference formation rates, which are calculated with a numerical aerosol model.

Referee: *Second the authors use wide ranges for the input parameters of ionization rate, relative humidity, preexisting aerosol H_2SO_4 condensational sink, temperature and sulfuric acid concentration for the simulations in chapter 8. In ACP we have the nice possibility to include color pictures and I would recommend that the authors should use this possibility and include in figures 4-7 a color-code to mark the most important parameters (e.g. sulfuric acid concentration below 10^7 cm^{-3} in blue and higher in red). This is of course not possible for all parameters and the authors have to focus on the parameters which are most interesting for each run.*

Authors: Done.

Referee: *In the conclusion it would be interesting to include some comparison of the calculated nucleation rates with the results from other published nucleation theories.*

Authors: Simply comparing the output of the different methods does not provide truly meaningful informations: On the one hand, none of the methods can be considered a standard a priori. On the other hand, the flaws in a given representation need not to crop up when it is used in an atmospheric model: The joint probability distribution of the parameters controlling aerosol formation needs not to be uniform in the atmosphere. Thus errors of a method would matter little if they were confined to conditions that occur infrequently, or that contribute little to overall aerosol production. Therefore, assessing and evaluating the various implementations should be done using an

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atmospheric model and comparing its output to observations.

Referee: *Further the authors should point out the importance of ion-induced nucleation of sulfuric acid and water for different areas and latitudes like e.g. land - ocean, mixed layer- free troposphere, tropics - northern latitudes.*

Authors: The relative importance of the different nucleation mechanisms is an open problem, although certain statements on the spatial distribution of their relevance can be made. We have added these in the Introduction section of the revised manuscript.

References

[Kerminen and Kulmala(2002)] Kerminen, V.-M. and Kulmala, M.: Analytical formulae connecting the "real" and the "apparent" nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Aer. Sci., 33, 609–622, doi: 10.1016/S0021-8502(01)00194-X, 2002.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2169, 2007.