

Interactive comment on “Simulation of the chemical evolution of biomass burning organic aerosol” by Georgia N. Theodoritsi and Spyros N. Pandis

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(1) *The Theodoritsi and Pandis manuscript reports on the predicted sensitivity of organic aerosol (OA) mass to biomass burning emissions, using a source resolved version of the chemical transport model, PMCAMX (PMCAMX-SR). Studies such as this one are important for understanding the potential air quality and climate effects of anthropogenic and biogenic biomass burning emissions, particularly since the representation of biomass burning-derived SOA is relatively undeveloped in most chemical transport models. The inclusion of biomass-burning derived SOA, particularly when including IVOC, leads to substantial contributions to total predicted OA. This study high-*

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lights the need to better constrain biomass burning emissions inventories, including the volatility distribution, and to better understand SOA formation potentials of those emissions. This manuscript is likely to be of interest to the ACP community, and publication is recommended upon addressing the following comments.

We do appreciate the constructive comments and suggestions of the referee. We have done our best to address them and revise the paper accordingly. Our responses (regular font) follow the comments of the referee (in italics) below.

Technical Comments

(2) *It is known that simulating the spatial and temporal distribution of OA, particularly SOA, can be challenging; compensating errors can obscure model performance. In the abstract and in section 7, in addition to the absolute performance statistics, it would be useful to report the change in performance with the expanded treatment of bb-OA (POA+SOA). Weaker performance in winter could be a function of the base simulation (emissions, chemistry, and/or meteorology) and not necessarily a function of the expanded treatment of bbOA.*

We agree with the referee that the performance of a model for total OA depends on a lot of factors (multiple sources, secondary production, removal, meteorology). The PMCAMx-SR performance is a little better for OA than that of the regular PMCAMx. PMCAMx has a similar weak performance in the winter suggesting, as the reviewer states, that this is probably due to other factors and not the expanded treatment of the bbOA. We have added a new section discussing briefly the differences in performance and also the above issues.

(3) *Line 58: It is recommended that it be emphasized that bbOA is added as a third category, and is not explicitly considered anthropogenic or biogenic, though bb emissions are characterized in the manuscript as anthropogenic (ag. and heating) or biogenic (wildfire).*

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We have clarified this point, following the reviewer's suggestion. Indeed, the model does not currently separate the anthropogenic (e.g., residential heating, agricultural burning) from the biogenic sources (wildfires) of bbOA.

(4) Line 111-113: How many model compounds are used to represent IVOCs and SVOCs, respectively? Was the SAPRC mechanism updated as part of this study? If so, the authors should provide further detail in the supplement. If not, a reference should be provided (may be the Environ reference, just needs to be moved).

In this work the IVOCs, SVOCs, and LVOCs are described with 9 volatility bins (from 10^{-2} to $10^6 \mu\text{g m}^{-3}$). Different lumped compounds are used for the fresh (primary) and secondary organic compounds. The simple reactions of these compounds (one volatility bin change for each reaction with OH) have been added the original SAPRC mechanism. The size distributions of the SVOCs and LVOCs in the particulate phase are described. This information has been added to the revised paper.

(5) Line 139: Was the May et al. volatility distribution applied to all bb emissions? The use of "wood burning" here implies only residential wood burning, but it is assumed that the bb volatility distribution was applied to all three categories of bb emissions. This needs to be modified/clarified.

The May et al. volatility distribution was applied to all categories of biomass burning emissions. The use of the term "wood burning" in line 139 is now replaced with "biomass burning". We also clarify that the same volatility distribution is assumed for all bbOA sources.

(6) Line 146: How does partitioning within this model framework depend on aerosol composition?

Organic gas-particle partitioning depends on aerosol composition according to gas-particle partitioning absorption theory. The model assumes that the organic com-
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pounds form a single pseudo-ideal solution in the particle phase and do not interact with the aqueous phase. This is now clarified in the paper and a reference is provided.

(7) Lines 153-155: The description of the Lane et al. VBS scheme is confusing as written. Given the generally widespread use of the VBS SOA model, it might be clearer to write that SOA is represented using 4 bins, and X number of VOC precursors that are tracked separately as either aSOA-v or bSOA-v. So the number of actual model surrogates seems like it would only be 4*aSOA-v,gas + 4*aSOA-v,p + 4*bSOA-v,gas + 4*bSOA-v,p, and is not dependent of the number of VOC precursors (as implied by 4 surrogate SOA compounds per VOC).

