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Mechanism of single ice crystal growth in mixed clouds

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ABSTRACT

At the Desert Research Institute, Reno, NV, USA, we investigated atmospheric ice crystal initiation, growth and interaction. We developed a mechanism of ice crystal growth in mixed clouds that is consistent with basic chemistry and physics, observations by others, and numerical studies of ice crystal growth from the vapor phase. The mechanism may provide insight into ice crystal morphology and charge separations in ice crystals growing in mixed clouds.

1. Introduction

By understanding the mechanism of atmospheric ice crystal growth, we expect to learn more about the nature of ice crystal growth and crystal growth in general. The focus of this article is to develop a mechanism of ice crystal growth in mixed clouds.

One archetype of a mixed cloud involves supercooled droplet concentrations of the order of 100 cm^{-3} as compared to ice crystal concentrations of the order of one per liter. The droplets in high concentration relative to ice crystals serve as a vapor reservoir, maintaining the environmental vapor density near water saturation in opposition to the effects of updrafts, downdrafts, and entrainment of dry air.

Growth of ice crystals in mixed clouds may be compared with and contrasted to droplet condensation in a supersaturated environment. Theoretical development of droplet condensation growth considers the balance of heat and mass between the droplet and its environment. Condensation of vapor to the droplet releases latent heat, which acts to warm the droplet. Convective diffusion of heat away from the droplet removes sensible heat and acts to cool the droplet. When the droplet grows in a supersaturated environment, a balance is achieved whereby the heat flux to the droplet from condensation balances the heat flux from the droplet from convective diffusion.

Theory for ice crystal growth in a mixed cloud is more complicated than theory for droplet condensation growth because: (1) the ice crystal growth process involves transition from the vapor to the solid phase, and consequently involves two phase changes, condensation and freezing; (2) ice crystal geometry is non-spherical and highly variable among ice crystals; (3) as ice crystals grow larger, they evolve from faceted hexagonal prisms to shapes that include needles, sector plates, and dendrites; and (4) there is a strong dependence of ice crystal habit on temperature at water saturation, with three transitions between *a*-axis growth (plates) and *c*-axis growth (columns) between 0 °C and −30 °C.

The lack of geometric symmetry and the need to consider two phase changes both complicate development of a mechanism of ice crystal growth in mixed clouds. The next section presents observations that were used to develop the mechanism.

2. Background

Studies of ice crystal habits (Kobayashi, 1961; Rottner and Vali, 1974) showed that temperature is the major driving force of habit, since it switches from plates (*a*-axis growth) above −4 °C to columns (*c*-axis growth) between about −4 and −8 °C, then back to plate-like growth between about −10 C and at least −18 °C, with columnar growth occurring again at lower

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temperatures. Whether crystals grow as columns or plates, the common property they have is a large, flat non-growing or slowly growing facet and a relatively small fraction of the surface where the ice crystal lattice is growing.

Pruppacher and Klett (1978) summarized results of dimensions of atmospheric plate-like ice crystals observed by several researchers (their Fig. 2–30), and provided a power–law relationship for c/a versus a. Larger plate-like ice crystals exhibit smaller c/a ratios, meaning that, as they grow, the population of plate-like ice crystals grows predominantly along their a-axes. They presented similar summary of dimensions of natural columnar ice crystals in Fig. 2–31. Larger columnar crystals exhibit larger c/a ratios, meaning that their growth is predominantly along their c-axes. The power–law relationships are regression lines to data obtained by several researchers under a variety of atmospheric conditions, classified into separate regimes for columnar and plate-like growth. The ice crystals sampled in snowfall by various researchers not only grew under a variety of conditions, but likely also experienced significant fall time outside the mixed cloud they grew in. The power–law relationships are therefore considered as guidelines of how atmospheric ice crystals appear at the surface.

Mason et al. (1963) reported hopper-structure crystal surfaces during growth or evaporation, and suggested that, for some growth conditions, neither the basal nor prism faces of crystals grow as flat surfaces. They found that ice crystal growth occurs by propagation of successive steppe-like ridges under appropriate conditions. The hopper growth mechanism requires vapor to predominantly condense at the tips and migrate to the growing steppe, where molecules prefer to freeze in to the ice lattice and thereby propagate the steppe front. When steppes propagate too far from the edge, new steppes are formed. They postulated that the steppe formation and propagation depended on average condensed water molecule migration distances on the different ice crystal surfaces.

