Response to Anonymous Referee #3

We thank the reviewer for the very careful review of our manuscript; your time, comments and suggestions are greatly appreciated. All the comments have been addressed and we believe they have improved our manuscript substantially. In the following please find the responses to your comments and the changes made to the manuscript.

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 NOx 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.

Response:

Specific Comments:
1) In general, loose terminology that needs to be defined
a. p. 12592, line 7: Define "low Ozone"

Response:

We have specified the range of the ozone concentration in the revised manuscript. Now it reads:

"…low ozone (O_3) concentration (<20 ppbV)."

b. p. 12592, line 15: Define "ambient relevant conditions"

Response:

We guess that the reviewer meant p.12595, line 15. We have specified the conditions in the revised manuscript.

Now it reads:

"In this study, we investigated the SOA formation and growth of several common monoterpenes, α-pinene, β-pinene and limonene, by OH oxidation at ambient relevant conditions (low NO_x (0.01-1 ppbV), low VOC (~ 4 ppbV) and low particle concentrations (sub $\mu g\ m^{-3}$ to several $\mu g\ m^{-3}$))."

c. p. 12595, line 16: Define "low O3 concentration" as relevant to atmospheric conditions

Response:

We have specified the O_3 concentration in the revised manuscript. Now it reads:

"…low O_3 concentration (<20 ppbV)…"

d. p. 12595-6, lines 29-1: Define "ambient relevant HO_2/RO_2 ratios"

Response:

We have elaborated this point in the revised manuscript by defining the ratios. Now it reads:

"Compared with other OH scavengers, mainly organics such as butanol, cyclohexane, etc., CO helps keep the RO_2/HO_2 concentration low since in the atmosphere HO_2 usually exceed or is close to RO_2 concentration (Hanke et al., 2002; Mihelcic et al., 2003), in contrast with many laboratory studies where RO_2 concentration is much higher than HO_2 concentration (Kroll and Seinfeld, 2008)."

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.

Response:

We have clarified this in the revised manuscript. Now it reads:

"OH concentration and total OH reactivity (k_{OH}) were measured directly, and through this the overall reaction rate of total organics with OH in each reaction system was quantified."

b. p. 12592, line 16: "…the reaction of OH with organics in a reaction system"

This is more precise, but still innately confusing

Response:

We have clarified this in the revised manuscript. Now it reads:

"…links particle growth to the reaction rate of OH with total organics in a reaction system."

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}(Org) vs. what you define here (interpreted as k_{OH}[OH]). See Comment 2e.

Response:

k_{OH}(Org) is the k_{OH} (OH reactivity) of total organics in Fig. 3. In the revised manuscript we have modified these to make it consistent. Now it reads:

"Direct derivation of the overall reaction rate of total organics with OH (product of OH reactivity of total organics and the OH concentration)…"

In the caption of Fig. 3, now it reads "…k_{OH}(Org) (OH reactivity of total organics)".

d. p. 12600, line 10: "…reaction rate of OH with organics for the first time"

Response:

We have changed this in the revised manuscript to keep it consistent. Now it reads:
"…reaction rate of OH with total 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

Response:

We have changed this in the revised manuscript to keep it consistent. Now it reads:
"…reaction rate of OH with total organics"

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.*

Response:

In our manuscript, we used k_{OH} only as OH reactivity, not as the rate constant for the reaction of individual species with OH. As the review suggested, we have added a brief statement to avoid confusion as follows.

"Note that k_{OH} denotes OH reactivity throughout this paper rather than the rate constant for the reaction of individual species with OH."

g. Fig. 3: k_{OH} *generally referenced as rate constant rather than reaction rate and legend entry* k_{OH}*(Org) is inconsistent with axis label as* k_{OH} *even though units are correct. Be clearer with notation.*

Response:

We have corrected the axis label as "k_{OH}(Org)" in the revised manuscript to make it consistent.

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?

Response:

The dilution affects the gas-particle equilibrium. Unfortunately due to the unknown identities, vapor pressure of the compounds and unknown amounts on the particle, it is not possible in this study to correct this effect. However, the compounds contributing to the particle growth here have very low vapor pressure (effective vapor pressure on the order of magnitude of $\mu g\ m^{-3}$), which makes the effect of dilution on the gas-particle equilibrium less significant.

In the revised manuscript, we have added a comment on this as follows (page 25, lines 20-25).

