We thank the reviewers for their comments. The response below and the corresponding changes to the manuscript have helped improve the manuscript. We have highlighted the reviewer comments in blue, our response in red and the changes in the manuscript in *italic red*.

Response to comments made by reviewer 1

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.

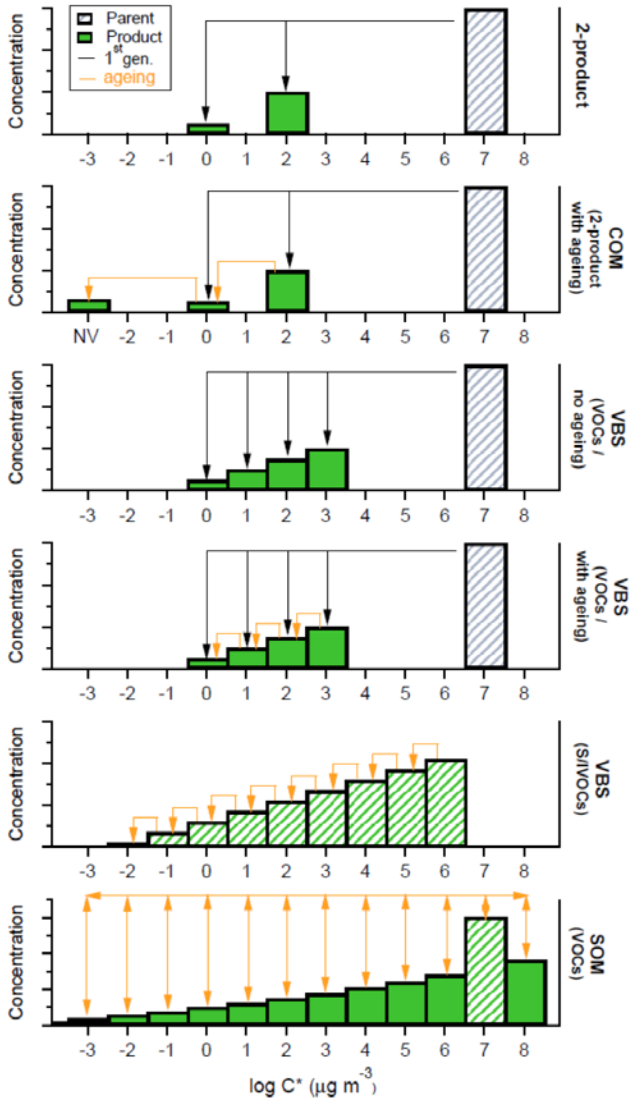

Figure 1: Schematic for different SOA models.

To assist the reader in understanding the modeling history and multitude of framework-approach combinations surrounding the treatment of organic aerosol (OA) in chemical transport models, we have included Figure 1 and a description of it in the supporting information. We agree with the Reviewer that this should be placed in SI since the work is focused on the findings from an application of a next-generation OA model rather than a review of OA models. The following text will be added to the supporting information as a figure caption:

“Schematic illustrating the differences between some of the different approaches for modeling SOA. From top to bottom: the 2-product model; the COM-type model, i.e. 2-product with ageing; the VBS as applied to VOCs with no ageing; the VBS as applied to VOCs with additional ageing; the VBS as applied to S/IVOCs; and the SOM. The black arrows indicate the formation of products directly from the parent VOC and the orange arrows indicate ageing reactions, i.e. reactions involving product species. For the SOM, all species are reactive and both functionalization and fragmentation are possible. In the other models that include ageing, only functionalization reactions are included, i.e. reactions that decrease compound vapor pressures.”

In the main text, we will point the reader to this schematic in the methods section stating:

"A conceptual schematic comparing the SOM oxidation and SOA formation scheme with other schemes (e.g. 2-product, COM, VBS) is provided in Figure. S.1."

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."

The reviewer is correct. We meant that sentence in reference to the chamber experiment already including the influence of multigenerational oxidation. We have revised the statement as follows:

"In summary, it is possible that the chamber-observed SOA formation accounts for the majority of the multi- generational oxidation reactions that contribute to SOA mass and hence, a two-product approach to model SOA formation would already include the mass-enhancement associated with multi-generational oxidation. However, such a two-product model may not necessarily accurately represent the chemical composition of SOA".

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.

