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Atmospheric Chemistry and Physics 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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Manuscript Title: Sensitivity of global cloud condensation nuclei concentrations to primary sulfate emissions parameterizations

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Reply to Referees' comments

The authors thank Jeffery Pierce for the thoughtful and constructive comments which help to improve the manuscript. Our point-to-point replies to the comments are given

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below. The manuscript has been revised accordingly. All the changes have been highlighted in the revised manuscript using MS word “Track Changes” tool.

Major issues 1. (a) Page 27696, 2nd paragraph . . .

To reflect this, we have modified the sentence as follows: Many of the above mentioned global aerosol studies assume that 15

(b) Page 27698, line 7 ...

Agree. The model assumptions of other primary particles are also important. This has been pointed out in the revised manuscript.

(c) Page 27698, line 8 . . .

Agree. The impact of primary sulfate emission on CCN depends on both nucleation schemes and emissions of other primary particles. This is what we want to emphasize in the Introduction. The sentence has been slightly modified.

(d) Page 27698, lines 19-21 ...

To avoid ambiguity, we deleted this sentence.

(e) Page 27708, Line 10; Page 27696, line 22 . . .

Good suggestion. We have modified the revised manuscript to address this.

(f) Page 27710, Line 1 ...

True. We have modified the paragraph to reflect this.

2. The authors compare their results from their simulations to measurements of CN10 . . .

A discussion of the uncertainties in CN10 predictions has been added to the end of Section 3.3. We also stated that the best fit of FS2.5FN5 is specifically for the assumptions used in the model. It is also stated in the abstract.

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.

Fair comments. We have modified the text to avoiding confusion and point out that other studies have considered the condensation of SOA although using a different scheme.

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 of fit they used.

Done.

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.

Yes, we mean that it increases growth rates compared to the original SOA formation scheme in the GEOS-Chem. We have clarified this.

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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.

Yes, it is a typo. Corrected.

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

It is a treatment of primary sulfate emission used to represent sub-grid sulfur oxidation and sulfate particle formation in anthropogenic SO2 plumes. The sulfate mass on accumulation mode particles is a result of condensation and thus shall not increase the number concentrations. This is a simplification to avoid adding number concentration to the accumulation mode particles. Usually, BC and primary OC particles so-emit in these plumes.

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

Yes, corrected.

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.

Standard deviation of error in this study is the sample standard deviation of the errors between simulations and observations. It shows how much variation or "dispersion" there is from the "average".

To calculate log(data) errors is a good idea and will reduce the impact of errors in the polluted points. The conventional approach is more relevant to the errors in total concentration deviations (or global burden). In this study we would like to use the conventional error analysis approach but will keep this in mind in our future analysis.

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 nonlinear 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.

We think that the "positive nonlinear enhancement" was caused by the non-linear dependence of nucleation rate on [H2SO4] which is affected by [SO2] and CS (Figs 1 2). Here (Fig. 4) we were comparing CN10. To condense the "accumulation mode" primary sulfate mass also onto the sulfate distribution (and not just OC and EC) are not expected to affect the total CN10. As to CCN, the sizes of accumulation mode become important. To condense the "accumulation mode" primary sulfate mass also onto the sulfate distribution may increase the sulfate CCN but at same time may decrease the OC/EC CCN. The net effect on total CCN is likely to be small.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 27695, 2010.

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