Some proposed ice crystal growth mechanisms at low vapor supersaturations have focused on surface kinetics and morphological instability. Kuroda and Lacmann (1982) combined properties of quasi-liquid layers on ice surfaces with concepts of ice crystal molecular steppes to develop kinetic rates of crystal growth. This mechanism does not have bulk liquid water present anywhere on the crystal surface. Gonda (1976, 1980) studied morphology dependence on vapor and heat diffusion, and Kuroda (1989) investigated morphology dependence on surface kinetics and summarized studies to characterize morphological instability of ice crystals.

Lamb and Hobbs (1971) studied growth rates of basal and prism faces of ice crystals under conditions where crystal growth was controlled by molecular events on the crystal surface, rather than by the rate of supply of vapor from the environment or the rate by which heat is removed. Their results, which reported prism face growth rates in excess of basal face growth rates above −5.3 °C and between about −9.5 and −22 °C, are consistent with observations of habits of natural ice crystals.

When ice crystals grow slowly, such as on substrates and with low vapor density excess, the conditions resemble those studied by Lamb and Hobbs and the growth mechanism is reflected in the rates of growth of the basal and prism faces. This may be the mechanism that governs ice crystal habit in the atmosphere. Yet, because their study involved both slow growth conditions and a relatively high rate heat conduction to the substrate, we suggest that additional studies are necessary to confirm its relevance to the atmospheric case.

Finnegan and Pitter (1988) reported observations of ice crystals growing in free fall in a mixed cloud within the DRI 6.7 m^3 cloud chamber. Microphotographs of ice crystals grown in free fall in the presence of supercooled water droplets (Finnegan et al., 1991) sometimes exhibited shadows that might be interpreted as steppes or ridges, but crystals photographed on edge revealed that the steppes occurred in the concavities, between two flat outer basal planes of plates, or near the tips, inside the flat prism faces of columns, rather than on the major outer surfaces of the crystal.

In concurrence with the observations reported by Mason et al. (1963), Finnegan et al. (1991) found hopper-like growth on ice crystals grown between −8 and −9 °C, when axis ratios were near the equilibrium value ($c/a = 0.8$). These ice crystals grew much more slowly while falling within a mixed cloud. Some of the ice crystals had concavities on both their basal plane surfaces and their prism face surfaces.

Nelson and Baker (1996) developed a theoretical model for ice crystal growth and sublimation. They postulated the vapor flux to be uniform over each flat crystal surface, and the surface water molecule movement limited to a few micrometers. Their interest was to formulate the processes that influence ice crystal shapes in ice clouds.

Nelson and Knight (1998) studied ice crystal growth on substrates. They observed the growth of basal and prism faces of ice from the vapor. For growth at low supersaturations with respect to ice, they observed non-faceted growth at all temperatures, which they attributed to be due to layer nucleation.

Studies investigating the nature of ice crystal growth and aggregation were conducted in the DRI 6.7 m^3 cloud chamber. Each experimental run was isothermal: the chamber was controlled at the same temperature throughout and the temperature was held constant through the run. The studies involved generation of a cloud of water droplets by ultrasonic nebulization of a dilute solution, typically 10^{-6} N, of a soluble salt, such as sodium chloride (NaCl) or ammonium sulfate ($(NH_4)_2SO_4$). Ice crystals were initiated in the chamber by adiabatic expansion of moist air. Ice crystals grew for several minutes in an isothermal mixed cloud. Microphotographs were taken of ice crystals that landed on chilled microscope slides in the chamber. Finnegan and Pitter (1988) observed a high incidence of T-shaped aggregates, where two ice crystals were joined with the tip of one crystal fused near the center of the other, and often the c-axes of the two crystals formed a right angle at the junction.

Dr. Finnegan recounted studies of ice crystal formation from supercooled fog that conducted in near-pristine conditions during the Yellowstone Experiment. When Vincent Schaefer use dry ice for ice nucleation, single ice crystals grew to large sizes without aggregation and produced spectacular optical (halo) effects. However, when Finnegan used flame-generated silver iodide aerosols ($2AgI \cdot NaI$) for ice formation, rapid aggregation followed and the halo effects were not observed. Moreover, the crystal aggregates were strongly oriented point-to-center (T-shaped).