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Collaborative Research on Aircraft Icing and Charging Processes in Ice

AFOSR-91-0376 (G)

C P R Saunders Department of Pure and Applied Physics UMIST, Manchester M60 1QD, UK

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COLLABORATIVE RESEARCH ON AIRCRAFT ICING AND CHARGING PROCESSES IN ICE

Final Technical Report

AFOSR-91-0376

Period 01 September 1992 - 31 August 1994

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Collaborative Research on Aircraft Icing and Charging Processes in Ice.

Introduction

This contract has permitted detailed considerations of thunderstorm charging processes involving ice-ice contacts resulting in charge transfer. The report comprises a review of thunderstorm electrification, concentrating on particle charging processes, and includes discussions of the recent findings made in the laboratory.

1) Thunderstorm Electrification

The thunderstorm.

Thunderstorms have been of interest and concern for centuries, so it is perhaps surprising that the processes leading to thunderstorm electrification are still under active debate. There is strong evidence that particles in thunderclouds carry electric charges and various charging mechanisms have been proposed. For the electric field to develop in storms, charges of opposite sign must be separated and this may be caused by gravitational forces acting on oppositely charged particles of different size and aerodynamic drag, or by convection currents that transport regions of charge throughout the cloud.

The conventional thunderstorm charge dipole, as identified by Wilson (1920, 1929), with its upper region of positive charge and lower region of negative charge, produces a vertical electric field. Intra-cloud lightning dissipates these charges and cloud to ground lightning from the negative charge center brings negative charge to ground. Positive point discharges are released from the ground by the high electric field. Thunderstorms leave behind them a vertical, fair weather, electric field (-100 V m⁻¹ near the ground) caused by the positively charged atmosphere and negatively charged ground. The thunderstorm charge structure turns out to be rather more complicated than this simple dipole model and the generation of the thunderstorm charges, their transport to the observed locations, and the details of the cloud charge structure are areas of continuing investigation.

Recent radar and charge center analysis has confirmed that thunderstorm charge centers are collocated with regions of precipitation in well defined temperature bands (Krehbiel, 1986; Williams, 1989; Lhermitte and Krehbiel, 1979; Krehbiel et al, 1979). The work has also confirmed the early observations of Reynolds and Brook (1956) that rapid electrical

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development is associated with the growth of precipitation, as shown by the rapid intensification of the radar echo. The fact that, in general, thunderstorms world-wide have their charge centers in similar and well defined temperature bands, colder than 0°C, suggests that the presence of ice is an important requirement of any thunderstorm charging mechanism. It is argued that field, laboratory and modeling results are consistent with thundercloud electrification by means of brief collisions between ice crystals and small graupel pellets (soft-hailstones). Figure 1 shows the location of the principal thunderstorm charge distributions, with a representation of charge separation by particle collisions. At high altitudes and low temperatures, graupel charges negatively and ice crystals carry a positive charge to the upper cloud regions. At lower, warmer levels, graupel charges positively and Jayaratne and Saunders (1984) suggest that the particles fall to form a lower positive charge center while the negative ice crystals are carried up and, together with the negative graupel falling from above, form a region of negative charge. In the mature stages of storm development, an additional, inductive, charging process may occur when water droplets rebound from falling graupel pellets in the existing high electric field.

Introduction to particle charging processes.

Mason (1972) set out some requirements of a tenable theory of thundercloud electrification in terms of the rates of electric charge separation by particle interactions required to account for the observed electric field development.

(i) The average duration of precipitation and lightning from a single thunderstorm cell is 20-30 min.

(ii) The average electric moment destroyed in a lightning flash is about 100 C km, the corresponding charge being 20-30 C. (C=Coulombs)

(iii) The charge is generated and separated in a volume bounded by the $-5^{\circ}C$ and $-40^{\circ}C$ levels with a radius of about 2 km.

(iv) The negative charge is centered near the -10°C isotherm and the main positive charge is situated some kilometers higher up; a subsidiary positive charge may also exist near cloud base, being centered at or below the 0°C isotherm.

(v) Sufficient charge must be generated and separated to supply the first lightning discharge within 10-15 min of the appearance of precipitation elements inside the cloud of radardetectable size, and to establish large-scale vertical electric fields of at least a few kilovolts



Figure 1. A schematic diagram of a thundercloud showing the charging of graupel pellets by ice crystal collisions. At altitudes above the reversal temperature level, the graupel charges negatively. Below the reversal temperature level it charges positively. The separation of the particles in the updraft leads to the development of the three charge centers.

per centimeter.

Mason also described details of some thunderstorm charging mechanisms involving interactions of graupel with ice crystals or water droplets. He favored an inductive charging process, as first proposed by Elster and Geitel (1913), in which ice crystals or supercooled droplets transfer charge during their rebound from pellets of graupel carrying polarization charges caused by the ambient electric field.

In 1981, Latham considered the relative merits of particle and convective mechanisms. He ruled out the convective mechanism because of a shortage of available ions and the long time scale involved in transporting ions into the cloud. Reynolds et al (1957) measured charge transfer when ice crystals interacted with simulated riming graupel pellets in the absence of an electric field and suggested that the temperature difference between the particles was driving the charge transfer; however, Latham favored a process driven by differences in surface potentials as first proposed by Buser and Aufdermaur (1977). Illingworth (1985) also supported non-inductive charge transfer between crystals and riming graupel, as first investigated by Reynolds et al (1957) and later by Gaskell and Illingworth (1980) and Jayaratne et al (1983). Williams (1985) considered cloud dynamics and the effect of air motions on convective and particle charge transfer processes. He was concerned that the energy of falling precipitation was generally insufficient to account for the electrical energy of storms unless the charging process was very efficient; but, confirmation of substantial charge on particles in regions of precipitation has come from observations of particle velocity changes following lightning by Krehbiel (1986).

