
Anonymous Referee #1

Received and published: 26 October 2015

The authors present a thoughtfully-conceived and well-written manuscript detailing their evaluation of several methods for predicting secondary organic aerosol formation and aging at the regional scale (8 km and 36 km grid resolution). The paper embodies a substantial step forward in the challenging effort to capture the complex atmospheric chemistry responsible for organic aerosol formation, and the scope/content of the paper are reasonably suited for publication in Atmospheric Chemistry and Physics. I find no errors in the science concepts or data presented and this work builds on a series of rigorous studies that lend confidence to the results. However, there are some issues
regarding language/presentation that would benefit from some more careful thought in addition to some technical corrections that should be made before I recommend the paper for publication. In particular, the explanations that refer to “generations” of chemistry are sometimes too vague and could use some added precision.

Language/Presentation:

1) Page 25841, lines 2-5: This sentence “Note that oxidation...two-product or VBS parameterization.” is one example of many times the authors try to describe an exceedingly precise detail of current OA models without going into enough detail to fully explain it to non-SOA or even non-experienced-VBS modelers. I think a schematic, perhaps in the supplemental information, is warranted here that shows explicitly the relationship of the gas-phase mechanism, emissions inventory and SOA calculations to each other under the different configurations they have brought up. Many users of CMAQ, particularly those interested in the ongoing conversation about how to model POA and SOA will be interested in this detail, even if they are not quite up to date on the complicated, rather fractured approach that many transport models currently employ.

2) Page 25849, lines 20-24: It seems awkward to refer to “the majority of the multigenerational oxidation reactions that contribute to SOA mass (but not necessarily to SOA composition)”. If the differences in SOA composition were driven wholly by oligomerization reactions (which are turned off in this case), I would agree. But the authors have explained that a lot of the composition differences are due to semivolatile species oxidizing further and creating both lower and higher volatility material. So the oxidation reactions that “contribute” to SOA composition will also contribute to SOA mass, just in a more complicated way than in the non-aging schemes. Could the authors be more precise here, and rephrase this sentence to something like, “In summary, it is possible that chamber-observed SOA formation, as parameterized with a two-product approach, accounts for the majority of the net mass produced by multigenerational oxidation reactions, even though the chemical properties of the SOA formed are probably not as well-reproduced.”
3) Page 25854, line 15-18: This is not technically true as I read it. Lots of studies using the VBS have separated the OPOA from the traditional SOA (Tsimpidi et al., 2010; Fountoukis et al., 2014; Matsui et al., 2014). Even the two studies cited (Murphy and Pandis, 2009; Jathar et al., 2011) do this. We published one study that divided the OPOA and explicitly tracked the NT-SOA from IVOCs separate from primary SVOCs (Murphy and Pandis, 2010). Are the authors trying to highlight the nonlinear effect of absorptive partitioning? Although I’m not asking the authors to specifically cite any of these papers, I think there is a possible contradiction in the language that should be cleared up.

4) Page 25855, line 5-8: I don’t think it’s surprising that the predictions using the SOM parameterizations agree so well with those using the BaseM parameterizations, especially since they are constrained with the same observations and experimental durations. This agreement might suggest that the BaseM is reproducing the correct mass increase as the authors say, but it also might mean the SOM is misrepresenting the continued aging. Could the authors please explain briefly and cite the evidence (from past SOM chamber studies presumably) that leads them to conclude the former? Is it from the PAM experiments?

5) Page 25855, lines 12-19: The use of the term “first-generation” is problematic already for SOA formation since, as the authors point out, so much chemistry is happening simultaneously. The term takes on a more practical definition when applied to current models that artificially segregate gas-phase chemistry (via carbon-bond or SAPRC mechanisms) from SOA aging. Still, to my knowledge, there is no clear definition of first-generation products for many or most precursors. Now, introducing the “second-generation” seems unwieldy. Are the authors referring to products formed later in a chamber experiment and distinguishing them from products formed in a real atmospheric system after the duration of a typical experiment has elapsed?

Also, the sentence “Alternatively…in fact, dominant.” doesn’t make much sense to me. This may be from my confusion about the term second-generation. If multi-generational
aging is dominant, how will a static representation ever be sufficient? It seems like this outcome would only be the case when the effects of the multigenerational aging exactly, or nearly, cancel. Even if this is the case, there is probably some time-dependence associated with the applicability of the static representation, and that timescale may be of similar magnitude to the atmospheric lifetime of the intermediates. The problem becomes even more complex in the actual atmosphere when temperature, oxidant level, deposition rates, and mixing states are all variable.

6) Page 25856-7, line 24-3: This line of reasoning seems unclear. How do the authors know the evaporated POA mass is balanced by the SOA mass arising from “recycling”? Also, how do “chamber experiments that include emissions of POA and IVOCs already include SOA products from their multi-generational oxidation”? And how do “those considerations”, the identity of which I’m also uncertain, imply them? Do the authors mean to say that “chamber experiments that inform emissions of POA and IVOCs…”? If so, it does not seem like there is enough evidence to say that the NEI, for instance, already includes the multigenerational oxidation product mass from evaporated POA, since this mass will depend on environmental conditions and the specific conditions of each emission test used for input to the inventory. Could the authors consider rephrasing this statement as an assumption they have made rather than a conclusion from previous work?

Technical Corrections:

1) Page 25841, line 9: The preceding sentence mentions “both research and regulatory groups”. The addition of the VBS to CMAQ could certainly be categorized as regulatory-focused. Please add a citation from a slightly more “pure” research-based effort, such as WRF-Chem (Ahmadov et al., 2012; Matsui et al., 2014), GISS II’ (Farina et al., 2010), EMEP (Bergström, et al., 2012) or PMCAMx (Fountoukis et al., 2014; Tsimpidi et al., 2010; Lane et al., 2008).

2) Page 25841, lines 13-14: This point is technically not true as the authors themselves
point out later in the manuscript; the infamous “biogenic aging off” configuration would embody essentially a different aging configuration, where fragmentation and functionalization are assumed to balance each other, from the perspective of mass enhancement (eerily similar to the major conclusion of this paper).

3) The authors make a strong case for using the SOM, or at least sticking with the chamber-derived yields and not adding a distinct multi-generational aging enhancement of OA mass. For those that will envision adding the SAPRC-11/SOM framework to a CTM, I think it is worth reporting the number of added species (is it 392 gas + 392*N_size aerosol as in Jathar et al., 2015?) and relative computing time increase compared to the CMAQ base case, at least in the supplemental information.

4) Page 25848, line 12: Fig. S1 does not seem to appear in the supplemental information.

5) Page 25852, line 5: I think the authors mean to cite Epstein et al. (2010). Grieshop et al. (2009) used something more like Hvap = 69 – 4*log10(C*).

6) Page 25852, line 24: Looks like figures 5 and 4 should be switched.

References:


Interactive comment on Atmos. Chem. Phys. Discuss., 15, 25837, 2015.