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Stabilized Criegee Intermediates from biogenic alkene ozonolysis and their role in atmospheric system

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Sulphuric acid (H2SO4), is the key compound in atmospheric secondary aerosol particle formation. In atmosphere, sulphur dioxide (SO_2) is partly converted to H2SO4 vapour in a reaction chain initiated by the SO_2 oxidation by hydroxyl radical (OH). In our very recent research (Mauldin et al., 2012) we have demonstrated a previously not considered pathway for atmospheric H2SO4 formation from SO_2 which is of special importance for forested areas of the world. This new formation pathway involves a reaction of stabilized Criegee Intermediates (sCI) with SO_2. These sCI originate from the oxidation of forest emitted alkenes, including monoterpenes, by ozone.

Here, we describe our recent progress in experimental investigations of sCI chemistry. We produced sCIs in a laminar flow tube by ozonolysis of a variety of biogenic alkenes abundant in forests. We report the sCI yields and lifetimes as well as reaction rate coefficients with SO_2 and compare our results with our earlier estimates (Mauldin et al., 2012). We apply the knowledge obtained from these laboratory experiments in order to assess the global relevance of sCI oxidation of SO_2 to global sulphuric acid, small particle and cloud condensation nuclei concentrations by means of global aerosol-climate modeling. Our results indicate that SO_2-oxidation by sCI has a measurable, but likely not dominant effect on atmospheric levels of above mentioned quantities. Besides SO_2-oxidation, we also discuss the other sCI related processes and their potential relevance for atmospheric system.

Mauldin et al., A new atmospherically relevant oxidant of sulphur dioxide, Nature, 488, 193-196, 2012.