

Simonen et al. present experimental evaluation of the new Tampere Secondary Aerosol Reactor (TSAR). Performance metrics of the TSAR are evaluated against the Potential Aerosol Mass (PAM) oxidation flow reactor: residence time distributions, particle losses, OH exposure, sulfuric acid and toluene SOA yields, and O/C and H/C of toluene SOA. The authors additionally demonstrate the application of the TSAR to resolve changes in the SOA formation potential of car exhaust during dynamometer measurements of a vehicle subjected to a transient driving cycle. The TSAR has potential applications for secondary aerosol formation potential studies of emissions sources. Overall, I was impressed. I would support publication in Atmospheric Measurement Techniques after consideration of my comments below.

1. The authors present evidence that the shorter residence time, higher particle transmission efficiency, and narrower residence time distributions of the TSAR relative to the PAM reactor provide several advantages, especially for transient drive cycle measurements and similar applications. However, in my opinion the short residence time (37 sec) also presents important limitations that are not adequately discussed in the current manuscript. In the absence of significant seed particle surface area, such a short residence time likely limits the condensation of oxidized organic species, to the point that SOA yields that are reported from the TSAR in principle represent lower limits compared SOA yields reported from oxidation flow reactors with longer residence times and/or smog chambers.

For example, the accommodation coefficient (α) has been shown to be approximately 0.1 for some oxidized organic vapors that condense to form SOA (e.g. Saleh et al., 2013). To the extent that $\alpha = 0.1$ is representative of other SOA types, this suggests that the relevant timescales for condensation of oxidized organic vapors ranges from minutes to hours (e.g. Lambe et al., 2015; Seinfeld and Pandis, 1998). This is likely less of an issue in the TSAR for species such as sulfuric acid ($\alpha \sim 1$), which may be why the authors obtain mass closure with sulfuric acid. The following results presented by Simonen et al. further support the hypothesis that condensation of oxidized organic vapors is limited by the short residence time:

- a. Particle size distributions of toluene SOA generated in TSAR and PAM reactor at comparable OH exposure (Fig. 7) show that the mode diameter of SOA particles generated in the TSAR is much smaller than in the PAM reactor (40 nm versus 100 nm).
- b. The maximum toluene SOA yield value obtained in the PAM reactor would increase from ~ 0.20 to ~ 0.33 (Fig. 8) if size-dependent particle loss corrections are applied (from statement on Page 14, Line 17, I assume the yields are not currently corrected for losses). This correction is obtained by applying a measured transmission efficiency ~ 0.6 for 100 nm dioctyl sebacate particles through the PAM reactor (Fig. 2; Lambe et al., 2011). The yield values obtained at other OH exposures would presumably also increase significantly, indicating that toluene SOA yields in the PAM reactor were $\sim 40\%$ or higher than toluene SOA yields measured in the TSAR. Importantly, the mean residence time of the PAM reactor operated in these studies was also significantly higher (~ 160 sec, as calculated from internal volume of 13.3 L and flow rate 5 LPM).

In my opinion, the authors should add a paragraph near the beginning of the paper that states this up front, and discusses the potential implications of a very short residence time – perhaps at the end of Section 2.1, and elsewhere in the manuscript as appropriate. I think it is also worth

attempting to constrain the magnitude of residence-time-limited condensational growth, so that reported TSAR SOA yields obtained with a 37 sec residence time can be corrected (with some uncertainty) to SOA yields that would be obtained with longer condensation times. For example, the authors might consider implementing the “LVOC fate correction” model described in Palm et al. (2016) or something similar to estimate the magnitude of condensable vapors that exit the TSAR before condensing elsewhere.

