

Interactive comment on “The atmospheric impacts of monoterpene ozonolysis on global stabilised Criegee intermediate budgets and SO₂ oxidation: experiment, theory and modelling” by Mike J. Newland et al.

Anonymous Referee #1

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In this paper, the authors performed a chamber experiment to understand the effect of humidity toward the SO₂ reaction of the ozonolysis product of the alpha-, and beta-pinene; and limonene. The 298 K experimental result was analyzed assuming the reaction of two different types of stabilized Criegee intermediate (SCI) forming from the ozonolysis. SCIA which favorably reacts with H₂O and SCIB which favorably unimolecularly decomposes. This two SCI model provided a good fit to the chamber results. Using this fits, they can obtain relative ratios between SCI bimolecular reaction rates with atmospheric trace gases. Results of quantum chemistry calculations are presented

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to provide support for the rates obtained from the experimental fits. These results are placed into the global simulation model to show that the SO₂ reaction only accounts for <1% of the global decay path for SCI obtained from ozonolysis of monoterpene. These experimental results confirm the theoretical prediction presented by one of the authors (PCCP, 19, 31599, 2017). All in all, I think these results are important and should be published. However, I have the following points I think the authors should clarify.

First, the authors should clarify the difference between the real atmospheric environment and their chamber. The real atmospheric environment has a range of temperature, relative humidity, and pressure; they should say which temperature, humidity, pressure range can be attained in their chamber. Second, on page 22, lines 13-16, the authors mention the issues concerning the non-linear results for the limonene results in Figure 2. Can the authors cool the limonene before entering the chamber to decrease the ozonolysis rate? Or can they try an experiment at lower temperatures, to obtain cleaner data for low humidity? Third, cyclohexane is used as an OH scavenger. Is the SCI reaction with cyclohexane slow that it will not interfere with their analysis?

Small points that can be fixed are as follows: In page 13 lines 11-13, the authors mention that water dimer reaction will be negligible at atmospherically accessible [H₂O]. However, it has already been shown experimentally that for anti-CH₃CHOO water vapor reaction, water dimer reaction will dominate the room temperature reaction at a relative humidity (RH) above 30%. (PCCP, 18, 28189-, 2016) On the other hand, the present chamber experiments were done at RH 0.1 to 28%. Therefore, the authors should change this part to “For the analysis of the present chamber results the water dimer reaction can be ignored.” In Page 21 lines 23-24, they mention the effective rates for the SCI water vapor reaction at RH 75%, 298 K, and discuss results, but their experimental chamber results are up to RH 28%, so I am not sure it is relevant to mention the results for such high RH.

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