2) Electrical Structure of Thunderstorms

Precipitation and charge center development

Benjamin Franklin found that charge from a thunderstorm brought to ground along the string of his kite was most often negative, although he sometimes found positive charges near cloud base. Confirmation that the lower charge center is usually negative came from studies of lightning by C T R Wilson who identified a vertical dipole within thunderstorms, the upper charge center being positive. Reynolds and Neill (1955) set up seven ground stations in New Mexico to detect electric field changes caused by lightning from which they located the charge centers in thunderstorms. The negative centers were generally at altitudes between 6 and 7 km in regions of temperature between $-6^{\circ}C$ and $-15^{\circ}C$ with positive centers

above. Negative centers as cold as -33° C were noted. A lower center of positive charge was also occasionally identified. Because the charge generation occurs in regions with temperatures below 0°C, they suggested that the thunderstorm electrification processes may be associated with a glaze-ice mechanism involving riming and vapor deposition.

Krehbiel (1986) determined the location of the charge centers for a series of lightning strokes in a Florida summer thunderstorm. He found that the negative center remains at a fixed altitude while the positive center moves up with the updraft at 8 m s⁻¹. This result suggests that a charge separation process is underway at around the -15°C level involving collisions of particles; the smaller, positively charged particles are carried aloft while the larger negative particles fall against the updraft and are effectively levitated at the -15°C level. A further implication is that the charging process, involving the collision of particles, must reverse the direction of the charge transfer between the particles at the level of the negative charge center. Above this level, graupel charges negatively and below it, graupel charges positively. The level is therefore associated with a particular temperature level in the cloud that has been labelled "the reversal temperature". This point is of great significance to laboratory simulations of particle charging processes, which also show that the sign of the charge transfer is dependent on temperature and cloud liquid water content. The importance of temperature to the location of the charge centers is shown by the observations of storms in different geographical locations by Jacobson and Krider (1976), Krehbiel et al (1979), Krehbiel (1981), and Brook et al (1982). It is notable that summer storms in Florida, and New Mexico and winter storms in Japan, despite their vastly different vertical extents, all possess negative charge centers in a temperature band between -10°C and -20°C.

Krehbiel et al (1979) gives us important evidence of the link between precipitation and lightning charge centers in a New Mexico thunderstorm. Radar reflected intensity contours reveal that thunderstorm charges reside selectively in the precipitation zones in the temperature range -9°C to -17°C. A single lightning flash is made up of multiple strokes which draw on charge spread across a wide extent of the cloud. In a typical sequence, the first stroke removed -20 C of charge and the last one transferred -2.6 C to ground with a total charge transfer over the whole flash of around -40 C. Lhermitte and Krehbiel (1979) studied three-dimensional motions within a developing storm in Florida by means of radar while simultaneously locating radio emissions from the cloud caused by discharges. They

noted that the updraft velocity increased rapidly to over 20 m s⁻¹ at an altitude of 6 to 7 kilometers (corresponding to temperatures in the range -10° C to -15° C) at the same time as the radiation rate increased. Simultaneously, a downdraft developed on the upshear side of the storm that brought down ice crystals while a high radar reflectivity at the -10° C level was associated with levitated graupel and hail growing by riming. These results led the authors to suggest that interactions between crystals and graupel are the source of thunderstorm charges. Cloud to ground discharges in seven Florida thunderstorms were studied by Maier and Krider (1986) who found a wide range of charge center levels, between -14 and -26°C. Analysis revealed charge centers, typically of 5 km diameter, that maintained a fairly constant altitude throughout a particular storm, again signifying the importance of a region of charged precipitation.

Measurements inside thunderstorms

The recent improved research capabilities of aircraft, instrumented rockets, balloon borne instrumentation and radars has enabled thundercloud microphysical, dynamical and electrical developments to be studied in detail. In a series of studies of the electric field inside New Mexico thunderstorms by means of instrumented rockets, Winn et al (1974) found that the field rarely exceeded 400 kV m⁻¹. Previously, higher fields than this were thought necessary to initiate a lightning discharge. They noted that the intense field and charge regions were concentrated in relatively small volumes. In France, Laroche (1986) found a maximum vertical field of 70 kV m⁻¹ in storms producing lightning. Of particular note are the aircraft studies in New Mexico and Florida using the Schweitzer airplane and the studies carried out in a variety of locations with the sailplane belonging to the National Center for Atmospheric Research. The in-cloud measurements of Gaskell et al (1978) were the first to record simultaneously the size and charge of precipitation particles, while the recent measurements of Weinheimer et al (1991) have provided charge, size and shape information from which particle types can easily be determined. A comprehensive study of thunderstorms by means of aircraft and ground based radars was made in Montana in 1981 (Co-operative Convective Precipitation Experiment, CCOPE) and in the Summer of 1991, in Florida, the Convection and Precipitation/Electrification Experiment, CaPE, involved airborne and ground based radars in studies of storm electrification and dynamics. Other important developments have been made with balloon-borne instruments that permit a fairly slow vertical sample to be

made at high resolution through clouds.

The Schweitzer airplane carried an electric field meter and a particle charge and size detector into thunderstorms in New Mexico; Gaskell et al (1978) and Christian et al (1980) reported vertical fields up to 100 kV m⁻¹ with charge densities on precipitation particles of 5 C km⁻³. Around -3.5° C a typical value of electric charge carried by a small graupel pellet was around ± 50 pC, but no systematic relationship was found between particle charge and size. Furthermore, smaller precipitation particles often carried charges in excess of values that could have been produced by the inductive charging mechanism in an electric field at breakdown strength.

Vali et al (1984) mounted a charge induction tube on a particle imaging probe (Particle Measuring Systems, 2D-C) and for the first time gathered simultaneous charge, size and particle type information. Details of the device are given by Cupal et al (1989). In flights in a Montana Summer thunderstorm at temperatures down to -10°C, they reported measurable charges on graupel particles. They noted that only a small fraction of particles carried detectable charges, but there was no correlation between size and charge and that some of the smaller particles (<200 μ m) carried charges near to the corona limit. Gardiner et al (1985) made measurements in the same cloud with a device that measured only particle charge, the observation of charged graupel in the above study permitted them to assume that the charges they measured were associated with the graupel detected simultaneously using a 2D probe. They flew at the -5° C level and found that the highest charge densities (-0.5 C km⁻³) were co-incident with regions of high graupel concentration. They also measured electric fields and found the highest values below regions with high radar reflectivities. These results are consistent with a particle based mechanism of thunderstorm electrification in which the charge separation interactions occurred at higher levels. They found that only about ten per cent of the particles were charged significantly (> ± 5 pC) and that these highly charged particles were present in the earliest stages of electrification. They could not verify whether the largest charges were carried on the largest particles because the charge and sign measurement systems were separate. The result that only a small number of particles are charged significantly is consistent with the Doppler radar observations of Williams and Lhermitte (1983) in which changes in the fa'l speed of charged particles following the removal, by lightning, of the levitating electric field, were rare. The question of why only some graupel pellets are charged significantly is an important area for future

study.

