

We thank the reviewers for their comments and concerns and we have addressed them point-by-point in this response. We have also highlighted changes made to the final manuscript based on the reviewer's comments. Reviewer comments are in blue, author comments are in red and manuscript changes are in *gray and italic*.

Reviewer 1

The outputs of this model (SOA concentrations and O:C ratios) are of major importance, but how well they represent the ambient atmosphere? There is no comparison with measured data. The authors should select domains and periods where ambient data from AMS are available. A comparison between the modeled SOA and total OOA extracted by PMF (i.e. sum of SV-OOA and LV-OOA species) is necessary for the evaluation of this model. The idea of handling together fragmentation and functionalization processes is important; however, it has to be evaluated against real ambient data. Though the predicted SOA concentrations are in the range between 0.3 and 2 $\mu\text{g m}^{-3}$, which is a logical range, the spatial concentration distribution could be very different compared to ambient measurements. Concerning the O:C ratio the authors use a short and very general statement (page 1877, lines 4-6) to justify their results, which is very weak. O:C also has to be compared with measured values. Even if the average O:C range is within what is globally seen it does not mean that spatially the model reproduces the correct values. This has to be proved. The authors should evaluate this model. In my opinion, without comparisons to ambient measurements and evaluation of the predictions, this paper is very weak.

We have evaluated the basic performance of the UCD/CIT air quality model using multiple aerosol components in a previous manuscript (under review at Atmospheric Environment) and we have included a summary figure of the model-measurement comparison below (see comparison for OA). Since the Base and the average of the SOM simulations produce the same SOA concentrations (see Figure 4 in current manuscript), we find similar performance for the model-measurement comparison between the SOM simulations and measured values. This is especially true in the South Coast Air Basin where model calculations predict little SOA formation from anthropogenic emissions (OA is dominated by POA).

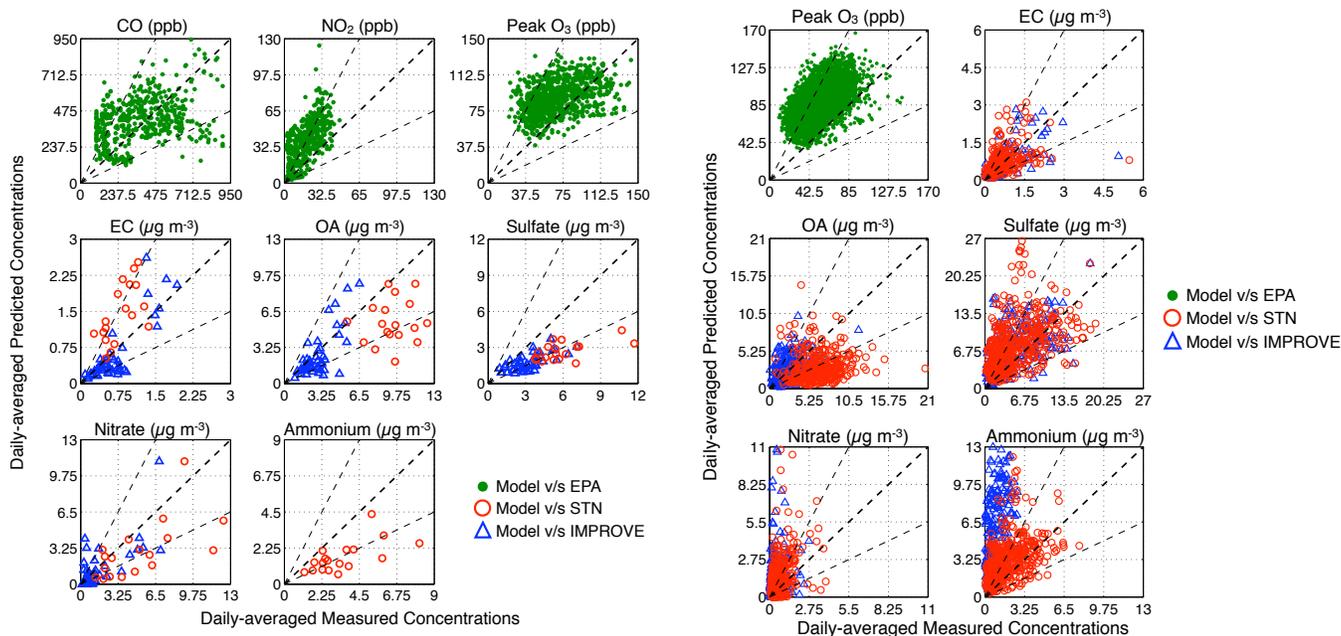


Figure 1: Scatter plots comparing model predictions to measurements made in the South Coast Air Basin (left panel) for Jul-Aug 2005 and eastern US (right panel) for Aug-Sep, 2006.

