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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 Dr. Jerome Fast for the detailed, 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.

*Page 14660, line 11: I assume that GEOS-Chem also provides the boundary conditions. The phrase regarding the spin-up time is not central to the manuscript and can be omitted from the abstract.*

Agree. We rewrote the sentence as follow: Size- and composition- resolved aerosol properties from GEOS-Chem + APM simulations are used to initialize and provide boundary conditions for the WRF-Chem + APM model.

*Page 14660, lines 13 – 14: The statement regarding the “reasonable agreements have been obtained” is very general. Can something more specific be summarized in the abstract?*

We rewrote the sentence as follow: The modeling results have been evaluated with the relevant measurements obtained during the INTEX-A field campaign in the summer of 2004. Model simulation captures the high concentrations of SO<sub>2</sub> and CN<sub>10</sub> at surface layer and source regions but under-predicts the values in the upper troposphere.

*Page 14661, lines 11 – 13: It is true that some studies have asserted that size distribution is more important than composition, but the issue is still open to debate.*

Agree. It has been rewritten as follow: Dusek et al., (2006) suggested that the aerosol number size distribution may play an important role in the determination of the particles’ ability to act as CCN.

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*Page 14661, line 25: Change “is the major publically” to “is a publically”. The sentence is awkward.*

Changed.

*Page 14661, line 27: Here and elsewhere, the authors have focused on comparing APM with MOSAIC; however, there are two other aerosol models with substantially different approaches in WRF-Chem. It is not clear why those are not mentioned, or how the statements on MOSAIC differ from those for the other two aerosol models.*

WRF-Chem v3.1.1 includes three choices for aerosol models which are GOCART, MADE/SORGAM and MOSAIC. GOCART does not include aerosol microphysics. MADE/SORGAM employs aerosol microphysics based on modal approach. Modal approach, while computationally efficient, is not very suitable for rigorous aerosol microphysics simulation. When particle nucleation and growth happens frequently, the modal approach loses a lot of details of aerosol microphysics (for example, the difference in the sizes of freshly nucleated particles of a few nanometers to aged secondary particles 50-100 nm, size-dependent growth and coagulation rates, etc.). MOSAIC considers aerosol microphysics based on sectional approach, the same approach employed by APM. Therefore, we feel APM and MOSAIC are more similar and more comparable than the other two models. We added some discussions on this in the revised paper.

*Page 14662, line 12: Suggest changing “is its size range and resolution” to “is its default size range and resolution”. MOSAIC was released with these default settings in anticipation of how most user’s would apply the model and have chosen a limited number of size bins because of the computational cost (considerations when code is made available in a community model). The authors imply that MOSAIC can only be*

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*run with 4 or 8 size bins. This is true by default, but the user could make a modest number of changes to the Fortran code to configure MOSAIC with a different number of size bins and change the size range of the bins (in fact, some investigators have done so). MOSAIC in its box-model version as described in Zaveri et al. (2008) can employ any number of size bins. In the next sentence, I do not understand the phrase “In addition to the modal approach” since MOSAIC employs a sectional approach, not a modal approach.”*

Changed. We rewrote the sentence as follow: MOSAIC can be implemented in the sectional framework where the aerosol size distribution is divided into 4 or 8 discrete size bins.

*Page 14662, line 23: I agree with the comments the authors have regarding using MOSAIC, with its default 4 and 8 size bins that cannot resolve small, freshly nucleated particles. The authors have not commented though on the modal aerosol model in WRF-Chem and whether that model is suitable to study nucleation.*

We do not think modal approach is suitable to study nucleation. The modal approach uses one log-normal distribution to represent nucleation mode (< 100 nm) particles. The problem emerges when nucleation (formation of particles of a few nanometers) occurs in air mass containing aged (or already grown) nucleation mode particles ( 30 – 100 nm). As we understand, the modal approach has to take average to get a mean modal size which reduces the sizes of aged nucleation mode particles while instantaneously increases the size of freshly nucleated particles. Similar problem also exists when air masses containing particles with significant different mode sizes mix during transport. The mode size averaging process in the modal approach, while has small effect on particle mass, is likely to lead to significant uncertainty in the predicted particle size distributions and number concentrations of particles that can act as cloud condensation nuclei (CCN).

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*Page 14664, line 10: The statement on ISORROPIA is simply not correct. MOSAIC does not employ ISORROPIA. An attempt was made to couple ISORROPIA to MADE, but it has never worked correctly in version 3.2 and earlier. MADE uses another thermodynamic equilibrium model by default.*

Yes, MOSAIC can simulate bulk equilibrium by itself, while MADE/SORGAM is coupled with RPMARES scheme by default. We deleted this statement here.