In further analysis of the same Montana storm, Dye et al (1986) reported that precipitation development and particle interactions, which could lead to charge transfer, occurred in a transition zone between the upraft and downdraft between -10° C and -20° C where there was liquid water present. There was limited field intensification in the early stages of cloud growth even though convection and growth of small particles was under way. Rapid field growth and lightning followed the formation of millimetre sized graupel particles with a concentration exceeding 10 Γ^1 . Negative charges accumulated near the -20° C level, associated with a high radar reflectivity region. Their conclusion from these studies was that the particle charging process leads to negatively charged rimed particles that grow and reside in the fringes of the updraft, while the smaller positive particles are carried aloft to form the classic thunderstorm charge dipole.

In measurements in storms in New Mexico, Dye et al (1988) noted a negative region of charge at the -12°C level associated with the updraft-downdraft transition zone. Another cloud showed a positive region at -20°C. Both charge regions occurred where particle collision rates were a maximum and close to radar reflectivity maxima; they possessed graupel and supercooled water, which could be associated with a precipitation charging mechanism. Dye et al (1989), in further studies of New Mexico storms, noted that the onset of significant electrification occurred only after convective growth when radar reflectivities at the -10°C level exceeded 40 dbZ and cloud tops exceeded the -20°C level; for lightning to occur, the cloud tops had to be colder than -20°C. These results provide further confirmation of the early work of Reynolds and Brook (1956) and Moore et al (1958) and are commensurate with particle growth processes in regions of supercooled water. The field data thus appear to be consistent with a charging mechanism involving precipitation.

Marshall and Winn (1982) used an instrumented balloon to probe New Mexico thunderstorms. Particle charge was detected with an induction tube and the particle size was determined from the vertical velocity. They noted that the measured charge density, of around -5 C km⁻³ in the negative charge center at -15°C, agreed well with that calculated from the gradient of the electric field, indicating that precipitation particles were carrying the charge.

The combined charge detector and two dimensional imaging probe has been developed further by Weinheimer et al (1991) and has been used in sailplane studies in New Mexico. In one such study, while the sailplane spiralled upwards, the field record showed a localised charge region rather than a charged layer. Peak charge densities were of order ± 10 C km⁻³ which are sufficient to account for the observed electric field strength. The bulk of the charge was carried on 2-4 mm graupel and the charge carried increased with particle size, as expected with a collisional charging mechanism. The individual charges ranged from the minimum detectable (i μ C) up to a few hundred pico-Coulombs. Two aspects of these studies need further investigation. The charge data showed that only a few particles carry the bulk of the charge, in agreement with Gardiner et al (1985). The result points to non-uniformities in the cloud or to different histories of the precipitation particles. Also, no pristine large ice crystals (>500 μ m) were observed in regions of graupel pellets where charging was occurring and so Weinheimer et al suggest that collisions between large and small graupel may lead to charge transfer. If this is so, then questions arise as to the charging mechanism which, apart from the inductive process, is thought to depend on differences in the surface properties of the interacting particles.

The increasingly widespread use of lightning detectors has provided some surprising results. Prior to their use, most thunderstorm electric field change studies were carried out in the summer months, but their introduction has permitted the study of field changes throughout the year. Lightning to ground in summer storms tends to bring negative charge to ground whereas Orville et al (1987) find that in the winter, the relative number of positive strokes to ground increases. Brook et al (1982), in studies of winter-time storms over the sea of Japan, found that positive ground strokes predominated. They attributed this to the extensive horizontal shear in those clouds that permitted the overhanging upper positive charged region to discharge directly to ground. Orville et al (1988) also has evidence of shear with bipolar charged regions aligned with the upper winds so that positive flashes emanate from regions downwind of the negative region.

Most of the analysis of thunderstorm electrification has been performed on isolated convective clouds, such as those in New Mexico. More complicated storms are now being studied, such as the mesoscale convective storms (MCS) that occur in Oklahoma. Hunter et al (1992) report that an instrumented balloon, Doppler radars, a lightning ground-strike location system and satellite information were among the main sources of a detailed study of an MCS. These clouds are complex because they are made up of regions that have been transported from adjacent systems and so it is hard to unravel the history of a particular

parcel of particles. For example, the balloon study revealed 11 distinct charge layers, ten of which were between the $+10^{\circ}$ C and -24° C levels. Charge densities up to 3.9 C km⁻³ were found, similar to the values in convective cells. Most of the cloud to ground lightning, which came from the anvil or the stratiform region, was positive. Further study of storms such as these is needed to ascertain whether their charging processes are consistent with particle interaction and charge separation processes.

A most important development in the range of tools available for the remote identification of cloud physical processes, is the differential reflectivity polarization radar technique used by Bringi et al (1984) and Illingworth et al (1987) to discriminate between regions of water or ice in cumulus clouds. Krehbiel et al (1991) have used the technique to provide evidence of the build-up and collapse of strong electrification inside storms by detecting the presence of electrically aligned particles at all levels above 0°C. Particle alignment is lost following a lightning stroke but is re-established when the field rebuilds, indicating the likelihood of a further stroke. Illingworth and Lees (1992) used a magnetic direction finding technique to locate the position of lightning in a summer thunderstorm in England while simultaneously observing the precipitation with polarization radar. They confirmed that lightning is collocated with the maximum precipitation echo, which itself was produced by graupel, and they concluded that the presence of graupel is a necessary condition for lightning.

