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## The reaction of HCl with calcite particles in the context of stratospheric aerosol injection

### Content

Climate intervention by stratospheric aerosol injection (SAI) has been proposed as a technique, which could rapidly and at low costs mitigate some of the adverse effects of climate change. The idea of SAI is to inject aerosols into the stratosphere to cool the climate by increasing the Earth's albedo, thereby reducing some risks which arises due to climate change. Submicron  $\text{CaCO}_3$ -particles have been proposed as an emission species due to their potential positive effect on stratospheric ozone (Keith et al., 2016). Calcite particles could react with acids such as HCl by creating  $\text{CaCl}_2$  on the surface, which could result in de-chlorination of the stratosphere and thus in increased ozone concentrations and a healing of the Antarctic ozone hole.

However, the interaction of calcite particles with stratospheric acidity (i.e., HCl,  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$ ) at stratospheric temperatures ( $< 220\text{ K}$ ) and relative humidity ( $< 1\%$ ) is poorly constrained, resulting in high uncertainties for the response of stratospheric chemistry. Following up from previous literature indicating a strong dependence of HCl uptake on  $\text{CaCO}_3$  particles on relative humidity (e.g., Huynh et al., 2020), the goal of this experiment is to measure cumulative depth profiles of the chemical composition of calcite particles exposed to stratospheric partial pressures of HCl in-situ under presence and absence of water at stratospheric temperature to quantify the magnitude and the efficiency of HCl uptake and thus the impacts on stratospheric chemistry.

We were capable to measure the initial uptake of HCl on a  $\text{CaCO}_3$  powder sample live and in situ over the course of two hours (Fig 1). The measurement conditions were 10% relative humidity (one order of magnitude higher compared to the stratosphere) and  $\text{pHCl}$  of  $1.25 \times 10^{-5}$  (two orders of magnitude larger compared to the stratosphere) at 250K (30K above stratospheric temperatures). The binding energies of the Ca 2p<sub>3/2</sub> peak and the Ca 2p<sub>1/2</sub> could be determined to 199.7eV and 198.1 eV, respectively, which is in agreement with literature. After this first success we measured at 4 different probing depths at 4 different conditions at 250 K: 1)  $\text{RH}=0.1\%$ ,  $\text{pHCl}=1.3 \times 10^{-4}$ , 2)  $\text{RH}=0.1\%$ ,  $\text{pHCl}=2.3 \times 10^{-5}$ , 3)  $\text{RH}=0.7\%$ ,  $\text{pHCl}=2.3 \times 10^{-5}$  and 4)  $\text{RH}=0.7\%$ ,  $\text{pHCl}=4.5 \times 10^{-4}$  (Fig 2). At 200eV kinetic energy, the most surface sensitive conditions, measured at the largest  $\text{pHCl}$  and  $\text{RH}$ , the largest increase of signal from chlorine Cl 2p was observed. However, also O 1s and C 1s were more significant than at the other kinetic energies. This indicates contributions from hydrates and possible dissolution of  $\text{CaCl}_2$  at the surface. Looking at the phase diagram of  $\text{CaCl}_2$  as a function of T and RH,  $\text{CaCl}_2$  hydrates can be observed already at very low RH1. This indicates, that measuring at lower temperatures is crucial to prevent the formation of hydrates.

Results of an upcoming continuation beamtime in which we will perform measurements at stratospheric temperatures will also be presented at the workshop.

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