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Interactive comment on "Understanding sources of organic aerosol during CalNex-2010 using the CMAQ-VBS" *by* M. C. Woody et al.

M. C. Woody et al.

woody.matthew@epa.gov

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We appreciate the reviewer's thoughtful and insightful comments. In response to the reviewer's comments, we have made efforts to more clearly define our terminology, most notably POA, SOA, SVOCs, and IVOCs. We believe this clarification helps to address many of the comments provided by the reviewer. Below follows responses to specific comments, with the reviewer's original comment followed by our response in bold.

Specific Comments: P26746, L12: What is meant here by "semi-explicit OA treatment" and "SOA lumped by parent hydrocarbon"? The products forming SOA in this model are semi-volatile, as they are in the VBS model, correct? And are not most species

treated via a 2-product framework, with the exception of isoprene?

We have revised the abstract, clarifying our definitions of the CMAQ-AE6 OA treatment and the CMAQ-VBS OA treatment. Specifically for CMAQ-AE6, we have revised the text to read: Traditionally, CMAQ treats primary organic aerosols (POA) as nonvolatile and uses a 2-product framework to represent secondary organic aerosol (SOA) formation.

P26748, L2: The authors seem to be implying that an O:C > 0.3 indicates a large contribution from SOA. If this is their intention, this thought should be made explicit.

We have clarified this statement, adding we believe O:C > 0.3 suggests significant contributions from SOA. The text now reads: "Average OA O:C ratios exceed 0.3 in southern California (Craven et al., 2013) suggesting significant contributions from SOA,"

P26751, L4: I do not entirely follow the arguments resulting from the statement that fragmentation is more important for biogenic SOA than for POA. I would think the appropriate comparison is between biogenic SOA and anthropogenic SOA from aromatic compounds, which are the species to which ageing is applied by default. The comparison between biogenic SOA and POA does not seem relevant here.

We agree that that the comparison for fragmentation from biogenic SOA and POA lacks relevance in this context. Our intent was to highlight the importance of fragmentation for biogenic SOA which we have revised the text to indicate without the comparison to POA. The text now reads: "In excluding aging of secondary biogenic SVOCs in all but our sensitivity simulation, we effectively assume that the net result of functionalization (aging) and fragmentation, an important process for accurate predictions of biogenic SOA (Donahue et al., 2012b), does not increase biogenic SOA concentrations (Fountoukis et al., 2011)."

P26751, L8: It would be could if the authors could clarify further what they mean when they say that "a portion...of the OA mass [is] shifted from the POA to the SOA set." It

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would seem more appropriate to me to have the “SOA” set include all vapors and the “POA” set to include only condensed-phase material.

We have clarified this in the text, indicating this is a modeling methodology to track O:C. “a portion. . .of the OA mass shifted from the POA to the SOA set (Koo et al., 2014). The transfer of oxidized primary SVOCs (i.e. POA) to secondary SVOCs (i.e. SOA) is used as a modeling technique to maintain accurate O:C ratios, a feature of the 1.5-D VBS used in CMAQ (Koo et al., 2014), using existing POA and SOA basis sets and avoid additional computational burden of added model species (e.g. oxidized POA basis set).” Also, in an effort to clarify what each of POA, SOA, SVOCs, and IVOCs refer to in our framework, we have made an effort to more clearly define each of these terms for our study. “Traditional CMAQ-AE6 nonvolatile POA is replaced in CMAQ-VBS with semivolatile POA, referred to here as primary SVOCs, comprised of primary gas and particle phase organics located in the primary anthropogenic basis set. In this framework, CMAQ-VBS POA is therefore primary SVOCs located in the particle phase.” “CMAQ-VBS also includes a formation pathway of SOA from the oxidation of IVOC emissions, where IVOCs represent gas phase compounds with volatilities between SVOCs and VOCs (C* values ranging from 104 to 106 μg m-3). Most of these compounds are generally considered to either be missing from emission inventories entirely or mischaracterized as non-SOA forming compounds. The inclusion of IVOCs represents an additional SOA precursor mass introduced into the model relative to CMAQ-AE6.” “CMAQ-VBS semivolatile SOA is represented using secondary SVOCs (gas and particle phase) located in the secondary anthropogenic and biogenic basis sets.”

