Atmos. Chem. Phys. Discuss., 10, C10500–C10506, 2010
www.atmos-chem-phys-discuss.net/10/C10500/2010/

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Chemistry
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Discussions

Interactive comment on "Sensitivity of global cloud condensation nuclei concentrations to primary sulfate emissions parameterizations" *by* G. Luo and F. Yu

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Received and published: 30 November 2010

Review of "Sensitivity of cloud condensation nuclei concentrations to primary sulfate emissions parameterizations" by G. Luo and F. Yu

This paper shows the sensitivity of predicted CN and CCN in a global chemical transport model to the representation of sub-grid sulfur-plume nucleation (a.k.a. primary sulfate). They explore changes of both the total mass of primary sulfate as well as the fraction that went into forming/growing new particles versus condensation onto existing particles.

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Discussion Paper

Overall, I think this is an interesting study of an important issue that deserves to be published in ACP. However, I feel that there are two major issues and several more minor issues with the paper that must be corrected/improved before it can be published.

Major issues

1. There are several cases in the paper where information about previous work is incomplete and comparisons to previous work are misleading.

    (a) Page 27696, 2nd paragraph: Spracklen et al., 2008 did not have any primary sulfate (see the changes described in Spracklen et al., 2006). Makkonen et al, 2009 assumes that all primary sulfate is at accumulation-mode sizes (based on the AEROCOM emissions paper, Dentener et al., 2007).

    (b) Page 27698, line 7: Adams and Seinfeld, 2003 had no primary particles other than primary sulfate, so the large increases in particle number came from switching from a nucleation only simulation to a simulation that included primary particles, so the impact of primary sulfate is overstated because of this. Similarly, Spracklen et al., 2005b only had sulfate and sea-salt (no OC, EC and dust). Thus, primary sulfate were the only continental source of primary particles.

    (c) Page 27698, line 8: Pierce and Adams, 2009. The first case mentioned here had no boundary-layer nucleation thus will overstate the sensitivity of primary particles. Also, the CCN concentrations DECREASED by 40% from the base case when primary emissions were DECREASED by 40%. Likely the percent change would be less than this if primary emissions were increased from the base case due to CCN saturation.

(d) Page 27698, lines 19-21: “It is clear... factor of up to ˜6”. Nucleation schemes definitely matter here (mostly strongly if some form of BL nucleation is used versus not used); however, this is likely not the most important factor in the variance between different studies. The Adams and Seinfeld, 2003 and Spracklen et al., 2005b have no other continental primary emissions (no OC, EC and dust), and Adams has no sea-salt. Therefore, these studies will have much higher sensitivities to primary sulfate.

I would guess that the primary-sulfate sensitivity difference between two identical simulations with different BL nucleation schemes (e.g. Activation vs. IMN) would vary by far less than a factor of 2. The Wang and Penner (2009) BHN_EMP compared with your FS2.5FN15 simulation is really the closest thing to an apples-to-apples comparison.

(e) Page 27708, Line 10; Page 27696, line 22; The Wang and Penner BHN_EMP is really the apples-to-apples comparison here. When you compare your FS2.5FN5 simulation to their BHN_EMP simulation, the difference is a factor of 3, but this can largely be explained by the fraction of the mass that goes into new particles (5% vs. 15%). When you compare your FS2.5FN15 simulation to theirs, the difference is <50% rather than a factor of 3. The difference between FS2.5FN15 and BHN_EMP is what you can attribute to the nucleation scheme, other primary emissions, scavenging etc. (you don't need these to explain the factor of 3-7 uncertainties that you mention).

(f) Page 27710, Line 1: It is inappropriate to compare directly to Pierce and Adams, 2009 here because we turned down ALL primary emissions by a factor of 3 rather than shutting off just primary emissions. Also, make sure you are comparing apples-to-apples when comparing to Wang and Penner, 2009 (see the point above).

2. The authors compare their results from their simulations to measurements of CN10. This is a good exercise to make sure that the model is in the realm of reality; however, it is a poor method for determining if a certain model input is better than another. There are many ways to get a good comparison of variables in global CTMs with aerosol microphysics.

