Enhanced and Oriented Riming of Growing Ice Crystals

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ABSTRACT

Geometrically oriented riming was found in Formvar resin replicas of columnar ice crystals collected in cumulus clouds at −6°C during an aircraft field program in Texas. Rimed cloud droplets were found either on the ends of the crystals or in a girdle around the middle. Oriented riming is attributed to preferential collection on growing ice crystals with charge separations between the crystal body and growing ends. Droplet attraction to separated charge regions of growing ice crystals results in enhanced riming and increases the rate of precipitation development. Effects of this process on cloud electrification depend on whether the cloud droplets carry net charges or are polarized. The impact of this oriented riming process on several cloud electrification scenarios is discussed.

1. Introduction

Laboratory studies have documented the existence of electric charge separations within growing single ice crystals containing low concentrations of atmospherically common ionizable salts. Growing ice crystals with charge separations obviously play a major role in many atmospheric processes. In precipitation development, the chain of events that occur between initiation of the ice phase and rain or snow on the ground is not well documented (Reynolds 1988; American Meteorological Society 1998a,b). An understanding of this chain of events is of importance in both natural and induced precipitation processes and is the major rationale for the continued interest in ice crystal processes. Further studies will, of necessity, have to include further field studies; laboratory studies are unable to adequately address events beyond simple two-crystal aggregation.

2. Background

Dilute supercooled solutions of certain inorganic salts develop electric charge separations between the ice and remaining liquid during freezing (Workman and Reynolds 1950). Cloud chamber experimentation has shown that growing single ice crystals that contain low concentrations of these inorganic salts also have electric charge separations (multipolar moments) between the crystal body and growing crystal ends or edges on which there are liquidlike layers (Finnegan and Pitter 1988). This was evidenced by observation of substantial concentrations of geometrically oriented, two-crystal, point-to-center (with 90° angles) aggregates in the experiments. When ice crystals formed in a cloud in which droplets contained sodium sulfate (a salt that does not induce a charge separation during bulk freezing), little or no aggregation was observed. When two nebulizers were used to generate a mixed cloud in the cloud chamber (half the droplets containing a salt that induces a negative charge on the ice side of the ice–solution interface on freezing, and half containing a salt that induces positive charge on ice during solution freezing), then both point-to-center and point-to-point aggregates (with 90° angles) were observed. These experiments confirmed that charge separations occur in growing single ice crystals containing appropriate salts, and that ice crystal aggregation is promoted by charge separations.

Further studies revealed that charge separations influence the morphology of growing ice crystals and promote secondary ice nucleation (Pitter and Finnegan 1990) and, through electron transfer processes, lead to reduction–oxidation (REDOX) reactions of included ions and the water–ice phases themselves (Finnegan et al. 1991, 2001; Pitter et al. 2003). These results concerning nucleation and chemical reactions further confirm the existence of charge separations within growing single ice crystals and illustrate the wide range of effects that result.

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The added ion inclusion mechanism hypothesized to lead to the formation of charge separations during bulk linear freezing of dilute salt solutions, as advocated by Workman and Reynolds (1950), is inconsistent with more recent observations. Inclusion of many different inorganic ions into the ice phase during solution freezing or single crystal growth is improbable, given the observed rejection of inorganic salts by the ice phase during freezing of aqueous solutions. An analysis of solution bulk freezing data established that the charge separation process is controlled by the pH of the freezing solutions or of the liquid or liquidlike layers at the growing ends or edges of single crystals (Finnegan and Pitter 1997). For example, freezing of basic solutions results in formation of charge separations with the ice phase negative. Freezing of acidic solutions results in positively charged ice on freezing; the remaining solution becoming negative. Sodium chloride is a salt of a strong acid and strong base, and its solutions are neutral. Nevertheless, the presence of dilute solutions of sodium chloride yields negatively charged ice during freezing. This occurs because the chloride ion Cl^- is oxidized to the hypochlorite ion OCl^- during freezing or the crystal growth process. Hypochlorous acid is a very weak acid, and solutions of its sodium salt are strongly basic. Charge separations disappear when the solutions are completely frozen or growth of the ice crystals ceases. Charge separations do not occur if salt concentrations are greater than about 10^{-3} molar.

