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## Hydrochloric acid droplets on ice crystal surfaces embedded in ice by vapor growth

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Ice crystal surfaces are covered with quasi-liquid layers (QLLs) and it leads great influence on the global environment whether ice surfaces are dry or wet. For example, since QLLs enhance various chemical reactions in ice clouds, the formation of QLLs by atmospheric gases has been studied intensively. In particular, chemical reactions on ice surfaces in the presence of hydrogen chloride (HCl) gas play important roles in ozone-depleting reactions [1].

However, such studies were performed using spectroscopy techniques, which did not have enough spatial and temporal resolution. After that previous method would not reveal the dynamic behavior of individual QLLs because we revealed by advanced optical microscopy that the QLLs are not static complete wetting but dynamic partial wetting on ice surfaces [2]. Therefore, we show direct visualization of QLLs on ice basal faces in the presence of 0.1% HCl gas by advanced optical microscopy.

We found that the HCl gas induced the appearances of liquid layers with a droplet shape in the temperature range of  $-15.0 \sim -1.5$  degC, where no QLL appears in the absence of HCl gas [3]. However, note that since the lowest temperature of our experimental setup is  $-15.0$  degC, the lowest temperature for the appearance of the droplet QLLs is still unknown. We concluded that the droplets were not bulk pure water but HCl solution because we observed various features of the HCl-induced droplets as described below, which were not observed in the absence of HCl gas.

Under undersaturated condition, the HCl droplets were irregularly moving and broke into smaller ones [3]. Because the HCl droplets could be assumed to be equilibrium with ice crystals, the HCl concentration is adjusted by melting and freezing of ice crystal so that its melting point equals to the ice temperature. Therefore, these phenomena are closely related to the evaporation of the HCl droplets and subsequent melting of ice crystals caused by the increase in the HCl concentration.

Under supersaturated condition, the HCl droplets were quickly embedded into ice crystals because growth of ice crystals preferential started from the droplets surface and spread in the horizontal direction. After that, when we gradually decreased the water vapor pressure to undersaturated condition, the embedded droplets reappeared on ice crystal surfaces by the evaporation of ice. The HCl droplets were equilibrium with ice crystals around the droplets because reappearance droplets were same size as embedded droplets. Therefore, when the temperature of ice crystals were  $-10$  degC, the HCl concentration of the embedded droplets were estimated at 7 wt.% by the melting point of HCl solution.

The new incorporation mechanism of HCl gas into ice crystals suggested the possibility that natural ice crystals also include soluble atmospheric gases. For example, this mechanism might explain a large amount of chloride ions stored in the polar stratospheric clouds that cause ozone depletion.

[1] McNeill et al. (2006) PNAS 103, 9422.

[2] Sazaki et al. (2012) PNAS 109, 1052.

[3] Nagashima et al. (2016) Cryst. Growth Des. 16, 2225.

### Significance statement

We succeeded in direct visualization of HCl-induced droplets on ice basal faces in the temperature range of  $-15.0 \sim -1.5$  degC by advanced optical microscopy. Because the HCl droplets were embedded in ice crystals by vapor growth, natural ice crystals might incorporate soluble atmospheric gases by this mechanism.

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