

This study investigated the OH-oxidation of α -pinene ozonolysis products by reporting the mass yields and elemental composition of SOA produced. The main finding of this study is that OH-oxidation of α -pinene ozonolysis products that is equivalent of 2-4 days of atmospheric OH exposure leads to 20-40% net increase in the SOA yields and an increase in the aerosol O:C ratio by up to 0.04. While highlighting the importance of multi-generation aging in the atmospheric evolution of SOA, a topic that has been extensively studied previously, this finding alone, however, does not warrant the scientific significance and publication of this manuscript on ACP. Substantial revisions are needed, as described in the individual points below.

General:

To what extent the experimental observations that 20-40% increase in SOA yields by further OH-oxidation could be applied to the actual atmospheric conditions? The authors used HONO as the OH precursor, which means that hundreds of ppbv levels of NO were present in the experiments conducted. Such an experimental condition is quite different from what is in the atmosphere, where most monoterpene emissions are from remote regions that encounter low NO air masses (e.g., a few ppb or less). As we know, NO significantly alters the VOCs oxidation mechanisms primarily by reacting with RO₂ radicals, leading to vastly different product distributions from those observed in the absence of NO, a chemical regime where RO₂+HO₂ and RO₂+RO₂ reactions dominate the fate of RO₂ radicals. That said, the experimental results presented in this study only represent a barely-seen scenario in the atmosphere and does not have any atmospheric relevance. The authors are suggested to conduct 'low-NO_x' experiments as well where H₂O₂, for example, can be used as the OH precursor. The observed increase in SOA yields, together with the high-NO experiments, can be used as upper and lower limits for the α -pinene SOA aging in the atmosphere.

The methodology of this study, including the chamber experimental approach, SOA yield measurements corrected by particle wall loss, O:C ratio calculations by AMS measurements, has been widely used in the community for decades. The main result of this study, i.e., SOA yields increase by 20%-40% upon OH-oxidation aging, is insufficient to warrant publication of this manuscript. The authors should endeavor to

explore more detailed mechanisms involved in the α -pinene SOA aging by thoroughly analyzing the AMS data, e.g., What functionalities/products have changed and contributed to SOA production during aging? Any indication of PMF analysis, for example, on the key processes involved in the chemical aging?

Specific:

Page 2, Line 35-36: There have been a number of studies investigating the SOA aging processes using both static chamber and flow tube reactors (e.g., Robinson et al., Science, 2007; Loza et al., ACP, 2012; Lambe et al., ES&T, 2012). The authors need to revise the statement ‘Most laboratory studies of SOA formation so far have focused on the ...’

Page 3, Line 64: References need to be given here.

Page 6, Line 154: What is the particle size range measured by SMPS?

Page 6, Line 171: The OH radical molar yield from ozonolysis of α -pinene is roughly 0.7. So technically, the measured SOA yield at first stage is already a result of certain OH aging. Have the authors estimated the fraction of α -pinene that was oxidized by OH radical in the ozonolysis experiments? Have the authors considered adding any OH scavengers (e.g., H₂O₂, CO, methanol) in the first stage of the experiments?

Page 7, Line 205: The assumption that the particle loss rate in the 300-600 nm range is the same potentially leads to underestimation of the corrected SOA yields, as bigger particles have higher wall loss rate due to gravity deposition.

Page 8, Line 236: The authors are suggested calculate the SOA density from the AMS measured O:C and H:C ratio of SOA (Kuwata et al., ES&T, 2012) and compare with the current value used.

Page 9, Line 237-238: Adding the ‘ $V(t)/V_{\max}$ ’ factor in the SOA yield correction procedure does not seem necessary. The authors stated that ‘deviations of $V(t)$ from V_{\max} are caused by the uncertainty associated in applying the size dependent wall loss corrections’. As we know, particle wall loss rate depends on a few parameters, including chamber size, eddy diffusion, static charges on the Teflon surface, and etc. For most experiments presented here that only lasted for a few hours, these parameters should be

fairly constant so that the particle wall loss rate should be quite consistent before and after one experiment. Where are the deviations originating from? I wonder if the assumption that particle loss rate in the 300-600 nm range is the same has any impact on the SOA yield correction here.

Page 27, Figure 5: Why the O:C ratio of SOA kept decreasing in the second dark period? What are the concentrations of NO_x and O_3 during this period? Is there any NO_3 -initiated chemistry going on?