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Analysis of gas and particle phase products in the ethylene ozonolysis using negative ion chemical ionization mass spect

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The reaction of ozone with alkenes is a significant loss process of both ozone and alkenes in the atmosphere and plays an important role in air pollution processes in urban areas. The alkene ozonolysis produces Criegee intermediates, which have relatively high reactivity and partly decompose to produce radicals such as OH, HO_2 and RO_2. Especially, the formation of OH in alkene ozonolysis can be important as a nighttime source of OH. Additionally, ozone-alkene reaction can contribute to the formation of secondary organic aerosols (SOAs). Despite of its importance, the reaction process of the ozone-alkene reaction is not fully understood. This is the case even in the ozonolysis of ethylene, which is the simplest alkene, widely used in industry, and one of the most significant volatile organic compounds (VOCs) released into the environment.

In this study, we investigated the ethylene ozonolysis in laboratory experiments with a Teflon bag by using negative ion chemical ionization mass spectrometry (NI-CIMS). NI-CIMS is a powerful tool with less fragment, high selectivity, and high sensitivity for analysis of compounds such as carboxylic acids and hydroperoxides, which are expected to be produced in the ethylene ozonolysis.

As gas-phase products, we detected oligomeric hydroperoxides composed of Criegee intermediates as a chain unit, as well as formic acid and hydroperoxides which are previously reported. Additionally, the formation of SOAs in the ethylene ozonolysis was observed and theirits components were analyzed using NI-CIMS. The oligomers composed of the Criegee intermediates were also found as particle components. The formation of gas-phase oligomers and SOAs was strongly suppressed by adding methanol as a Criegee scavenger, clearly indicating that the Criegee intermediate plays a main role in the formation of oligomers and SOAs in the ethylene ozonolysis. The sequential addition of Criegee intermediates to hydroperoxides was proposed as the oligomer formation mechanism.

Keywords: ozonolysis, ethylene, SOA, hydroperoxide, oligomerization