3) Thunderstorm charging processes

The fact that some clouds become highly electrified is not surprising. When an insulating particle rubs against a similar or dissimilar particle, electric charge is transferred by piezoelectric, thermoelectric or other triboelectric effects. When the two particles are conductors, charge transfer in an electric field is simply explained; however, precipitation particles have a wide range of surface conductivities that may limit the transfer of charge. On the other hand, collisions between particles may not always be necessary for the production of substantial electric fields leading to lightning, for example, cloud particles can capture ions and then be carried by convection currents to form regions of accumulated charge. What is perhaps surprising is that it is taking so long to solve the problem of thunderstorm electrification. The actual mechanisms responsible for charge transfer between colliding particles have not been universally agreed, and the convective motions that transport charges in the cloud have not been fully traced. The processes considered here occur during

particle collisions in which charge is transferred independently of the local electric field strength and so involve only non-inductive charge transfer.

Ice crystal interactions during riming

Large charge transfers have been detected during the interactions of vapor grown ice crystals with graupel pellets in the presence of super-cooled water droplets. Confidence in the effectiveness of this process to thunderstorm electrification follows from extensive laboratory work and from data obtained during penetrations of thunderstorms by research aircraft. These particle interactions were first studied in cloud simulation experiments by Reynolds et al (1957) who calculated that the charge separated by ice crystals colliding with a riming ice sphere is adequate to account for thunderstorm electrification. They performed their experiments at temperatures around -25°C with realistic liquid water contents and found that ice spheres, representing falling graupel pellets, became negatively charged while the colliding crystals removed positive charge. In a thunderstorm, this charge transfer process leads to the observed vertical dipole when the oppositely charged particles separate under gravity. They noted that without crystals present, droplets colliding with graupel did not separate charge, within their detection limit. These results led them to suggest that the charge transfer was due to a temperature difference between the interacting particles; the crystals remain essentially at the air temperature while the graupel pellets capture supercooled droplets and are warmed by the release of latent heat during droplet freezing. Later, Marshall et al (1978), Gaskell and Illingworth (1980) and Jayaratne et al (1983) showed that the charge separated between interacting particles is not dependent on their temperature difference; also, the theory of temperature gradient driven charge transfer (Latham and Mason 1961) predicts charge transfers considerably below those noted by Reynolds et al. Church (1966) performed related experiments to those of Reynolds et al and found that at -15°C the ice sphere charged positively due to crystal collisions unless the liquid water content in the cloud was low, when it charged negatively. In 1978, Takahashi noted a similar result, while Gaskell and Illingworth (1980) bounced frozen ice spheres off a riming ice target and found positive target charging at temperatures of -5°C and -10°C and negative charging at -15°C and -20°C, as shown in Figure 2. These results form the basis of a viable charging mechanism in which ice crystals charge riming graupel negatively at low temperatures and positively at higher temperatures, but the sign of the charging is also influenced by the presence of the liquid water captured by the graupel.



Figure 2. Charge transferred to a riming ice target by collision with 100 μ m ice spheres, in a laboratory cloud of various liquid water contents at four temperatures at a velocity of 8 m s⁻¹. • -5°C; \bigcirc -10°C; + -15°C; * -20°C. (Gaskell and Illingworth, 1980.)

Detailed laboratory studies of the above charging processes have been carried out over the last ten years in apparatus similar to that shown in Figure 3. Ice targets in a supercooled droplet cloud become covered in rime (by moving the target or by drawing a supercooled droplet cloud past the target) and ice crystals growing in the cloud interact with the riming target. The charge transferred per crystal separation event can then be determined from measurements of the charging current and the ice crystal concentration together with a knowledge of the crystal/rimer collection efficiency. Jayaratne et al (1983) confirmed that graupel charging reverses sign as a function of temperature, as shown in Figure 4, and that this "reversal temperature" moves to higher (warmer) values with decreased cloud liquid water content. They also found that the amount of charge transferred depends on impact velocity and on the size of the small ice crystals used. Keith and Saunders (1990) extended the previous work by using larger ice crystals, up to 800 μ m diameter, and found that the charge transfer increases rapidly with crystal size for small crystals but increases at a lower rate for larger crystals. They suggested that high values of charge transfer are limited by the reverse charge transfer of some of the charge residing on the surfaces when the particles A study of crystal/graupel interactions in the dark revealed light emission separate. associated with this reverse charge transfer in the form of corona (Keith and Saunders, 1988). From their charge transfer experiments, they formulated relationships between charge transfer, crystal size, and impact velocity for positive and negative charging situations.

The numerical dependence of crystal/graupel charge transfer on the liquid water content (LWC) in the cloud was formulated by Saunders et al (1991) from a series of cloud chamber experiments under controlled conditions. In earlier experiments, Jayaratne and Saunders (1985) showed that the effective liquid water content (EW), made up of those droplets large enough to hit the riming target, is important in controlling the charge transfer. Figure 5 shows the positive and negative rimer charging regimes as a function of EW and temperature. The authors presented parameterized equations combining the charge dependence on temperature, crystal size and impact velocity with EW. The equations permit the determination of charge transfer over a range of cloud conditions representative of those in the particle charging regions of thunderstorms and are suitable for inclusion in numerical models of thunderstorm electric field growth. An example of the use of these results comes from a study by Dye et al (1988) in a New Mexico summer thunderstorm. They found a build up of negative charge below the $-12^{\circ}C$ level implying a charge sign reversal



Figure 3. Cloud chamber for study of riming electrification. Droplets and ice crystals interact with a moving rimed target and charge transferred during the ice crystal collisions can be measured.



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Figure 5. The sign of the charge transferred to a riming target by ice crystal collisions as a function of the effective liquid water content and the temperature. The Regions shown correspond to those in Table 1.

temperature close to that level. For the measured LWC of around 0.8 g m⁻³, the reversal temperature, determined from the laboratory experiments, is -13° C and a reversal temperature of -11° C corresponds to the average LWC encountered in the cloud of around 0.5 g m⁻³. The negatively charged precipitation falls below the level of temperature reversal and so is detected at temperatures higher than the reversal temperature itself.

