

Ahlberg et al. investigated the yield of secondary organic aerosol (SOA) generated from the photooxidation of an m-xylene/ $\alpha$ -pinene mixture in an oxidation flow reactor (OFR) as a function of seed particle surface area, type, and phase. At the operating conditions that were used, they found that the SOA yield increased by a factor of 2-3.5 as a function of increasing seed concentration from 0 to 3000  $\mu\text{m}^2 \text{cm}^{-3}$  (condensation sink = 0 to  $0.1 \text{ s}^{-1}$ ), with the highest increases observed in the presence of ammonium nitrate and wet ammonium sulfate seeds. The seed dependence on SOA yield was modeled using the “LVOC fate correction” model published by Palm et al., ACP, 2016, and hypothesized to be due to residence-time-limited condensation of oxidation products in the OFR. The effect of SOA elemental ratios (H:C and O:C) was also characterized as a function of seed particle concentration. Given the emergence of OFRs as a technique to characterize SOA formation, and the previously established dependence of SOA yields on available condensation sink, I would support publication of this manuscript in ACP after consideration of my comments below.

1. Somewhere in the manuscript, perhaps in conclusions, the authors should add a paragraph to discuss recommendations for (i) seed particle composition and (ii) seed particle surface area concentration for SOA yield measurements in OFR studies. For example:
  - a. Should ammonium nitrate seeds be used because ammonium nitrate is less humidity-dependent than ammonium sulfate? Are there deficiencies with using either AN or AS that could be addressed with other seed types?
  - b. It seems that  $CS > 0.06 - 0.07 \text{ s}^{-1}$  is required at the conditions that were used in these studies (e.g. PAM, 160 sec residence time). It would be useful to model or parameterize the CS that is required, as a function of residence time, in graphical or equation form.
  - c. The work would have greater impact if the result can be generalized to other OFR designs that might for example have different surface-to-volume ratio than the PAM reactor and consequently different CS required to promote condensation.
2. I think it would be better to plot Figure 2 later in the paper so that the fractional LVOC loss pathways for the data points shown in Fig. 3 can be added to Figure 2. Additionally, the justification for assuming an OH reaction rate of  $1\text{E-}11 \text{ cm}^3/\text{molec}/\text{sec}$  is never provided; thus, the associated level of uncertainty in the modeling is unclear. At the very least the model should be run over a wider range of OH rate constants—perhaps a factor of 2 or 5 in either direction relative to  $1\text{E-}11$ —to evaluate the sensitivity of the model result to the specified kOH.
3. I prefer that the authors not refer to their operating conditions as “atmospherically relevant.” It’s already implied that atmospherically relevant conditions were used, but the reader can decide the atmospheric relevance themselves in the context of specific atmospheric conditions or source regions. In the instances below, for example, it may not be generally true that  $\text{RH} = 60\%$ ,  $\text{ORG} = 5 \text{ ug}/\text{m}^3$  are always “atmospherically relevant” conditions:
  - a. “using an oxidation flow reactor operated at an **atmospherically relevant RH of 60 %**” (P1, L12-L13)
  - b. “A relatively low, and **atmospherically relevant, SOA mass concentration of  $\sim 5 \mu\text{g m}^{-3}$** ” (P3, L27)

- c. “liquid-liquid phase separation rarely occur at O:C ratios higher than 0.7 in systems containing **atmospherically relevant** organics” (P8, L9-L10)
4. Introduction, and elsewhere as appropriate: Jathar et al., ES&T, 2017 and Zhao et al., ES&T, 2018 should be cited and discussed as their work also investigated SOA yield dependence as a function of particle concentration / condensation sink.
5. P1, L18: “low volatile” → “low volatility”
6. P2, L24: I suggest adding “the yields of **SOA generated from m-xylene...**” or similar.
7. P3, L28: Please show a representative subset of SMPS & AMS size distributions of the seed/SOA mixtures as a function of seed concentration. How much of the SOA condenses on the seeds and how much homogeneously nucleates to form new particles? This is not clear at present. The size distributions could be shown in the supplement.
8. P5, L6: The authors state: “Before experiments, the reactor was run without seeds or VOCs until the volume concentration was below  $0.2 \mu\text{m}^3 \text{cm}^{-3}$ ”. Please clarify whether the lights were turned on for generating OH/O<sub>3</sub> during these periods?
9. P6, L5: What is the mass-/volume-weighted mobility diameter? (possibly the more critical value in assessing how much of the mass is not transmitted through the AMS inlet lens?)
10. P7, L1-2: This statement is not clear: “which is similar the residence time of the reactor shortcircuit”
11. P8, L9: problem with citation formatting.
12. P8, L20-L24: The authors state: “*The O:C value increases with increasing SOA mass concentration (and seed particle concentration since these are connected), which is opposite to what is expected since more oxidized molecules tend to be less volatile (Shilling et al. 2009).[...] Increasing the seed particle number concentration also decreases the SOA mass fraction. It follows that SOA mass then is spread out on more particles, leaving less organics per particle, which could enhance the partitioning of more volatile material to the gas phase. This would leave more time for gas phase oxidation, and consequently a higher O:C ratio, provided the molecules partition to the particles.*” There are a lot of nuances to this discussion. At the least, it is unclear whether the logic that is presented here is fully supported by the measurements. It also seems self-contradictory-for example, how can higher seed particle concentrations promote condensation of oxidized organic vapors (the main result of the paper) while simultaneously “[enhancing] the partitioning of more volatile material to the gas phase”? These statements need to be reevaluated and modified in order to provide a more cohesive interpretation of the data.

13. Figure 3-4 and 6 should indicate the SOA precursors that were used (α-Pinene + m-Xylene), preferably in the figure axis labels or captions or both.
14. Figure 4: what are the propagated uncertainties in yield ratios, condensation sink and in LVOC fate model outputs?

### **References**

S. H. Jathar, B. Friedman, A. A. Galang, M. F. Link, P. Brophy, J. Volckens, S. Eluri, and D. K. Farmer. Linking Load, Fuel and Emission Controls to Photochemical Production of Secondary Organic Aerosol from a Diesel Engine. *Environ Sci. Technol.*, 51(3), 1377-1386, 2017.

Y. Zhao, A. T. Lambe, R. Saleh, G. Saliba, and A. L. Robinson. Secondary Organic Aerosol Production from Gasoline Vehicle Exhaust: Effects of Engine Technology, Cold Start, and Emissions Certification Standard. *Environ. Sci. Technol.*, 52 (3), 1253–1261, DOI: 10.1021/acs.est.7b05045, 2018.