FIELD OBSERVATIONS OF ICE CRYSTAL FORMATION IN CLOUDS AT WARM TEMPERATURES

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ABSTRACT. A field study of the effect of treating a shear line convective cloud with a 20% aqueous solution of ammonium carbonate is described. The results indicate that the presence of certain soluble salts in growing ice crystals may be sufficient to initiate ice crystal multiplication in clouds at temperatures of -4 to -5 C, as measured by a Rosemount instrument, or -3 C, as measured by a reverse flow thermometer. The field results are consistent with laboratory experiments conducted in the Desert Research Institute's cloud chamber, where ice multiplication was observed at temperatures as warm as -4 C. In the cloud chamber experiments, ice multiplication only occurred when crystals were growing rapidly and aggregating. Aggregation was more readily observed when certain soluble salts were added to the water which formed the supercooled cloud. A postulate is advanced as an explanation of the ice multiplication observed in the laboratory and field studies.

1. INTRODUCTION

Weather modification is typically conducted by inducing ice formation in supercooled clouds, either by introducing ice nucleating aerosol particles or by causing homogeneous nucleation with dry ice or liquid propane. A novel departure from this concept, treating clouds with sodium chloride to enhance the formation of large drops, was attempted in the Salt Shaker tests during 1967 (Schock, 1968) and the Cloud Catcher tests from 1969-1972 (Dennis et al., 1974).

Our recent laboratory results revealed an interesting phenomenon occurring in supercooled clouds. Ice multiplication occurred at temperatures from -4 to -30 C, without the presence of graupel particles. The study also found a strong correlation between the soluble salts contained in the water and whether a particular experimental run developed ice multiplication. When multiplication occurred, there was a strong correlation between the salt used and the rate with which ice multiplication occurred. Since the soluble salts were added to distilled, deionized water at 10^{-4} normal concentration, consistent with those found in cloud and precipitation water, we attempted a field experiment to see whether ice multiplication could be initiated in a cloud with a sufficient quantity of an appropriate soluble salt. The field experiment, described in the following section, dispensed a 20% solution by weight of ammonium carbonate [$(NH_4)_2CO_3$] in water into a shear line convective cloud at -4 C. The results, described in Section 3, reveal a high correlation between the detection of the treated volume, using sulfur hexafluoride as a tracer, and the presence of both some large, aggregated ice crystals and many small (less than 100 micrometers long) ice crystals. Section 4 describes the laboratory experiments which led to the field study, and Section 5 presents our postulate of how ice multiplication might have occurred. Section 6 presents a conclusion of the study and its impact.

2. PROCEDURE

During the NOAA-North Dakota field program at Dickinson, ND in June, 1987, arrangements were made for a seeding aircraft to dispense a concentrated solution of ammonium carbonate (20 percent by weight in water) from one wing, using a Lohse acetone-silver iodide generator with the flow restrictor removed. In this manner, the aircraft dispensed a few kilograms of solution on 1 July, 1987 between 11:34:41 and 11:40:24 while flying at -4 C in a cloud formed by shear line convergence. The drop size distribution produced by the pressurized spraying was not measured.

Simultaneously, 22.9 kg of sulfur hexafluoride gas were released from the aircraft to serve as a tracer for the treated volume of air. After dispensing the two agents, the seeder aircraft left the scene and the North Dakota Citation aircraft equipped with cloud physics instrumentation and a Scientech LDS-3 sulfur hexafluoride monitor repeatedly penetrated the cloud in search of the seeding signature.

The cloud selected for the experiment was a weak shear line cloud, selected primarily because of the unavailability of any isolated cumuli clouds during the short field season. Although the selected cloud was not considered ideal from a treatment/effect viewpoint, it allowed excellent repeated examination of the treated cloud volume.

The North Dakota Citation aircraft contained the usual cloud physics instrumentation: temperature, pressure, etc., and a 2D-C probe which was used in the identification of small ice particles.