We did not mean imply anything about the ability of the 3D model to track model species that represent various flavors of oxidized products of POA and SOA; as the reviewer points out, plenty of 3D models (particularly PMCAMx in addition to the current UCD/CIT model) have tracked each source/process separately. Rather, we intended to convey the idea that the scientific theories surrounding the volatility of POA, emissions of IVOCs and aging of traditional SOA, POA vapors and IVOCs have not been dealt with in isolation. In other words, COM-type models have tended to subscribe to all theories simultaneously and have not examined in detail the isolated role of experimentally-constrained aging in a 3D model (which is the focus of this work). We have clarified the sentence as follows:

"Previous studies that have examined the influence of multi-generational oxidation of traditional VOCs using COM-type models have typically combined the effects of VOC ageing and IVOC and POA vapor oxidation (e.g. Murphy and Pandis, 2009; Jathar et al., 2011) together and have not investigated the role of each process separately."

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?

The reviewer raises an important question: how well constrained is the SOM with respect to "ageing", i.e. the implementation of multi-generational chemistry. As discussed in Cappa and Wilson (2012) and in the supplemental material of Zhang et al. (2014), the SOM was developed based on physical and chemical principles that govern the oxidation of organic species. As noted in Zhang et al. (2014), the rate coefficients used to describe the reactivity of SOM product species were developed through explicit fitting to the output from GECKO-A simulations (Aumont et al, 2005). GECKO-A is an explicit chemistry model that uses structure-reactivity relationships to calculate rate coefficients. Additionally, we can consider the various data sets used in fitting SOM. For all compounds, with the exception of benzene, the number of oxidation lifetimes was > 1, suggesting that at this point species beyond the first generation would have been produced and thus the effects of "ageing" are intrinsic. Further, SOM accurately captured the "turnover" in the SOA mass concentration that was observed for isoprene (both low-NOx and high-NOx) and for a long-time experiment conducted using m-xylene (see Figs. S9 and S10 in Zhang et al. (2014)). Such a turnover results from continued ageing well-beyond the point where first-generation products dominate the product distribution. Although not direct evidence, this suggests that SOM can capture the effects of continuous ageing within the parameterization. Finally, during some of the initial SOM development, the SOM was explicitly fit to the output from simulations of dodecane + OH using the GECKO-A model. Specifically, the SOM was fit to the total OA mass concentration from simulations conducted using very small concentrations of precursor VOC (1 ppt) in the presence of a comparably large amount of absorbing seed (1 ug m^{-3}) with [OH] = 2e6 molecules cm^{-3}. After fitting to *only* the SOA concentration data, the SOM performance was evaluated by comparing the SOM 'species' time-evolution (e.g. $C_{12}O_4$) to the GECKO-A output. The GECKO-A species (which are actually real molecules) were binned

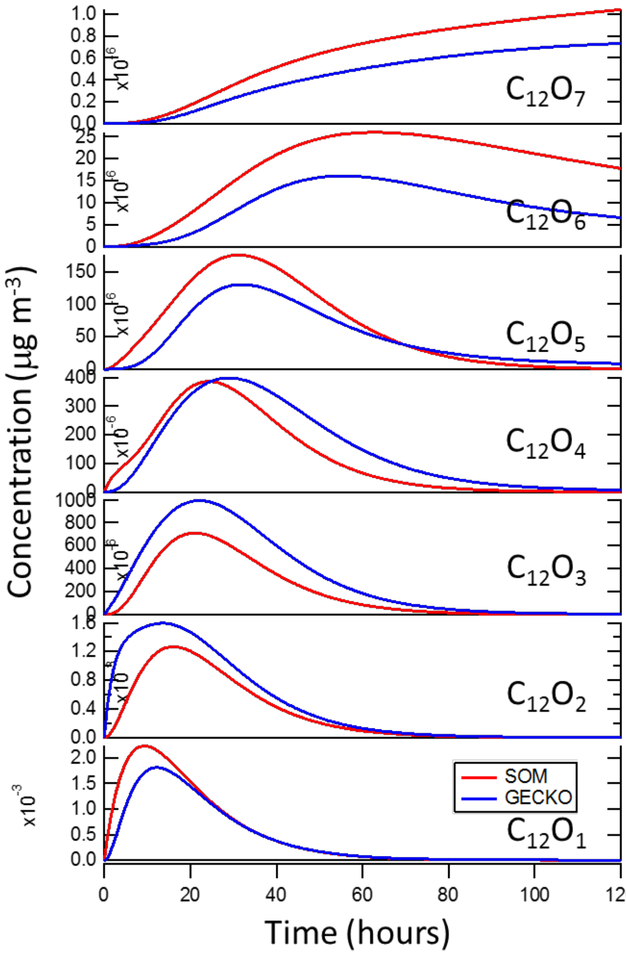

Figure 2: Simulated concentrations of SOM gas-phase 'species' with 12 carbon atoms and varying numbers of oxygen atoms compared to the output from simulations using the GECKO-A model. Simulations are for dodecane + OH under zero NO_x conditions.