We have followed the reviewer's suggestion and provided additional information to avoid confusion. A new figure has been added depicting the various surrogate compounds used and the corresponding chemical aging reactions. Based on the original work of Lane et al. (2008a), SOA from VOCs is represented using four volatility bins (1, 10, 10^2 , $10^3 \mu\text{g m}^{-3}$ at 298 K). As the reviewer suggests the model uses 4 surrogate compounds for SOA from anthropogenic VOCs (aSOA-v) and another 4 for SOA from biogenic VOCs (bSOA-v). These can exist in either the gas or particulate phase so there are two variables from each. There additional surrogate compounds for the oxidation products of anthropogenic IVOCs and SVOCs. PMCAMx-SR includes additional SOA surrogate compounds from biomass burning. We have followed the suggestion of the reviewer and clarified the SOA VBS-scheme in the revised manuscript.

(8) Lines 162-170: The description of chemical aging is also somewhat confusing. It might be clearer to refer to the volatility bin, rather than "vapors" and "semi-volatile SOA". Do the POA and SOA aging reactions both result in an increase in OA mass (line 170)? Is this independent of the mass increase associated with a shift to a semi-volatile bin? Does the OA mass increase apply to the biogenic SOA aging, even though no change in volatility is assumed?

We have rephrased these sentences to clarify that the aging reactions are for the material of each volatility bin that is in the gas phase. We clarify that all these aging reactions (both POA and SOA) are assumed to reduce the volatility of the reacted vapour by one order of magnitude which is linked to an increase in OA mass by approximately 8 percent to account for added oxygen. For the biogenic SOA aging is assumed to lead to no net change of volatility and OA mass. This is also clarified now.

(9) *To clarify the volatility distributions and aging, a figure such as 5-2 in the CAMx user's guide would be very helpful.*

This is a good idea. We have added a new figure to the paper (Figure 2) depicting the various OA components simulated and their chemical aging reactions.

(10) *Section 2.2: It is recommended that the authors consider the publication by Alvarado et al. (2015), which also evaluated volatility distributions of bb emissions. It may be beyond the scope of the manuscript to repeat the model runs using the Alvarado volatility distribution, but it would be useful to consider it in the introduction and discussion, and include it in the Figure 1 panels. The Alvarado et al. study also attempted to account for IVOC emissions not included in two published volatility distributions (including May et al.). Overall, there is significantly more mass (or higher fraction of bb-POA emissions) in the 10^5 and 10^6 bins in the subject manuscript (base case) than in Alvarado et al.*

We have followed the recommendation of the reviewer and added some discussion of the Alvarado et al. volatility distribution. We also added the results of the Alvarado study in the discussion of the role of IVOCs from biomass burning in SOA formation.

(11) *Also, while scaling the anthropogenic POA EF by 1.5, which gives a sum of fractions >1, has been well described in current literature, it is not clear that the same rationale applies to the biomass burning emissions used in this work. While the IVOC*

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bins are not constrained by data and thus absent in the published VBS distributions, this is not equivalent to missing mass in the bb-POA emissions totals. It seems that some scaling of the May et al. fractions may be needed to include the IVOC bins without giving a sum of fractions >1 (e.g., as done in Alvarado et al.). This probably needs a bit more discussion/clarification in the methods, as the mass attributed to the IVOC bins has a significant effect on predictions of bb-SOA (as demonstrated by the sensitivity case). Reference: Alvarado et al., ACP, 15: 6667-6668, doi:10.5194/acp-15-6667-2015.

This is often a confusing point. We have added discussion clarifying it. We do underline the difference with the Alvarado et al. emissions in the corresponding discussion. We also include the absolute emission rates for all bins in Table S1 to make sure that there is no confusion about the used input in our simulation.

Editorial Comments

(12) *In general, it is recommended that the authors check carefully for use of abbreviations. In many instances, an abbreviation is introduced but then not used consistently throughout the manuscript (e.g., organic aerosol (OA) in section 2.1). In a few cases, an abbreviation is introduced but not defined (e.g., AMS line 100).*

We have reviewed all the abbreviations used in the manuscript and made the corresponding corrections.

(13) *Line 16: Oxidation products of the bbOA? Or of bb emissions? If the latter, sentence needs revision.*

We have revised this sentence. These are the oxidation products of biomass burning emissions.

(14) *Line 22: Suggest removing "same" before contribution. It is a little confusing as*

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written.

Corrected.

(15) Line 50: What does “their” refer to?

It refers to the organic compounds. We have rephrased this sentence.

(16) Lines 54 and 57: Suggest using “or” rather than “and”, to indicate OA can be primary or secondary and of anthropogenic or biogenic origin.

Done.

(17) Lines 217-232: The discussion about the emissions is a bit unorganized. Are the anthropogenic biomass burning emissions from a source other than GEMS or the Pan-European inventory? If not, recommend to add “including anthropogenic biomass burning emissions” (line 203 or 210). Line 218-219 is then not needed. It is also recommended to move line 217 to the previous paragraph in which the other emissions inventories are described (likely before the introduction of MEGAN).

We have made the corresponding corrections to organize better the discussion about the emissions used in this study.

(18) Italicize variables in equations.

Done.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-1166>, 2018.