

***Interactive comment on* “Simulation of particle size distribution with a global aerosol model: contribution of nucleation to aerosol and CCN number concentrations” by F. Yu and G. Luo**

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Review of “Simulation of particle size distribution of a global aerosol model: contribution of nucleation to aerosol and CCN number concentration” by F. Yu and G. Luo

This article describes the implementation of aerosol microphysics into the GEOS-CHEM global CTM. This model is then used to simulate the fraction of total particle number and CCN number that is primary versus secondary throughout the troposphere. The total number of particles larger than 10 nm (CN10) are compared to

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observations, and the sensitivity of this comparison to “primary sulfate” is found. The size distributions above several sites are also shown to give insight into the relationship between vertical structure and aerosol processes.

The paper presents a new approach to the global nucleation-CCN question. Where previous groups have modified the global nucleation rates and seen how CCN concentrations have changed (thus finding the sensitivity of CCN to nucleation), none explicitly determined the fraction of CCN that began as secondary particles. Rather than finding the sensitivity of CCN to nucleation as done in previous papers, Yu and Luo have a model configuration that explicitly tracks the nucleated particles and determines the contribution of these particles to CCN. Thus, the paper contributes new information the global aerosol-cloud-climate community. The paper is high quality and deserves to be published in ACP once the following concerns have been addressed.

General comments

1. I do not feel that using an average global supersaturation of 0.4% is the best way to assess the impact of nucleated particles on clouds. Stratus clouds that cover large areas and are most susceptible to changes in albedo due to aerosols have maximum supersaturations of 0.1-0.2%. Convective clouds are generally less susceptible to cloud albedo changes (except for shallow cumulus), but may have dynamical feedbacks due to changes in CCN. Maximum supersaturations have large ranges in these clouds, but often reach as high as 1% or greater.

Furthermore, for wet removal you are using a cutoff diameter that depends on the type of clouds. Therefore, in areas dominated by large-scale wet removal (with an activation cutoff of about 0.2%), the CCN(0.4%) assessment is including ultrafine particles that have not yet able to activate in local clouds and be subject to wet removal. The lifetime of these particles that have critical supersaturations between 0.2 and 0.4% could be much longer than those with critical supersatura-

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tions lower than 0.2% and thus would bias your CCN estimates in favor of these particles. Conversely, in regions where convective wet removal dominates, you are not including the smallest activated particles.

If you want to take into account the potential affects of aerosols on convective systems, it would be better to add panels to Figures 9 and 10 and show results from two supersaturations, one corresponding to your large-scale activation diameter (which should be a supersaturation of about 0.2%) and the other corresponding to your convective activation diameter (which should be a supersaturation of about 1%). If you only are concerned with the clouds most susceptible to albedo changes, then it would be best not add extra panels, but to lower the supersaturation to 0.2%, the cutoff for activation in your large-scale clouds.

2. I believe this is the first time a single-moment sectional aerosol microphysics algorithm has been used in a global model. I believe you have configured the algorithm to conserve number during microphysics; however, this introduces some numerical diffusion to the shape of the size distributions. You do have a large number of size sections, so it is likely that this error may be low enough to ignore, but it would be useful to either include a benchmark comparison to systems with analytical solutions for condensation and coagulation, or reference a previous paper that has done this or similar.

Specific comments

1. Page 10598, Line 12: A question of philosophy: Why constrain primary emissions, but not nucleation, deposition SOA etc.? All have large uncertainties.
2. Page 10599, Line 23: “substantial fraction” to me implies “nearly all” or at least “more than half”. I think saying “non-trivial fraction” is more precise because, while you cannot ignore the contribution of nucleated particles to CCN (as you