P26751, L10: It is not clear here what is meant by “oxidized POA”. Does this refer to “POA” vapors that are oxidized? It would be useful if the authors were to clarify the distinction between gases and condensed-phase material more explicitly. I am finding the POA/SOA distinction here to be somewhat difficult to follow. Along these same lines, the terms SVOC and IVOC could be more explicitly defined for the reader

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(especially in the context of P26751, L6-14 and how this relates to the information on the previous page). I think that it could be helpful to move the paragraph starting on Line 15 on P26752 to earlier in this section, probably before discussion of the SOA treatment.

Please see the response to the previous comment, which clarifies our definitions of POA, SOA, SVOCs, and IVOCs. We have also moved the paragraph starting on Line 15 of P26752 to before the discussion of the SOA treatment as recommended.

P26753: The discussion of some of the caveats associated with estimating S/IVOC emissions, especially source-specific emissions, is very helpful, although it would be even better if the authors were to put this in the context of more recent measurements and constraints on S/IVOC emissions in addition to the relationship with other model treatments. Some of this is provided later in the manuscript (P26763), but it would seem appropriate to include some discussion here.

We have added the following text to the manuscript in an effort to provide better context to S/IVOC emission estimates across modeling studies: "Therefore, modeled S/IVOC emissions can range from 2.5 to 7.5x existing POA inventories to match measurements (which makes direct comparisons to existing inventories difficult) and remains a source of uncertainty in conducting and comparing models that include S/IVOCs."

Figure 1: It would be useful if the authors were to put a box around the LA area to guide the reader to that location.

We have added a box around Downtown LA and Pasadena to this figure and updated the figure caption to reflect the update.

Table 3: It would be useful if the authors were to provide the equations used to calculate the statistical metrics.

We have updated the table to include equations for the statistical metrics.

Regarding the supplemental figures, it would be useful if the authors were to (a) have

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the figures included in the main text in order and (b) to include mention of all supplemental figures in the main text. The reader should be made aware of the content of the supplemental within the main text.

We have referenced each of figures and tables located in the supplement in the main body of the manuscript.

P26755, L27: The authors note that larger model-measurement gaps were seen during photochemically active periods when OOA concentrations were higher. In looking at Fig. 2, this is not abundantly apparent to the reader. After estimating some numbers off of the figure, it seems to me that this is not universally true. For example, the observed/model ratio on 4-June (when OOA is higher) is only âĹij2.3 but is âĹij3.3 on 8-June when OOA is lower. It would be useful if the authors were to formalize this thought through an explicit demonstration, for example by plotting the observed/model ratio as a function of observed OOA concentrations.

We had intended the model to measurement gaps to refer to the difference in OA mass. However, as the reviewer points out, when one instead uses the ratio of observed to modeled concentrations, apparent occurrence of large "gaps" changes. In order to clarify, we have revised the text to read: "with the largest differences in modeled to measured OA mass generally occurring during photochemically active periods (e.g. 4 to 7 June).

Fig. 4 vs. Fig. S8: To facilitate comparison, it would be useful if the Fig. S8 were modified to separate the CMAQ HOA and CIOA.

Fig. S8 does separate CMAQ-VBS HOA and CIOA. We believe the reviewer may instead be referring to Fig S9. That said, Fig. S8 was a combination of the data from Fig. 4 a and b (CMAQ-VBS vs. AMS data), with no new data being presented on Fig. S8. Therefore we have removed it from the supporting information. We have also moved the information on Fig. S9 (CMAQ-AE6 vs. AMS data) to Fig. 4 c and d to help facilitate comparisons. While ideally we would separate CMAQ-AE6 HOA and CIOA

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similar to CMAQ-VBS, CMAQ-AE6 does not track these separately and instead only reports POA.

P26756, L21: I find the meaning of this sentence to be somewhat unclear in terms of what is meant by "theoretical partitioning" in the context of "AMS measured OA". Do they mean that the larger CIOA concentrations lead to lower concentrations of semivolatile vapors due to enhanced partitioning? I think this is the case, but it could be stated more explicitly.

