

Point-by-point responses to Reviewer 3

1. Line 193-196. Detecting products at their parent-ion peaks cannot be considered as evidence for minimal fragmentation and thermal decomposition, because the parent-ion peaks certainly should be detected. The key evidence should be whether fragment-ion peaks were also detected, and if yes, how big they were, in comparison to the parent-ion peaks.

The plausible fragment-ion peaks for oligomers and organic acids are the monomeric ion-peaks and $[M-17]^+/[M-45]^+$ from α -cleavage, respectively. According to our detected particle-phase products (Fig. 2C), non-fragmental oligomers/NCO and organic acids represented the dominant forms, while negligible monomer (e.g., $m/z = 73$) and fragments for organic acids (e.g., $[M-17]$ or $[M-45]$) were detected, confirming minimal fragmentation by TD-ID-CIMS measurement. The above pointed has been elaborated in revision on pp. 9-10.

2. Regarding my prior comment #2, I think the authors misinterpret my point. I did not mean that the "equilibrium gas-particle partitioning" is the correct modeling approach for this work, but I do think volatility certainly needs to be considered. The authors said in their response that "scientific research shall be judged solely based on the available evidence, but not on a prevailing view of one's perception". There has been a great amount of evidence showing that volatility plays a big role in gas-particle partitioning (unnecessarily in rapid equilibrium). These evidence should not be neglected. The authors pointed out some "overwhelming" evidence to argue that volatility is not important in their system. For example, the significant increase in particle size and constantly varying particle properties. But these only suggest that m-xylene+OH has a high SOA yield. The continuous particle growth during the two hours may be due to that m-xylene was continuously consumed during the two hours (Figure 1a). None of these can be used as direct evidence against volatility. A most direct evidence should be whether the volatile COOs were largely present in the particle phase in their own form. If yes, then the authors are correct, if not, then the volatility is correct. Such a piece of evidence has been shown in the manuscript to suggest the opposite view. A major relatively volatile product, methylglyoxal (m/z 73), was observed in large amount in the gas phase, but not in the particle phase (Figure 2c). The authors' conceptual misunderstanding is that they seems to think volatility has to link with equilibrium partitioning. But they do not mean the same process. Volatility matters, but this does not mean the partitioning has to be in equilibrium or fast equilibrium.

Indeed, there is confusion concerning the aspects of equilibrium partitioning and volatility leading to SOA formation. We agree with the reviewer that SOA growth is ultimately dependent on the volatility of the aerosol-phase products. Evidentially, there are two pathways to form low-volatile compounds, i.e., gas-phase oxidation to yield non-volatile COOs and multiphase reactions of volatile COOs to form non-volatile products in the aerosol-phase. In our study, the gas-to-particle conversion from m-xylene oxidation occurs dominantly via reactive uptake for dicarbonyls and organic acids, with a relatively small contribution from condensation by low-volatile PAQs and nitrophenols. The above points have been incorporated into the text on p. 22.

Finally, the authors need to go one step further and understand the kinetic processes. Part of the reason that the authors observed the continuous uptake and appear to be able to model the behavior using an effective uptake process, is not because gas-particle partitioning does not happen, but more likely because the uptaken dicarbonyls forms non-volatile oligomers in shorter timescales than evaporation. These processes should be thoroughly discussed in the manuscript,

rather than just saying that they have evidence that gas-particle partitioning does not matter in this system.

The overall gas uptake onto liquid or solid particles is commonly described by an empirical framework on the basis of an effective uptake coefficient (γ), which accounts for the gas-phase diffusion, the interfacial process (i.e., accommodation), and the aerosol-phase processes. The aerosol-phase processes include thermodynamically driven surface or volume-limited partitioning (including condensation for low-volatile species or solubility for volatile species) and chemically driven reactivity relevant to the functionality (Gomez et al., 2015). In our study, the gas-to-particle conversion from m-xylene oxidation occurs dominantly via reactive uptake for di-carbonyls and organic acids, with a relatively small contribution from condensation by low-volatile PAQs and nitrophenols. The above points have been incorporated on p. 22.