*Page 14665, line 11: I think a bit more description on IMN and how nucleation in this model is different from previous approaches. Since this is the main topic of the paper and so that readers do not have to search elsewhere. Later in the manuscript, one is lead to believe that SO<sub>2</sub> is the primary pathway for nucleation, which in itself is not new. Are amines or other compounds that are also thought to affect nucleation included? If not, please discuss the possible implications in the text.*

More detailed description on IMN was given in the Introduction and Section 2.3.3. The current IMN considers the binary H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O system only. Amines and other compounds may affect nucleation (both neutral and ion-mediated nucleation processes), with the magnitude of the effect depending on the concentrations of these species in the atmosphere. Further research is needed to develop theoretical nucleation models so that the potential impact of amines and other compounds on nucleation in the atmosphere can be assessed. This has been clarified in the revised manuscript.

*Page 14666, line 6: Please include a description on how size distribution is handled in the emissions. The NEI inventory only provides users total PM<sub>2.5</sub> mass.*

It has been included.

*Page 14666, line 25: “leaving a high resolution for the size range” is awkward.*

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We rewrote the sentence as follow: providing a high resolution for the size range

*Page 14667, line 3: BCOC is not defined.*

BCOC is defined at Page 14666, line 5.

*Page 14667, line 17: I count 82 tracers based on the previous text. Not sure where 85 comes from.*

There are 82 APM tracers. The other 3 tracers are nitrate, ammonium and other inorganics aerosol.

*Page 14667, line 17: This section needs to state that the model does not account for SOA. What implications does this have on simulations presented later in the paper? Based on the description of the different number of size bins used for different species, I assume that an external mixture is assumed. Is that correct? Also include a discussion on what type of mixing assumption is employed, since this will affect aerosol optical properties and aerosol activation. Although these feedback processes are not included here, it is stated at the end of the paper that it would be the next logical development.*

Good suggestion. We stated that the model does not account for SOA. Particles are semi-externally mixed in the APM. Via coagulation and condensation, some of secondary species become part of primary particles (i.e., coating). The coating of primary particles by secondary species is simulated using 4 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. We have added some discussions to clarify these in the revised paper.

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*Page 14668, line 4: I have two thoughts at the end of this paragraph. Does the good performance imply that we know everything about nucleation? If so, there are many studies indicating possible missing or unknown processes that are not accounted for by current models. Second, the authors quote that the IMN scheme accounts for nucleated particle number within a factor of 2, yet later in the text there is a factor of 10 or more difference between the observations and simulations. This does not seem consistent.*

The good or reasonable performance indicates that the IMN scheme is likely to be important in the atmosphere. However, it does not imply that we know everything about nucleation. Under certain conditions (especially in the polluted urban environments), species other than H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O may be involved in both neutral and ion-mediated nucleation processes. A factor of 2 refers to previous global study comparing annual mean values. The present study compares instantaneous values and large fluctuation (or scattering of data) is expected, as a result of uncertainties not only in nucleation scheme but also other processes.

*Page 14669, lines 1 – 4: Is the methods employed here the same as what is already employed by other aerosol schemes in WRF-Chem? The Wesley and Zhang et al. schemes are already part of other aerosol models in WRF-Chem.*

Yes, we employed the same methods.

*Page 14669, lines 8 – 11: The text implies that wet removal is included for some types of aerosols, but not others. Why? Again, the text seems to imply external mixing is assumed although not explicitly stated.*

Wet removal is included for all types of aerosols here. We only use the parameterization developed by Henzing et al. (2006) to deal with size-resolved washout rate for sulfate and sea salt in APM. Particles are semi-externally mixed here. BC sulfate, OC

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sulfate, sea salt sulfate, and dust sulfate are scavenged with BC, OC, sea salt, and dust, respectively.

*Page 14670, line 1: “WRF-Chem + CBM-Z has ” is redundant. Can probably just change it to “CBM-Z has” or “CBM-Z in WRF-Chem”. This usage later throughout the text seems awkward to me as well since WRF-Chem would not be WRF-Chem without some photochemical or aerosol model. Perhaps “WRF + CBM-Z” or “WRF with CBM-Z” is more accurate.*

Changed to “WRF-Chem using CBM-Z”.