In an effort to clarify, we have revised the text to read: "To determine if partitioning alone explained the underprediction in modeled midday CIOA concentrations, we considered two potential scenarios. In the first scenario, we removed model OA bias by replacing modeled OA with AMS measured OA and then calculated the theoretical partitioning of modeled semivolatile CIOA vapors. Using the higher AMS OA concentrations, more semivolatile CIOA vapors partitioned to the particle phase and increased modeled CIOA concentrations by approximately 10% in the afternoon. In the second scenario, we treated the modeled CIOA as nonvolatile. . ."

P26757, L3: The authors conclude that the underprediction of CIOA even using nonvolatile CIOA indicates that emissions were low. Couldn't an overestimate of the boundary layer height, especially at night, also be a contributing factor when considering absolute values? The authors might consider normalizing by background corrected CO, as they do with the SOA, to account for issues of dilution.

We agree that modeling bias in the boundary layer heights could have a role in model performance for CIOA. However, Kelly et al. (2014) found CMAQ model performance for the boundary layer was generally good with the exception of the evening transition. During these hours, CMAQ underpredicted mixing, which increased concentrations. In recognizing that low emissions may not be the only reason for the underprediction, we have also added underpredictions may also be attributable to too high of dispersion, similar to one of the factors we considered for SOA.

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P26757, L18-21: The authors conclude based on the nighttime underprediction that low emissions are likely the reason for the underprediction during the daytime, rather than photochemistry. However, if the diurnal emissions profile is incorrect, it may be that the daytime underprediction is due to photochemistry (and low emissions) while the nighttime underprediction is due to low emissions alone. I suggest that the authors need to make a stronger argument as to how the nighttime underprediction truly constrains the reason for the underprediction during the daytime given an uncertain emissions timing. It is argued that the slower growth of the CIOA during the daytime compared to SV-OOA is suggestive of low emissions, as opposed to photochemical influence, but isn't it possible that the reaction rates are different, leading to differences in the timing when coupled with transport?

We agree that it is difficult to determine directly from the data if emissions or photochemistry are more likely the cause for the underprediction of CIOA. Therefore, we have revised the text to indicate both as plausible causes.

Figure S11: The meaning of "POA" in this figure is unclear. Is this CIOA + HOA (total POA) or just the HOA-type? In the main text, "POA" seems to be used to mean "non-CIOA POA" (Section 3.2.2). Similarly in Fig. S12 This should be clarified and terminologies used consistently throughout. Perhaps the authors could adopt the terminology "oPOA" to indicate "other POA" aka non-CIOA POA.

In an effort to clarify, we have defined non-CIOA POA as other POA (POA-o) throughout. We chose not use "oPOA" since it is often used to mean oxidized POA.

P26759, L2: The "opposite" behavior of the modeled non-volatile oPOA compared to the observed HOA is similar to that of the semi-volatile oPOA, correct? As stated, this makes it seem that such "opposite" behavior is only for the non-volatile treatment, but Fig. 4 suggests that there is a similar "opposite" behavior for the semi-volatile case.

We agree that the semivolatile other POA diurnal pattern, albeit muted, is similar to the nonvolatile other POA diurnal pattern. We have therefore revised the text to read:

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"The resulting diurnal pattern (Fig. S3) was higher in the morning and evening, with a minimum in the afternoon, similar to the more muted diurnal pattern of the semivolatile treatment (Fig. 4a) but opposite the AMS measurements (lower in the morning and evening, peaked in the afternoon)."

P26759, L17: Is this discussion associated with "ageing" in the context of POA indicating that including some SOA as POA would help? This is not entirely clear to me, I think in part due to my not entirely understanding the model definitions of POA and how/whether the "POA" category includes SOA (here, I think, referred to as oxidized POA). It would be useful if this could be clarified here and/or within the methods section.

We have attempted to clarify this both here and in the methodology section. In the methodology section, we have more clearly defined SVOCs, IVOCs, POA, and SOA and how oxidized POA is represented (66-90% POA depending on the level of oxidation). We have specifically revised the text here to read: "Alternative aging schemes. . .generally produce more OA mass. . .and if applied to primary SVOCs could better represent the POA-o midday peak (Hayes et al., 2015) since the majority of aged primary SVOCs (i.e. oxidized POA) remains as primary SVOCs/POA."