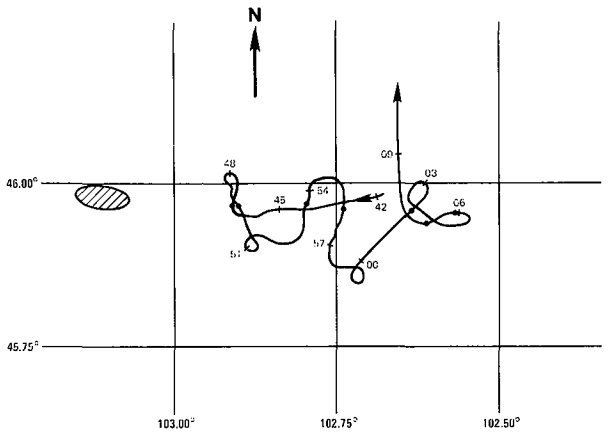
The Citation investigated the cloud from 11:40 to 12:07. In that period, it detected strong pulses of the SF_6 tracer seven times. Although there is an unavoidable separation with time between a gas plume and a plume of water droplets released simultaneously, due to the terminal velocity of the drops, the SF_6 plume served as a tracer for the treated volume of the cloud. Thus, we sought differences in the nature of the ice particles between portions of the cloud tagged with SF_6 and otherwise.

3. FIELD STUDY RESULTS

The flight track (Figure 1) indicates that the seven intersections with the SF_6 plume were detected. The SF_6 monitor is sensitive to parts per trillion concentrations and has a response time of three seconds. Extrapolation of the air parcels where SF_6 peaks were detected backward in time, using the aircraft-computed winds, resulted in placement of the sampled parcels in close proximity to the treated parcel in space and time (shaded oval to west of flight path). In this sense, the selection of a weak shear line was probably optimal for allowing multiple plume intersections.

Table 1 summarizes ice particle sizes and concentrations detected during the experiment. During each of the seven plume intersections, the 2D-C probe detected ice crystals present in concentrations exceeding 1 per liter (as high as 70 per liter). In six of the seven intersections, the average ice particle size was 500 +/- 50 micrometers diameter, while the seventh event exhibited a mean ice particle size of about 1160 micrometers. By comparison, three occurrences of ice particles in excess of 1 per liter were found unassociated with SF_6 peaks. The ice concentrations varied from 1 to 6 per liter in these events, and the mean ice particle diameters varied from 840 to 1730 micrometers, with an average value of 1220 micrometers.

FIGURE 1



Flight track of cloud physics aircraft, with time pips (beginning at 1142 local time and ending at 1209) and circles to indicate where the sulfur hexafluoride plume was detected. The shaded oval to the west indicates where air parcels in which sulfur hexafluoride was detected would be extrapolated to, based on aircraft-detected winds, at the time of the tracer release. This region corresponds with the position of the seeder plane at the time of release.

TABLE 1

COMPARISON OF ICE CRYSTAL CONCENTRATIONS IN AND OUT OF SF_6 PLUMES

IN PLUME

TIME	TEMP. C	L.W.C. g/m^3	ICE CONC. $liter^{-1}$	MEAN ICE PARTICLE DIAM., um
11:46:48	-4.8	0.05	1 - 5	500
11:49:10	-5.0	0.05	35 - 40	500
11:53:26	-4.7	0.06	1 - 5	480
11:55:58	-4.6	0.10	5 - 10	450
12:01:47	-4.6	0.07	15 - 20	450
12:06:02	-4.6	0.12	65 - 70	1160
12:07:01	-5.2	0.00	20 - 25	550

OUTSIDE PLUME

TIME	TEMP. C	L.W.C. g/m^3	ICE CONC. $liter^{-1}$	MEAN ICE PARTICLE DIAM., um
11:51:20	-4.4	0.09	1 - 5	1730
11:56:50	-5.0	0.04	1 - 5	1110
12:04:30	-5.0	0.21	5 - 10	840

The Citation data was first analyzed by Dr. T. Grainger of the University of North Dakota, who brought attention to the strong coincidence of both high ice particle concentrations at -4 to -5 C and smaller particle sizes with the detection of SF_6.

The plume intersections occurred between six and 33 minutes after treatment, yet the ice particles in the treated parts of the cloud averaged about 500 micrometers diameter, independent of elapsed time from treatment. If the ice crystals were nucleated at the time of treatment, then we would expect an increase in size of the ice particles with time. Rather, the 2D images reveal that both larger, aggregated ice crystals and smaller (less than 200 micrometer) ice crystals present.

There is some similarity in the 1987 field experiments and one conducted in 1970, in which one end of a convergence line stratocumulus cloud was seeded with sodium chloride (Biswas and Dennis, 1971). The 1970 experiment released 150 kg of powdered salt just beneath cloud base in the southern part of the cloud. Although some communications (Havens, 1972; Blanchard, 1972) pointed to the complicating facets of prior silver iodide seeding and a cloud top temperature of -10 C, Biswas and Dennis (1972) indicated that the initial silver iodide treatment was ineffective.

