Point-by-point response to reviewer comments

Manuscript ID: egusphere-2024-3849
Title: "Secondary Organic Aerosol Formation from Nitrate Radical Oxidation of Styrene: Aerosol Yields, Chemical Composition, and Hydrolysis of Organic Nitrates"
Author(s): Wang, Yuchen; Xiang Zhang; Yuanlong Huang; Yutong Liang; Nga L. Ng

We thank the editor for the thoughtful comments. Each of the comments has been addressed and detailed in our point-by-point response below. The exact comment from the editor is in black and italic style while our response is in blue and normal format in this document. The revised text as it appears in the revised manuscript is in green.

Comment

1. Did you consider a potential bias of sulfate mass concentrations from AMS measurements due to coating of the seed particles and corresponding change in the collection efficiency? How could this affect your analysis?

Response

According to previous work by Bahreini et al. (2005, DOI: 10.1021/es048061a), the sulfate signal in HR-ToF-AMS increased following SOA formation, indicating improved collection efficiency (CE) of organic-coated seed particles. However, in this work, the SOA yields are calculated from the SMPS data. We did not calculate collection efficiency (CE) as we did not need to use this value in our analysis.

2. Figure 3a: Please add the relative abundance of monomeric and dimeric products (or CHO, CHON, CHON2) as a function of initial styrene concentrations and discuss the result in the context of typical atmospheric styrene concentrations.

Response

We added a new figure (Figure S4) to show the relative percentage contributions of CHO, CHON, $CHON_2$ as well as dimers and monomers. We also added the following to the main text in lines 216-218: "The relative percentage contributions of CHO, CHON, $CHON_2$, as well as dimers and monomers, remained relatively stable across various experiments with differing initial styrene concentrations (Figure S4)." We also mention in lines 435-438: "As the fractions of monomers dimers are relatively constant in experiments spanning a wide range of styrene concentrations, this may suggest that dimeric ONs are also important products when NO_3 reacts with typical atmospheric levels of styrene."

3. Figure 5a: Explain why the hydrolysis lifetimes are: 18.8 ±1.9 min at 70% RH (Shorter lifetime despite slower decay rate?) and 29.5 ±8.7 min at 50% RH (Longer lifetime despite faster decay rate?) although Figure 5a shows a faster decay of the pON/OA ratio at 50% RH compared to 70% RH?

Response

Since no sudden, drastic change in pON/OA was observed except for a few initial data points (Figure R1), the rate of pON hydrolysis may be fast enough such that the decay trend of pON compared to OA is not visibly manifested, as we mentioned in our previous hydrolysis paper (Takeuchi and Ng, 2019, DOI: /10.5194/acp-19-12749-2019). To avoid misunderstanding, we revised the sentence in lines 266-272: "As illustrated in Figure 5a, the time series of $_pON/OA$

stabilizes fairly quickly, similar to what we have observed previously for monoterpene systems (Takeuchi and Ng, 2019). If we utilize pseudo first-order rate equations to assess the hydrolysis lifetimes at 70% RH and 50% RH, the corresponding hydrolysis lifetimes are 18.8 ± 1.9 minutes and 29.5 ± 8.7 minutes for 70% RH and 50 % RH, respectively. Considering that a drastic change in the pON/OA ratio is not observed except for a few initial data points, the rate of pON hydrolysis may be fast enough that the decay trend of pON relative to OA is not visibly apparent. Therefore, we report the hydrolysis lifetime to be less than 30 minutes (Takeuchi and Ng, 2019)."
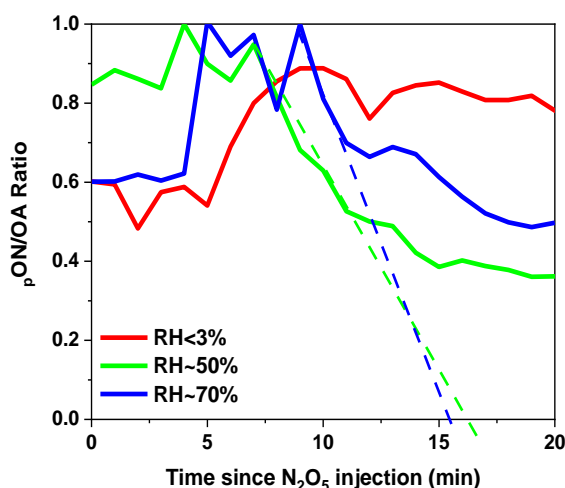

Figure R1. Time series data of the ratio of particulate ONs ($_p$ON) to total organic aerosols (OA) in Exp. 2 (RH<3%), Exp. 11 (RH~50%), and Exp. 12 (RH~70%) in first 20 minutes.

4. Figure S6: Consider changing the organic mass axis (e.g. log scale) to make the atmospheric relevant range better visible and discuss a potential extrapolation of your results to this range. Indicate how the lines given are calculated also in the caption.

Response

Thanks for the suggestion. We have added a new panel with log scale to Figure S6 (now Figure S7 in the latest version). As seen in the figure, the organic mass concentrations in our experiments (covering up to highly polluted areas) are much lower than those used in prior studies. We have revised the figure caption: "Comparison of SOA yields of styrene oxidation systems in this work and in literature (Yu et al., 2022; Tajuelo et al., 2019; Ma et al., 2018; Na et al., 2006; Schueneman et al., 2024) (a) on a linear scale and (b) on a logarithmic scale. The SOA yields and ΔM_O in Schueneman et al., (2024) and in Tajuelo et al., (2019) are extracted by WebPlotDigitizer. The higher end of the ΔM_O range in this study corresponds to highly polluted environments. The lines are SOA yield curves obtained by fitting the yield data to the Odum two-product model (Odum et al., 1996, 1997): $Y = \Delta M_O [\frac{\alpha_1 K_1}{1+K_1 M_O} + \frac{\alpha_2 K_2}{1+K_2 M_O}]$, with coefficients either taken directly from the published papers (Tajuelo et al., 2019; Na et al., 2006) or determined by ourselves using the published yield data points (Yu et al., 2022; Ma et al., 2018; Schueneman et al., 2024)."