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Uptake and reaction of HOBr on frozen and dry NaCl / NaBr surfaces between 253 and 233 K

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Abstract. The uptake and reaction of HOBr with frozen salt surfaces of variable NaCl / NaBr composition and temperature were investigated with a coated wall flow tube reactor coupled to a mass spectrometer for gas-phase analysis. HOBr is efficiently taken up onto the frozen surfaces at temperatures between 253 and 233 K where it reacts to form the di-halogens BrCl and Br_2, which are subsequently released into the gas-phase. The uptake coefficient for HOBr reacting with a frozen, mixed salt surface of similar composition to sea-spray was <approx> 10^{-2}. The relative concentration of BrCl and Br_2 released to the gas-phase was found to be strongly dependent on the ratio of Cl^- to Br^- in the solution prior to freezing / drying. For a mixed salt surface of similar composition to sea-spray the major product at low conversion of surface reactants (i.e. Br^- and Cl^-) was Br_2.

Variation of the pH of the NaCl / NaBr solution used to prepare the frozen surfaces was found to have no significant influence on the results. The observations are explained in terms of initial formation of BrCl in a surface reaction of HOBr with Cl^-, and conversion of BrCl to Br_2 via reaction of surface Br^-. Experiments on the uptake and reaction of BrCl with frozen NaCl / NaBr solutions served to confirm this hypothesis. The kinetics and products of the interactions of BrCl, Br_2 and Cl_2 with frozen salt surfaces were also investigated, and lower limits to the uptake coefficients of >0.034, >0.025 and >0.028 respectively, were obtained. The uptake and reaction of HOBr on dry salt surfaces was also investigated and the results closely resemble those obtained for frozen surfaces. During the course of this study the gas diffusion coefficients of HOBr in He and H_2O were also measured as (273 ± 1) Torr cm^2 s^{-1} and (51 ± 1) Torr cm^2 s^{-1}, respectively, at 255 K. The implications of these results for modelling the chemistry of the Arctic boundary layer in springtime are discussed.

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