The principal difference between the 1970 experiment and the present results lies in interpretation of the cloud physics mechanisms which were operating. Biswas and Dennis (1972) estimated that the salt seeding involved considerably fewer than 7×10^{14} condensation nuclei, while the induced shower consisted of roughly 2×10^{15} raindrops, and thus concluded that some chain reaction (drop breakup) operated in the cloud. They did not penetrate the cloud with aircraft, so they were unable to ascertain that ice was not present. We attribute the present results to ice multiplication, on the basis of the high concentrations of small ice particles at warm temperatures. We suggest that ice multiplication also may have occurred in the 1970 experiments.

The initiation of ice at temperatures warmer than -8 C by heterogeneous nucleation is rare, meteorologically, due to the scarcity of suitable ice nuclei. Silver iodide works poorly at -6 C, and more readily at temperatures of -8 C and below. Treatment with ammonium carbonate was not expected to yield ice forming nuclei. Additionally, studies with gaseous SF_6 dispensing during these NOAA-North Dakota experiments revealed no tendency for the SF_6 to nucleate ice crystals in clouds (personal communication, Don Griffith, North American Weather Consultants). Although the possibility of aircraft-produced ice particles (APIPs) (Rangno and Hobbs, 1983) exists, Rangno and Hobbs found APIPs only at much colder temperatures than were involved in the present study. They noted APIPs at between -12 and -20 C, with one exception at -7 C in descending virga. Their results don't indicate whether or not APIPs might be generated above -5 C.

The field study did not collect enough data to test a hypothesis with statistical rigor. Rather, it attempted to determine whether or not a signal could be sensed. It was designed after laboratory experiments indicated that ice multiplication occurs under conditions of rapid ice crystal growth and aggregation, and is enhanced by the presence of certain soluble salts initially dispensed within the cloud water. The field test demonstrated that higher concentrations of smaller crystals were found associated with the trace gas peaks, although the treatment was conducted with a 20 per cent solution of ammonium carbonate in water, which is not an ice nucleating agent. The following section details the laboratory experiments which led to the field study.

4. LABORATORY EXPERIMENTS

Laboratory experiments were conducted in the Desert Research Institute's 6.7 m^3 cloud chamber, which is described by Steele et al. (1981). The chamber was operated at ambient pressure (about 850 mb) and constant temperature during each experimental run. Temperatures investigated ranged from -2 to -30 C. The chamber contained a supercooled liquid water cloud of 1.5 to 3.0 g m^{-3}, generated by ultrasonic nebulizers, and was then nucleated by adiabatic expansion of moist air, similar to the technique described by Vonnegut (1948).

The water used to form the supercooled cloud was distilled, deionized water or water containing 10^{-4} normal solutions of dissolved salts. The concentrations of sodium nitrate ($NaNO_3$), sodium chloride (NaCl), ammonium sulfate [$(NH_4)_2SO_4$], or ammonium carbonate [$(NH_4)_2CO_3$] used are typical of the ionic concentrations found in clouds and precipitation.

The experimental results indicate that, under appropriate conditions, ice multiplication occurs between -4 and -30 C. The conclusions were based on several independent observations. The observations are as follows:

1. Time of ice presence -- the ice particles grow rapidly and settle out of the limited volume. Depending on the temperature (which controls the habit and drag characteristics), the ice particles fall out of the chamber within 3 to 8 minutes. However, experiments often lasted more than 10 minutes, occasionally more than an hour. One explanation for longer experiments is the action of ice multiplication generating new ice crystals.

2. Laser transmissivity characteristics. Petrushin (1983) noted that non-spherical ice particles scatter electromagnetic radiation of wavelength approximately their diameter much more strongly than other wavelengths. The laser transmissometer used in the experiments for measuring liquid water content (Gertler and Steele, 1980) always responded with increased attenuation just after the initial ice nucleation, as the crystals grew through 10.2 micrometers diameter (the CO_2 laser wavelength). Transmissivity then would normally increase as the liquid water cloud was consumed by the growing ice crystals. However, the longer-lasting experiments exhibited sporadic attenuation pulses in the laser, which were interpreted as effects of populations of newly-formed small ice crystals growing through the 10 micrometer size.

3. Dual-wavelength laser attenuation -- in addition to the CO_2 laser, a 0.55 micrometer wavelength HeNe laser, usually used to aim the CO_2 laser, was employed. During the longer experimental runs, the attenuation pulses of the CO_2 laser were preceded several seconds by similar attenuation pulses in the HeNe laser, indicating that crystals were first growing through the 0.55 micrometer size, and subsequently growing through the 10 micrometer size.

