Atmos. Chem. Phys. Discuss., 15, C11783–C11788, 2016
www.atmos-chem-phys-discuss.net/15/C11783/2016/

Atmospheric Chemistry and Physics Discussions

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

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Received and published: 20 January 2016

We appreciate the reviewer's time and effort to provide thoughtful and thorough comments. Below are responses to the reviewer's specific comments, with the reviewer's original comment shown followed by our response.

The abstract is generally a good representation of the paper, from the perspective of the presentation of a lot of results with limited synthesis and interpretation. The abstract is quite long, and it is suggested that some of the details be omitted. In addition, it is suggested that a synthesis of the results and their implications be included to highlight significance of the work.

As recommended by the reviewer, we have reduced the length of the abstract by omit-

ting details. We have also attempted to synthesis the results and provide some implications by concluding the abstract with the following text: “Overall, the CMAQ-VBS incorporates semivolatile treatment of POA and improves SOA model performance (though SOA formation efïňĄciency is still 1.6-2× too low). However, continued efforts are needed to better understand assumptions in the parameterization (e.g. SOA aging) and provide additional certainty in how best to apply existing emission inventories in a framework that treats POA as semivolatile, which currently degrades existing model performance at routine monitoring networks. The VBS and other approaches (e.g.AE6) require additional work to appropriately incorporate intermediate volatility organic compounds (IVOCs) emissions and subsequent SOA formation.”

p. 26749, line 17-18: The focus on the degree to which processes and/or sources characterized by CMAQ are at play in the ambient atmosphere is not particularly well connected to the focus on sources of OA as written.

In an effort to clarify, we have revised the text to read: “Our analysis focuses on the degree to which processes and/or sources characterized in CMAQ v5.0.2 may be responsible for OA observed as part of CalNex.”

The first three paragraphs of the introduction present many prior studies that are relevant to the research from the perspective of reporting previous OA measurement/modeling results in California, including during CalNEX. However, the information is poorly organized and it does little to build the motivation and need for the specific work presented. It is suggested that the authors consider reorganization of the introduction to better support their efforts in analyzing process/source contributions to measured and modeled OA.

We have reorganized the introduction as recommended to provide a clearer motivation for our work.

p. 26749, line 9: “indicates” should be “indicated” or “indicate”

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We have made the suggested revision.

p. 26750, line 1-2: In the abstract and throughout the results, CMAQ-AE6 is differentiated from CMAQ-VBS; here however, they are described as the single approach to simulate aerosols (aerosols 6 module with the VBS approach). This needs to be reconciled.

We have clarified the aerosol treatments here by revising the text to read: "Aerosols were simulated using the traditional aerosols 6 (AE6) module (CMAQ-AE6) and with an alternative version of AE6 which uses the volatility basis set (VBS) approach (Donahue et al., 2006) to model OA (CMAQ-VBS)."

It is suggested that the authors consider restructuring the methodology such that the paragraph starting on p. 26752, line 15, continues the discussion of SVOCs and POA from the paragraph starting on p. 26752, line 6; and then is followed by discussion of the emissions inventory and then modeling domain.

We have restructured the methodology section to better group related topics.

p. 26750, lines 10-20: The purpose of the additional references following Murphy and Pandis is unclear. In some cases, it seems as if the authors would like to reference the original data source, however, that is not made clear. What is the Koo et al. reference for? And Carlton et al. 2010? These are confusing given that the authors note that yields are based on Murphy and Pandis 2009.

Our intent was to reference the original data source but we have removed these references (with the exception of Hildebrandt et al., 2009) to avoid confusion since all relevant details are provided in Murphy and Pandis. The Koo et al. and Carlton et al. references were meant to point back to the CMAQ-VBS and CMAQ-AE6 descriptions of OA treatment. However, since these details are provided more clearly elsewhere, we have also removed these references here.

p26750-2751: The authors need to make it clearer that the order of magnitude reduc-

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tion in volatility (e.g., line 25, p26750) occurs at each step.

We have clarified that each oxidation step for both POA and SOA reduces volatility by an order of magnitude. “Primary SVOCs are aged/oxidized in the gas-phase by reactions with OH. . .with each oxidation step lowering volatility by an order of magnitude...” “Anthropogenic SOA aging reactions form products with a vapor pressure reduced by one order of magnitude (10x) for each oxidation step.

p. 26755, line 25: It is suggested that the authors remove the imprecise language such as “were in reasonably good agreement”, given that quantitative metrics follow.

We have made the recommended revision by removing imprecise language (e.g. “were in reasonably good agreement”) when quantitative metrics are provided.

p. 26758, line 22: It is not clear whether “estimated” here means calculated or concluded based on simulations. Please clarify.

We have clarified by replacing “estimated” with “calculated”.

p. 26763, line 6: Can the authors clarify what is meant by production efficiency? I was not expecting missing/mischaracterized IVOCs to be linked to production efficiency as is written.

SOA production efficiency is the efficiency per unit precursor at a given age. However, because of differences in how IVOC emissions are estimated, uncertainty in yields, and lack of SOA produced by IVOCs in CMAQ-AE6, we have categorized SOA formed from IVOCs as production efficiency.

p. 26763, line 25: Can the authors expand on what is meant by obtaining agreement for the wrong reasons? Can they give examples of what else would lead to the same conclusions?

We did not want the reader to interpret S/IVOC emissions accounted for all underpredictions in SOA production efficiency, though a factor of 7.5x would suggest that it does.

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Instead, other missing (or underrepresented) pathways (e.g. low yields, missing precursors, oligomerization, etc.) could represent part of the underprediction in formation efficiency that increased S/IVOC emissions accounts for. Therefore, we have revised the text to read: “However, the approximate agreement may be for the wrong reasons as increased S/IVOC emissions may account for SOA from other missing (or underrepresented) formation pathways and should not be over-interpreted as direct evidence of the presence of SOA formation efïňĄciency of S/IVOCs.

p. 26767, line 11: Replace “biogenic” with “biogenics”, or add “species”/”compounds”/etc.

We have revised the text to read: “. . .biogenic VOCs. . .”

p. 26768: It is not clear what is added by the application and discussion of the SIMPLE parameterization.

Our intent on the inclusion of SIMPLE was to answer the question as to how best can we improve CMAQ in the short term given many of the shortcomings highlighted in our work. We have clarified this by revising the text to read: “Given the limitations in CMAQ-AE6 and CMAQ-VBS to accurately predict SOA at Pasadena and uncertainty in how best to improve predictions raises the question as to if other parameterizations can improve CMAQ performance in the near term. To this end, we have applied a simplified SOA parameterization. . .”

Largely absent from the results and conclusions is the role of oligomers (e.g., see review by Ziemann and Atkinson, 2012) and their lack of representation in models.

In recognizing the formation of SOA from oligomerization and the absence of a formation pathway in CMAQ-VBS, we have included the following paragraph in the SOA results section: “Note that CMAQ-VBS does not include an oligomerization formation pathway in which heterogeneous/multiphase reactions form SOA (Ziemann and Atkinson, 2012). The lack of this pathway could account for underpredictions in production

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efficiency though it is plausible the SVOC aging parameterization already accounts for some of the mass formed through oligomerization. CMAQ-AE6, which does include an oligomerization formation pathway (Carlton et al., 2010), estimates approximately 20-25% of SOA at Pasadena is comprised of oligomers (Fig. S5), though because CMAQ-AE6 significantly underpredictions SOA, this equates to only a small amount of total mass (0.06 μg m-3 on average)."

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 26745, 2015.