according to their numbers of carbon and oxygen atoms (e.g. $C_{12}O_4H_{24}$ = $C_{12}O_4$) and the total abundance of the binned SOM-equivalent ‘species’ determined. The results of this previously unpublished comparison for the evolution of the gas-phase species is shown in the figure above. It is clear that SOM captures the general evolution of the GECKO-A species very well. These results increase confidence that SOM represents the ‘ageing’ processes associated with multi-generational chemistry in a reasonably realistic manner. (The authors thank Julia Lee-Taylor, Alma Hodzic and Sasha Madronich of NCAR for the GECKO-A simulations.) Since the comments and responses are archived and publically available, we do not plan to add this specific information to the manuscript, as we believe that it would be distracting from the main points.

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.

The reviewer raises an important issue about the exact definition of first (and subsequent) generations of reaction. We believe there is not a uniform agreement on this issue in the SOA community, but we feel that we have used this concept consistently with our favored definition. In all our manuscripts with the SOM (Cappa et al., 2015;Jathar et al., 2015a, b), we refer to first generation products as the first set of stable products arising from a series of radical-based reactions. For example, one could imagine the first generation of oxidation from a hydrocarbon to involve hydrogen abstraction leading to formation of an alkyl radical, reaction with oxygen to form a peroxy radical and subsequent formation of the first-generation functionalized products. This definition is consistent with the definition of a ‘generation of oxidation’ used in chemistry textbooks. We therefore feel strongly that (1) there is a clear definition for ‘generations of oxidation’ and (2) our use of the phrase ‘generation of oxidation’ is clear and consistent. A concise description of an oxidation generation is offered in our previous manuscript in GMD:

“We should note that the representation of the reaction chemistry in the SOM, in contrast to an explicit gas-phase mechanism like SAPRC, MCM or GECKO, is significantly simplified to capture the average chemistry. Further, each oxidation step in the SOM is an aggregation of numerous individual reaction steps, i.e. intermediate radical species are not explicitly simulated. For example, in reality each oxidation reaction is initiated through hydrogen abstraction to yield peroxy/alkoxy radicals. These radicals can go on to react (with HO_2, RO_2 or NO) or undergo isomerization to form low-volatility products such as organic nitrates, peroxides and hydroxy carbonyls, or can decompose leading to production of oxygenated fragments. These intermediate steps are not explicitly simulated, only the formation of the resulting stable product species.”

The reviewer seems to argue for a different definition of first and second generation of oxidation that is sometimes (in our opinion, incorrectly) labeled to think about oxidation reactions in smog chamber experiments. In the case of ozone-driven experiments, it is easy to think of ‘first generation’ products, as the oxidant only reacts with the parent VOC. However, in photooxidation, at any given point in time there is a distribution of products across multiple generations because the oxidant can react with both the parent VOC and the product species (see, for example, Wilson et al. (2012) or the figure above). This is true even very early on in an experiment. The term ‘generation’ can be easily understood as the number of times that something has reacted with an oxidant to get to its current state. We would suggest that the SOA modeling community should move away from using “oxidation generations” to describe in-chamber versus out-of-chamber chemistry and adopt a definition that aligns more with fundamental chemistry.

With respect to the reviewer’s second point, a static representation of SOA expressed through the two-product model can only capture the multi-generational chemistry observed directly in chamber experiments, with the relative yields of the product species fixed. As a simple analogy, this is equivalent to fitting a linear (straight line) relationship to an inherently non-linear (has curvature) system. The straight-line relationship does a reasonable job of explaining outcomes under some conditions but fails to fully represent the outcomes over all possible conditions. The results of this study appear to suggest that the “straight-line” relationship does a reasonable job of predicting outcomes over the conditions experienced in Southern California and the eastern US during typical air pollution episodes.

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?