4. Microscopic observation -- crystals which fell onto glass microscope slides typically revealed ice crystals of similar size and shape during short experimental runs, indicating that they were nucleated together and grew at the same rate. During the longer runs, the ice crystal populations were polydisperse, with small crystals always present. This presence of smaller crystals is interpreted as evidence of new nucleation, at a later time, and hence to ice multiplication in the course of the run.

5. Microscopic observation -- signs of ice multiplication were only noted when aggregation of ice crystals also occurred.

6. Temperature and the solute present in the cloud water affected the probability of ice multiplication occurring during any particular experimental run.

Of significant importance, ice multiplication was only noted when existing ice crystals were rapidly growing and aggregating. The soluble salts contained in the supercooled cloud water exerted strong influence on whether or not ice multiplication occurred during any particular experimental run. Ice multiplication was also noted more readily at temperatures close to -6 and -16 C, where thin needles and dendrites are produced. The results were consistent with our previous studies of the factors which give rise to the greatest electric potentials across growing ice crystal interfaces, and led us to postulate that the intense electric fields which could result during the aggregation of such ice crystals might be capable of nucleating new ice crystals. The theory is expanded in the following section.

5. THEORETICAL BASIS

The North Dakota field studies indicate that small ice crystals may be continually generated by some process at warm temperatures due to the presence of the 20% ammonium carbonate solution liquid water plume, as traced by gaseous SF_6. The laboratory studies presented above substantiate the field experiments and further emphasize the effects of certain soluble salts on ice multiplication. The authors know of no existing theory that explains nucleation or ice multiplication under the observed conditions. Therefore, we take the liberty to postulate how ice multiplication may have occurred.

Finnegan and Pitter (1986) postulated the existence of electric multipoles in growing ice crystals due to the differential incorporation rate of anions or cations in the ice crystal matrix, an extension to three dimensions of the commonly-observed freezing potential called the Workman-Reynolds effect (Workman and Reynolds, 1950; Gross, 1968). This effect results in a potential difference of several to several hundred volts across a growing interface of ice and water. The potential difference is dependent on the rate of interface advance and the soluble ions present.

The electric multipole postulate states that the ice phase acquires an electric charge of one sign as a result of differential incorporation of ions, and the liquid-like layer of the growing crystal acquires charge of the same magnitude but opposite sign, principally in the regions where the linear rate of growth of the ice crystal lattice is greatest. When growth ceases, the charge separation collapses due to recombination effects.

Manifestation of the effect of electric multipoles was first realized in the T-shaped aggregates known to be produced under appropriate conditions (Odencrantz et al., 1968; Magono and Tazawa, 1972). Other manifestations of the effect of electric multipole include

enhanced aerosol scavenging by growing ice crystals (Pitter and Finnegan, 1986).

The multiplication of ice in the cloud chamber parallels a phenomenon known in solution crystallization as crystal breeding, or false grain, whereupon colloidal suspensions of high concentrations of growing crystals (i.e., sugar) initiate spontaneous nucleation of new, smaller crystals (Van Hooke, 1961). The mechanism responsible for crystal breeding is unknown, but electrostatic effects are suspected.

6. CONCLUSIONS

We interpret the aircraft data to substantiate our laboratory experiments which indicate ice multiplication occurring under appropriate growth conditions in a manner analogous to crystal breeding or false grain reported in the crystallization literature. An important component in this mechanism is the presence of soluble salts in the cloud water which provide strong freezing potentials. This result, if substantiated, provides a method for modifying clouds which are too warm to treat with silver iodide, and additionally provides a long-lasting, self-perpetuating mechanism for propagating ice.

A link between crystal breeding from solution crystallization and ice multiplication in atmospheric clouds has been postulated, with laboratory and field evidence in support of an effect caused by the presence of soluble salts which result in greater potential differences across growing interfaces of ice. This postulate of a new ice multiplication mechanism requires additional study before it can be established as fact, but our preliminary results are encouraging in this respect.

These results need to be substantiated by further experimentation before the technique is operationally used for weather modification. Potential benefits of the use of ammonium carbonate include (1) ease of dispensing -- the experiment utilized existing silver iodide generator equipment with slight modifications; (2) inexpensive; (3) absence of silver; (4) environmentally safe (ammonium carbonate is a fertilizer); (5) ease of handling (non-toxic); (6) effective at -4 C; and (7) long-lasting, self-perpetuating effect.

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