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Reactive halogen chemistry in the Arctic boundary layer over snow during spring: A 1D modelling case study

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Polar halogen chemistry has long been known to be active, especially in spring, and has an important influence on the lifetime of some volatile organics, ozone, and mercury. Reactive chlorine and bromine species, produced from snow and aerosols, can have significant impacts on the oxidative capacity of the polar boundary layer. However, halogen production mechanisms from snow remain highly uncertain, making it challenging to include descriptions of halogen snow emissions in models and to understand the impact on atmospheric chemistry. In this work, we investigate the role of Arctic chlorine and bromine emissions from snow on boundary layer oxidation processes using a one-dimensional atmospheric chemistry and transport model (PACT-1D). We explore the impact of halogen snow emissions and boundary layer dynamics on atmospheric chemistry by modelling primary emissions of Cl_2 and Br_2 from snow, and heterogeneous recycling reactions on snow and aerosols. We present a two-day case study from the 2009 Ocean-Atmosphere-Sea Ice-Snowpack (OASIS) campaign at Utqiagvik, Alaska.

The model reproduces both the diurnal cycle and high quantity of Cl_2 measured, along with the observed concentrations of Br_2, BrO, and HOBr. Due to a combination of chemical emissions, recycling, vertical mixing, and atmospheric chemistry, reactive chlorine is confined to the lowest 15 m of the atmosphere, whilst bromine impacts chemistry up to the boundary layer height. Following the inclusion of halogen emissions and recycling, HO_x concentrations (HO_x = OH+HO_2) increase by as much as a factor of 30 at the surface at mid-day. Consequently, volatile organic compound (VOC) lifetimes are significantly reduced within a shallow layer near the surface, due to chlorine atoms from Cl_2 snow emissions and increased HO_x attributable to halogen chemistry.