   In a previous paper (Yu, F., G. Luo , T. Bates , B. Anderson , A. Clarke , V. Kapustin , R. Yantosca , Y. Wang , S. Wu, Spatial distributions of particle number concentrations in the global troposphere: Simulations, observations, and implications for nucleation mechanisms, J. Geophys. Res., 115, D17205, doi:10.1029/2009JD013473, 2010.), the authors use this technique to determine if different nucleation schemes are appropriate or not. In the current paper they use the same technique to determine the correct amount of primary sulfate and the fraction of this mass to nucleated particles. The effect of nucleation schemes and primary-sulfate assumptions on predicted CN10 concentrations in the continental BL are very similar. Thus, one may get a good fit to observations from lower nucleation and higher primary sulfate, or visa-verse.

   Furthermore, there are many more uncertain factors that affect CN10 concentrations (such as wet deposition, dry deposition, OC and EC emissions rates and sizes).

   I feel that the authors do not clearly discuss these uncertainties and imply that 2.5% primary sulfate with 5% to the nucleation mode is definitely the

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best value to use. It may end up being that this is a robust value to use for many peoples' models, but it may also end up being an artifact of the specific assumptions currently being used in this model.

Please add a discussion of the uncertainties in CN10 predictions to the end of Section 3.3 and make sure to state the best fit of FS2.5FN5 is specifically for the assumptions used in the model.

Also, please state this briefly in the abstract (p. 27696, l. 18).

Other comments

1. Page 27696, line 9; Page 27699, line 9; Page 27709, line 3: Saying that the APM scheme is the first to consider condensation of low-volatility vapors to nucleated particles is misleading. Pierce and Adams, 2009, Spracklen et al. 2008 and all recent microphysics papers out of Carslaw's group all assumed that SOA was non-volatile and condensed SOA onto all particles (including freshly nucleated particles). Saying that this is the first scheme to have SOA condensation onto nucleated particles implies that the previous studies missed this important source of growth. What the APM has that is new is that it dynamically ages semi-volatile SOA to low-volatility SOA, which is a nice advancement, but what was stated implies something different.

2. Page 27698, line 15: "the empirical nucleation mechanism". It would be more descriptive to call this empirical activation-type nucleation since this is the type

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Discussion Paper

of fit they used.

3. Page 27700, line 16: This is related to two points above. “The treatment significantly increases the growth rates of nucleated particles...”. Increases compared to what? The papers listed in the above point used a simple scheme that condensed ALL SOA as non-volatile onto the surface area. The new scheme used in this paper would actually give slower growth (for the same amount of SOA condensation) because not all SOA would be treated as low or non volatile. I'm guessing you mean that it increases growth rates compared to schemes that treat SOA as semi-volatile and partitions to the aerosol mass distribution.

4. Page 27701, line 23: Why are lognormal-mode parameters given for the accumulation mode (and not just the nucleation mode) when it is being condensed? I think this is a typo since the listed modal parameters are the same as the nucleation mode.

5. Page 27701, line 23: Why is condensing sulfate only condensed onto BC and OC and not sulfate, sea salt and dust?

6. Page 27704, line 28: “freshly nucleated mode, nucleation mode and accumulation mode”. Did you mean, “freshly nucleated mode, Aitken mode and accumulation mode”?

7. Table 2 and Page 27706: What is standard deviation of error? Also, since the observation data spans well over an order of magnitude and you've plotted the

comparison on a log-scale, I find that errors in log(data) to be much more useful. With the linear errors that you are giving now, errors in the polluted points (high CN concentrations) are going to dominate the error.

8. Page 27706, line 28: Often the non-linear processes in microphysics cause a dampening of changes in CN10 and CCN to changes in model inputs (e.g. A factor of 1.5 change in an input variable would cause a change in CN10 of a factor of 1.2). Can you please discuss further the reasons for the positive non-linear enhancements you saw? I am curious to know if you condensed the "accumulation mode" primary sulfate mass also onto the sulfate distribution (and not just OC and EC) if you would still get such a discrepancy between FS5FN5 and FS2.5FN15. This would increase the growth rates of sulfate particles to CCN sizes greatly in the FS5FN5 case.

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