

Comment on “Secondary organic aerosol formation from smoldering and flaming combustion of biomass: a box model parametrization based on volatility basis set” by Giulia Stefenelli et al. (2019)

Summary/recommendation:

This paper is an interesting study on secondary organic aerosol formation from biomass burning. The authors conducted 14 experiments under two burning conditions (flaming, smoldering and flaming) and with different types of stove. Emissions from the burned stoves were sampled and were aged via OH-oxidation reactions to investigate the secondary organic aerosol yields and the chemical properties of the oxidation products from smoldering and flaming. A box model and a genetic algorithm approach were used to quantify the contribution of the VOC oxidation products to SOA yield and to better explain the SOA formation process.

However, many portions of the paper were made difficult to follow due to missing details. I recommend that this study be published but with minor revisions. I request that the authors consider the following points as they revise this manuscript:

General comments:

1/ As the title indicates, this paper should focus mainly on SOA formation from smoldering and flaming combustion of biomass. However, few information is given in that issue. The authors should better describe the burning conditions of smoldering and flaming by adding more details especially on how the authors reproduce experimentally the flaming and smoldering combustion.

2/ According to Majdi et al. (2019), Koo et al. (2014), Konovalov et al. (2015) and Ciarelli et al. (2017), Intermediate and Semi Volatile Organic Compounds (I-SVOCs) are considered as one of the most important SOA precursors emitted by biomass burning. Why did the authors focus only on SOA from VOCs ?

Specific comments:

1/ Page 4, lines 31-32: Why did the authors choose these experiments? How could the authors study the effect of smoldering/flaming combustion of biomass when the other experimental parameters (OH exposure, temperature of the smog chamber, stove....) change at the same time ?

2/ Page 5, line 27: Flaming combustion occurs at high temperature. Can the authors give an order of magnitude of this high temperature ?

3/ Page 5, line 31: Why did the authors choose to combine flaming and smoldering emissions in set 1?

4/ Page 7, line 7: What did the authors mean by “other processes”?

5/ Page 9, line 6: Can the authors give more information about this common set of 263 ions used to identify the most important SOA precursors?

6/ Page 11, line 22-23: Why did the authors choose to set the number of volatility bins to 6 ? How was the volatility C^* determined ? Did the authors measure the volatility of each species ?

7/ Page 11: More clarification is needed in section 3.3.2. Did the given chemical properties of surrogates represent the average of the compounds that are classified in a determined volatility bin ?

8/ Page 12, line 11: How did the authors assume this single ΔH value for all surrogates? Did the authors consider any assumptions to determine this value?

9/ Page 13, section 3.3.4 is very short. Can the authors clarify how did this algorithm work to identify the optimized parameters and what did the authors mean by the process of “natural selection”?

10/ Page 13, Figure 2: The 'others' VOC emissions show the highest relative contribution mainly in set 1. What are these 'others' VOCs emissions?

11/ Page 14, line 11: “Relevant compounds in the latter class are benzene, toluene and xylene.” According to Bruns et al. (2016) experimental SOA yields, benzene is the third principal contributor to SOA after phenol and naphthalene. Did the authors characterize these SAH compounds?

12/ Page 14, line 20: “The SOA/POA ranges between 2 and 6, similar to ratios observed in previous studies.” Please add references.

13/ Page 14, lines 22-23: “Such inconstancy in behavior is either due to differences in the chemical composition within the same class.” How did the authors defined these chemical compositions? And how can these compositions change within the same class? Can this be explained by differences in the burned stoves? How can the authors compare SOA contribution from the different classes if the chemical composition can change? Please explain this further.

14/ Page 18, lines 17-18: “In general for both phases studied, higher contributions to SOA formation was found for cresol and phenol species and chemically similar compounds.” This conclusion is not well supported and not discussed in the paper. Please clarify or add a reference.

Technical comments:

1) Figure 7 caption: Please replace “the model tend to underestimate ... ”by “the model tends to underestimate...”.

2) Eq (1) page 7: Is there any missing bracket in the equation ? Please verify.

3) Figure 2 caption: Did Set1 represent only smoldering phase or smoldering and flaming as defined in page 5 line 31? Please correct.

References:

Bruns, E., El Haddad, I., Lowik, J., Kilic, D., Klein, F., Baltensperger, U., and Prévôt, A.: Identification of significant precursor gases of secondary organic aerosols from residential wood combustion, *Sci. Rep.*, <https://doi.org/10.1038/srep27881-2016>, 2016.

Ciarelli, G., El Haddad, I., Bruns, E., Aksoyoglu, S., Möhler, O., Baltensperger, U., and Prévôt, A. S. H.: Constraining a hybrid volatility basis-set model for aging of wood-burning emissions using smog chamber experiments: a box-model study based on the VBS scheme of the CAMx model

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Koo, B., Knipping, E., and Yarwood, G.: 1.5-Dimensional volatility basis set approach for modeling organic aerosol in CAMx and CMAQ, *Atmos. Environ.*, 95, 158-164, <https://doi.org/10.1016/j.atmosenv.2014.06.031>, 2014.

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