As supported by these cloud chamber studies, it is reasonable to assume that charge-separation-induced processes of enhanced aggregation, crystal morphological changes, secondary nucleation of ice crystals, and chemical reactions also occur in the atmosphere. Conditions similar to those simulated in the chamber often are observed in natural clouds. Many of the salts that induce charge separations during freezing of dilute solutions [including NaCl, $(NH_4)_2SO_4$, and $CaCO_3$] are common cloud condensation nuclei. Field verification of the cloud chamber experiments is described below.

3. Field program

A field program for the Texas Natural Resources Conservation Commission was conducted in the plains of west Texas during August 1999. The major goal of the program was to gather evidence on the influence of inorganic salts in cloud water on cumulus cloud precipitation processes. Operational objectives of the field program were to (a) verify the existence of charge separations in growing single ice crystals in young cumulus clouds not influenced by cloud seeding operations, (b) collect water samples from these clouds, and (c) analyze the samples for ion species and their concentrations.

The aircraft used for the program was a U.S. Navy S-2 Tracker under contract to the Desert Research Institute. Eleven research flights were conducted between 5 and 25 August 1999. Multiple penetrations of three or more clouds were performed during each flight. Ice crystal data was collected from 10 separate clouds during six of the research flights. Ice crystals were sometimes encountered in environments where they were not growing, and the effects on multipolar charge separations were not always evident. Cloud water samples were collected on riming rods only at $-6°C$ (due to aircraft performance limitations and high cumulus cloud bases in west Texas in August). Ice crystal replicas were obtained using a Formvar resin-coated moving film instrument at the aircraft's cruising speed (Turner 1996).

4. Field data analysis

The concentration of ice crystals was low in the young cumulus clouds in this study. Therefore, there were few aggregates and little data from these flights to evaluate the effects of multipolar charge distribution on ice particle aggregation. On the other hand, there were many geometrically oriented rimed columns and prisms on which the rime ice appeared preferentially on the ends or middle of the crystals (Figs. 1 and 2).

Analysis of collected cloud water samples (Fig. 3) showed that inorganic salts that induce the Workman–Reynolds ''effect'' of charge separation during freezing were present in appropriate concentrations of 10^{-5} to 10^{-4} molar. Analyses were conducted using ion chromatography for the anions; atomic absorption spectrometry for sodium, calcium, and magnesium cations; and autocolorimetry for the ammonium ion. Based on ion species detected, the cloud water samples contained sodium chloride NaCl, calcium carbonate $CaCO_3$, and occasionally ammonium sulfate $(NH_4)_2SO_4$, and magnesium carbonate $MgCO_3$. The presence of sodium chloride and two alkaline earth carbonates in growing ice crystals results in the acquisition of a negative charge by the central body of the ice particle while the growing ends or edges carry a positive charge. Due to their acidic nature, $(NH_4)_2SO_4$ solutions yield positively charged ice with negatively charged ends or edges.

The presence of charge separations in growing ice crystals in the atmosphere is hypothesized to account for orientation of the riming. Understanding the mechanism or mechanisms producing oriented riming requires consideration of the net charge and polarization of cloud droplets riming growing ice particles.

5. Discussion

Ice crystals falling through a supercooled water droplet region grow by vapor deposition and riming. Rapidly growing ice crystals develop charge separations if their formation is initiated by freezing of cloud droplets containing appropriate concentrations of soluble, ionizable salts that promote freezing potentials. Alternatively, freezing potentials may develop in growing crystals rimed by cloud droplets containing these salts. Neither of these charge separation mechanisms has been con-