P26759, L26: Regarding the conclusions associated with the NEI, again I think that it would be helpful if the authors were to more explicitly define their categorizations of what counts as POA. I find it somewhat difficult to understand how underestimates of SVOCs leads to underestimates of POA, unless oxidation of SVOCs produces POA. I think that it does in this model, but I am finding the terminology to be difficult to follow. I think that the authors could make this work more easily readable by adopting more precise language. For example, if their POA really equals POA + some fraction of oxidized SVOCs, then a better name would be POA+SVOCOA (or something like that, as I realize that is a cumbersome terminology). But even that might not be sufficient, because if I am understanding correctly some fraction of SVOCs contributes to a different SOA category. Ultimately, I strongly encourage the authors to rethink their overall

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terminology to make it more easily accessible to the reader.

We have made efforts to more clearly define our definitions of POA, SVOCs, SOA, etc in the methodology section which we believe will provide clarity here.

P26760, L9: It would be useful if the authors were to point the reader to a figure or table at this point. Perhaps Fig. 3 and Fig. 4. Along these same lines, it would be useful if the authors were to move some of the additional information (specifically the LV-OOA and SV-OOA diurnal profiles) in Fig. S8 to Fig. 4b.

We have added a reference to Fig. 4b here as well as added diurnal profiles of LV-OOA and SV-OOA from Fig. S8 to Fig. 4b.

P26760, L12-13: It would be useful if the authors were to note that the cited studies were not for LA but for other urban regions.

We have noted these studies were not specific for LA by revising the text as follows: "This is consistent with many regional air quality studies (Volkamer et al., 2006; De Gouw et al., 2008), including CMAQ (Foley et al., 2010), which often underpredict urban SOA. Although those studies are not speciïňĄc for LA, the similarity of tracer-normalized SOA concentrations across urban areas (e.g. De Gouw and Jimenez, 2009; DeCarlo et al.,2010; Hayes et al., 2015; Zhang et al.,2015) supports the occurrence of a general urban SOA under-prediction with models.

P26762, L2: The authors use –log(NOx/NOy) to estimate photochemical age and compare observed values at Pasadena to their modeled values. They conclude that the modeled photochemical age was too small by a factor of 1.5. However, doesn't this analysis also rely on the spatial distribution of NOx emissions being correct? NOx is converted to NOy over time, but if the NOx inputs are too low or too high along the transport pathway then the photochemical age will be incorrect. It could potentially be useful to also consider the NOx/CO ratio. Or, potentially, the toluene/benzene ratio (although the timescales may be too short for this to be a useful photochemical clock).

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Also, I find the second half of this sentence to be difficult to follow, in particular the section starting "...but not on SOA." I suggest the authors might be more explicit here.

We agree that NOx/NOy as a proxy for photochemical age has limitations. However, photochemical age estimated by measurements (Hayes et al., 2013) was similarly estimated using NOx/NOy. Therefore, it provided the best means to compare model and measurements. With regard to second half of the sentence, we have revised the text as follows: "...which helps explain part of the underprediction in SOA concentrations (Figs. 3 and 4) but not underpredictions of SOA production efĭňĄciency (Fig. 5) (i.e. the efficiency per unit precursor at a given age)."

Fig. 5: What should the reader take away of the apparent intercept in Fig. 5a not being 0? If the fit were performed without constraining the fit to go through zero, a steeper slope would likely be obtained, correct? In Fig. 5b, is there a reason that the authors chose to not present the model results in the same manner as in Fig. 5a, i.e. using a Gaussian density kernel estimate to colorize the points? There seem to be a lot of points in the CMAQ-VBS simulations that fall along a line with much lower slope than the fit slope, which was forced through zero. The authors should also note in the main text (and/or the caption) that the fits were forced through zero.

Though the presentation of point density appears to suggest otherwise, we made no alterations to the slope or y-intercept (0.0002) of the best-fit line for CMAQ-VBS. We chose not to include the Gaussian density kernel estimate of density on Fig. 5b since it includes points for both CMAQ-VBS and CMAQ-AE6 and adding another dimension (i.e. colors for density) would make the plot difficult to read and interpret.

