Atmospheric
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Discussions

Interactive comment on "A 3D particle Monte Carlo approach to studying nucleation" *by* Christoph Köhn et al.

Anonymous Referee #3

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Review on the manuscript "A 3D particle Monte Carlo approach to studying nucleation" by Christoph Köhn, Martin Bødker Enghoff, and Henrik Svensmark.

The present manuscript deals with simulation on growth of sulphuric acid clusters. The authors call the modelling method Monte Carlo. I would avoid using such name in this context. Technically, method employs random number generator, so it can be called Monte Carlo method. However it does not give any hint on details of modeling. Furthermore, in molecular physics the term Monte Carlo simulations is reserved for the group of methods modelling equilibrium distribution functions of particular variables. In the manuscript such examples are given: Kusaka et al.(1998), and Kathmann and Hale (2001). I would call the employed method: simulation of random walk governed by diffusion equation. This remark is just a suggestion and has no influence on the

referee's decision.

There is some confusion in description of the method. It is mentioned in the abstract and Eq. (6) says that the velocity of the merged particle is defined by conservation of momentum. On the other hand, according to the description of the method the initial velocities are not assigned to the molecules; neither positions nor the probability of sticking depend on the velocities. The particle positions are completely defined by diffusion equations and the velocity is just unnecessary detail in the simulations.

It is not clearly explained how the evaporation is incorporated into modeling. The correct procedure is compare the value $\exp(-\gamma \Delta t)$ to the generated random number r between 0 and 1. If $\exp(-\gamma \Delta t) > r$, the evaporation doesn't happen, otherwise – yes. Perhaps, it is done so but just not said.

As far as I could deduce from the text all values are obtained from one single run (one realization of the random process). The spatial and size distributions and size from one single run are not comparable to experiments. It is necessary to have big number of realizations (I recommend 10 000 - 100 000 for smooth results) with different initial positions of molecules. The procedure for the calculating of the average nucleation rate is correct since the ensemble averaging can be substituted with the time averaging.

The major flaw in the manuscript is that movement of particles is considered as diffusional. It is well known that if ratio L/R (Knudsen number) much more than 1, the collision frequency between the particles is defined by the rate from the gas kinetic theory rather than by the diffusional rate constant; here L is the mean free path, R is the size of the particle. Despite that most of the way particles move in the diffusional regime at large Knudsen numbers the limiting stage for collision is the last step when the particles move in the free-molecular regime. Fuchs ("Mechanics of Aerosols", Pergamon Press, London, 1964) discusses this problem at length in the book. For particles as large as 0.85 nm, the pressure of 1 bar, and temperatures 200-300 the Knudsen number is roughly 40 − 60.

The method employed in this study is not novel. If it were applied properly, it should give results identical to the ones from the solution of Smoluchowski problem since no new physics is introduce into the model.

I do not recommend the manuscript for publication in the journal Atmospheric Chemistry and Physics.