Recent work in UMIST has shown that Figure 5 requires modification. The figure was prepared on the assumption that it applied at all rimer/crystal velocities so long as the cloud was adjusted to keep the Effective Liquid Water Content (EW) constant. (EW is that portion of the droplet cloud that accretes on a rimer - at high speed EW approaches the LWC, but at low speed, or with small droplets, they can differ by a factor of ten). Further experiments were performend in the cold room by drawing the droplet crystal cloud past a stationary target and by impacting crystals on a rotating target. There have been significant differences between the charge transfer results from different research groups and these have been attributed in the past to differences between moving and stationary targets, and between experiments performed at low and high velocity. A typical speed in the UMIST work has been 3 m s⁻¹ which is representative of the fall speed of millimeter sized graupel, while other workers have used 9 m s⁻¹ which is applicable to large graupel or small hail. Figure 6 shows the results of experiments at a range of velocities for crystal impacts with a fixed icing target. The results with a moving target were similar showing that both simulations of a graupel pellet falling through a thunderstorm are equivalent. The results show clearly that speed can affect the sign of the charge transfer to the riming target. This velocity effect is associated with the extra collection of cloud droplets at higher speed and is also associated with the heat balance of the rime and the increased collection of larger droplets at higher speeds, which affects the density and structure of the rime. Conceptually, the collection of more liquid water favors positive charging, hence increasing the speed also favors positive charging as shown in the figure. Furthermore, the quantity that should have been conserved in these experiments at a range of velocities, is the rate of accretion of rime (at higher speeds, the vapor source must be decreased). The experiments to obtain Figure 5 were done at 3 m s⁻¹ and because the rime accretion rate is proportional to EW x V, Figure 5 may have its vertical scale re-captioned "EWxV/3" with no other changes. Obviously at 3 m s⁻¹ the figure is the same as Figure 5, but at 9 m s⁻¹, less cloud water is required to reverse the charge transfer from positive to negative at any particular temperature. This re-assessment

of Figure 5 is too recent to have been included in numerical models of thunderstorm electrification rates, but it means that in mature storms with large graupel falling at high speed, they are more likely to charge positive than under the original Figure 5. This may present a problem for the generation of the conventional charge dipole. It certainly presents a problem for the reconciliation of the results of Takahashi (1978) and Saunders et al (1991), for at -20°C, Takahashi has charge sign reversal (from negative to positive with increase in cloud water content) at 4 g m⁻³, whereas, the modified Figure 5, (used at 9 m s⁻¹ to match Takahashi's data and converted to LWC) has charge sign reversal at a LWC of 0.45 g m⁻³. There is some evidence from other experiments in UMIST that Takahashi has substantially overestimated his LWC, maybe by as much as ten times. However, the new Figure 5, based on the results of Figure 6, implies that graupel charges negatively only under conditions of low liquid water content and for small graupel falling at relatively low speeds.

Ice sphere interactions during riming.

A different experimental approach has been adopted by Avila and Caranti (1991, 1992) who have continued the studies of Gaskell and Illingworth (1980) by using individual 100 μ m ice spheres to impact riming ice surfaces at 5 m s⁻¹. This procedure may model charging during collisions between large and small graupel particles. In their experiments, charge transfers from single collisions were noted in the range +40 to -80 fC. For a droplet cloud with an EW of 0.2 g m⁻³, most of the charge transfers to the rimer were positive at temperatures above -18°C and negative below -24°C, (see Figure 7) implying a reversal temperature around -21°C. Increasing the LWC or temperature favored positive rimer charging in agreement with the ice crystal charge transfer results summarised above. The equations described above may be used to compare the values of ice crystal and ice sphere charge transfers, although it must be borne in mind that the charge transfer mechanisms are possibly different and that there is a problem with a temperature reversal at -21°C at this value of EW, as discussed below. At -24°C, for EW equal to 0.2 g m⁻³ with 100 μ m ice crystals interacting with a riming target at 5 m s⁻¹, the predicted average crystal/graupel charge transfer is -7.5 fC (from Saunders et al, 1991). The average charge noted by Avila and Caranti (1992) for these conditions was -22 fC. The larger average charge transfers with 100 μ m ice spheres compared with similarly sized ice crystals may be caused by their higher momentum or may be due to a different charging mechanism.



Figure 6. Charging current to a fixed riming target at a range of velocities following initiation of ice crystals at t=0.

Construction And



Figure 7. Charge transfer histograms obtained when 100 μ m ice spheres interact with a riming target. The effective liquid water content is 0.2 g m⁻³. (Avila and Caranti, 1992.)

The charge sign reversal temperature in the Avila and Caranti experiments is not in good agreement with the reversal temperature found in the experiments with ice crystals. When Avila and Caranti (1991, 1992) perform experiments near their revers 1 temperature of -21°C with EW equal to 0.2 g m⁻³, they find that an increase in either the liquid water content or temperature changes the predominant sign of target charging from negative to positive. From Figure 5, for ice crystal interactions around -21°C, the corresponding critical value of EW for charge sign reversal from negative to positive with increase in EW or temperature is 0.9 g m⁻³. This difference in the required liquid water content for temperature reversal may indicate that the charge transfer mechanism in the individual ice sphere experiments is differences, which need to be resolved, between the conditions on the riming surfaces or in the cloud in the two sets of experiments. A consequence of these results, which point to most charge transfers causing positively charged graupel, unless the LWC is very low, is the increased difficulty in explaining the predominance of negative graupel in thunderstorms.

4) Charge transfer mechanisms

The precise mechanism by which charge is transferred between two interacting ice particles remains under active debate. The processes outlined below do not have to overcome the problem of short contact times that limit the inductive charging process because of its need to conduct charge from distant surface towards the contact point. In the following processes, the charge is available at the point of contact.

Relative Growth Rates and the Liquid-Like Layer

From their crystal/graupel charging experiments, Baker et al (1987) were able to explain charge sign reversal in terms of the relative growth rates of the interacting particles. The riming target collects supercooled water droplets and also grows by diffusion from the environmental vapor provided by the local water droplet cloud; it also grows from the vapor released when droplets freeze on the surface. On the other hand, small ice crystals in the cloud grow only by diffusion from the environmental vapor and thus the two types of particle have different diffusional growth rates. The freezing time of a droplet on the rimer surface is temperature dependent and so at low temperatures, with rapid droplet freezing, there is less vapor released than at higher temperatures. Qualicatively, the calculations showed that the interacting crystals and the graupel pellet have relative growth rates that may reverse direction at the reversal temperature and that the faster growing particle charges positively. Quantitative determinations await more detailed analysis of the heat balance of the surface of riming graupel.

