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Shedding light on tropospheric H_2SO_4 production from Criegee intermediates + SO_2: a comprehensive laboratory chamber study using the highly instrumented HELIOS platform

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Although the dominant source of H_2SO_4 in the atmosphere is generally considered to be the reaction of SO_2 with OH, it is probable that the rapid reactions of Criegee intermediates (CIs) with SO_2 can contribute significantly to the tropospheric H_2SO_4 budget under certain conditions. CIs are produced from alkene ozonolysis, and the vast quantities of unsaturated biogenic and anthropogenic volatile organic compounds emitted could provide a large and diverse flux of CIs to the atmosphere. There remain several key uncertainties regarding the global importance of CIs towards SO_2 oxidation, which are principally related to the ambient concentrations of CIs and the competition between CI reaction with SO_2 against the many other bimolecular and unimolecular loss processes. This is especially true of the larger, more complex CIs that are produced from terpene ozonolysis.

We present experimental studies of the ozonolysis of tetramethylethylene, α-pinene and limonene, using the HELIOS chamber. HELIOS is a highly instrumented large-scale outdoor atmospheric simulation chamber and consists of a hemispheric 90 m^3 Teflon-foil reactor, which is interfaced to a variety of on-line measurements including FTIR, PTR-ToF-MS, FIGAERO-ToF-CIMS, OH/H_2SO_4-CIMS, Aerolaser HCHO, LOPAP and SMPS, together with several GC-MS/FID and LC-MS instruments and a suite of monitors (NO, NO_2, O_3). Equipped with this range of instrumentation we are able to conduct alkene ozonolysis under near-ambient conditions, whilst we also have a high coverage of key reactive species in the systems of interest.

From our results, we are able to provide new information regarding kinetic and mechanistic behaviour of several atmospherically important CIs and their reactive intermediates, providing new constraints on the role of CIs on the tropospheric H_2SO_4 budget.

Keywords: ozonolysis, Criegee Intermediate, sulfur dioxide, sulfuric acid, kinetics