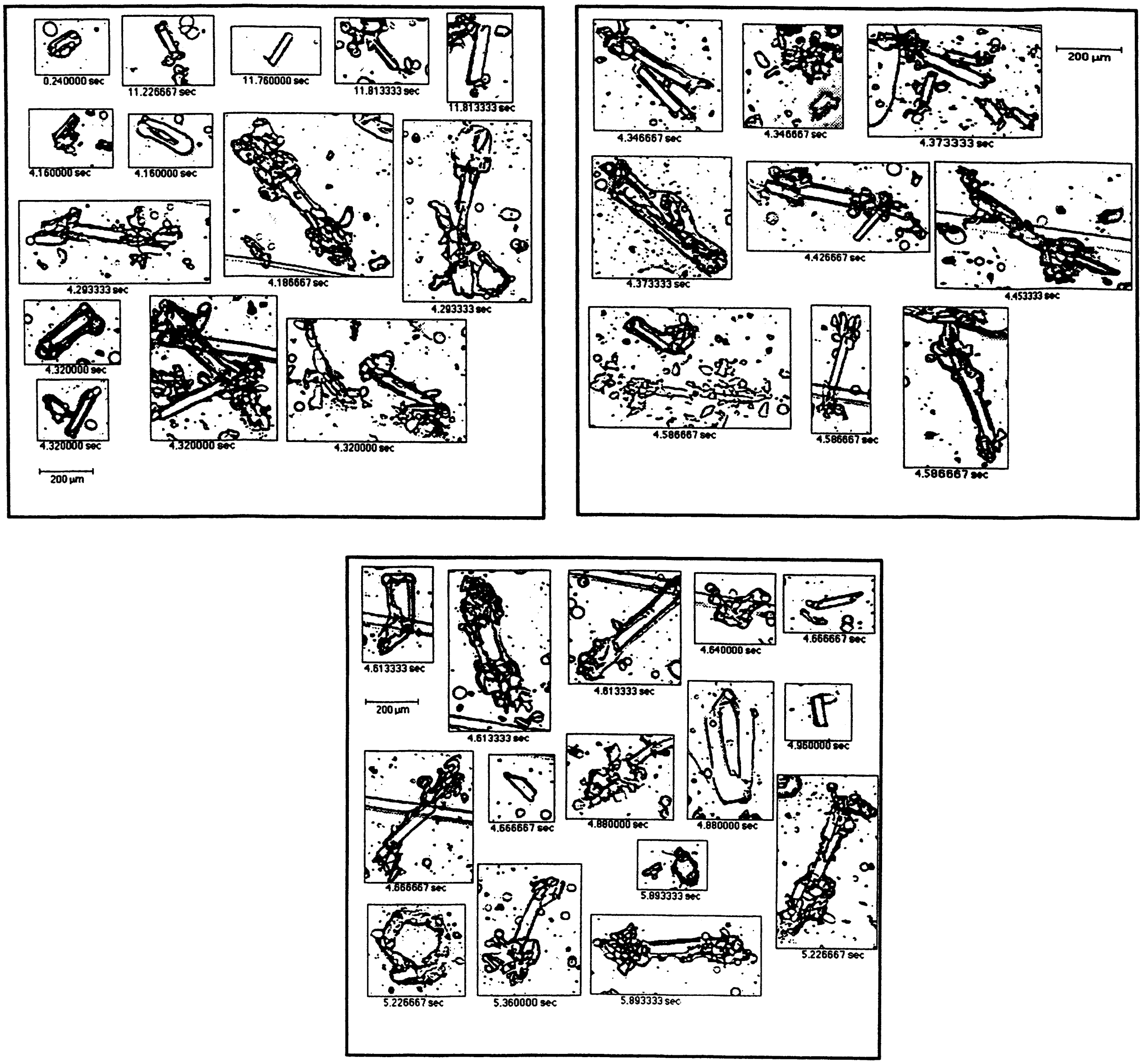

FIG. 1. Examples of geometrically oriented rimed ice crystals from a Texas field program (Huggins et al. 2000).

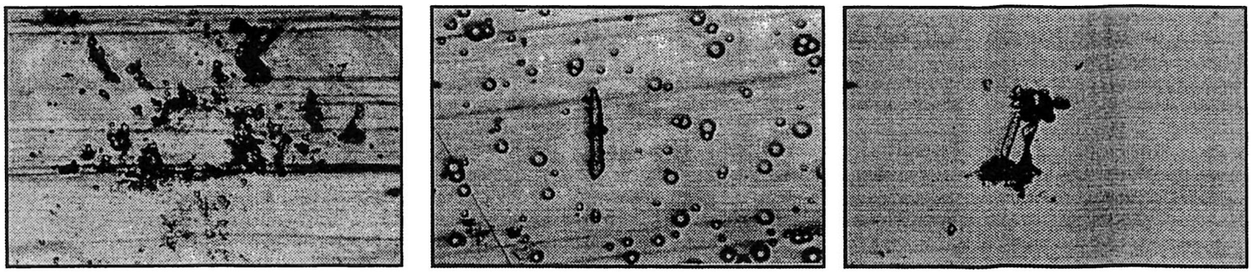

FIG. 2. Geometrically oriented rimed ice crystals showing a column with rime drops attached either (left and center) near the center or (right) at both ends (Huggins et al. 2000).