We have revised the manuscript to compare the episode averaged SOA concentration and O:C value for SOA against the AMS data gathered at Riverside, CA during the SOAR study in the summer of 2005 (Study of Organic Aerosols at Riverside). The comparison is described in the revised manuscript as follows: *“Predictions from the SOM model were compared to measurements made by the aerosol mass spectrometer (AMS) during the Study of Organic Aerosols at Riverside (SOAR) in the summer of 2005 (Docherty et al., 2011;Docherty et al., 2008). Over the two-week simulation, the SOM model under predicted total OA by 40% at this location. This suggests that important atmospheric processes and/or emissions sources upwind of Riverside are omitted from the model framework. The SOM model predicted an average SOA concentration of $0.50 \mu\text{g m}^{-3}$ (average of low yield and high yield), which is 10% of total OA. The campaign-averaged (30 day) oxygenated OA (OOA) concentration measured by the AMS (sum of the semi-volatile OOA, medium-volatility OOA and composite low-volatility OOA) was $7.1 \mu\text{g m}^{-3}$ (80% of total OA). Since the model-predicted OA at Riverside is dominated by POA (~90%), the O:C is controlled by the O:C of the emitted POA (~0.1-0.2) and is lower than the campaign-averaged O:C of 0.31 inferred from the AMS data. The under-prediction (in SOA concentrations and O:C) is typical of predictions in regional (Carlton et al., 2010) and global models (Farina et al., 2010) and arises mostly from an incomplete understanding of the sources and pathways of OA. Numerous factors may contribute to the under prediction of O:C at Riverside, including missing emissions sources for SOA precursors, semi-volatile and reactive behavior of POA (Robinson et al., 2007), SOA formation from unspciated emissions (Jathar et al., 2014), aqueous production of SOA in cloud, fog and aerosol water (McNeill, 2015) and multi-generational aging (Donahue et al., 2012). The SOM model provides a framework to test these production pathways of OA as our understanding about these processes matures.”.*

The reviewer recommends that we compare our model predictions of SOA and O:C against measurements at multiple locations. At this point, these can be inferred only with the aerosol mass spectrometer (AMS), which tends to be operated at one of or a few locations in major field campaigns. There are about 15 or so AMS datasets available over the South Coast Air Basin and the eastern US in the past 15 years. The comprehensive evaluation requested by the reviewer would require us to simulate different spatial domains for different time periods (aka different episodes), an activity that would need us to develop meteorological inputs and SAPRC-SOM specific emissions inventories for each episode, especially challenging if we were to compare with observations from aircraft (which can provide for spatial understanding). Clearly the authors are interested in this topic and will pursue this comparison in future papers, but it is beyond the scope of this introductory paper describing the regional SOM model. The objective of this work was to couple a semi-explicit OA mechanism (SOM) with a gas-phase chemical mechanism (SAPRC), implement it in a 3D model and demonstrate its usefulness by simulating a short episode. Further, the above exercise will only help evaluate the model at specific sites during specific time periods and they will not help constrain spatiotemporal predictions of SOA and O:C. A detailed spatiotemporal model-measurement comparison, as suggested by the reviewer, is not possible at this time with the currently available AMS data.

Reviewer 2

1. My major objection concerns SOA formation in SOM as a function of NO_x levels. Two SOM grids are available, one for low NO_x conditions (high SOA yields) and another for high NO_x conditions (low SOA yields). These two grids are used successively over the full domain. Using

two sets of results based on these two distinct grids is not satisfactory in the perspective of model applications. The paper states that the “current configuration does not allow for continuous variation in the dependence of SOA on NO_x” (p1870, I27). This is clearly a major weakness of this model configuration. This paper being devoted to the implementation of SOM in 3D models, the authors should explain how SOM can ultimately be used to simulate the range of NO_x conditions (i.e. from high to low NO_x) encountered in a given domain. This point is a critical issue that should be examined in this paper devoted to SOM implementation, i.e. how could the model configuration be improved, can a “unified” parameterization (from low to high NO_x conditions) be designed based on a these two grids.

