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Competitive reaction of CH_2OO with SO_2 and water vapour and the thermal lifetime of CH_2OO at 293 K

Torsten Berndt (1), Heikki Junninen (2), Roy L. Mauldin III (2,3), Hartmut Herrmann (1), Markku Kulmala (2), and Mikko Sipilä (2)

(1) Institut für Troposphärenforschung, Department of Chemistry, Leipzig, Germany (berndt@tropos.de), (2) University of Helsinki, Department of Physics, Helsinki, Finland, (3) University of Colorado at Boulder, Colorado, USA

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T. Berndt (1), H. Junninen (2), R. L. Mauldin III (2,3), H. Herrmann (1), M. Kulmala (2), and M. Sipilä (2)

(1) Leibniz Institute for Tropospheric Research, Permoserstr. 15, 04318 Leipzig, Germany; (2) University of Helsinki, Department of Physics, 00014 Helsinki, Finland; (3) University of Colorado at Boulder, Department of Atmospheric and Oceanic Sciences, Boulder, Colorado 80309, USA

H2SO4 represents a key substance in the process of atmospheric nucleation. The importance of gas-phase products from olefin ozonolysis other than OH radicals, most likely stabilized Criegee Intermediates (sCIs), for the process of atmospheric SO_2 oxidation to H2SO4 has recently been discovered.
Subject of this work are investigations on H2SO4 formation from CH_2OO + SO_2 as a function of the water vapour content and the measurement of the CH_2OO steady state concentration starting from the ozonolysis of ethylene used for formaldehyde oxide generation. Measurements have been conducted in an atmospheric pressure flow tube at 293 K using NO_3–CI-APi-TOF mass spectrometry for H2SO4 detection.
The experiments show a square-dependence in H_2O for the kinetics of the reaction CH_2OO + H_2O indicating that likely the water dimer $(H_2O)2$ governs the reaction with CH_2OO rather than the water monomer. This finding is in line with results from quantum chemistry. Furthermore, a sCI yield (CH_2OO) of 0.40 ± 0.18 can be deduced from the H2SO4 measurements in accordance with results from other experimental techniques. A CH_2OO thermal lifetime > 1s was found as a result of CH_2OO steady state measurements for different reactant concentrations at 293 K.
The importance of H2SO4 formation from CH_2OO + SO_2 for atmospheric conditions is discussed based on kinetic parameters obtained in this study.