"In addition, the dilution may also affect particle mass concentration through altering the gas-particle equilibrium. Due to the unknown identities, vapor pressure of the compounds and unknown amounts on the particle, it is not possible in this study to correct this effect. However, the compounds contributing to the particle growth here have very low vapor pressure, which may make the effect of dilution on the gas-particle equilibrium less significant."

4) p. 12598, line 6: How would considering a density change during course of the experiment affect interpretation of your results?

Response:

We had the AMS data and from its PToF (particle time-of-flight) mode we could obtain the densities throughout the experiments. However, the low particle concentration in the study makes the density characterization highly uncertain. From our previous studies, the density was found to be relatively constant throughout the whole experiment (Salo et al., 2011; Saathoff et al., 2009). Therefore, we assume the density change has little effect during the course of the experiment. In the revised manuscript, we have added this explanation (page 7, lines 16-18).

5) p. 12599, line 7: What is the LDL of the NOx box used for these experiments?

Response:

For a time resolution of 90 s the detection limits of the NOx analyzer were 5 and 10 pptV and the accuracies 5% and 10% for NO and NO_2, respectively. In the revised manuscript, we have added this description.

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?

Response:

The initial particle growth was due to the nucleation although there were some pre-existing particles but with fairly low concentration before the experiment started (see Table 1).

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?

Response:

Here we mean that most of the first generation compounds are saturated for monoterpenes that have only one carbon-carbon double bond, and OH can still react with them, while for ozonolysis once the double bond reacts with ozone, the products do not react with ozone any more. In the revised manuscript, we have elaborated this (page 9 lines 13-16).

8) Mathematical derivation
a. p. 12601, line 7: For conservation of C_t^p*, does this not assume that i+ is a resulting product with sufficiently low vapor pressure to stay in the particle phase?*

Response:

We consider C_t^p to be constant in each time step since the change in each time step is minor compared to C_t^p.

We have modified this sentence in the revised manuscript. Now it reads:

"C_t^p is assumed to be constant in each time step because the change in each time step is minor compared to C_t^p, and additionally loss of i is compensated by gain in i+ when the vapor pressure of i+ is sufficiently low to be on the particle phase and thus C_t^p is approximately conserved."

b. p. 12602, line 15: How do you reconcile use of an average molecular weight, M, when there is fragmentation as well?

Response:

We assume that the molecular weight of i+ is similar with that of i, i.e., neither functionalization nor fragmentation change the molecular weight dramatically. In the case of fragmentation, it is possible that molecular weight changes significantly if the fragmentation happens in the middle of the molecule. In this case we keep the molecular weight of each species.

Eq. 14 becomes:

$$\frac{dm_t}{dt} = \sum_i R_{OH,i} m_t \left(\frac{\frac{M_{i+}}{M}}{C_{i+}^0} - \frac{\frac{M_i}{M}}{C_i^0}\right)$$

Eq. 17 becomes:

$$GE_{OH}(t,i) = \frac{\frac{M_{i+}}{M}}{C_{i+}^0} - \frac{\frac{M_i}{M}}{C_i^0}$$

M_i and M_{i+} can be incorporated in the definition of the overall vapor pressure with a slight change.

$$\frac{\sum_i R_{OH,i} \cdot \frac{\frac{M_{i+}}{M}}{C_{i+}^0}}{\sum_i R_{OH,i}} = \frac{1}{\overline{C}_{i+}^0}$$

$$\frac{\sum_i R_{OH,i} \cdot \frac{\frac{M_{i+}}{M}}{C_i^0}}{\sum_i R_{OH,i}} = \frac{1}{\overline{C}_i^0}$$

In the revised manuscript, we have added these equations to the Appendix A.

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?

Response:

The individual saturation vapor pressures of multi-generation species here were only used to derive an average saturation vapor pressure and final relationship between particle growth and the reaction of total organics with OH (Eq. 22 and 23). Since the reaction system is a complex mixture of various organics with known or unknown identities and the vapor pressures of individual species are unknown, an average saturation vapor pressure was used in this study.

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?

Response:

Gas phase and particle phase are assumed to be in equilibrium as stated in the manuscript (p. 12600 line 21), i.e. evaporation and condensation are inherently included and are in dynamic equilibrium.