The reviewer’s comment is well received. SOA formation varies with NO_x levels and needs to be appropriately represented in 3D models. While a “unified” parameterization is desirable, we believe that the SOA modeling community does not have sufficient data to realistically represent the NO_x dependence of SOA formation in 3D models, particularly when simultaneously modeling multigenerational oxidation.

Take for example, the NO_x dependence in the Community Multiscale Air Quality (CMAQ) model (Carlton et al., 2010). Only aromatic precursors are considered to be sensitive to NO_x; long alkane and biogenic precursors do not have any NO_x dependence. Recent data from the California Institute of Technology (Chhabra et al., 2011;Loza et al., 2014) clearly shows that long alkanes and biogenic VOCs have NO_x dependent SOA yields. A separate issue is that long alkanes show the opposite trend as aromatics and biogenics to NO_x levels, i.e. high NO_x results in high SOA formation and vice versa (Loza et al., 2014). The NO_x dependence in CMAQ is represented through the competition of the peroxy radical (RO₂) to react with HO₂ (low NO_x) or NO (high NO_x). While predictions of NO in the model can be validated by comparing against measurements, it is unclear whether the model accurately reproduces concentrations of HO₂. Since the NO_x dependence is very sensitive to the predictions of HO₂, the reliance on HO₂ adds uncertainty to the production of SOA. Further, CMAQ only models the first generation of NO_x dependent products and it is unclear how one needs to model the NO_x dependence of multi-generational oxidation. For instance, to accomplish this, one would need chamber data that simulates transport and processing of biogenic/anthropogenic SOA from low/high NO_x environments to high/low NO_x environments. To the best of our knowledge, these data do not exist.

Our modeling approach acknowledges that we do not completely understand the dependence of NO_x on SOA formation, especially as it relates to the multi-generational oxidation of SOA. Instead of presenting a unified parameterization, we provide an upper and lower bound for SOA production that represents production through low NO_x and high NO_x pathways respectively. In our opinion, this approach acknowledges the uncertainty inherent in predictions of SOA and provides a more accurate accounting of uncertainty in the calculations than models that make large assumptions to achieve a “unified” framework for all NO_x conditions. In summary, while we agree with the reviewer that we need to account for the NO_x dependence of SOA, we disagree that we have the data to develop and deploy a realistic unified parameterization at this time. By simply using what was available beforehand (namely the parameterization from Henze et al. (2008)), we are ignoring the uncertainty prevalent in such unified parameterizations.

2. The authors claim that the SOM version tested in this study does not account for oligomerization or heterogeneous reactions (e.g. p1869, I22-27). This statement is misleading. Indeed, the SOM approach is empirical, based on chamber observations in which these processes very likely occur. Even though no transformation in the condensed phase is explicitly represented in the design of SOM, fitting SOM to match chamber observations somehow

implicitly account for these processes. The links between SOM design, chamber observation fits and 3D applications deserve additional discussion in the paper.

To address the reviewer's comment we have revised the statement accordingly: *"While the SOM framework can be adapted to explicitly model other production and loss processes (e.g., oligomerization (Yatavelli et al., 2012), heterogeneous reactions (Shiraiwa et al., 2013) in the atmosphere, in this work we consider parameterizations developed that consider only the multi-generational gas-phase oxidation of SOA precursors and their subsequent products. As with all existing SOA parameterizations that are used in 3D models, inherent in the parameterization are the effects of condensed-phase (and other unaccounted for) processes. As improved understanding of the kinetics and reaction chemistry of key heterogeneous and condensed-phase processes is developed they will be incorporated into the SOM framework."*

3. Minor comment: The sentence p1878, l21 is ambiguous. The high NO_x and low NO_x grids were alternatively used over the full domain, without considering the simulated NO_x field (see also comment above). How can the authors state that the "product distribution is different under different NO_x levels"? A "unified NO_x parameterization" should be used to answer this question.

We have modified the statement accordingly: *"With the SOM we are also able to quantify the distribution of the SOA mass in carbon and oxygen space and find that the predicted product distribution is different under the two simulated NO_x levels"*.

What we mean here is that final product distribution of SOA in SOM space is different for low and high NO_x conditions. We do not intend to imply anything for the production distribution of atmospheric OA. Atmospheric OA might exhibit a mixed product distribution that can be simulated only when a scientifically rigorous unified parameterization is developed (see earlier comment about why we have decided to undertake two simulations with two different NO_x assumptions instead of a unified parameterization).

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