P26763, L1-5: The authors concluded that the SOA production efficiency was underpredicted by a factor of 1.6-2. They note that Zhang et al. (2014) indicated a potential underestimate in SOA production by a factor of 2-4 from losses of SVOCs to chamber walls, but also that this was only for alkanes and toluene and was specific to the chamber used. They then conclude that SVOC wall loss does not likely account for

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the entire underestimate of SOA production efficiency. I do not entirely see how this conclusion is justified based on the statements given. As a hypothetical, what if the losses were greater in other chambers? Also, a factor of 2-4 is greater than a factor of 1.6-2. I suggest that the authors revisit the justification for this conclusion.

Our interpretation of the Zhang et al. (2014) results was that the 2-4 factor was an upper bound estimate based on other studies. However, we failed to reference these other studies in support of our conclusion. We have revised the text to include these references and provide further justification for our conclusion: "However, the factor of 4 is for alkane systems (speciated long alkanes are not considered SOA precursors in CB05) and toluene and specific to the smog chamber used in Zhang et al. (2014). Other studies have generally reported lower values, ranging from 1.2 to 4.1 for low-NOx conditions and 1.1 to 2.2 for high-NOx conditions (Ng et al., 2007; Chan et al., 2009; Chhabra et al., 2011; Loza et al., 2012; Cappa et al., 2013). Therefore, the 2-4 factor likely represents an upper bound and SVOC wall loss does not likely account for the entire underestimate of SOA production efficiency."

P26763, L13: The authors note that the use of the results of Jathar et al. (2014) to update the IVOC emissions and parameterization in CMAQ-VBS could help to bridge the gap between model and measurements, and then go on to perform some sensitivity tests by scaling up the S/IVOC emissions. However, it would be useful if the authors were to more specifically make a connection with the Jathar et al. work. What does that work imply about what is correct? Are the test simulations consistent with those results?

Our hypothesis was that better constrained IVOC emissions from Jathar et al. (2014), who indicated SOA formed from IVOCs dominated total SOA formed from combustion emissions, and yields could account for some of the missing CMAQ-VBS SOA mass. Our goal was to test how much of the missing SOA mass S/IVOC emissions could account for using upper bound S/IVOC emissions. However, this connection was unclear. In an effort to clarify our intentions, we have revised the text to read: "Current

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CMAQ-VBS IVOC emissions are scaled to primary SVOC emissions (1.5x) based on the results of a diesel generator (Robinson et al., 2007) and could potentially be updated to utilize more recent results, such as those reported by Jathar et al., 2014 who indicated unspeciated organics (S/IVOCs) dominated SOA mass formed from combustion emissions. Future work is needed to explore if better constraining IVOC emissions and yields in CMAQ would help improve model performance, but it would likely not account for the entire missing SOA mass based on sensitivity simulations using upper bound S/IVOC emissions. In these simulations, S/IVOC emissions were increased. . .”

P26764, L11: Are the measurements referred to here from Baker et al. (2015) or Zotter et al. (2014)? This should be clarified. Also, it would be useful if the authors could indicate (a) by how much >1 the non-fossil fractions were and (b) why contributions form a medical waste incinerator would lead to an estimated non-fossil fraction > 1.

The measurements were from Baker et al., (2015). To clarify, we have seperated the clauses referencing Baker et al. (2015) and Zotter et al. (2014). The text now reads: “to compare CMAQ-VBS. . .against filter-based measurements collected at Pasadena (Fig. 6) (Baker et al., 2015). Those measurements indicated, on average, a near even split of non-fossil (48%) and fossil (52%) carbonaceous mass (Baker et al., 2015). The Baker et al. (2014) non-fossil measurements were also consistent with other collocated 14C measurements collected during the same time period (51% non-fossil) (Zotter el al., 2014).” The values for the non-fossil fractions > 1 ranged from 1.1 to 3.3. The fractions are based on an assumed non-fossil 14C concentration of approximately 1.2 x 12-12 14C/C (Buchholz et al., 2013). Medical incinerators emit 14C tracers and can therefore produce 14C concentrations > 1.2 x 12-12, biasing the non-fossil fraction to values > 1. We have updated the text to reflect these clarifications as follows: On 6 days the measured non-fossil fraction was >1 (values >1 ranged from 1.1 to 3.3). . .The non-fossil fraction estimates assume a non-fossil 14C concentration of 1.2 x 12-12 14C/C and emissions from medical incinerators, which contain 14C, can bias the 14C/C ratio (Buchholz et al., 2012).