The effects of dilution and particle wall loss on particle concentration have been corrected as stated in the manuscript (p.12597 line1-4). The effect of vapor wall loss is not corrected since it is hard to determine directly due to the unknown identities, vapor pressures and its contribution in the particle phase etc. In our study, we used a large reaction chamber with low surface to volume ratio (surface/volume ratio ~0.88 m^{-1}), which is favorable for reducing the wall loss of vapor. We have estimated the uncertainty of particle mass concentration due to the vapor loss to be approximately 17%. This is estimated using the measured wall loss rate of pinonaldehyde and assuming the lost vapors have the same particle yield as in the reaction system.

The particle-phase photolysis is not included in this derivation, which could also potentially affect the gas-equilibrium.

In the revised manuscript we have elaborated these items including the limitation of our analysis similarly as above (page 14 lines 15-19; page 25 lines 20-25).

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?

Response:

The multi-generation products discussed here have fairly low vapor pressure. Unfortunately neither PTR-MS nor GC-MS is able to measure these compounds because of the significant loss by adsorbing to the sampling line and possible degradation in the drift tube of the PTR-MS.

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)

Responses:

The formation of low-volatility compounds from monoterpene oxidation such as 3-methy butane tricarboxylic acid (3-MBTCA) has been found in our previous studies (Emanuelsson et al., 2013). This has also been reported by a number of studies from the oxidation of monoterpene and its first generation products (Hallquist et al., 2009; Jaoui et al., 2005; Szmigielski et al., 2007; Claeys et al., 2007; Muller et al., 2012; Kristensen et al., 2014). Since these compounds have multi-functional groups, each of which can reduce the vapor pressure dramatically (Kroll and Seinfeld, 2008; Ziemann and Atkinson, 2012), these compounds with multi-functional groups are estimated to have very low vapor pressure.

As in the response to comment 6, there was nucleation in these experiments, which introduced particles available for the condensation.

In the revised manuscript, we have elaborated this part similarly as above, adding more explanations and references.

c. p. 12607, line 19: Do you mean "…saturated vapor pressures [increased]" instead?

Responses:

The reviewer is correct. We have corrected this mistake in the revised manuscript.

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.

Response:

As we explained in the manuscript (page 12607 lines 7-9), for α-pinene the reaction was stopped by closing the louvre before particle size reached the plateau phase (due to the experiment time limit). Therefore, before closing the louvre particles showed continuous growth.

We have added additional comments in the revised manuscript (page 17 lines 2-4).

"For α-pinene, particle growth did not reach the plateau phase. This is because the reaction was stopped by closing the louvre when particles were still growing."

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?

Response:

As in the response to the reviewer 1, before nucleation there were some pre-existing particles present introduced during humidification which had relatively large diameter (median diameter 40-60 nm). When the nucleation started, the median diameter shifted to a small size, manifesting a seemingly drop in the particle diameter. In the revised manuscript, we have left out the particle size data before nucleation in case this may mislead the readers and also added the note about this data process.

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.

Response:

We had some wording problems here. We meant that when O/C reaches 0.4, $GE_{OH}(t)$ has dropped dramatically to a low value. In the revised manuscript, we have re-written this sentence. Now it reads: "$GE_{OH}(t)$ had decreased dramatically to a much lower value when O/C ratio increased to around 0.4 and leveled off. Accordingly, H/C started to decrease from the beginning of the reaction and then leveled off at the same time as O/C. The decrease of $GE_{OH}(t)$ reflects the increasing role of fragmentation. "

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?

Response:

Here we took β-pinene system as an example for the reference purpose. In the revised manuscript, we have added the values for α-pinene and limonene system.

h. p. 12613, lines 19-21: Can this statement about carbonyl incorporation during oxidation be backed up mechanistically to support the calculated elemental ratios?

Response:

In the reaction of monoterpene with O_3, taking α-pinene as an example, the $-CH_2-$ group can be converted to -C=O group which reduces the H/C and increase O/C. One path way is shown as follows, which is now added to the revised manuscript. Monoterpene reacts with O_3 producing $RO_2\cdot$ radical, which can undergo internal hydrogen shift forming another $R_1O_2\cdot$ radical (Ehn et al., 2014). The $R_1O_2\cdot$ radical can react with other $RO_2\cdot$ radical forming -C=O group at the same time losing two hydrogen atoms.

In the revised manuscript, we have elaborated this point.