Baker and Dash (1989) searched for a mechanism that would account for this growth rate dependent charge transfer and suggested that the orientation of surface molecules causes an excess of negative ions in the disordered melt layer (liquid-like layer) on the surface of ice. (Figure 8). The charge in the surface is dependent on the growth rate: the faster growing particle has a thicker surface layer and so provides more negative charge during particle interactions. The mechanism relies on the transfer of charged material from one particle to another during contact and confirmation that this can occur is awaited. A difficulty with the concept is that optical techniques have shown that a liquid-like layer on ice exists only down to -4°C (Furukawa et al 1987). This result is consistent with recent charge transfer results of Dong and Hallett (1992) in which an ice layer, at temperatures down to -4°C, behaves, in certain charging experiments, the same as a water surface; below -4°C its surface charging behavior is that of ice. There is evidence, however, (Dash, 1989) that the surface layer can exist to much lower temperatures. A further problem with this mechanism is that the surface layer thins when the temperature is lowered and yet substantial crystal/graupel charge transfer has been observed in laboratory studies performed below -30°C; furthermore, in thunderstorm penetrations, Weinheimer et al (1991) find an increase in the negative charge on precipitation particles as the temperature decreases.

Together with the need for measurements of mass transfer during the charge transfer process, the effect of impurities in the ice needs to be studied. Jayaratne et al (1983) found that the sign and magnitude of crystal/graupel charging was affected by impurities in the cloud water droplets that formed the rime ice; sodium salts enhance negative rimer charging, and ammonium salts increase positive charging. These impurity results have not been satisfactorily explained by any charging process and the first to do so would be favored.

Recent unpublished work in the UMIST laboratory has thrown an extra difficulty in the way of accepting the liquid-like layer hypothesis. Interaction of ice particles with an ice surface separates charge with the sign depending on the thickness of the liquid-like layer. According to the theory, the layer is negatively charged and so the surface with the thicker layer will have material removed by a smaller interacting particle and so will charge



Figure 8. Charge transfer associated with negative charges in surface liquid like layers on a riming graupel pellet and an ice crystal. (After Baker and Dash. 1989.)

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positively. This ties in with the general idea that charging depends on growth rate, and the particle growing faster will have a thicker liquid-like layer and so is more likely to charge positive. Therefore, it was not found surprising that in new experiments, interactions between 200 μ m sand particles and an ice surface caused the ice surface to charge positively whether it was growing or sublimating. The sand particles have no liquid layer, therefore, of the two interacting particles, the ice has the thicker layer and so charges positively. However, tests with other materials were not so clear cut - small aluminum particles sometimes charged the ice negative. The answer here may be connected with effective work function differences, but given the difficulty in accounting for ice/ice charge transfer, it may be unhelpful to consider interactions involving other types of particles.

In this connection comes the work of Cals and colleagues at ONERA in their studies of charged layers at ice/gold interfaces. Their objective is to understand more about the adhesion of ice, particularly to aircraft and they are examining the electrical aspects associated with a junction between metal and ice. The work also has relevance to the charge distribution in the polymer skins of space-craft, which may cause electrical interference, (Cals et al, 1989). It was suggested that the technique is applicable to finding the charge density, sign and position of the charges in other substances, including ice. The technique involves a rapid high energy laser pulse incident upon the charge layer, which causes it physically to move. The movement is detectable by electrodes near the surface connected to amplifiers that produce nano-second pulses whose time of arrival and pulse shape indicate the characteristics of the charge layers. The early work in this field proved that the techniques had severe signal/noise limitations. Later, the gold/ice interface was studied and the results provided evidence of an electric field created by the double layer at the interface with a thickness less than that of the pressure pulse itself, of the order of 30 μ m. The experiments were performed at -15°C where there is maximum adhesion between ice and the substrate. The sensitivity of the technique is such that if charges are detected, then they are likely to be of significance in thunderstorm electrification. However, the ice/gold interface is, by itself, not entirely relevant to the atmosphere, although it may tell us something about the effective work function of ice. Of more relevance, is the possibility of studying the effect on the ice/gold sample of riming with supercooled water droplets so that the Caranti rime ice pseudo-contact potential (discussed below) can be investigated further. Also, the detection of a second charge layer at the ice/rime ice interface would be highly informative.

This awaits further study.

Charging of Growing or Sublimating Ice Surfaces

Experiments conducted with ice crystals in the absence of a cloud of water droplets have shown that when the crystals collide with an ice surface growing by vapor diffusion, the surface charges positively; conversely, evaporating (sublimating) surfaces charge negatively (Buser and Aufdermaur 1977, Gaskell and Illingworth 1980, Jayaratne et al 1983). Further experiments without ice crystals present, in which parts of an ice surface were blown off in an air stream, proved that the above results were caused by the removal of growing or sublimating surface (Saunders et al, 1993). However, Griggs and Choularton (1986) found that even low density rime ice is too strong to be broken by 100 μ m frozen spheres or by ice crystals impacting at realistic velocities as used in the charging experiments; they did find that dendritic growths may be broken off in agreement with Latham (1963). This leads to the possibility that all the charge transfers noted by Caranti and co-workers, who used 100 μ m ice spheres, are caused by the breaking of frost on their target. Unfortunately these data are not compatible with the results of Jayaratne et al (1983) and Saunders et al (1991) who noted that small ice crystals, only 20 μ m in size, do lead to charge transfer when they collide with an ice target when such small crystals are unlikely to lead to breaking of frost. In recent experiments, this interaction was simulated using 20 μ m lycopodium spores to represent the crystals. The spores impacted at 9 m s⁻¹ on a recently rimed ice target and any fragments of ice dislodged would have been detected in a supercooled sugar bath. However, there was no evidence of fragmentation. So, if small crystals can lead to charge transfer, yet small spores cannot break off any ice surface, the likelihood is that the breaking off of surface is not a thunderstorm charging mechanism.

