

General Comments

Zhao et al. presents a mathematical framework to link particle growth rate with OH reactivity of total organics from three monoterpene (i.e. alpha-pinene, beta-pinene, limonene) systems. They compare the OH-initiated oxidation of these compounds to ozonolysis (using CO as OH scavenger) experiments and find comparable efficiency in particle growth. The paper is generally well-written and clear (great motivation in the introduction) and the approach in methodology (low organic aerosol loadings, low VOC and NO_x concentrations) for chamber experiments is commended, but the discussion of results did not highlight new results in context of the initial motivation and atmospheric relevance. The derivation of the “novel method which quantitatively links particle growth to the reaction of OH with organics” seems to be overly simplified (comments below). While attempts are made to chemically describe the SOA formation and resulting particle growth observed in the experiments, I find treatment of the chemistry in general to be cursory and/or mostly speculation. In particular, attempts are made to explain observations of particle growth in terms of functionalization and fragmentation processes using elemental analyses (H/C, O/C) using an aerosol mass spectrometer (AMS), though more defensible chemical arguments should be made by making a better effort to explain results in the context of chemical mechanisms (examining expected oxidation products from these systems that have been studied and characterized in numerous previous studies). This paper could merit publication after addressing the following comments.

Specific Comments:

- 1) In general, loose terminology that needs to be defined
 - a. p. 12592, line 7: Define “low Ozone”
 - b. p. 12592, line 15: Define “ambient relevant conditions”
 - c. p. 12595, line 16: Define “low O₃ concentration” as relevant to atmospheric conditions
 - d. p. 12595-6, lines 29-1: Define “ambient relevant HO₂/RO₂ ratios”
- 2) In general, use of reactivity, reaction rate with OH, k_{OH} , R_{OH} , etc. is confusing.
 - a. p. 12592, line 9: “...overall reaction rates of organic compounds with OH were quantified”

This gives a false sense that reaction rates with OH were determined for multiple organics in each system. Clarify up front here that you have lump summed total OH reactivity of all organic in each experiment for a monoterpene system.
 - b. p. 12592, line 16: “...the reaction of OH with organics in a reaction system”

This is more precise, but still innately confusing
 - c. p. 12595, lines 20-21: “Direct derivation of the overall reaction rate of organics with OH (product of OH reactivity from organics and the OH concentration)...”

Now you introduce “reaction rate” and “reactivity”. How does this definition differ with what is presented in Fig. 3 legend $k_{OH}(\text{Org})$ vs. what you define here (interpreted as $k_{OH}[\text{OH}]$). See Comment 2e.
 - d. p. 12600, line 10: “...reaction rate of OH with organics for the first time”
 - e. p. 12605, line 10: “...reaction rate of OH of all organics”

Now you state this for “all organics” – maybe just keep wording to be “total organics” to be consistent

- f. p. 12605, line 17: k_{OH} is for all organics in the system, right? This is confusing compared to general use of k_{OH} referenced as rate constant for individual species. Should state this to make the distinction early on.
 - g. Fig. 3: k_{OH} generally referenced as rate constant rather than reaction rate and legend entry $k_{OH}(\text{Org})$ is inconsistent with axis label as k_{OH} even though units are correct. Be clearer with notation.
- 3) p. 12597, line 1: With regard to dilution of the chamber over experiment duration, authors claim that dilution “applies equally to suspended particles and gases,” but what about dilution source affecting gas-particle equilibrium?
 - 4) p. 12598, line 6: How would considering a density change during course of the experiment affect interpretation of your results?
 - 5) p. 12599, line 7: What is the LDL of the NO_x box used for these experiments?
 - 6) P. 12599, line 23: “...oxidation products are generated, which condense on the particle phase resulting in particle growth.” Are the initial condensing surfaces on pre-existing particles what remains in the chamber between cleanings? No seed particles added, or do you attribute initial particle growth to nucleation?
 - 7) p. 12600, line 1: What do you mean here “...OH also reacts with saturated compounds”? Are you assuming that the first generation products are saturated compounds?
 - 8) Mathematical derivation
 - a. p. 12601, line 7: For conservation of C_i^p , does this not assume that $i+$ is a resulting product with sufficiently low vapor pressure to stay in the particle phase?
 - b. p. 12602, line 15: How do you reconcile use of an average molecular weight, M , when there is fragmentation as well?
 - c. p. 12604, line 15: What potential implications are there considering individual saturation vapor pressures of multi-generational species rather than an average saturation vapor pressure which allows simplification and derivation to equations 22/23?
 - d. How are the following factors accounted for in this derivation: 1) evaporation, 2) dilution, 3) vapor and particle-phase wall loss, 4) as well as particle-phase photolysis?
 - 9) Chemistry
 - a. p. 12606, line 13: Does the PTR-MS or use of GC-MS allow for identification of multi-generation products that you can monitor to support this claim?
 - b. p. 12607, line 10: How do you support chemically the claim that low-volatility compounds (functionalization) were actually generated here? How do you know their vapor pressure? What particles are available for condensation of these apparent low-volatility products? (see comment 6)
 - c. p. 12607, line 19: Do you mean “...saturated vapor pressures [increased]” instead?
 - d. P. 12607, lines 23-25: Additional explanation should be given for why alpha-pinene continues to grow and beta-pinene plateaus as shown in Fig. 2.
 - e. Fig. 2A: What do you attribute the drop in particle diameter at the beginning of the alpha-pinene experiment when text suggests that beginning is a period of functionalization/particle growth?
 - f. p. 12608, lines 17-19: It does not seem like $GE_{OH}(t)$ drops dramatically when O/C reaches 0.4 for beta-pinene system. The drop after this point is not really any different than the drop prior to reaching this O/C level.

