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Comment on "Volatility basis-set approach simulation of organic aerosol formation in East Asia: implication for anthropogenic-biogenic interaction and controllable amounts"

The authors present a thorough report of their effort to simulate organic aerosol formation and transport in the East Asian domain with up-to-date algorithms implemented into the WRF-Chem model. This work should be published in ACP as it amounts to a step forward in the community. However, I have some reservations about various parts of the manuscript that lacked adequate discussion and context (detailed below). With these additions, I have no hesitation recommending the manuscript for publication.

### Major Points:

1) Page 6210, lines 18-26: This simplification is a promising solution to a vexing problem in modeling OA in large-scale CTMs and maintaining some amount computational efficiency. The authors explain the approach well enough, but don't mention the possible numerical consequences. Much of the OA loading is admittedly accumulation mode, but not all. I would expect much of the POA material to be at low sizes, more characteristic of combustion processes. Moreover, the authors are working with an on-line, coupled MET/CTM; what impact does this simplification have for cloud activation? Are there any previous examples of a model trying this approach? The authors should, at the very least, warn the reader that the effects of this approach are very uncertain.

2) Page 6211, lines 1-10: The assumption of  $(\text{SVOC} + \text{IVOC}) / \text{POA} = 7.5$  is very uncertain and claiming consistency with the Mexico City studies is not enough for justification. Are there any volatility measurements from East Asia to even get a small peek at how reliable this assumption might be? Are the combustion activities (i.e. vehicle types, energy usage, etc) similar enough between all of East Asia and Mexico City to warrant such an assumption without exploring its uncertainty? I would expect to see some kind of sensitivity study focusing on this parameter. The disclaimer at the end of Section 2 (that these uncertainties were discussed in Tsimpidi et al. (2010) ) does not quite hold up since that work focused on entirely different geographical scales, energy generation profiles, and socio-economic contexts. Moreover, noting that POA emissions were constrained with measurements, it is very likely that some of the POA measured consisted of semivolatile compounds. If this is the case, then multiplying POA by 3 to get the SVOC emissions would overpredict POA concentrations. Should not the POA emissions be rescaled to account for this?

3) As seen in Fig. 8a and Fig. 10, the large controllable fraction estimate is significantly related to the huge OPOA concentrations present when anthropogenic aging is turned on, as the authors point out. This is hugely uncertain. It is probably an upper limit and should be stated somewhere.

4) In general, the authors rely heavily on timeseries comparisons to evaluate the output from the model. Table 5 presents statistical evidence of mixed results. Figure 5 though, begins to address other types of comparisons that may be more telling for the capabilities of this model. Seeing as how there is only one take home point from Fig. 5 (that the model with aging performs much better than that without aging), I wonder if there are more comparisons along “chemical coordinates” that would be helpful here. For instance, why didn’t the authors look at model performance versus temperature, time of day, black carbon or CO concentrations, precipitation, OOA/OA ratio, etc to try to characterize the ability of the model to predict SOA relative to its ability to predict other model components (in addition to sulfate)? As an example, comparing OA to sulfate allowed the authors to attempt to control for photochemical activity, while comparing to black carbon would control for some important anthropogenic and biomass burning emissions.

5) I was surprised in general that the OA concentrations were so low, given the notoriety of Beijing and other East Asian cities. They seem to compare approximately similar to results obtained for the US (Lane et al., ES&T, 2008) and Europe (Fountoukis et al., 2011). Are these results only including PM<sub>1</sub> concentrations, in order to be more comparable to the AMS results? I recommend it be stated very clearly what size cutoff is being used so reader’s know how to contextualize the results. Since the authors are taking the next step to quantify a “controllable” fraction of OA, I think it would be very beneficial to mention the up-to-date estimates for average total particulate matter concentrations in various parts of the region. Readers in the general aerosol community will be interested to know what impact controlling these sources would have on the total loading (at different sizes), not just that of the OA.

### **Minor points:**

Page 6205, lines 15-25: Not all “traditional” models underestimated OA and/or SOA concentrations using the Odum approach. For example, see Simpson et al. (2007) where OA was modeled over Europe. Many would argue that the underestimation could just as likely have come from using unrealistically low yields, rather than from not including aging or using the VBS.

Page 6205, Line 28: I would strongly urge the authors to refrain from using the acronym SVOCs and instead refer to both groups separately, i.e. “SVOCs and VOCs.” While I understand the motivation for streamlining the terminology in the manuscript, it is my opinion that combining these terms will lead to confusion and doesn’t add much value.