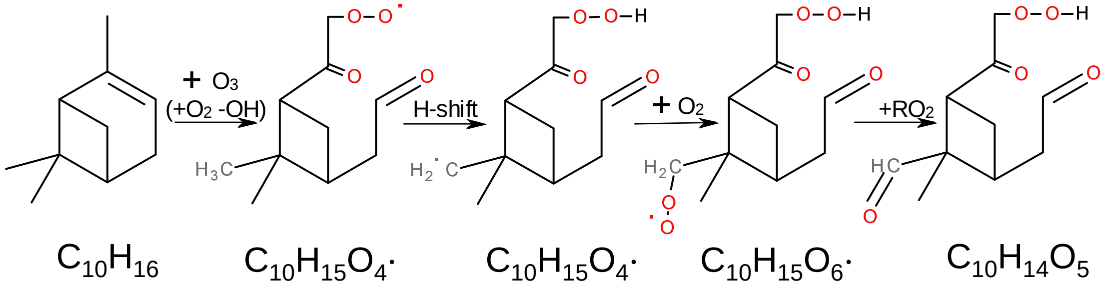


Figure S7. Schematics illustration of one possible pathway of reducing H/C for α-pinene ozonolysis

i. p. 12613, lines 26-29: Are you implying that after O_3 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?

Response:

After O_3 has reacted with monoterpenes containing only one carbon-carbon double, the reaction products have no carbon double bond left and do not react with O_3 anymore. However, H/C relates to not only the carbon-carbon bond but also other functional groups such as carbonyl, hydroxyl etc. During the reaction of monoterpene with O_3, carbonyl function can be formed from $-CH_2-$ (as shown in Fig. S7), therefore reducing the H/C. Based on the H/C from AMS measurement, such a process reducing the hydrogen efficiently seems to be more important in the ozonolysis of monoterpene compared to OH oxidation.

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.

Response:

The review is right in that particle size and particle mass can be affected by other factors. As in the response to comment 8 d, some of the factors have been considered or corrected in our analysis, and others are not.

We have changed this sentence in the revised manuscript. It reads now:

"In the OH oxidation, we found the transition of functionalization and fragmentation correlated with the evolution of particle size and particle mass as a function of OH dose."

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?*

Response:

The reviewer is correct. Isolation of ozonolysis by OH scavenger is not directly transferable to the atmosphere. However, the technique to scavenge OH in ozonolysis experiments to isolate the effect of ozone is an accepted, standard procedure in mechanistic studies as here. Moreover, in the atmosphere there are always "OH-scavengers" around, because the matrix e.g. of VOC is more complex than in our chamber experiment. Therefore an experiment without scavenger can be also unrealistic as too much dark OH is available compared to a real atmosphere. So "somehow" scavenging reflects some realism, however not in a way that can be easy related to the real atmosphere. But this debate is outside the scope of this paper.

The point we could not address is the oxidation of ozonolysis primary products by OH. This is likely the same as formation of higher generation products from primary OH products. We would estimate that if the total turnover of a VOC with ozone and OH increases, the growth rate after first oxidation step (which is determined by both OH and O_3 oxidation) increase similar as in OH oxidation experiments.

In the revised manuscript, we have added more comments on this aspect as follows.

"In this study, we designed the experiment to study mechanistically the particle formation and growth; therefore we used two extreme cases: pure OH oxidation and pure ozonolysis case. We did not do experiments with both OH and O_3 (in the dark). In the atmosphere, where both OH and O_3 are present, products from the reaction monoterpene with O_3 can further react with OH, hence the chemical composition of aerosol (in terms of elemental composition) may keep evolving continuously and turning more and more to the OH path. In the atmosphere, both OH oxidation and ozonolysis of monoterpene are important pathways for the particle formation and growth, with their relative importance depending on the specific ambient conditions."

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?

Response:

The relationship of overall reaction rate of the total organics with OH with the particle growth rate applies well in the well-characterized system, like in chamber experiments. Such relationship is being planned to be used in more VOC systems in the chamber experiment. For applicability in the atmosphere, the approach must be tested in atmospheric models and compared with other approaches like 2-product model or vapor pressure basis set. It could be seen as independent consistency test to compare growth rates otherwise generated by models with that expected from total turnover (OH reactivity in the model). To generalize the VOCs speciation must be extended. Different VOC types (such as sesquiterpene or isoprene or linear alkenes) at given overall reaction rate of total organics with OH may have fairly different particle growth efficiencies, thus resulting in different particle growth rates.

In the revised manuscript, we have added the following comments:

"The relationship of overall reaction rates of the total organics with OH with the particle growth rates applies well in well-characterized chamber systems. Such relationship is being planned to be tested using more VOC systems in the chamber. . For the atmosphere, it is much more complex to apply such method. Different VOC types (such as sesquiterpene or isoprene or linear alkenes) contribute to overall reaction rate of total organics with OH but may have different particle growth efficiencies resulting in different particle growth rates. This has still to be characterized in experiments. ”

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?

