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Formation of secondary organic aerosol and oligomers from the ozonolysis of enol ethers

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Abstract. Formation of secondary organic aerosol has been observed in the gas phase ozonolysis of a series of enol ethers, among them several alkyl vinyl ethers (AVE, ROCH=CH₂), such as ethyl, propyl, *n*-butyl, *iso*-butyl, *t*-butyl vinyl ether, and ethyl propenyl ether (EPE, C₂H₅OCH=CHCH₃). The ozonolysis has been studied in a 570 l spherical glass reactor at ambient pressure (730 Torr) and room temperature (296 K). Gas phase reaction products were investigated by in-situ FTIR spectroscopy, and secondary organic aerosol (SOA) formation was monitored by a scanning mobility particle sizer (SMPS). The chemical composition of the formed SOA was analysed by a hybrid mass spectrometer using electrospray ionization (ESI). The main stable gas phase reaction product is the respective alkyl formate ROC(O)H, formed with yields of 60 to 80%, implying that similar yields of the corresponding excited Criegee Intermediates (CI) CH₂O₂ for the AVE and CH₃CHO₂ for EPE are generated. Measured SOA yields are between 2 to 4% for all enol ethers. Furthermore, SOA formation is strongly reduced or suppressed by the presence of an excess of formic acid, which acts as an efficient CI scavenger.

Chemical analysis of the formed SOA by ESI(+)/MS-TOF allows to identify oligomeric compounds in the mass range 200 to 800 u as its major constituents. Repetitive chain units are identified as CH₂O₂ (mass 46) for the AVE and C₂H₄O₂ (mass 60) for EPE and thus have the same chemical compositions as the respective major Criegee Intermediates formed during ozonolysis of these ethers. The oligomeric structure and chain unit identity are confirmed by HPLC/ESI(+)/MS-TOF and ESI(+)/MS/MS-TOF experiments, whereby successive and systematic loss of a fragment with mass 46 for the AVE (and mass 60 for EPE) is observed. It is proposed that the oligomer has the following basic structure of an oligoperoxide, -[CH(R)-O-O]_n-, where R=H for the AVE and R=CH₃ for the EPE. Oligoperoxide formation is thus suggested to be another, so far unknown reaction of stabilized Criegee Intermediates in the gas phase ozonolysis of oxygen-containing alkenes leading to SOA formation.

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