Notwithstanding the above argument, a mechanism of charge transfer has been put forward by Caranti et al (1991) and Avila and Caranti (1991). They note that surface features growing by deposition on a rime surface experience a temperature gradient with the outer tips being warmed by latent heat release. When small ice particles collide with the surface, they break these surface growths and Caranti et al propose that the protons on the hydrogen bonds tend to remain on the colder side of the fracture so that the negatively charged tips are removed and the target ice is left with a positive charge; see Figure 9. Conversely, a sublimating structure has a cold tip relative to its base and so it charges the surface



Figure 9. Charge transfer associated with temperature gradients in surface growths on a rimer. (After Avila and Caranti, 1992.)

negatively when removed. Delicate structures on sublimating ice surfaces have been noted by Cross and Speare (1969) and these could be responsible for the negative charging when removed by glancing ice crystals or by an air jet, as in some of the above experiments. Latham (1963) found that the removal of frost deposits led to charge transfer but rejected the process because calculations of temperature gradient driven charging by Latham and Mason (1961) proved inadequate to account for thunderstorm electrification. This new proposal of Caranti et al (1991) relies on considerably more charge transfer under a temperature gradient than is accounted for by the Latham and Mason theory and has yet to be worked through in detail. In particular, the fact that the proton tunnelling time is orders of magnitude faster than the time to break off surface features makes it hard to follow how ordered charge transfer will accompany surface breakup. Besides, the new sand impact results referred to above show that sand grains charge an ice target positively, yet an immediate following experiment with small aluminum particles charges the surface negatively. Surely, both of these types of particle will be capable of breaking off features from the ice surface, so the result seems to militate against surface break-up as a viable charge transfer mechanism.

There is a further problem with the temperature gradient, surface removal theory; experiments performed by Jayaratne et al (1983) in which ice crystals carried along in a heated air stream impacted an ice target should have caused positive charging because of the removal of surface fibers with warmed, hence positively charged outer tips; however, the ice target, which was sublimating, charged negatively. This result is consistent with ice charging caused by the surface interactions of colliding particles such that the particle that is sublimating charges negatively; conversely the particle that is growing, charges positively. Furthermore, the relative growth rates are important; the particle growing faster than the other, charges positive.

Contact potential charging

Caranti and Illingworth (1983) reported that riming an ice surface caused it to develop a negative pseudo-contact potential compared with the unrimed surface. Their results are shown in Figure 10. The contact potential became more negative with decreasing temperature to reach a fairly steady value of around -400 mV at temperatures below about -20°C. They hypothesised that an ice crystal colliding with a rimed surface will lead to negative charging of the rime because of a contact potential difference between the crystal



Figure 10. Change in the contact potential of an ice surface after riming, as a function of temperature. The unrimed ice surface is assigned an initial potential of 0 Volts. (Caranti and Illingworth, 1980. Reprinted with permission from Nature).

and the rime surface. A simple calculation shows that for a contact area of 100 μ m², a potential of 100 mV and a separation between the surfaces of 1 μ m, the charge transfer is of order 10 fC, in agreement with observations. Caranti et al (1985) found no change in contact potential associated with a change from growth to evaporation of an ice surface, thus ruling out this mechanism as an explanation for charge sign reversal associated with growth and evaporation. In the experiments in which ice spheres interacted with a riming surface by Avila and Caranti (1991, 1992), the contact potential mechanism would not have been active because both interacting surfaces would have the same contact potential having been formed by the rapid freezing of supercooled water.

Charges on dislocations

Takahashi (1978) proposed that during the impact of an ice crystal with dendritic branches on a rimer, a pair of free protons and negatively charged dislocations are created. While the rime branch is being broken off, the charges separate under the action of the local temperature gradient and account for the positive charging of graupel under low liquid water content conditions. The breaking of surface features has already been questioned above.

An alternative dislocation mechanism that may account for negative rimer charging is provided by some evidence that dislocations in the structure of ice carry a positive charge and that the charge densities are temperature and growth rate dependent. Keith and Saunders (1990) used these data in calculations of the charge available for transfer during crystal/graupel collisions and found that there is adequate charge to account for the experimental observations. For example, for 5x10⁹ m⁻² dislocations per unit area having a charge per length of $+6x10^{11}$ C m⁻¹, the charge available on an area of 55x55 μ m² is +50 fC. This is the right order of magnitude to explain the observed charge transfers; furthermore, variation in velocity and crystal size will alter the contact area between the interacting particles which will affect the magnitude of the charge transfer. McCappin and Macklin (1984) found that the density of dislocations in rime ice increases with decrease in temperature, while McKnight and Hallett (1978) noted that slowly growing crystals have low dislocation densities. Thus the observed charging can be explained in terms of a difference in dislocation concentration on the surfaces of the interacting particles such that during particle interactions, the smaller particle physically scoops out surface material from the larger particle.

The dislocation mechanism can provide charge densities on the ice surfaces that are the "right" order of magnitude to account for the observed charge transfers as explained above, however, if a positively charged dislocation is trapped in the ice lattice, then local mobile charges will be electrostatically influenced and any positive charge center will soon be neutralised by surrounding opposite charges. Thus any interacting particle is just as likely to scoop out positively charged material as negative.

Application of the particle charging mechanisms.

Disregarding for a moment the problems outlined above with many of the charging processes, there follows an analysis of how the various charging mechanisms tie in with laboratory observations. The various charging regions shown in Figure 5 may be associated with specific rimer surface conditions controlled by EW and temperature. Table 1 identifies possible charging mechanisms associated with each charging region for the ice crystal/rimer experiments. Positive charging of graupel may be due to the removal of rimer surface, either surface growth from the vapor, rime growth or part of a liquid-like layer. Negative rimer charging may be caused by either a contact potential difference between rime and the interacting ice crystals, by a difference in concentration of charged dislocations on the interacting surfaces, or by removal of a charged liquid-like layer. It is likely that all charge transfer processes work simultaneously, but one or the other dominates depending on the temperature and liquid water content.