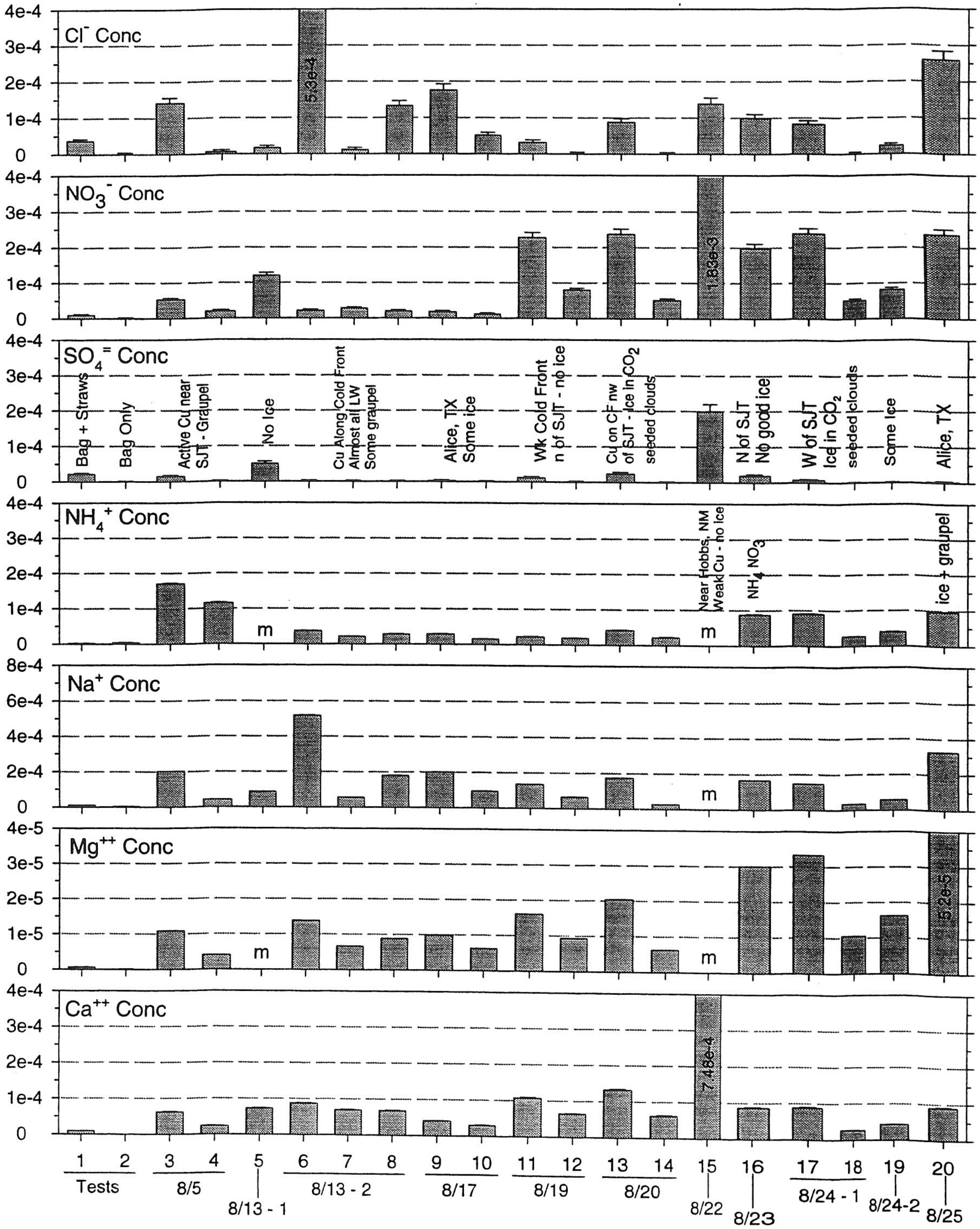

FIG. 3. Concentrations of seven ions found in 18 cloud water samples (Nos. 3–20) collected during a Texas field program (Huggins et al. 2000).

sidered in any published cloud microphysical or atmospheric electrical studies. Further, the possible impact of these multipolar moments on rates of aggregation, riming, and cloud electrification have not been considered in any model simulations. The following processes may occur due to multipolar moments in the interaction of charge-separated, growing ice crystals and supercooled droplets.

