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Atmospheric Chemistry and Physics Discussions

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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General comments:

The authors present an interesting study comparing and contrasting the behavior of the APM model in a regional and global model. Some comparisons between observations and regional-scale model predictions for SO2 and CN10 are presented. Since the introduction and conclusion provides discusses the motivation for accurately simulating fine particles for CCN, it would be useful to include one or more plots that evaluate the ability of APM to simulate the size distribution compared with the INTEX-A observed size distribution. In addition, it would also be useful to show the performance of the

model in simulated sulfate, since this will also indirectly evaluate SO2 predictions. The meteorological conditions during INTEX-A were wetter and cooler than normal, and cloud-processing of SO2 might be important. However, it seems that the present model does not account for this process that could be significantly affect the conclusions drawn from the WRF simulation.

The description of the performance is often overstated, and I point out a few examples in the specific comments below. All that is needed is some rephrasing to put the results into the proper context. For example, IMN has been used in GEOS-Chem and has usually been evaluated by comparing annual observed and simulated aerosol number in previous papers. This is a necessary, but not sufficient condition. A model could perform well for a long-term average, but the day-to-day performance could still vary significantly. This study attempts to take the next step by using a higher spatial resolution model and comparing model presentation at specific times and locations with aircraft data, which is a much more stringent and challenging criteria for models. One could expect a bit more scatter between observed and simulated quantities at these temporal and regional spatial scales, but it is difficult to attribute the causes for the differences. Errors in the number of freshly nucleated particles could be due to errors model predictions of the meteorological conditions (e.g. transport, vertical mixing, relative humidity), emissions of primary gases and aerosols, or to the assumptions in IMN. So it becomes problematic to evaluate the performance of IMN that is not isolated from other factors in the model simulation. There is some discussion in this regard, but more discussion is needed.

There have been numerous modeling studies (meteorological, photochemical and aerosol) of INTEX-A, yet virtually none of them are mentioned. It would be useful to discuss at certain points throughout the text where appropriate how the present model performance compares to other studies.

Specific Comments:

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.

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

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.

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

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.

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 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.

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.

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.

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 SO2 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.

Page 14666, line 6: Please include a description on how size distribution is handled in the emissions. The NEI inventory only provides users total PM2.5 mass.

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

Page 14667, line 3: BCOC is not defined.

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

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.

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.

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.

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.

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.

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.

Page 14671, lines 18 and 20: I do not think it is necessary to list the actual file name used. The web address and reference to Singh et al. is sufficient.

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

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.

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

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.

Page 14672, line 26: Change "0.542" to "0.54". The last digit is not significant.

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

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

Page 14673, line 18: I cannot see how the model is capturing the horizontal and vertical gradients in SO2 from the figure that has been provided. Vertical gradients can be determined in a gross sense.

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

Page 14673, line 24: The authors list only one, of several possible reasons for the under-prediction in SO2 in the upper troposphere. More insight is needed in the text. First, boundary conditions could contribute to this. Does GEOS-Chem under-predicted SO2 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.

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

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.

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.

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.

Page 14676, line 20: Please indicate how CCN concentrations at supersaturation val-

ues are calculated.

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.

Page 14667, 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.

Page 14668, line 17: Does this imply that other factors proposed in the literature that might affect nucleation are not as important as SO2? How does one know?

Page 14669, lines 2-3: As with my previous comment, the error in convective transport is misleading since there are likely other factors as well.

Page 14669, 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 important for over the eastern U.S., are not considered.

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.

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.

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.

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