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Interactive Comment

Interactive comment on "Representation of nucleation mode microphysics in global aerosol microphysics models" *by* Y. H. Lee et al.

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We thank the reviewer for their constructive comments and suggestions and have made several changes to the paper to address the issues raised. Reviewers' comments are shown in italics with our response shown after each.

General comments:
In this work the authors investigated the sensitivity of simulated particle number and CCN concentrations to using different lowest size cutoffs and model time steps for aerosol microphysics. They found that using parameterized microphysics results in higher formation rate of CN10 from nucleated particles and shorter coagulation lifetimes of ultrafine mode particles than the model with explicit aerosol dynamics.

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While the particle number simulation is less sensitive to the choice of the microphysics timestep than to the choice of cutoff size.
The manuscript is clear and well structured. The topic is of interest to aerosol modelers and fits the scope of GMD well. On the other hand, I agree with reviewer 1 that more information should be provided regarding the computational cost of each experimental setup. Also, in my opinion, the relationship between the adaptive time step (for each aerosol microphysical process) and the 10min/1h time step (for process coupling) should be mentioned/described in the introduction section, so that the reader can understand the motivation of doing such sensitivity experiments better. I recommend publication of this manuscript once the authors have addressed the specific comments listed below.

Specific comments: P893: Results presented in present work are based on simulations with one global model with sectional microphysics. It's better to revise title to something like: "Representation of nucleation mode microphysics in a global aerosol model with sectional microphysics".

RESPONSE) We agree with the referee and changed the title to be "Representation of nucleation mode microphysics in a global aerosol model with sectional microphysics".

P896L27: Previous studies using box model simulations already provided some useful hints on how to solve the production-condensation-nucleation system more accurately. See section 2.3 of Herzog et al. (2004) and section 3 of Kokkola et al. (2009). Are there similar studies that investigated this issue using box model simulations with the sectional method?

Herzog, M., D. K. Weisenstein, and J. E. Penner (2004), A dynamic aerosol module for

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global chemical transport models: Model description, J. Geophys. Res., 109, D18202, doi:10.1029/2003JD004405.
Kokkola, H., Hommel, R., Kazil, J., Niemeier, U., Partanen, A.-I., Feichter, J., and Timmreck, C.: Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions, Geosci. Model Dev., 2, 97-112, doi:10.5194/gmd-2-97-2009, 2009.

RESPONSE) We think this comment is not related to the sentences in P896L27. Nevertheless, here is our response. The similar study has been done by Pierce and Adams (2009), and the gas-phase H2SO4 in this version of TOMAS is based on their work and assumed to be in pseudo-steady state equilibrium between chemical production and condensational/nucleation losses. This information was provided in Section 2.

Pierce, J. R. and Adams, P. J.: A computationally efficient aerosol nucleation/condensation method: pseudo-steady-state sulfuric acid, Aerosol Sci. Tech., 43, 216–226, 2009.

P897L14: It would be nice to let the reader know how high the additional computational

RESPONSE) We provided the information in Section 2.1 in the revised manuscript.

"Regarding the computational burden of each model configuration, based on a one-month simulation with binary nucleation using a 600 Mhz single processor of an SGI Origin 300. For TOMAS-30, it takes about 58 hrs. For TOMAS-36, the 1-hour time step run takes about 73 hrs, and for the 10-min time step run, about 122 hrs. Finally, for TOMAS-40, the 1-hour time step run takes about 79 hrs, and for the 10-min time

step run, about 123 hrs."

P902L5-9: Is there any boundary nucleation scheme included in the model?

RESPONSE) This version of TOMAS does not have any additional boundary nucleation scheme such as cluster-activation nucleation scheme. The ternary-based nucleation occurs in the boundary layer. The following sentence is added in the Section 2.1 in the revised manuscript.
"We would like to note that the TOMAS model used in this study does not include any additional boundary nucleation scheme such as the cluster-activation nucleation scheme because it has been shown that the ternary nucleation scheme used here does predict frequent nucleation events in the boundary layer (Jung et al., 2006; 2008; 2010)."

P902L26: How is CCN calculated in TOMAS?

RESPONSE) We provided the CCN calculation method in Section 2 in the revised manuscript.

"For in-cloud scavenging, the model uses modified Kohler theory to determine whether or not a particle is a CCN. The large-scale and convective clouds assume to have supersaturations of 0.2% and 1.0%, respectively. The activation calculation is performed in each grid cell and time step with precipitation based on instantaneous aerosol size and composition. CCN concentrations presented in this paper are computed based on the monthly mean aerosol size and chemical composition fields (Pierce et al., 2007; Lee et al., 2009 for more details). The particles larger than the activation diameter at 0.2% supersaturation is counted as CCN(0.2%). When the activation diameter fall

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in between size boundaries, we perform interpolation to determine what fraction of particles in a size section activate."

P902L27 and P917 Fig2: The pressure levels presented are strange. It would be hard to compare this result with those from other models. If possible, please show the nucleation rates on standard pressure levels. Also, since J10 is a very important quantity in the discussion, it would be helpful to provide J10 fields in the figure.

RESPONSE) The pressure levels shown in the figures are the mid-point of each vertical layer – Note that the pressure boundaries are provided in Section 2 in the revised manuscript. We agree that J10 can be useful quantity, but unfortunately we do not have J10 fields from TOMAS-36 and TOMAS-40 models. Since the primary emissions are fixed, we believe the CN10 maps can provide an indication of the differences in J10 fields between the simulations.