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Section 3.3: After reading through this section a few times, I suggest that addition of a summarizing table could be quite helpful that has entries for the different combinations of species considered (e.g. CMAQ-VBS fossil fraction with EC and without EC).

We have added a table summarizing the CMAQ-VBS and observed non-fossil C, fossil C with EC, and fossil C without EC.

Fig. 8: It would be very useful if the authors were able to split their "A_AGE" category into aged SOA originating from VOCs versus that from IVOCs. Also, I do not see the "B_IVOC" category in the figures, although one is indicated in the caption. To which species does this refer? Finally, it would be useful if the authors were to include the total AMS OOA on this figure, for comparison and reference to Fig. 4. Clearly, the predicted OOA is still greatly underpredicted even after addition of ageing of biogenic species.

We have split the "A_AGE" category into aged SOA originating from VOCs and IVOCs. The reference to "B_IVOC" was a typographical error and it has been removed from the figure caption. We agree that including AMS OOA on the figure would help facilitate comparisons. However, given the large underprediction, including the measured OOA would alter the scale of the y-axis and compress the modeled contributions to the point they would be difficult to discern.

P26767, L21: While I agree that the ageing scheme represents a "technique to increase model SOA yields," I find the words "similar to" in the context of the Zhang et al. (2014) findings to be a bit awkward as these refer to different physical processes. Additionally, Zhou et al. (2015, ES&T, 49, 2245−2254) demonstrate that the addition of ageing on top of existing parameterizations leads to an overprediction of SOA concentrations in chamber experiments, suggesting that such increases in SOA concentrations may result for the wrong reason. I suggest instead the authors state something to the effect of "The results indicate the majority of SOA was formed from aging, representing a technique to increase model SOA yields. Although via a different

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process, the resulting outcome is similar to that obtained if SOA yields are increased to account for SVOC losses to chamber walls, as proposed by Zhang et al. (2014) and used with CMAQ-AE6 in Baker et al. (2015). Also, although the inclusion of ageing reactions leads to an increase in SOA concentrations, this may be for the wrong reason as recent model-measurement comparisons with chamber experiments suggest that including ageing reactions on top of existing parameterizations can lead to overprediction of SOA concentrations (Zhou et al., 2015)." In other words, I think that a similar caveat as was included for the scaling up of S/IVOC concentrations is required.

We agree that the wording, particularly the use of "similar to", in the context of these two different physical processes could be improved. Based on the reviewer's recommendation, we have revised the text to read: "Although via a different process, the resulting outcome is similar to that obtained if SOA yields are increased to account for SVOC losses to chamber walls, as proposed by Zhang et al. (2014) and used with CMAQ-AE6 in Baker et al. (2015). Also, although the inclusion of aging reactions leads to an increase in SOA concentrations, the model parameterization may overemphasize the contribution from aging as recent model to measurement comparisons with chamber experiments suggested the addition of aging reactions on top of existing parameterizations can lead to overpredictions of SOA concentrations (Zhou et al., 2015)."

P26767, L25: I suggest that this is reworded. The models do not, in my view, "utilize comparable SOA yields." The yields from the models are not constant values, but the result of specification of semi-volatile product yields that produce SOA. I suggest instead that they simply change "utilize" to "produce." Here, also, the Figure numbers should be given and the figures reordered.

We agree that "produce comparable SOA yields" is a more accurate statement and have revised the text to reflect this change. We have also provided the figure numbers for the yield curves in the supplement and reordered the figures.

SIMPLE model: I suggest that a line for the SOA/CO slope from the simple model be

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added to Fig. 5.

We have added the SIMPLE model slope to Fig. 5 as recommended.

General note on Figures: Many have relatively small axes labels. I encourage the authors to make sure that the labels are sufficiently large such that when formatted into a final article they remain easy to read.

We have increased the size of axes labels.

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