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Interactive comment on "Simulation of particle formation and number concentration over the Eastern United States with the WRF-Chem + APM model" *by* G. Luo and F. Yu

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Simulation of particle formation and number concentration over the Eastern United States with the WRF-Chem + APM model
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Reply to Referees' comments

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The authors thank the referee for the thoughtful and constructive comments which help to improve the manuscript. Our point-to-point replies to the comments are given below. The manuscript has been revised accordingly. All the changes have been highlighted in the revised manuscript using MS word "Track Changes" tool.

How does the model distinguish between secondary formed and primary CCN? I could not easily deduct are the particles handled completely externally mixed? What happens in mode-to-mode coagulation?

In our study, activated CCN from sulfate particles (containing nitrate and ammonium through equilibrium uptake or partition) is determined as secondary formed CCN. In the APM, secondary particles are separated from primary particles but the coating of secondary species on primary particles is considered (we termed such an aerosol system as "semi-externally mixed"). The coagulation scavenging of secondary sulfate particles by primary particles are simulated using four separate tracers (BC sulfate, OC sulfate, sea salt sulfate, and dust sulfate) to keep track of the bulk sulfate mass associated with BC, OC, sea salt, and dust, respectively. Condensation (of sulfuric acid) and in-cloud SO2 oxidation also contribute to BC sulfate, OC sulfate, sea salt sulfate, and dust sulfate. When some of BC, OC, sea salt, and dust are removed from the atmosphere via dry and wet deposition, the corresponding portion of sulfate associated with these particles are also removed. We distribute the sulfate (and associated nitrate, ammonium) to the bulk populations according to the surface areas of primary particles.

The APM explanation (2.2) needs to be reduced. After line 3 in 14665, there is very little relevant information for this paper, as this work should be considering what is done, not what is possible to do with APM

Reduced.

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The authors claim VOC as one of the emissions considered (ln 4, pg 14666). How does the VOC oxidation take a part in the system? They claim that it does not take a part in the nucleated particle growth, but do SOA from SORGAM module any way influence the system?

VOC oxidation and its impact on photochemistry are considered by CBMZ chemical scheme in the system. Because we are still looking for a suitable SOA formation scheme for WRF-Chem, we do not consider the contribution of SOA to the particle growth in the present study. Recent study indicates that the production of low volatile organics from oxidation aging of secondary organic species is necessary in order to properly account for the contribution of SOA to particle growth (Yu, 2011). Further research is needed to include such a SOA formation mechanism in WRF-Chem + APM.

What are the consequences of removing all anthropogenic sulphur emission in comparison to INTEX-A measurements? This is important issue, not only from direct nucleation point-of-view, but for the overall growth to CCN or removal by coagulation point-of-views. I would urge the authors to consider a small sensitivity analysis of this.

We do not remove all anthropogenic sulfur emission in our study. We assumed that all anthropogenic sulfur emission is emitted as SO2 and no anthropogenic sulfur is emitted as primary sulfate particles. Actually, primary sulfate particles in anthro-pogenic sulfur emission are secondary particles formed in sub-grid plumes. When model resolution is low and no nucleation scheme can be employed to simulate sulfate nucleated in sub-grid plumes, primary sulfate emission rate has to be assumed in these investigations. However, primary sulfate emission parameterization is expected to have large uncertainty, and presently it is difficult to determine the correct value for sub-grid secondary sulfate particle formation (Luo and Yu, 2011; Yu, 2010b). In this study, because WRF-Chem + APM has high model resolution (27 km x 27 km) and the IMN nucleation scheme has been employed, the absence of primary sulfate

particles in anthropogenic sulfur emission can be compensated by nucleated sulfate particles. The sensitivity analysis of primary sulfate emission parameterization done by Luo and Yu (2011) indicated that the inclusion of primary sulfate emissions (2.5% of anthropogenic sulfur emission emitted as primary sulfate and 5% of primary sulfate mass in the nucleation mode) can enhance the CN10 and CCN0.2 up to 10-20% over the Eastern United States.