Response:

The HC consumed did not change linearly with reaction time, and so did OH dose. That is the reason why HC consumed and OH dose were used as x-axis in Fig. 1 and Fig. 2, respectively. In Fig. 3-5 the reaction time was used.

In Fig. 2A only the data for photooxidation period is shown, i.e. before the Louvre close. The Louvre closing did not coincide with the change in rate of aerosol concentration and diameter.

In the revised manuscript, we have added the figures showing the OH dose and HC consumed as in Fig.1 and Fig.2 using reaction time as x-axis.

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.

Response:

Accepted. As in the response to 10.a, a figure showing the time series of aerosol concentration and HC has been added to the revised manuscript so that the changes in Fig. 1 can be easily compared with those in Fig. 2-5.

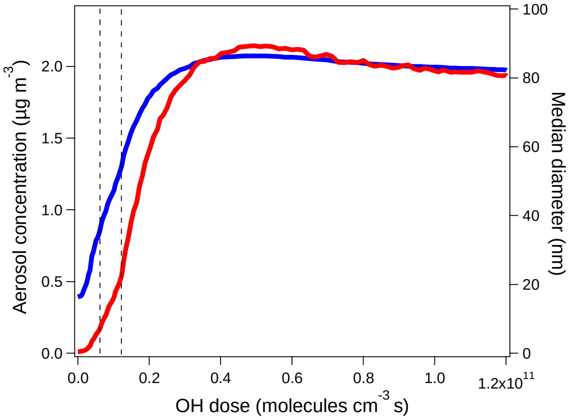
c. Fig. 2: Additional explanation for general shape of curves would be appreciated. Why in Fig. 2A are there so many changes in rate of aerosol growth? Why in Fig. 2C do the particle growth rate and mass growth rate seem parallel and never cross compared to the case of alpha- and beta-pinene?

Response:

Accepted.

The initial drop in the aerosol size in Fig.2A is due to the shift from background particles to particle nucleation and subsequent growth the as explained in the response to comment 9 e. The other changes in the aerosol growth in Fig.2A were mainly due to the significant fluctuation of OH concentration due to the cloud coverage which also caused the significant fluctuations in the reaction rate of all organics with OH in Fig. 4A. In the revised manuscript we have added the similar explanation (page 16 lines 10-13).

In Fig. 3C, the seeming parallel was due to the scaling of particle concentration and particle size. These are two independent parameters. When the scale was just, the two curves could also cross (shown in the following figure).



d. Fig. 3A: What is the explanation for all of the variation in $GE_{OH}(t)$ *for the case of α-pinene experiment?*

Response:

As in the response to comment 10.c), the variations in the $GE_{OH}(t)$ for the α-pinene case are due to cloud coverage change that day. This caused the fast and significant fluctuations in the OH concentration and reaction products concentrations.

We have added more explanations in the revised manuscript.

Technical Corrections:

1) p. 12593, line 17: Recommend changing "…thus on climate depends…" to, "…thus depends on…"

Response: Accepted.

2) p. 12595, line 18: Change "…of whole the…" to "…of the whole…"

Response: Accepted.

3) p. 12606, line 9: Delete "the" in "difference of VOC and the particle concentration."

Response: Accepted.

4) P. 12607, line 14: Change "promote" to "promoting."

Response: Accepted.

5) p. 12610, line 22-23: Use same notation for particle number concentrations reported.

Response: Accepted.

6) Fig. 2 caption: Add "of" before "each monoterpene and change "in respect to" to "with respect to"

References

Claeys, M., Szmigielski, R., Kourtchev, I., Van der Veken, P., Vermeylen, R., Maenhaut, W., Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., and Edney, E. O.: Hydroxydicarboxylic acids: Markers for secondary organic aerosol from the photooxidation of alpha-pinene, Environ. Sci. Technol., 41, 1628-1634, 10.1021/es0620181, 2007.

Ehn, M., Thornton, J. A., Kleist, E., Sipila, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I.-H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurten, T., Nielsen, L. B., Jorgensen, S., Kjaergaard, H. G., Canagaratna, M., Maso, M. D., Berndt, T., Petaja, T., Wahner, A., Kerminen, V.-M., Kulmala, M., Worsnop, D. R., Wildt, J., and Mentel, T. F.: A large source of low-volatility secondary organic aerosol, Nature, 506, 476-479, 10.1038/nature13032, 2014.

