Study of the Ozonolysis of Ethene and 2,3-Dimethyl-2-Butene Using Cavity Ring-down Spectroscopy

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Ozonolysis is one of the main oxidation channels of alkenes in the atmosphere, as well as a significant source of secondary organic aerosol (SOA) and hydroxyl and organic radicals. Carbonyl oxides, also known as Criegee intermediates (CIs), are key products in the ozonolysis of alkenes, being produced with a broad internal energy distribution. While CIs with high internal energy can dissociate and produce OH, RO, and other organic radicals, CIs with low internal energy (known as stabilized CIs) react rapidly to form other carbonyl compounds involved in the SOA production. In this work, indirect measurements of the yield of stabilized Criegee intermediates (SCIs) from the ozonolysis reaction of ethene and 2,3-dimethyl-2-butene were carried out at low pressures using cavity ring-down spectroscopy (CRDS). Determination of the yield of SCIs was performed by chemical titration using SO_2. In the case of ethene ozonolysis, the formation of formaldehyde by reaction of SCI with SO_2 was also monitored. Chemical kinetic modeling was performed to estimate the kinetic rate constants of the reactions between SCIs and SO_2. The yield of SCIs was found to decrease with decreasing pressure and reach a minimum value at the low pressure limit, which corresponds to the fraction of CI formed with internal energy below that of dissociation.