*Page 14670, line 12: I appreciate the comparisons of computational costs among models. However, the comparisons presented here are not exactly fair or complete. First, APM is not calculating cloud-aerosol interactions whereas MOSAIC is. A more fair comparison would be to compare APM with MOSAIC without aerosols. Please include an estimate of MOSAIC without aerosols for this particular configuration. It would also be useful to include the computational cost of MADE/SORGAM – a modal aerosol model in WRF-Chem, which has fewer species than APM. It would be also important to remind the reader that computational cost is one factor and that accurate is another factor to consider.*

During the comparison of computational costs among different chemistry and aerosol schemes in WRF-Chem, we closed aerosol feedback via the switch of aer ra feedback in namlist.input. The estimation of computational cost of MADE/SORGAM and time costs for each aerosol in WRF-Chem + APM, WRF-Chem + MOSAIC, and WRF-Chem + MADE/SORGAM have been added into the revised paper.

*Page 14671, lines 18 and 20: I do not think it is necessary to list the actual file name*

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used. The web address and reference to Singh et al. is sufficient.

Changed.

*Page 14671, line 26: WRF outputs instantaneous values by default, not hourly-average ones. Are the authors sure they have outputted hourly-averaged values.*

Meteorology factors, gas phase tracers and aerosol tracers are outputted every 20-min and averaged to an hourly-mean. Aerosol microphysics information (nucleation rate, CN10 number concentration, and CCN number concentration) is accumulated and averaged by time steps.

*Page 14672, line 9: "lapse of temperature" is awkward. Change to "lapse rate of temperature". Also change "gradient of wind speed" to "vertical gradient of wind speed". I assume that vertical gradient is what is implied.*

Changed.

*Page 14672, line 13-14: I cannot tell which numbers go to temperature or wind speed. Please re-write the sentence to clarify.*

We rewrote the sentence as follow: The Normalized Mean Error (NME) for temperature and wind speed is -0.003 and 0.004, respectively. The corresponding Normalized Mean Absolute Error (NMAE) is 0.007 and 0.325, respectively.

*Page 14672, lines 19-20: This sentence is strange and needs to be changed. Wind speed is always influenced by the pressure gradient (not usually). Local weather conditions is also affected by previous three factors given.*

The sentence has been rewritten as follow: Usually, the high wind speed is dominated

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by the pressure gradient.

*Page 14672, line 26: Change “0.542” to “0.54”. The last digit is not significant.*  
Changed.

*Page 14672, line 27: I am not sure about RH being “acceptable”. At what level is “acceptable” reached? As the authors state in the next sentence, errors in RH will affect aerosol microphysical processes. In this study RH is too low on average, and therefore aerosols would likely not uptake as much water as reality (if RH is high enough). This will be more important when APM is coupled to aerosol optical properties*

Agree. RH, which is associated with the process of water vapor in the water cycle, is hard to be re-produced by the model. In this study, simulated RH is lower on average than observation. We deleted this sentence.

*Page 14673, line 6. As with my previous comment, it is not necessary to list the file name used.*  
Changed.

*Page 14673, line 18: I cannot see how the model is capturing the horizontal and vertical gradients in SO<sub>2</sub> from the figure that has been provided. Vertical gradients can be determined in a gross sense.*

It has been corrected as follow: The model simulations capture the major characteristics of vertical gradients of SO<sub>2</sub> along the flight paths.

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Page 14673, line 21: Change “-0.44” to “-0.44 ppt”.

Changed.

*Page 14673, line 24: The authors list only one, of several possible reasons for the under-prediction in SO<sub>2</sub> in the upper troposphere. More insight is needed in the text. First, boundary conditions could contribute to this. Does GEOS-Chem under-predicted SO<sub>2</sub> as well? Second, the authors do not mention whether a parameterization for vertical transport associated with convection affects the APM aerosols? The convective scheme in WRF applies only to the meteorological quantities. Some of the aerosol schemes have been coupled with the Grell convective transport (not MOSAIC), but it is not clear whether APM has been coupled in this way. If so, the model description needs to include this topic. If the convective transport parameterization does not affect APM aerosols, than the 27 km grid spacing will impact the vertical transport of aerosols. A smaller grid spacing would be needed to better simulate vertical motions associated with convective clouds.*

Agree. It is no surprise that the model cannot well reproduce boundary layer process, for it is one of the least understood areas of meteorology. Boundary conditions could contribute to this under-prediction of SO<sub>2</sub> in the upper troposphere. We rewrote this part and added some insight discussions.

Because the outputs of GEOS-Chem are daily averaged, it is hard to compare directly GEOS-Chem simulations with aircraft observations. However, we compared monthly averaged vertical profiles of GEOS-Chem and WRF-Chem. The comparison indicated that simulated SO<sub>2</sub> by GEOS-Chem is higher than that of WRF-Chem in the upper troposphere. It implicated that GEOS-Chem may show better performance than WRF-Chem as WRF-Chem significantly under-predicted SO<sub>2</sub> in the upper troposphere.