The reviewer is correct on all counts. We have not considered multigenerational aging of POA vapors or IVOCs in our simulations, although we plan to do so in proposed future work. We were speculating about the implications of our study on the semi-volatile and reactive behavior of POA and the SOA formation from IVOCs. What we meant to say was that our simulations that assume non-volatile and non-reactive POA could be assumed to be identical to a simulation where the evaporation from semi-volatile POA would be offset by the SOA production from POA vapors and IVOCs (this ignores the fact that the spatial patterns could look very different between these two simulations). Clearly the statement is confusing and as the reviewer points out could imply that we have done simulations to show the offsetting effects. We have not yet done this work and we will remove that sentence and replace it with the assumptions we have made regarding POA. Specifically, we plan to modify the paragraph to read:

“In this work, we consider POA as non-volatile and non-reactive and do not consider SOA contributions from IVOCs or semi-volatile POA vapors. Oxidation of IVOCs and semi-volatile POA vapors (i.e. SVOCs) can lead to the production of new SOA mass, but evaporation of POA

leads to a decrease in the total OA mass. To some extent, these effects are offsetting (especially for SVOCs, which do not contribute new carbon mass to a model). To the extent that the loss of POA is balanced exactly by the formation of SOA from IVOCs and 'recycling' of semi-volatile POA vapors, the simulations here represent a scenario in which the total OA mass is conserved, although possibly with the wrong spatial distribution (Robinson et al., 2007). Most efforts to incorporate SOA formation from IVOCs and SVOCs have simulated their oxidation using a version of the VBS model in which multi-generational ageing is implicit, but highly underconstrained and structured in such a way that the ultimate (long time) SOA yield is greater than unity because all mass is converted to low-volatility products and oxygen addition is assumed. The SOM framework provides a way to explicit account for the influence of multi-generational chemistry in SOA formation experiments that include semi-volatile POA vapors and IVOCs (Gordon et al., 2014a;Gordon et al., 2014b;Gordon et al., 2013;Grieshop et al., 2009a;Grieshop et al., 2009b;Hennigan et al., 2011;Miracolo et al., 2011;Miracolo et al., 2012;Platt et al., 2013;Platt et al., 2014;Nordin et al., 2013;Chirico et al., 2010;Heringa et al., 2011;Tkacik et al., 2014), and thus should be useful for constraining the contribution of these compound classes to the ambient OA budget. In addition, the simulations here do not consider the influence of vapor wall losses on SOA formation. Such losses can influence SOA yields in chambers, and consequently the parametrizations that result from fitting of such chamber data. The influence of vapor wall losses on simulated ambient SOA and OA concentrations within the SOM framework is examined in a companion paper (Cappa et al., 2015). Ultimately, models like the SOM can be applied to chamber experiments to better understand the role and contribution of POA, IVOCs and vapor wall-losses to total OA."

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).

We have added a subset of the suggested references.

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).

We have rephrased that sentence to

"Second, they assume that the multi-generational oxidation of products of different anthropogenic VOCs (e.g., alkanes versus aromatics) or different biogenic VOCs (e.g., isoprene versus monoterpenes) share the same reaction mechanism."

In terms of the 'eerie similarity' mentioned by the reviewer, although we agree one could take away such a conclusion, we emphasize the fact that the choice of having ageing reactions on for aromatics but off for biogenics is not based on any physical or chemical intuition, but from what amounts to 'model tuning' in that the decision to do so was made by Farina et al. (2010) after comparing 3D simulations to some observations in Lane et al. (2008). Others have generally followed suit, but this dichotomy has never been validated outside of the 3D model framework. Further, one could also conclude from the current paper that the same holds true for aromatics, but this is counter to what is done by many others who include ad hoc ageing. In other words, we strongly suggest that rather than using ad hoc ageing schemes on top of existing parameterizations, models should instead be revised to explicitly include ‘ageing’ reactions within the parameterization framework (such as is done with SOM and more recently within the VBS framework by Zhao et al. (2015)), as is noted in the reviewer’s next comment.

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.

We have pointed out the computational expense of running the SAPRC+SOM mechanism in our paper in GMD. We have added the following brief statement to the manuscript in Section 2.3

“Simulations were performed for 19 days with the first 5 days used for spin up. For the SoCAB, each simulated day using the SOM required approximately 4 h of elapsed time (on 40 Intel i5-3570 processor cores) so a 19-day episode was simulated in less than 4 days. For the eastern US, each simulated day required approximately 9 h of elapsed time so a 19-day episode was simulated in about 8 days. The SOM simulations were approximately four times slower than the BaseM simulations on account of the large number of model species.”.

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

We apologize for the mistake. It was a version problem. Figure S.1 has been added to the supporting information, but is now labeled as Figure S.2 to accommodate the insertion of a new Figure S.1..

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*).

Yes, that is correct. The reference has been corrected.

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

We have made the correction.

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