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Simulating the Atmospheric Impact of Criegee Intermediates: Implementation of new understanding in atmospheric chemical mechanisms

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Unsaturated hydrocarbons - alkenes - account for about 90% of global VOC. Stabilized Criegee Intermediates (SCI) are thought to be formed in the atmosphere mainly from reactions of unsaturated hydrocarbons with ozone.

SCI have been shown in laboratory and chamber experiments to rapidly oxidise SO_2 and NO_2, providing a potentially important gas phase oxidation route for
these species in the atmosphere. They have also been implicated in the formation of aerosol and organic acids. However, the importance of SCI reactions with traces gases is critically dependent on the relative ratio of the rate constants for the reactions of the SCI with these and other trace gases, with H_2O, and for unimolecular decomposition, which vary between SCIs, and between geometric isomers.

The selection of reactions and rate constants is critically important in determining the calculated impact of SCI processes upon atmospheric composition and chemistry. Since the recent resurgence in interest in this chemistry, a number of model studies have been performed, with SCI mechanisms of varying comprehensiveness and accuracy, as the understanding of the community has evolved from new laboratory, theoretical and chamber studies, and field observations.

Here we present an assessment of the dependence of modelled SCI abundance, behaviour and impacts upon the Criegee mechanism adopted, in the context of (a) the accepted status quo prior to the laboratory and field studies of Welz et al. and Mauldin et al., (b) changes to the SCI mechanism reflecting new kinetics for key bimolecular reactions, e.g. with SO_2 and NO_2; (c) emerging understanding of the interactions of SCI with water vapour and their unimolecular decomposition and (d) reactions with other atmospheric trace gases. The modelled SCI behaviour is compared with the results from recent chamber studies, and the resulting calculated SCI abundance and impacts evaluated for urban and forested atmospheric boundary layer scenarios.