In this study, the Grell convective transport scheme is coupled with APM. However, the under-prediction of both SO<sub>2</sub> and SO<sub>4</sub> indicated that the vertical transport scheme needs to be improved.

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*Page 14674, line 16: As with other laltitude vs height plots, I cannot make the connection between the text description in this line and the plot.*

The figure has been changed, and the associated discussion has been updated.

*Page 14674, line 28: How is “acceptable job” being defined? Acceptable in terms of what? Since the authors discuss the importance of simulating particles in this size range that may act as CCN, it is unclear as to the potential impact of the simulated errors on the impacts of aerosols on clouds. Please elaborate.*

Yes, “acceptable job” is too vague. We have deleted it. The potential impact of the simulated errors on the small particles and its impacts on clouds have been discussed at the beginning of the paragraph.

*Page 14675, line 5: Again the use of “reasonable agreement” is rather vague. Please be more specific. The authors should refer to other modeling studies (that include WRF) of INTEX-A (and there have been many) to put the current performance into the context of other studies would help.*

We did some literature search and didn’t find any relevant modeling studies that we can compare to. To address the referee’s concern, we have changed the sentence to “the WRF-Chem simulations can capture the major characteristics of observed meteorological factors and the spatial-temporal distribution of atmospheric components”.

*Page 14676, line 12 – 14: This sentence could be much stronger, had the authors also show statistics comparing GEOS-chem with observations, similar in Figs. 2 and 3. I suggest putting such a comparison in a table. It is not unexpected that the present simulation at higher spatial resolution will produce more spatial variability.*

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The present version of GEOS-Chem uses the NASA Global Modeling Assimilation Office (GMAO) GEOS-5 operational data product (v5.2.0) with 6-hour temporal resolution (3-hour resolution for surface fields and mixing depths). Such an averaged meteorological fields plus relatively lower horizontal resolution (2x2.5) make it difficult to directly compare GEOS-Chem global simulations with in-situ instantaneous aircraft observation. Therefore, our comparisons with GEOS-Chem results are generally based on monthly mean or annual mean values.

*Page 14676, line 20: Please indicate how CCN concentrations at supersaturation values are calculated.*

We assumed that the activation diameters for supersaturation values of 0.8%, 0.4% and 0.2% are 33 nm, 57 nm, and 82 nm, respectively. When particles are larger than a given activation diameter, they are accounted as CCN at the corresponding supersaturation value.

*Page 14677, lines 8 – 9: I am a bit confused by what is meant by “their interactions” since it has not been established if the aerosols are externally mixed or not.*

We use semi-externally mixing in APM to study the interactions between secondary particles and primary particles.

*Page 14677, line 19: The objective listed is a good one, but “improvement” implies that the new technique is better than an existing one. However, this study does not show how other aerosol models in WRF-Chem perform for this case and against APM. The authors should consider rephrasing this text.*

We rewrote the sentence as follow: The main objective of this study was to simulate the aerosol processes in WRF-Chem by incorporating an advanced particle microphysics

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(APM) model into the framework of WRF-Chem.

*Page 14678, line 17: Does this imply that other factors proposed in the literature that might affect nucleation are not as important as SO<sub>2</sub>? How does one know?*

It is now established that H<sub>2</sub>SO<sub>4</sub> molecules oxidized from SO<sub>2</sub> are involved in the atmospheric nucleation process. Other species (such as ammonia, amines, organics) may be involved in the nucleation process as well but their role is likely to be secondary (compared to that of H<sub>2</sub>SO<sub>4</sub>). Currently robust nucleation models considering the effect of these species are not available.

*Page 14679, lines 2-3: As with my previous comment, the error in convective transport is misleading since there are likely other factors as well.*

We also discussed other factors which may contribute to the errors in the revised paper.

*Page 14679, lines 3-4: This is the first time that secondary organic aerosols are mentioned. It should be noted in the model description that SOA, which is likely to be important for over the eastern U.S., are not considered.*

Yes, we moved the discussion to model description and emphasized here SOA is likely to be important for over the eastern U.S., especially during summertime.

*Figure 1. The 3-D panels provide very little useful information to the reader regarding model performance. I suggest replace these panels with percentiles with values binned by height.*

We color coded the scatter plots (right column) with measurement height. The 3-D panels have been removed.

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*Figures 2 and 3. I have the same comment as Fig. 1, change (c) and (d) to have percentile plots binned by height for both the observations and simulated values.*

Scatter plots have been color-coded by altitude. The height vs latitude plots have been removed.

*Figure 4. It might be useful to include another panel that averages the WRF results to the same resolution as GEOS-Chem. This would provide another means of assessing the impact of subgrid scale variability in terms of global model, as discussed in Qian et al. ACP, 2010.*

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

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

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