Response to Reviewer 1

We gratefully thank you for your constructive comments and thorough review. Below are our responses to your comments.

(Q=Question, and A=Answer)

General Comments:

Q1. The introduction is far too long. Also, significant quantities of introductory material appear in the results (lines 264-285 and 325-340). This material should all appear in the intro, which itself needs to be written more concisely. The authors should also try and use better paragraph structure in places and use shorter paragraphs in general.

A: Thanks for your suggestion. All of the introductory materials shown in the Sect. 3.2, Sect. 3.3, and Sect. 3.4 are now moved to the introduction. The introduction has been restructured and shortened.

Q2. Another general point is that the methods do not provide enough detail. For each technique discussed, the authors should provide detection limits, precision and accuracy.

A: We follow the advice of the reviewer. The detection limits, precision, and accuracy have been provided for the methods of detecting peroxides and carbonyls in the revised text. We also provide the diagrams of the flow tube reactor, the coil collector, and the Horibe tube in the Supplement

Q3. Throughout, there is a tendency to talk about things being higher, lower or different. It would be better if such expressions were quantified where possible.

A: Thanks for your suggestion. We have checked our manuscript and changed such expressions.

Q4. Finally, the experiments are carried out under conditions far removed from most ambient atmospheres, both in terms of concentrations of limonene and ozone, but also temperature (277K). Given this, I was missing a statement of the applicability of these results to real atmospheric conditions, or indeed as alluded to in the

manuscript, indoor environments.

A: We regret that we did not clarify well enough. All of our experiments were conducted at 298 K as stated in the Sect. 2.2. The temperature of 277 K was just used to keep the SOA solution in order to maintain the stability of samples and prolong their storage time. We have explained this in the revised manuscript. As for the concentrations of reactants used in our experiments, the concerns of the reviewer are reasonable. To get enough products for analysis in a short reaction time, both of the concentrations of limonene and ozone in this study were obviously higher than those in the real atmospheric conditions, which might have influence on the gas-phase and particle-phase chemistry. So the effects of the reactants concentrations on the experimental results are discussed below and this part is now added in the Supplement.

A major impact of the high concentrations of reactants is the increased RO_2 concentration. In recent years many studies reported that the autoxidation processes formed highly oxidized RO_2 radicals, which reacted with HO_2 and other RO_2 radicals forming highly oxidized multifunctional organic compounds (HOMs) (Jokinen et al., 2014; Richters et al., 2016a, b). The production of HOMs is controlled by two competing processes, i.e., RO_2 autoxidation vs. RO_2 reaction with HO_2 and other RO_2 radicals. Zhang et al. (2015) found that at low α-pinene levels, the longer lifetime of RO_2 radicals favored the isomerization pathways and consequently led to enhanced ELVOC dimers production. They estimated that the corresponding lifetime of RO_2 radicals decreased by less than an order of magnitude when the initial α-pinene mixing ratio increased from 10 ppbv to 150 ppbv, which was not sufficient to perturb the dynamics of overall RO_2 chemistry. In our experiments, where the limonene concentration was below 200 ppbv, we speculated that although the RO_2 chemistry was affected to some extent it would not bring huge influence on the results. When the RO_2 concentration is high, the reactions of SCIs and RO_2 radicals might happen in the system (Sadezky et al., 2008). Zhao et al. (2015) found that the reactions of SCIs and RO_2 radicals played a key role in particle formation in *trans*-3-hexene ozonolysis, while for large

alkenes such as terpenes and sesquiterpenes such reactions might be unimportant. Thus, although the concentrations of reactants used in this study were higher than that in the real atmospheric condition, the SCIs + RO_2 reactions did not have a huge effect on the reaction system. The SOA yield of limonene ozonolysis observed in this study was in the range of the values reported before (Ahmad et al., 2017; Chen and Hopke, 2010; Pathak et al., 2012). It is true that when the mixing ratios of reactants are high, the gas-particle partitioning processes of semi-volatility and low-volatility products are promoted resulting in higher SOA yield, yet we think that it may not have great impact on the representativeness of the products we investigated in particles.

Specific Comments:

Q6. I am not going to list every grammatical error, but I will mention a few here. I think that when the authors talk about 'SOA property' they mean SOA composition. This phrase should be replaced throughout with something more informative. Similarly, I am not clear what they mean by 'oxidants transition'. Again, clarification is needed.

A: Thanks for your suggestion and we have changed "SOA property" to "SOA composition" throughout the text. Besides, "oxidants transition" is replaced by "oxidation regime" in the revised manuscript. In this study, the objective of investigating the oxidation regime of alkene ozonolysis is to understand the formation of the oxidizing products in limonene ozonolysis. These compounds, including OH radicals, stabilized Criegee intermediates, and peroxides, are critical to atmospheric oxidation processes since they own the power of oxidizing other species. We clarify this in the revised introduction.

Q7. 'Particulate unstable peroxides' is an awkward expression.

A: We have changed that to "unstable peroxides in particles".

Q8. Line 64 – 'eaters' - esters?

A: Yes, we have revised it.

Q9. Line 542: Assume these are Kp coefficients - should specify. I don't understand the explanation for the finding that Kp is much bigger than estimated, line 556 (and lines 579-580). The language needs to be improved. What impact could experimental conditions have (low T and high precursor concentrations).

A: We have specified that these are the calculated gas-particle partitioning coefficients. As for the explanation for the finding that the measured K_p is much bigger than the predicted K_p, we have clarified it better in the revised text. A plausible explanation for the large difference between the measured K_p and the predicted K_p was that carbonyl compounds were easy to polymerize and react with other species on particles, resulting in that these carbonyls existed in forms of hydrates and oligomers (Corrigan et al., 2008; Hastings et al., 2005; Kroll et al., 2005; Volkamer et al., 2007). The hydrates and oligomers of carbonyls have much lower vapor pressures than their precursors, and they could reversibly return to their carbonyl monomers during analysis (Healy et al., 2008; Ortiz et al., 2013; Toda et al., 2014). The temperature used here was 298 K and the effect of precursor concentrations was analyzed. When the mixing ratios of reactants are high, the gas-particle partitioning processes of semi-volatility and low-volatility products are promoted resulting in higher SOA yield. However, when we calculated the gas-particle partitioning coefficients, the effect of the concentration of total suspended particulate matter was taken into consideration as shown by Eq. (2), so the higher SOA concentration caused by higher precursor concentrations would not impact the partitioning behaviors of carbonyls. Another effect of the high precursor concentrations was that it might promote the reactions of carbonyl compounds on particles, yet the concentrations of most carbonyls detected in our reactions were usually several ppbv, which were just a little higher than those in the real atmosphere, so we speculated that this impact on the partitioning coefficients was also limited.

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