- g. p. 12609, lines 8-13: This is the only species specific vapor pressure validation done. Can you do this for other systems like alpha-pinene as well?
- h. p. 12613, lines 19-21: Can this statement about carbonyl incorporation during oxidation be backed up mechanistically to support the calculated elemental ratios?
- i. p. 12613, lines 26-29: Are you implying that after O₃ reacts with monoterpenes, there is no additional oxidation because the products are non-reactive with O₃? If this presumably also means products that are more saturated as in comment 7), should H/C not be higher?
- j. p. 12614, lines 9-11: I do not think you can claim to know functionalization vs. fragmentation "...through the analysis of the evolution of particle size and particle mass." They may be correlated, but changes in particle size and particle mass can be affected by dilution, evaporation, condensation, particle photolysis, and other processes not related to particle-phase reactions leading to functionalization/fragmentation.
- k. In the conclusion I would like to see additional comment on placing the results described in p. 12613, lines 26-29 along with the result of comparable particle growth efficiency between OH-oxidation and ozonolysis described in the context of atmospheric relevance. Understandably, OH scavenger in ozonolysis experiments was used to isolate oxidation by O₃, but in the atmosphere, with both O₃ (and secondary OH formation) and OH, how do you synthesize the experimental observations? What if you did not put in an OH scavenger during an ozonolysis experiment so you can see the effective growth due to ozonolysis with OH?
- l. Where do we need to go further if overall reaction rate of total organics with OH is an important parameter towards getting effective growth rate of particles—can this be applied to the atmosphere?

10) Figures

- a. Figs 1-5: I find the x-axes on these figures to be inconveniently inconsistent. Can both metrics of time/reaction time be shown on figures (e.g. OH dose and HC consumed) so different events can be compared across all timelines presented in these figures? (e.g. does the Louvre closing coincide with the change in rate of aerosol concentration and diameter in Fig. 2A?)
- b. Fig. 1: The "hook"/vertical portion in the time dependent growth curve can be interpreted as purely multi-generation products with the initial HC concentration completely consumed. It would be nice to see how this times with events in Figs. 2-5 when you know that particle evolution is due purely to multi-generation products.
- c. Fig. 2: Additional explanation for general shape of curves would be appreciated. Why in Fig. 2A are there so many changes in rate of aerosol growth? Why in Fig. 2C do the particle growth rate and mass growth rate seem parallel and never cross compared to the case of alpha- and beta-pinene?
- d. Fig. 3A: What is the explanation for all of the variation in $GE_{OH}(t)$ for the case of a-pinene experiment?

Technical Corrections:

- 1) p. 12593, line 17: Recommend changing "...thus on climate depends..." to, "...thus depends on..."
- 2) p. 12595, line 18: Change "...of whole the..." to "...of the whole..."

- 3) p. 12606, line 9: Delete “the” in “difference of VOC and the particle concentration.”
- 4) P. 12607, line 14: Change “promote” to “promoting.”
- 5) p. 12610, line 22-23: Use same notation for particle number concentrations reported.
- 6) Fig. 2 caption: Add “of” before “each monoterpene and change “in respect to” to “with respect to”