Page 6207: The authors present a nice discussion of previous work. However, they leave out an emerging concept that I would expect to be important for future work with this model: the role of “anthropogenic water” (Carlton and Turpin, 2013). Improving the model with an aqueous formation pathway and repeating all of the experiments is well outside the scope of this manuscript, which already discusses a lot of issues. I would

expect, however, to see this pathway discussed in the introduction though, and I would expect the results to be discussed at some point in the context of this possibly important interaction. Figure 6 suggests that there is plenty of sulfate around. Would a large interaction seem plausible? To what magnitude?

Page 6211, lines 14-16: I'm glad to see the authors point to the uncertainties inherent in dry deposition parameterizations. Please also mention for the reader what has to be assumed physically to effect a change in the dry deposition velocity by a factor of 2. I assume solubility is driving this calculation? If so, what is the range of solubilities that results in a factor of 2 difference?

Page 6211, lines 18-24: I did not quite understand what the authors have chosen to do for in-cloud aerosol. Is there a separate size distribution for this material? Does the model simulate regeneration of aerosols after cloud drop evaporation? If so, is the chemical composition information (i.e. fractional contribution of ASOA, BSOA, POA, etc) maintained through the process of activation/uptake followed by evaporation/regeneration? Table 3 suggests that in-cloud aerosol is modeled explicitly, but it is unclear how this works.

Section 4.3 is a little light for such a deep topic. Could the authors add some discussion about the choices of aging rate constants? Do they believe these bound the possibilities of aging rate constants in East Asia or just probe the space as in previous work? In many past studies, different aging rate constants were applied to SOA and POA. The authors also explore some past assumptions that ASOA and BSOA age with different rate constants. However, it appears as if they always age anthropogenic POA and SOA compounds similarly. Is there a composition effect when increasing or decreasing the rate constant? More specifically, Table 6 shows that increasing the rate constant by a factor of 4 enhances OPOA concentrations more relative to ASOA. I take it this is because there is a huge source of OPOA material sitting at intermediate volatility? It could be helpful to discuss this result in the context of previous studies that had far less fractional contribution of OPOA to the total (i.e. Tsimpidi et al., 2010; Fountoukis et al., 2011) using  $4 \times 10^{-11} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$ . Is this pointing to overzealous emissions? Please consider moving the last two paragraphs of section 5.1.1 to section 4.3.

Page 6221, line 15: Using the aggregated OM:OC ratio of 1.6 seems unnecessary here given the model detail available. The authors have enough information to use 1.4 for the POA and 2.0 (or even 2.2) for SOA species, as an example. This would better reflect the oxygenated nature of the SOA. Of course, if I understand the model-measurement comparison correctly, then the model will underestimate the measurements to an even greater extent. Also, in line 16, "OA concentrations" should read "OC concentrations" I think.

Figure 6 and Section 3.2: What was the collection efficiency for the AMS in this data and what effect does that have on the comparisons?

Figure 7: Please consider adding a panel for OPOA distribution to the left side. Even though there won't be one for the right side, it is instructive for the reader to see the contribution of this species. I was surprised the bars for OPOA were so high in Fig. 8.

Table 6 and Figure 7: How was the boundary layer height determined for this calculation? When the authors write “~1 km,” are they implying that they chose a fixed number of WRF-CHEM layers and used that for the entire analysis, or was the number of layers allowed to fluctuate with meteorology? If so, was temperature or vertical mixing used to diagnose this behavior? If done incorrectly, this could certainly bias the results. Why not use the bottom layer concentration to compare to the measurements, for instance?

**Technical Points:**

Page 6205, line 15: Please revise to read “parent VOCs (for example, isoprene and ...)” since these are not the only compounds used for modeling SOA sources.

Page 6206, line 7: I would not consider OOA to be a “measure of SOA” in a strict sense, although I understand what the authors are getting at. Please consider rephrasing to something like “(OOA, thought to be analogous to SOA) concentrations” or “(OOA, likely reflective of SOA) concentrations”.

Page 6206, line 20: Remove “of”.

Page 6217, line 9-10: Please fix the units for  $6.3 \times 10^6 \text{ cm}^3$  and on the next line.

Fig. 5: Please, if possible, add the quantitative slopes of these lines to the figure in some way.

Fig. 8: The colors changing meaning between the panels is confusing. Please consider rearranging the bar graph so that simulation runs are grouped together and species are color coded. Then you could use the same colors that are used in panels b and c.

Page 6223, line 4: Please consider changing “ANaging” to “AN-aging” throughout the text.