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- show in this paper), I would assume that fewer than half actually grow to CCN sizes (globally averaged).
3. Page 10600, Line 14: The primary sulfate particles in these studies were used to represent sub-grid nucleation. You should mention “these studies” in that sentence because the papers with using the ECHAM5-HAM model (e.g. Stier et al., 2005 and Makkonen et al., 2009) add the anthropogenic sulfate to accumulation mode and coarse mode. In the Makkonen paper, it says primary sulfate is used to represent sub-grid nucleation; however, unless they’ve changed the size distributions from Stier et al., 2005, the particles are much larger than those used in the other global studies mentioned. (Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 5, 1125-1156, 2005.)
 4. Page 10600, Line 24: The Pierce and Adams (2009) paper has simulations with activation-type nucleation in the boundary layer also. These results are very similar to the simulation with primary sulfate in Wang and Penner (2009).
 5. Page 10606, Lines 17-end of page: Again, unless the primary sulfate emissions are changed in Makkonen from that of Stier et al., 2005, the primary sulfate is very different from the values you’ve listed. Also, in Spracklen et al., 2006, they have turned off their primary sulfate emissions. In their 2008 paper they don’t address primary sulfate, so I assume that it’s still shut off. It might be worth asking people from these groups what they actually have been using.
 6. Page 10609, Line 27: Why is the primary sulfate that condenses only condensed onto BC and OC rather than also onto existing sulfate, sea salt and dust? This condensation could be particularly important for growing ultrafine sulfate to CCN sizes. However, I do see some difficulties condensing this material in general because the plume where the sulfate is condensing does not fill the entire gridbox.

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- Therefore, some of the existing particles should grow, while others should not. This is a difficult issue.
- Page 10611, Lines 17-18: I guess it is implicitly assumed that the hydrophobic OC and BC are externally mixed from all other species here (including sulfate) since they can't act as CCN. Is there an aging timescale from hydrophobic to hydrophilic? (It is possible that the aging timescale is mentioned somewhere else in the paper, but I missed it.)
 - Page 10612, Line 16: When any particle from your sulfate bins coagulate with your bulk populations (e.g. OC), the sulfate mass becomes associated with the bulk population? I believe this may bias the contribution of nucleation to CCN low. If a 200 nm sulfate particle that formed through nucleation coagulates with a 40 nm OC particle, you then add the sulfate mass to the OC population even though it was the sulfate particle that had already grown to CCN size. I realize that you plan on adding these other species into the microphysics in the future, but its probably worth mentioning this numerical difficulty here.
 - Page 10612, Line 17: Each process (coagulation, condensation and particularly aqueous oxidation) will add sulfate to different sized particles (e.g. aqueous oxidation will only add sulfate to activated particles). How do you distribute the sulfate (and SOA) across the bulk populations.
 - Page 10614, Line 2: Why “especially nucleated particles”? This may be the case if the nucleated particle is mostly sulfate and you're looking at the partitioning of ammonia; however, the case might be very different if you are looking at the partitioning of relatively hydrophobic SOA (relative to inorganics) that wants little to do with water and inorganic ions.
 - Page 10614, Line 22: Have you operator split the chemical generation of H₂SO₄, nucleation and condensation processes with each acting for 15 minutes, or do

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you integrate these three processes together up to an overall timestep of 15 minutes? In cases with high aerosol surface area, condensation may deplete H₂SO₄ gas-phase concentrations significantly during the timestep, so that the operator splitting method won't work for these long overall timesteps.

12. Page 10615: Is coagulation called every 15 minutes and is done separately from the condensation/nucleation calculations? In polluted areas, the coagulation timescales for 1 nm particles may be significantly shorter than 15 minutes. This means that particles may be formed and grow to larger sizes (where they have longer coagulation timescales) before coagulation has been called.
13. Page 10617, Line 19: Is there a model spin-up period?
14. Page 10619, Lines 5-7: Aerosol removal or transport may also have errors leading to errors in the comparison.
15. Page 10619, Lines 11-15: It would be useful to put some quantification of average bias and/or error for the two model simulations to summarize the difference between the simulations overall.
16. Figure 6 and Section 3.2: How much do the number of particles and the fraction of particles that are secondary change when you look at CN10 rather than CN3. Please add a figure or at least discuss this in the text. It will be useful to know how many particles are trapped in that size gap.
17. Figures 3-10: Please add a sentence to the captions about whether the units in the figures are normalized to STP or not. It is mentioned in the text but not for all figures, and it would be useful to have it in the caption.
18. Section 3.4: How sensitive is the fraction of particles that are secondary to the primary sulfate emissions? Its probably not worth it to add a figure, but it would be interesting to see a brief discussion in the text.

19. Page 10626, Line 15: Please use “non-trivial” rather than “substantial” here, again. You did not assess the fraction of secondary particles that make it to CCN sizes.
20. Page 10626, Line 29: It would be more precise to say “primary sulfate emissions” or “subgrid nucleation sulfate particles” here because you did not change the other primary emissions.

Spelling or grammar corrections

1. Page 10605, Line 26: “have” should be “has”.
2. Page 10608, Line 14: “latitudes” should be “altitudes”.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 10597, 2009.

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