

Interactive comment on “Modeling the Formation and Composition of Secondary Organic Aerosol from Diesel Exhaust Using Parameterized and Semi-Explicit Chemistry and Thermodynamic Models” by Sailaja Eluri et al.

Anonymous Referee #2

Received and published: 8 February 2018

Eluri et al. modeled their OFR experiments with diesel engine exhausts by VBS and SOM. Their work is detailed. They also considered a number of sensitivity studies and meticulously articulated how the studies were performed. The authors obtained a lot of interesting results to the OFR community, including both those confirming previous observations (e.g., the importance of S/IVOC in SOA formation and the possible kinetic limitation for OA growth in OFR) and new ones (e.g., minor impacts of RTD and OH spatial heterogeneity on SOA formation). This study fits well in the scope of ACP and is by and large of good quality. The paper will be acceptable into ACP after the issues

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below are addressed.

Major issues:

- Peng and Jimenez (2017) have published a characterization of whether the chemistry in OFRs with high initial NO is similar or very different from that in the atmosphere. The results on Figure 4 of that paper can be used to approximately determine whether the cases studied here fall in the good / risky /bad regions and whether the chemistry is predominantly in a low or high NO regime. This is important information that should be included in the paper.

- High-NO_x parametrizations were used in this study, while Peng and Jimenez (2017) found that high-NO_x conditions are very unlikely to be realized in OFR by initial NO injection only. (This can be verified with the previous suggestion) Low-NO_x SOA yields are often significantly higher than high-NO_x ones and SOA formed under low-NO_x conditions tends to have higher O:C (the results on elemental ratio in this study might be an evidence for this). The paper acknowledges that OA is 65% higher in a sensitivity test when low-NO_x yields were used. The validity of the modeling in this study is thus questionable, and the agreement in e.g. Figure 1 appears to be for the wrong reasons. The authors should show their simulation results for experiments using low-NO_x parametrizations and correct their conclusions if needed.

Specific comments:

Line 209 and elsewhere: Ahlberg et al. (2017) did systematic experiments and VBS modeling to show that SOA growth in OFR is kinetically limited at low OA mass loading. This paper also needs to be cited for discussions on the kinetic limitation of SOA growth.

Line 292: The POA VBS has more bins than the SOA ones shown in Table 2. How are the extra POA bins treated? Do they participate in aging?

Line 360: The reason why $\alpha=0.1$ is assumed is not clear to me until I arrive at the

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end of Section 3.2. A brief explanation or a reference to Section 3.2 is needed.

Line 386: Why do curves in Panels a and b of Figure 1 look slightly different? E.g. the measurement point at highest age is located between the VBS and SOM predictions in Figure 1a while it overlaps with the VBS one in Figure 1b.

Line 388: Why does an overprediction of x3 occur at low ages? This should be discussed further.

Line 431: Palm et al. (2016) reported that a low value of the accommodation coefficient ($\ll 1$) was inconsistent with their ambient OFR SOA formation experiments. A more direct measurement of the accommodation coefficient of SOA (compared to the more indirect measurements on the publications cited here) has been recently published by Krechmer et al. (2017), with measured values in the range 0.5-1. Both of these results support the use of higher values for accommodation coefficient and thus support the conclusions here.

Line 443: In Figure 3b, Curves (iv) appear to be higher than Curves (ii). However, the condensational sink of (ii) is always that at the end of the growth while that of (iv) starts from 1 nm particles (identical number concentration). The average of the latter is obviously lower than that of the former and hence less OA will condense in the case of (iv) in theory. Why does Figure 3b show the opposite?

Line 564: Figure 6 is supposed to convey important information. But it is impossible for me to tell data points for similar carbon numbers, so that I cannot verify relevant claims made in Sections 3.4. I suggest using different markers/patterns instead of point size to distinguish different carbon numbers.

Technical corrections:

Table 1: the 3rd column from the right is OH exposure, not OH.

Table 1: the initial NO and NO₂ concentrations should be given here, given their importance.

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Tables 2 and 3: many references in these tables are not listed on the reference list.

Equation 3: a parameter, A_s , seems to be explained as “A3” in the text. Please maintain the consistency.

Line 330: although spelled out in a table caption, “DPF+DOC” still needs to be spelled out here.

Line 455: a fractional bias $< -100\%$ does not look appropriate, although I understand what the authors mean. I suggest comparing them in log scale or using ratio instead of bias.

Figure 7d: the colors of the dashed lines are not correct.

Figure 8a: the exponent of “10⁻¹” in the x-axis is covered.

References:

Ahlberg, E., et al: Secondary organic aerosol from VOC mixtures in an oxidation flow reactor, *Atmos. Environ.*, 161, 210–220, doi:10.1016/j.atmosenv.2017.05.005, 2017.

Krechmer, J. E. et al. Direct measurements of gas/particle partitioning and mass accommodation coefficients in environmental chambers. *Environ. Sci. Technol.*, 51, 11867–11875, doi:10.1021/acs.est.7b02144, 2017.

Palm, B. B., et al.: In situ secondary organic aerosol formation from ambient pine forest air using an oxidation flow reactor, *Atmos. Chem. Phys.*, 16, 2943-2970, <https://doi.org/10.5194/acp-16-2943-2016>, 2016.

Peng, Z., et al.: Non-OH chemistry in oxidation flow reactors for the study of atmospheric chemistry systematically examined by modeling, *Atmos. Chem. Phys.*, 16(7), 4283–4305, doi:10.5194/acp-16-4283-2016, 2016.

Peng, Z. and Jimenez, J. L.: Modeling of the chemistry in oxidation flow reactors with high initial NO, *Atmos. Chem. Phys.*, 17(19), 11991–12010, doi:10.5194/acp-

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