2. **Page 3, Section 2.1:** It may be useful to briefly state why the authors choose to operate the TSAR as an “OFR254” instead of an “OFR185”. I assume that “OFR254” is more practical/safer because the lamps are external to the reactor, and the 185 nm light is not transmitted through the quartz walls of the reactor? Are there additional reasons? Is the UV intensity inside the TSAR varied at all, or is the OH exposure varied by changing the O₃ and/or H₂O concentrations?
3. **Page 5, Lines 9-10:** The authors state: “the only significant loss of SO₂ in the oxidation reactor is due to the reaction with OH radicals.” Was this assumed, or verified experimentally? Was the transmission efficiency of SO₂ measured in the absence of OH? Please clarify.
4. **Page 6, Line 4:** The authors state: “injecting pressurized air, SO₂, humidified air and ozone into the TSAR”. This is a confusing – isn’t the pressurized air also humidified? Figure 1 suggests that some fraction of the carrier gas is sent through a humidifier, another fraction is sent through the ozone generator, and both are mixed prior to entering the residence time chamber.
5. **Page 6, Line 27:** Consider deleting “which represent atmospheric oxidation” -- in my opinion there are open questions as to how representative all laboratory simulations of atmospheric oxidation chemistry are (flow reactors and smog chambers).
6. **Page 7, Line 9:** Please explain why the PAM reactor was run in “OFR185” mode and not also “OFR254” mode to more closely represent the TSAR operating conditions.
7. **Page 8, Line 2:** Consider replacing “theoretical residence time” with “calculated mean residence time” or “mean plug-flow residence time”, or something similar.
8. **Page 10, Lines 5-9:** The added convection caused by UV lamp-induced temperature gradients can be modeled by Taylor dispersion (e.g. Lambe et al., 2011; Huang and Coggon et al., 2016). I suggest applying a Taylor dispersion model to see if this more closely represents the measured residence time distributions that are presented in Figure 3.
9. **Page 11, Line 9:** I did not notice where the output of the UV lamps is discussed. Please report the UV actinic flux that is currently used in the TSAR. This is needed to get at least a qualitative sense of the relative importance of photolysis versus OH oxidation pathways for selected species (e.g. Peng et al., 2016). It should be possible to constrain the UV actinic flux from the measured OH exposures and input ozone and water vapor mixing ratios using the model described in Li et al. (2015) or something similar.

10. **Page 11, Section 3.4:** The authors might consider combining some of the text from Section 2.5 with Section 3.4, i.e. Page 5 Lines 25-29, Page 6 Lines 1-2 and Lines 10-20 to consolidate the discussion of sulfuric acid yield measurements.
11. **Page 11, Section 3.5:** The authors might consider combining some of the text from Section 2.6 (Page 6, Lines 22-30) with Section 3.5.
12. **Page 12, Lines 5-7:** This argument is speculative and not supported by the measurements. If it were true that “the PAM always outputs SOA with both high OH_{exp} and low OH_{exp} , [thereby reducing] the sensitivity to OH_{exp} ”, I would argue that the range of O/C and H/C of PAM-generated toluene SOA should also be narrower than in the TSAR. Figures 9 and 10 show that this is not the case – the range of O/C and H/C is similar in both reactors. I would consider deleting this statement. A more plausible argument, in my opinion, is that the residence-time-limited condensation of oxidized vapors in the TSAR (mean residence time = 37 sec) compared to the PAM reactor (mean residence time ~ 160 sec) results in higher sensitivity to changes in yields of condensable species at the lowest and highest OH exposures that were studied.
13. **Page 12, Line 29 – Page 13, Line 5:** Here, it may be useful to know the UV actinic flux in both reactors (see Comment #9). For example, at $\text{OH}_{\text{exp}} \sim 1.3 \times 10^{12} \text{ molec cm}^{-3} \text{ s}$, if “F254/ OH_{exp} ” (as defined by Peng et al., 2016) is significantly different in the TSAR and in the PAM reactor, one might hypothesize that direct UV photolysis of the SOA is more important in one system than the other, and perhaps this is correlated with the observation that $\text{OSc} \sim 1.1$ in the TSAR and $\text{OSc} \sim 0.6$ in the PAM reactor (Fig. 10). It is hard to tell without knowledge of the actinic fluxes in both reactors. It may also be useful to add error bars to represent the approximate uncertainties in OH exposure in both systems.
14. **Page 9, Section 2.7, Lines 4-6 and Pages 15-16, Section 3.6.2:** In Section 2.7, the authors state: “the amount of secondary aerosol mass produced in the TSAR was determined by subtracting the primary mass from the mass measured when using the TSAR.” The absolute secondary-to-primary aerosol mass enhancement is actually never discussed. I think it would be useful to show time series of the primary mass concentration and the secondary-to-primary emission ratio in Figure 13 (perhaps as additional subpanels), discuss in Section 3.6.2, and briefly compare with results from relevant literature studies such as Platt et al. (2013), Tkacik et al. (2014) and Karjalainen et al. (2016).

References

- R. Saleh, N. M. Donahue, and Robinson, A. L.: Time scales for gas-particle partitioning equilibration of secondary organic aerosol formed from α -pinene ozonolysis, *Environ. Sci. Technol.*, 47, 5588–5594, 2013.
- B.B. Palm, P. Campuzano-Jost, A.M. Ortega, D.A. Day, L. Kaser, W. Jud, T. Karl, A. Hansel, J.F. Hunter, E.S. Cross, J.H. Kroll, A. Turnipseed, Z. Peng, W.H. Brune, and J.L. Jimenez. In situ secondary organic aerosol formation from ambient pine forest air using an oxidation flow reactor. *Atmospheric Chemistry and*

Physics, Atmos. Chem. Phys., 16, 2943-2970, doi:10.5194/acp-16-2943-2016, 2016. Source code available for download at <https://sites.google.com/site/pamwiki/hardware/estimation-equations>.

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