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Reactive chlorine in the marine boundary layer in the outflow of polluted continental air: a model study

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Chlorine in the MBL is important for the oxidation of volatile organic compounds, the acidity budget, and the sulfur cycle. There is an increasing amount of indirect and direct measurements of inorganic chlorine compounds (e.g., HCl, Cl_2, Cl, HOCl) in the marine boundary layer (MBL). We present results from a state-of-the-art one-dimensional computer model that are based on data collected during the CHAiOS/ICARRT campaign on the Isles of Shoales off the New England coast in summer 2004. Our goal is to analyze the chemical processes leading to the release of chlorine, the resulting mixing ratios in the MBL and consequences for tropospheric chemistry.

With our model we can reproduce Cl atom concentrations on the order of 1.0 $\times 10^5$ cm^{-3} to 1.7 $\times 10^5$ cm^{-3} which is in the range of indirectly measured Cl concentrations during that campaign. These numbers suggest a substantial contribution of daytime Cl-radical chemistry to the oxidation of volatile organic compounds under these conditions.

As expected, continental pollution that is advected over the ocean leads to substantial release of HCl by acid displacement from newly emitted sea salt aerosol. We could show that oxidation of the HCl by OH and cycling between the gas and aerosol phase lead to a build-up of nighttime Cl_2 mixing ratios of up to 90 pmol mol^{-1} in our base scenario and 125 pmol mol^{-1} in a sensitivity study. Therefore the recirculation of continental airmasses over the ocean and back to the coast might explain previous measurements of high nighttime Cl_2 in onshore winds (Spicer et al., 1998).