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Aerosol-halogen interaction: Formation of halogenated compounds in the particle phase and change of aerosol optical properties

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Reactive halogen species, released by sea-salt activation, offer a class of reactants of utmost importance for heterogeneous reactions with organic aerosols. To study the interaction of organic aerosols with these halogen species, secondary organic aerosol (SOA) was formed from α-pinene, catechol and guaiacol in a 700 L glass smog-chamber and in a 3500 L Teflon smog-chamber at defined environmental conditions (with and without simulated sunlight and ozone at various humidities). The well-characterized and aged organic aerosols were exposed to molecular halogens in the presence of UV/VIS irradiation and halogens released from simulated natural halogen sources to study the halogen-SOA interaction process.

Various spectroscopic methods were used to investigate optical and structural changes of the organic aerosols. Diffuse reflectance UV/VIS spectroscopy was employed to characterize optical properties in the UV/VIS spectral range. With FTIR (Fourier Transformation Infrared) spectroscopy both, the aerosol formation and transformation process and, using ATR (Attenuated Total Reflectance) spectroscopy, the appearance of functional and structural elements of the particulate phase were characterized. Using Temperature-Programmed-Pyrolysis Mass-Spectroscopy (TPP-MS) and ultra-high resolution Ion Cyclotron Resonance Fourier Transform Mass Spectroscopy (ICR-FT/MS), degrees of halogenations and single halogenated molecules could be determined.

Overall, the optical properties of organic aerosols appear to be significantly changed due to the reaction with those reactive halogen species. While the interaction with chlorine leads to some bleaching (corresponding to a blue-shift), the reaction with bromine shifts the absorption in the UV/VIS range to the red. Also transformation of relevant functional groups and formation of carbon-halogen bonds was detected. While chlorine forms gaseous species like HCl and phosgene and leads to destruction of carbon-hydrogen bonds, bromine is mainly integrated into the organic aerosols. The reaction of halogens with secondary organic aerosols leads to changes of the different physico-chemical features of the aerosol, influencing their chemical composition, structural elements, volatility and their optical properties. Additionally, due to the chemical changes, also a change of the physical properties related to the atmospheric environment is expected, e.g. the ability to act as CCN, the potential to adsorb other low-volatile gases, or even the contribution to radiative forcing.