1) Shattering and splintering of ice crystals during riming have been suggested as a secondary ice formation mechanism (Hallett and Mossop 1974; Mossop and

Hallet 1974). When a charge-separated, growing ice crystal undergoes riming under conditions where rime fragments are breaking off, the fragments are hypothesized to be charged. Those breaking off from the tips would be charged oppositely from those breaking off from the centers, and the tip fragments would predominate in number. The growing crystal would then have a net charge opposite that of the splinters and would fall relative to the splinters, resulting in vertical charge separation. The sign of this vertical separation would depend on the chemical composition of the ice particles and riming droplets.

2) Sartor (1954) suggested that cloud droplets are polarized by the ambient electric field. Under this hypothesis, charge-separated, falling crystals would have an increased chance to collide with polarized cloud droplets (compared to unpolarized ones). Most cloud droplets would remain frozen to the ice particle after the collision, and a geometrically oriented rimed ice particle might develop. This mechanism would not change the net charge on the ice particle, improve the efficiency of future ice and cloud droplet collisions, or enhance cloud electrification. Since the mechanism would remove polarized cloud droplets more efficiently, however, it might slow cloud electrification via other mechanisms involving polarized cloud droplets. Enhanced collision efficiency between polarized cloud droplets and charge-separated, falling crystals also might result in a higher rate of splinter production that could enhance cloud electrification.
3) If ice and liquid water particles separate after collision in the presence of a downward-directed electric field (positive charge above, negative charge below), then negative charges on the upper part of the polarized droplets would be transferred onto the ice crystals to neutralize positive charges, leaving the ice particles negatively charged. This mechanism would leave the droplets positively charged. Since the droplets would most likely be carried by updrafts to the upper levels of the cloud, this inductive charging mechanism would enhance cloud electrification. Positive charges would build in the upper levels of the cloud and negatives changes in the lower half.
4) Grenet (1947) and Vonnegut (1953) independently conjectured that fair weather positive space charge is carried by updrafts into the bases of convective clouds, and Vonnegut et al. (1962) demonstrated the validity of this theory. The positive space charge attaches to cloud droplets nucleating at cloud base and growing in the updraft, and causes them to become positively charged. Numerical simulations of this process suggest that the mechanism is weak and unlikely to lead to sufficient charge separation to initiate lightning within the lifetime of a typical thunderstorm. (Chiu and Klett 1976; Helsdon et al. 2002). However, in the mixed phase region of clouds above the freezing level, there will be enhanced collision efficiency between positively charged droplets and negatively charged regions of the charge-separated ice crystals growing there. Geometrically oriented rimed ice particles should be observed. The net positively charged rimed ice particles, relative to cloud droplets, will fall, increase positive charge concentration toward the base of the cloud and accelerate vertical charge separation within the cloud. This mechanism should be included in numerical models of thunderstorm electrification in the future. It may be shown that convective electrification is a stronger mechanism than it has heretofore been understood to be.

These mechanisms may enhance or slow precipitation growth, cloud electrification, or both. All four involve interaction of cloud droplets and charge-separated, growing ice crystals; but two of the four likely will result in the formation of geometrically oriented rimed ice particles. Since the Texas experiments showed that cloud water in the region mainly contains NaCl and $CaCO_3$, negatively charged ice with positively charged liquid growing ends or edges would be expected. Under these chemical conditions and the second mechanism (collection of neutral but polarized cloud droplets by neutral but multipolar ice crystals), riming would occur on the growing ends or edges of ice crystals. Under the same conditions and the fourth mechanism (collection of positively charged droplets by neutral but multipolar ice crystals) riming would form around the centers of growing ice crystals containing NaCl or $CaCO_3$. For columns, the rime would form a girdle around the center.

6. Conclusions

A field program was conducted in Texas to obtain information on oriented aggregation of growing ice crystals in convective cumulus clouds. Rime ice was collected and analyzed for the presence of inorganic salts that induce potential differences between ice and liquid phases during freezing of dilute solutions. Results of this program supported laboratory studies on the existence of charge separations in growing single ice crystals. Specifically, analysis of cloud water samples confirmed the presence of inorganic salts with appropriate compositions and concentrations to induce charge separations in growing ice crystals.

Numerous columnar prisms with oriented riming also were observed at −6°C. The rime ice was primarily at the ends of the columns, but some crystals were observed with the rime ice in a girdle around the middle of the column. Given the conclusion that charge separations exist in growing single ice crystals, the presence of enhanced and oriented riming suggests that formation of electrical multipoles on growing ice crystals might play a role in cumulus cloud electrification. Four mechanisms that may influence charge separations within cumulus clouds are suggested.

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