Title: Kinetics of dimethyl sulfide (DMS) reactions with isoprene-derived Criegee intermediates studied with direct UV absorption
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General Comments

The authors address an important result from the 2015 *ACP* paper of Newland *et al.*: dimethyl sulfide (DMS) reacts with Criegee intermediates (CIs) a factor of ~3 faster than SO_2 does with CIs; the CI + DMS rate constant is thus ~1 x 10^{-10} cm^3 s^{-1}. Based on their time-dependent measurements of CI concentration with UV absorption, the authors estimate upper limits for the CI + DMS rate constant of ~10^{-15} cm^3 s^{-1} for CH_2OO and ~10^{-14} cm^3 s^{-1} for methyl vinyl ketone oxide (MVKO). Transition state theory (TST) calculations based on CCSD(T)//M06-2X quantum chemical data estimate the rate constant to be ~6 x 10^{-19} cm^3 s^{-1}.

The quality of the experimental and theoretical work is high and the results should be reliable. Where the manuscript could be strengthened is in its discussion of the previous work by Newland 2015. In this, I agree with the comments from Rickard and from Blitz. There should also be a slightly broader computational treatment of the relevant reactions along with a more detailed representation of the computational results.

Specific Comments

- The presentation of the theoretical predictions in Section 3.5 and Figure 5 would be much easier to follow if there were some graphical representation of the chemical structures being discussed.
- One reason for the difference is the current results and the results reported in Newland 2015 may be the impact of DMS on the MVKO + SO_2 reaction. It is not necessary to perform calculations on this reaction, but some mechanistic discussion would be pertinent.
- It could be enlightening to provide a brief context by comparing the title reaction with the previous study of CH_3SH + CH_2OO (*J. Phys. Chem. A* **2019**, *123*, 4096-4103).
- **Line 224**: What is the evidence for the CH_2OO-DMS adduct having "very strong zwitterionic character?"
- **Supplemental Information S20-S21**: The authors should present some calculations on the MVKO. In particular, it would be worthwhile to consider how DMS might affect the cyclization of the *anti* conformer of MVKO to the dioxole (see *J. Am. Chem. Soc.* **2018**, *140*, 10866). Here, I reiterate the comment of Rickard that it would be useful for the authors to estimate the relative amounts of the *syn* and *anti* conformers of MVKO.

Technical Corrections

- **Lines 232-233**: "We did not examine more exotic CI reaction such as insertion in the DMS C–H bonds, as these are known to have comparatively high barriers." This statement should have a reference.

- **Supplemental Information S20-S21**:The authors should tabulate the relative energies predicted by the M06-2X/cc-pVDZ calculations.