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Aerosol-halogen interaction: Halogenation of secondary organic aerosols

Johannes Ofner (1), Natalja Balzer (1), Sergej Bleicher (1), Joelle Buxmann (2), Hinrich Grothe (3), Heinz-Ulrich Krüger (1), Ulrich Platt (2), Philippe Schmitt-Kopplin (4), and Cornelius Zetzsch (1)

(1) Atmospheric Chemistry Research Laboratory, University of Bayreuth, Germany (johannes.ofner@uni-bayreuth.de), (2) Institute of Environmental Physics, University of Heidelberg, Germany, (3) Institute of Materials Chemistry, Vienna University of Technology, Austria, (4) Institute of Ecological Chemistry, Helmholtz Zentrum Munich, Germany

The release of reactive halogen species from sea-salt aerosol contributes a new class of reactants for heterogeneous reactions. These heterogeneous reactions have been overlooked so far, although they may occur with internal and external mixtures of sea-salt aerosol and organic aerosol and organic matter in soil. Such reactions might constitute sources of gaseous organohalogen compounds or halogenated organic aerosol in the atmospheric boundary layer.

To study the interaction of organic aerosols with released halogen species, secondary organic aerosol (SOA) was produced from α-pinene, catechol and guaiacol using an aerosol smog-chamber and characterized. Those aerosols were exposed to molecular halogens in the presence of UV/VIS irradiation and to halogens, released from simulated natural halogen sources like salt-pans, to study their behavior against those reactive trace gases.

The entire organic aerosol and its physico-chemical transformation process were monitored using various spectroscopic methods: Long-path-absorption- and Attenuated-Total-Reflectance (ATR-)-FTIR were used to determine the changes of functional groups and structural elements of the macromolecular aerosols. The optical properties in the UV/VIS range were monitored using Diffuse-Reflectance-UV/VIS-Spectroscopy. The evolution and change of the aerosol size distribution by the reaction with halogens was observed using an electrostatic classifier coupled to a condensation nuclei counter. Finally, Temperature-Programmed-Pyrolysis Mass-Spectroscopy (TPP-MS) and ultra-high resolution Fourier-Transform-Mass-Spectroscopy (ICR-FT/MS) were used to determine the degrees of halogenation and single halogenated molecules.

The heterogeneous reaction of reactive halogen species with those model aerosols leads to different gaseous species like CO_2, CO and small reactive molecules like phosgene. Methyl groups (and possibly other C-H containing groups) on the aerosol particles are destroyed to form HCl. Also carbon-oxygen bonds are affected by the reaction with these halogens, and a significant formation of C-Br bonds could be verified in the particle phase. Overall, the optical properties of the processed organic aerosols are significantly changed. Further, the aerosol size distribution of the organic aerosols is strongly influenced by the heterogeneous reaction. Studying the reaction of organic aerosols with molecular halogens at different simulated environmental conditions allowed us to allocate changes of the described physico-chemical parameters to specific reactive species and to interpret the complex changes shown by the reaction with halogens released from the simulated salt-pan.

The heterogeneous reaction of secondary organic aerosols with atmospheric halogen species leads to changes of several physico-chemical features of the aerosol, influencing their chemical composition, structural elements, their optical properties and size distribution. Changes of the physical aerosol properties are carried out by the chemical reaction, e.g. changing the ability to act as CCN or the contribution to radiative forcing.