Why are the sectional bins so different in different particle types? How do the coagulation work between these types and do you consider that e.g. sea-salt particles covered with SOA or sulphate? Are the particles always externally mixed? The section 2.3.2 is rather difficult to read.

One important feature of APM is that its bin resolution can be varied for particles of different types, so we can use a higher resolution for the size range of our interest. This enables us to use a reasonable number of bins to cover a wide size range while optimizing size resolution. The model allows the different microphysical processes to be calculated at different time steps and automatically decides the optimum time steps for computing efficiency without sacrificing accuracy.

We considered the self coagulation of size-resolved sulfate and sea salt particles, as well as the scavenging of secondary particles by sea salt, dust, BC, and POC particles. These secondary species scavenged by primary particles become part of primary particles as coated materials and are tracked. We have added some discussions to the section to clarify.

line 20, pg 14668: Are the sulphate particles ONLY scavenged by primary particles? Not at all by (quite numerous?) CCN sized grown particles from NPF? What are the consequences of such choice?

Sulfate particles can also be scavenged by NPF by self coagulation of size-resolved

sulfate in APM.

As the authors do not consider primary sulphate emissions, this might be of less importance, but do the emission rates of primary number emissions take into account sub-grid scale coagulation (e.g. Pierce et al, 2009). If not, some discussions on the importance of such processes are needed.

As we pointed out that the absence of primary sulfate particles in anthropogenic sulfur emission can be compensated by nucleated sulfate particles. In APM, nucleation, self-coagulation, condensation/evaporation and interaction with background aerosol are included. Sub-grid scale coagulation of primary particles can be accounted by APM aerosol microphysics simulation. Luo and Yu (2011) indicated that the emission rates of primary sulfate emissions are determined by oxidation and nucleation of sulfur in the plume, while sub-grid scale coagulation and other aerosol microphysics processes will contribute to the size distribution of primary emitted particles.

I think it is good that the authors considered the ability of the nudged CTM to actually produce the measured meteorology (3.2.1). However, as the BL processes are not well re-produced by the model (not surprising), the discussion on the NPF should also include BL evolution as an error source of NPF, especially as most of the nucleation seems to (?) happen near BL.

Agree. We added that the uncertainty of BL process could be expected as an error source of NPF in the revised paper.

The CN10 discrepancies between measurements and model results could also be strongly affected by the growth/removal in the 1-10 nm part. Here the absence of organics can be crucial. Where does the NPF happen in the model and how would the

increase of growth rates in BL from organics affect the results?

NPF happens in the lower troposphere associated with high concentration of sulfur acid gas and upper troposphere due to the low temperature there. Yu (2011) studied the contribution of successive oxidation aging and kinetic condensation of organic compounds to particle growth and pointed out that this process can significantly increase the growth rates of nucleated particles associated with the condensation of low-volatile SOG. SOA and its contribution to particle growth are likely to important in boundary layer over the eastern U.S., especially during summertime. In the future, we are going to include the recently developed extended SOA formation mechanism (Yu, 2011) into WRF-Chem + APM.

Figure 1 two leftmost columns are rather hard to distinguish where the changes actually are. One way would be to color code the scatterplots (right column) with measurement height.

Good suggestion. Changed as suggested.

as in the above point, figures 2 and 3 scatterplot (b) should be color-coded by altitude. Then (c) and (d) could be replaced with modelled top view (as in (a))

Scatter plots have been color-coded by altitude.

The resolution based comparisons (Fig 4 and 5) are interesting. I would like the authors to include (for direct comparison) an additional figure, at least on the map figures: How would the WRF-CHEM-APM results look, if they would be averaged to the GEOS-CHEM-APM grid? This would also show if the "small" scale processes of WRF would actually have an important effect on the cloud processes in the GEOS scale. If we would then consider WRF simulations as "accurate", this would work as

*some sort of approximation on sub-grid scale variation effects of the processes in the
global scale model?*

Good suggestion. We added the figures of WRF-Chem + APM regrided to the
GEOS-Chem + APM scale. Associated discussions have been updated.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14659, 2011.

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