Emanuelsson, E. U., Hallquist, M., Kristensen, K., Glasius, M., Bohn, B., Fuchs, H., Kammer, B., Kiendler-Scharr, A., Nehr, S., Rubach, F., Tillmann, R., Wahner, A., Wu, H. C., and Mentel, T. F.: Formation of anthropogenic secondary organic aerosol (SOA) and its influence on biogenic SOA properties, Atmos. Chem. Phys., 13, 2837-2855, 10.5194/acp-13-2837-2013, 2013.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, T. F., Monod, A., Prevot, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155-5236, 2009.

Hanke, M., Uecker, J., Reiner, T., and Arnold, F.: Atmospheric peroxy radicals: ROXMAS, a new mass-spectrometric methodology for speciated measurements of HO2 and Sigma RO2 and first results, Int. J. Mass Spectrom., 213, 91-99, 10.1016/s1387-3806(01)00548-6, 2002.

Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., and Edney, E. O.: Identification and quantification of aerosol polar oxygenated compounds bearing carboxylic or hydroxyl groups. 2. Organic tracer compounds from monoterpenes, Environ. Sci. Technol., 39, 5661-5673, 10.1021/es048111b, 2005.

Kristensen, K., Cui, T., Zhang, H., Gold, A., Glasius, M., and Surratt, J. D.: Dimers in alpha-pinene secondary organic aerosol: effect of hydroxyl radical, ozone, relative humidity and aerosol acidity, Atmos. Chem. Phys., 14, 4201-4218, 10.5194/acp-14-4201-2014, 2014.

Kroll, J. H., and Seinfeld, J. H.: Chemistry of secondary organic aerosol: Formation and evolution of low-volatility organics in the atmosphere, Atmos. Environ., 42, 3593-3624, 10.1016/j.atmosenv.2008.01.003, 2008.

Mihelcic, D., Holland, F., Hofzumahaus, A., Hoppe, L., Konrad, S., Musgen, P., Patz, H. W., Schafer, H. J., Schmitz, T., Volz-Thomas, A., Bachmann, K., Schlomski, S., Platt, U., Geyer, A., Alicke, B., and Moortgat, G. K.: Peroxy radicals during BERLIOZ at Pabstthum: Measurements, radical budgets and ozone production, J. Geophys. Res.-Atmos., 108, 8254, 10.1029/2001jd001014, 2003.

Muller, L., Reinnig, M. C., Naumann, K. H., Saathoff, H., Mentel, T. F., Donahue, N. M., and Hoffmann, T.: Formation of 3-methyl-1,2,3-butanetricarboxylic acid via gas phase oxidation of pinonic acid - a mass spectrometric study of SOA aging, Atmos. Chem. Phys., 12, 1483-1496, 10.5194/acp-12-1483-2012, 2012.

Saathoff, H., Naumann, K. H., Mohler, O., Jonsson, A. M., Hallquist, M., Kiendler-Scharr, A., Mentel, T. F., Tillmann, R., and Schurath, U.: Temperature dependence of yields of secondary organic aerosols from the ozonolysis of alpha-pinene and limonene, Atmos. Chem. Phys., 9, 1551-1577, 2009.

Salo, K., Hallquist, M., Jonsson, A. M., Saathoff, H., Naumann, K. H., Spindler, C., Tillmann, R., Fuchs, H., Bohn, B., Rubach, F., Mentel, T. F., Muller, L., Reinnig, M., Hoffmann, T., and Donahue, N. M.: Volatility of secondary organic aerosol during OH radical induced ageing, Atmos. Chem. Phys., 11, 11055-11067, 10.5194/acp-11-11055-2011, 2011.

Szmigielski, R., Surratt, J. D., Gomez-Gonzalez, Y., Van der Veken, P., Kourtchev, I., Vermeylen, R., Blockhuys, F., Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., Edney, E. O., Seinfeld, J. H., Maenhaut, W., and Claeys, M.: 3-methyl-1,2,3-butanetricarboxylic acid: An atmospheric tracer for terpene secondary organic aerosol, Geophys. Res. Lett. , 34, L24811, 10.1029/2007gl031338, 2007.

Ziemann, P. J., and Atkinson, R.: Kinetics, products, and mechanisms of secondary organic aerosol formation, Chem. Soc. Rev. , 41, 6582-6605, 10.1039/c2cs35122f, 2012.