

## Surface melting on ice surfaces visualized by advanced optical microscopy

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Ice is one of the most abundant crystals on the earth, and hence its phase transitions exert enormous influence on the global environment. Surface melting of ice crystals is one of such phase transitions: ice crystal surfaces are covered with thin liquid layers, so called quasi-liquid layers (QLLs), even below the melting point (0°C). Since QLLs enhance various chemical reactions in ice clouds, presence or absence of QLLs is also important for atmospheric chemistry.

Recently, we and Olympus Engineering Co., Ltd. have developed laser confocal microscopy combined with differential interference contrast microscopy (LCM-DIM) by which we succeeded in the direct visualization of 0.37-nm-thick elementary steps [1] and QLLs [2] on ice for the first time. The direct visualizations revealed that QLLs with two types morphologies partially cover the ice surface [2,3] although QLLs had been thought to cover the whole ice surface uniformly. And we found that the appearance of QLLs depended on not only the temperature but also water vapor pressures [4,5] and species of ambient gases [6].

For example, we found that hydrogen chloride (HCl) gas strongly induced the appearances of QLLs. The droplet shape QLLs were observed in the temperature range of -15.0 ~ -1.5°C, where no QLL appears in the absence of HCl gas [6]. These results indicate that HCl gas adsorbed on ice crystal surfaces probably changed the surface structure of ice crystals and then induced the subsequent melting of ice surfaces. The long-term (one-hour) existence of the droplet QLLs under the undersaturated conditions and their behaviors strongly suggest that the droplet QLLs were thermodynamically-stable HCl solutions. In addition, we found that the HCl induced droplets were embedded into ice crystals by growth of ice crystals and the embedded droplets appeared again by evaporation of ice crystals. These results show the possibility that ice crystals can store large amount of gas components as fluid inclusions.

[1] Sazaki et al. (2010) *PNAS* **107**, 19702.

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

[3] Asakawa et al. (2015) *Cryst. Growth Des.* **15**, 3339.

[4] Asakawa et al. (2015) *PNAS* **113**, 1749.

[5] Murata et al. (2016) *PNAS* **113**, E6741.

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

Keywords: Ice, Surface melting, Quasi-liquid layer, Hydrogen chloride gas, Advanced optical microscopy