P903L7-8: It's surprising to me that the primary particle emissions can contribute so much to the Aitken mode number concentration (or CN10) near the surface. What's the emission size parameter (mass to number ratio) you use in the model?

RESPONSE) We feel it is important to provide the emission size assumptions used in the model, so the following paragraph is added in Section 2.

"Emissions of primary particles are treated as in previous TOMAS studies and are briefly summarized here. The primary sulfate is emitted in two modes; fifteen percent of the mass is emitted to the first mode (number median diameter (NMD) of 10 nm; geometric standard deviation (GSD) of 1.6) and the rest is emitted to the second mode (NMD of 70 nm; GSD of 2) (Adams and Seinfeld 2003). The size distribution of biofuel

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and biomass burning carbonaceous particles is assumed to have a NMD of 100 nm with a GSD of 2. Fossil fuel carbonaceous emission is assumed to have two modes based on Ban-Weiss et al. (2010). About 1.8% of the mass (64% of the total number) is emitted to a smaller mode (NMD of 17.5 nm and GSD of 1.6) and the rest is emitted to a larger mode (NMD of 60 nm and GSD of 1.9). Sea-salt emission parameterization is based on Clarke et al. (2006), and mineral dust emission parameterization is based on Ginoux et al. (2001) and Marticorena and Bergametti (1995). ”

P903L25-28: Why is the nucleation rate overestimated for ternary nucleation but underestimated for binary nucleation?

RESPONSE) The lower nucleation rate in TOMAS-30 and TOMAS-36 is expected as their J10 is overpredicted and can lead to higher condensational sink (i.e. less sulfuric acid available for nucleation). So what we saw in the binary nucleation simulations is expected. In case of the ternary nucleation, we believe the lower nucleation rate shown in TOMAS-40 is due to a condensational sink by the nucleation mode particles (1-3 nm). Although their condensational sink is generally very small compared to the total condensational sink, it can become important under very fast nucleation event (e.g. fast ternary nucleation). Supporting this hypothesis, we found that the fast ternary nucleation areas show a lower sulfuric acid concentration in TOMAS-40. The following is added in Section 3.1.

“We would like to note that the underestimated Jnuc in 10 nm simulations for binary nucleation can be explained with the higher condensational sink (i.e. higher J10). However, it is not the case for the ternary nucleation. We believe that only during a very fast nucleation event by ternary nucleation the small nucleation mode particles (1-3 nm) can be a relatively important condensational sink as their number concentration is very high when nucleation occur, resulting lower nucleation rate. Except this case, their

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condensational sink is generally very small. This hypothesis can be also supported by the lower sulphuric acid concentration in TOMAS-40 over areas showing very fast nucleation rate (not shown). We think this partly explains why the J10 overprediction is too much worse in the 10 nm simulation with the ternary nucleation."

P904L10-12: Why is the ultra-fine mode lifetime in the 3nm simulation even larger than that in the 1nm simulation?

RESPONSE) First of all, we would like to mention that the higher lifetime is pronounced for the ternary nucleation. As shown in Figure 3d, most upper/free troposphere shows the increase of the CN10 concentration in the 3nm simulations, and these regions show higher CN10 concentration with ternary nucleation (see Figure 2b and 2e). Increasing CN10 in these pristine areas relative to the polluted areas (polluted region near surface and near continents where has higher coagulation loss rates) can lead to the longer lifetime of ultrafine mode particles.

P904L6-7: Do you mean the difference between the 3nm run and the reference simulation is around a few percent to 25%?

RESPONSE) Yes. As shown in Table 1, the difference in the ultrafine mode budgets is up to 25% between the 3 nm run and the 1 nm run. We rewrote the following sentences to clarify.

OLD: "In the 3 nm simulations, the aerosol burden and lifetime of the ultrafine mode are from a few percent to 25% of the reference case, and the 10 nm simulations show much larger errors compared to the references cases than do the 3 nm simulations"

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NEW: "Compared to the reference simulations, the aerosol burden and lifetime of the ultrafine mode in the 3 nm simulations differ by a few percent up to 25%, and the 10 nm simulations show much larger errors than do the 3 nm simulations."

P905L14: It would be helpful to include numbers for the reference simulations in table2, otherwise the reader have to turn back to table 1 for detailed comparison.

RESPONSE) We followed the referee's suggestion.

P906L11-12: Why is there underestimation of CN10? These regions are over pristine area and J10 may possibly be overestimated.

RESPONSE) The underpredicted CN10 areas in the binary nucleation simulations have no nucleation event, and those in the ternary nucleation have particularly lower nucleation rates and free ammonia concentrations in TOMAS-30 compared to TOMAS-40 model. For ternary case, we think the nucleation rate in TOMAS-30 might be too low to lead higher CN10. The following sentence is added in the revised manuscript. The new part is shown as bold.

"Unlike the 3 nm simulations, the 10nm simulations (shown in Fig. 3a, c) show underpredicted CN10 in some parts of the tropics for both binary and ternary nucleation schemes. **The underpredicted CN10 areas show no nucleation event for the binary nucleation and, for the ternray nucleation, noticeably lower nucleation rates and lower free ammonia concentrations in TOMAS-30 than TOMAS-40.**"

Interactive comment on Geosci. Model Dev. Discuss., 6, 893, 2013.