Atmos. Chem. Phys. Discuss., 9, C1351–C1354, 2009
www.atmos-chem-phys-discuss.net/9/C1351/2009/

Atmospheric Chemistry and Physics Discussions

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

F. Yu and G. Luo

yfq@asrc.cestm.albany.edu

Received and published: 30 May 2009

The authors thank Jeffery Pierce for the constructive and detailed comments. Our responses to the comments are given below.

General comments

1. Agree. We will revise Figures 9 and 10 to provide CCN information corresponding to stratus and convective clouds.

2. The numerical diffusion is generally small when the number of bins per decade of size space is > 10 (Jacobson et al., Atmos. Environ., 28, 1327, 1994; Yu, Thesis,

1998). For the size range important to CCN (<100 nm), we use $\sim$ 15 bins per decade of size space and thus the numerical diffusion is expected to be small (compared to other uncertainties associated with various parameterizations and model resolutions).

Specific comments

1. Because we have reasonable observations of primary emissions. As pointed in the text, the nucleation mechanism used in this study has been constrained by multiple-instrument characterization of nucleation events observed in the boreal forest. If there exist reasonable measurements of deposition and SOA processes, the model should be constrained in these aspects as well (which could be good future research topics).

2. Based on dictionary, substantial means “ample, sizeable”. Nevertheless, to address the referee’s concern, we will change “substantial” to “non-trivial” as suggested.

3. Will modify the sentence as suggested.

4. We will change the sentence to “The global aerosol simulations reported in Spracklen et al. (2008), Makkonen et al. (2009), and Pierce and Adam (2009), based on . . .”

5. See the point. To be more accurate, we will delete the references in question (“Spracklen et al., 2008; Makkonen et al., 2009”) from line 17 on page 10606.

6. To put the primary sulfate of the condensation mode onto BC and OC is an approximation. We agree with the referee that the approximation may underestimate the growth of ultrafine sulfate particles to CCN sizes. How to represent the sub-grid plume scale sulfate formation process in the global model is a subject that needs further research.

7. See page 10608, lines 4-10.

8. Good point. Yes, we added the sulfate mass to primary particles (BC, OC, dust, sea-salt) after coagulation even when the sulfate particles are larger than the primary

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particles. This might have underestimated the contribution of secondary particles to CCN. We will point this out in the revised manuscript.

9. We distribute the sulfate (and associated SOA, nitrate, ammonium) to the bulk populations according to the surface area of primary particles.

10. The nucleated particles are generally small and thus the condensation (or equilibrium uptake) can significantly change their sizes. In contrast, primary particles have much large sizes (and mass) and the relative changes of their sizes associated with condensation (or equilibrium uptake) are generally smaller.

11. The three processes are integrated together for the time step (15 min).

12. Coagulation time step depends on the particle concentrations (i.e., variable) (line 19, page 10615).

13. The original model (54 tracers) was spun up for one year and then the coupled model (127 tracers) was spun up for two additional months.

14. Agree. Some of these errors must have contributed to a factor of two differences shown in Figure 5.

15. The quantitative difference between the two model simulations have been shown in Figures 3-5. More comprehensive analyses of the impact of primary sulfate particle emission parameterizations will be carried out and reported in a separate paper.

16. The concentrations of particles between 3 and 10 nm depend on the locations. Interested readers can get some information from Figure 7 and by comparing Figure 3a with Figure 6c. Considering the paper length, we would like not to add an additional figure on this.

17. All the results presented in the manuscript are not normalized to STP. Instead of repeating the statement in each caption, we will point this out in the first paragraph of Section 3.

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18. The effect of primary sulfate emission is limited to source regions in the BL (also see Figures 3-5). Will add a brief discussion on this.

19. Will modify as suggested.

20. In addition to primary sulfate particles, we also used observations to constrain the emission of BC and OC. BC and OC particles contribute significantly to total particle number concentrations in many regions and it is important to use a reasonable size parameterization.

Spelling or grammar corrections

Will modify as suggested.

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