In the case of the liquid-like layer mechanism, the faster growing surface charges positively during crystal/rimer contacts. The charging process depends on the removal of charged surface when the crystal makes glancing collisions; the contact area, being dependent on velocity and crystal size, controls the magnitude of the charge transfer. In Region 1 of Figure 5, the rimer accretes locally at a higher rate than the crystals but the consequent rime surface heating means that over most of the surface area, the rimer grows by vapor diffusion at a lower rate than the crystals, leading to negative rimer charging. The mechanism may also account for the positive charging in Region 2 where the high accretion rate maintains a thicker surface layer on the rimer surface will sublimate because of local heating, although, over much of the surface, the liquid layer will be thicker than on the crystals. This hypothesis is consistent with the observation that the sign of the rimer charging is positive

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	RIMER	CONTACT	DISLOC-	LIQUID	TEMP.
	CHARGE	POTENTIAL	ATIONS	LIKE	GRADIENT
	SIGN			LAYER	
REGION 1	NEGATIVE	1	1	5	Х
REGION 2	POSITIVE	Х	X	1	1
REGION 3	POSITIVE	X	X	1	1

Table 1. A table showing the applicability of the various charge transfer processes to the results for ice crystal collisions shown in Figure 5.

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for all EW values up to wet growth, but the point needs further consideration. The positive charging in Region 3 may be due to the fact that the droplet accretion rate, although low, is sufficient to provide vapor so that the rimer grows faster than the crystals, but the heat released is too small to cause surface sublimation. This mechanism is particularly sensitive to the details of the heat and vapor fluxes on the rimer surface and these need further investigation.

Temperature gradients in rime structures may account for the positive charge transfers. Latham and Mason (1961) calculated that under a temperature gradient along an ice sample, the mobile protons migrate to the colder end leading to a weak potential difference between the warm and cold ends. This potential was found to account for the charge transfers observed by Latham (1963) associated with breaking frost (typically 10² fC) but is too weak to account for the large charge transfers noted in the ice crystal and ice sphere experiments. The charge transfers measured by Caranti et al (1991) are of order 10 fC and the authors invoke an enhanced temperature gradient driven charging process in which ice fracture results in the breaking of hydrogen bonds; the tunnelling protons tend to remain on the colder side of the break. So, if ice crystals can break surface features off a riming graupel pellet, charge transfer will result with a dependence on impact velocity and crystal size. In Regions 2 and 3 of Figure 5, the rimer surface grows from vapor diffusion from both the environment and the freezing drops and so the surface structure will have warmer, negative, tips than bases; removal by a passing crystal charges the rimer positively. In Region 1 of Figure 5, the negative charging could be accounted for if the surface structures were sublimating so that crystals remove the positively charged rime tips. However, according to the heat balance analysis of a riming graupel pellet (Macklin and Payne, 1967 and Saunders and Brooks, 1992) surface sublimation does not necessarily occur in Region 1 and so it cannot account for the negative charging, which therefore must be due to another mechanism.

If EW is increased sufficiently, then parts of the surface of the rimer will be warmed sufficiently to cause local sublimation leading to negative charge transfers to the rime when the surface growths are broken by crystal collisions. However, Saunders and Brooks (1992) found that the positive charging in Region 1 extends up to the wet growth limit (for example, $EW > 5 \text{ g m}^{-3}$ at -20°C) indicating that sufficient rimer surface must continue to grow even at high EW to cause net positive charging by this mechanism. This is consistent with the results of Avila and Caranti (1991, 1992) who find both signs of charge transfer, with one

sign being dominant, in a series of individual particle collisions.

The analysis of the contact potential mechanism shows that the rimer charges negatively because its contact potential is more negative than that of the ice crystals. The fact that in the ice crystal experiments, below the reversal temperature, negative charge transfer was independent of temperature is consistent with the results that below about -20°C the contact potential tends to a steady value. Thus the mechanism can account for the negative charging in Region 1 of Figure 5. Increasing EW does not change the rime contact potential but the contact potential of rime formed at a higher temperature is reduced, which permits a positive charging process to dominate in Region 2.

In the dislocation driven charging mechanism, the rimer surface grows more rapidly than the ice crystals and so has a higher concentration of positively charged dislocations. During contact, the rimer surface charges negatively following the mass transfer of a region of positive dislocations. With increase in temperature or LWC, the droplet freezing time increases and so the rimer dislocation concentration decreases and the mechanism becomes less effective. The mechanism is included here because dislocation charges and concentrations on ice single crystals, measured by X-ray topography, show that there is sufficient charge available to account for the observed charge transfers. It is not clear, however, why the positive dislocations in the ice lattice do not become surrounded by negative charges that would also be available for transfer during particle contact.

The classic temperature gradient theory, as enumerated by Latham and Mason (1961), is inadequate to account for the observed charge transfers. The enhanced temperature gradient mechanism of Caranti et al (1991) is unproven; proof that the mass transfer of charged surface can occur presents a formidable experimental problem. The concentrations and charges on dislocations on atmospherically realistic ice crystals and graupel are unmeasured. The controlling factor in the mechanism of charge transfer may simply be the availability and freezing time of supercooled droplets on the rimer surface. At relatively high temperatures, the droplets freeze slowly, and so have time to bathe the surrounding area with vapor whereas at low temperatures, higher freezing rates limit the vapor available. The situation is made more complicated by the heat released from the freezing droplets; the heat diffuses through the rimer surface and in turn controls the vapor diffusion rate to areas of the surface surrounding the freezing droplets. These ideas warrant a considerable research effort in order to resolve the outstanding problem of the mechanism of thunderstorm charge

generation.

5) Discussion

Despite the considerable efforts that have been made in recent years to solve questions concerning thunderstorm electrification, there are still outstanding problems. Laboratory studies of the charging of riming graupel pellets during ice particle collisions give results that are specific to the experimental techniques used, so to ensure that the experiments simulate the real cloud situation as closely as possible it is important to feed information back from field studies concerning particle types, concentrations, locations, velocities, temperatures and liquid water contents. The heat balance of a riming graupel pellet needs laboratory and theoretical work to ascertain the detailed behavior of the riming surface where the impacting ice crystals transfer charge. Is there evidence that surface material, which may carry charge, is transferred during the contact process? The inductive charging of graupel pellets by means of supercooled water droplets and ice crystals, although ruled out in the past because it is unable to account for observed electrification in the early stages of thunderstorm development, may be important in the later stages.

Another problem to be accounted for is the observation by Curran and Rust (1992) that low precipitation thunderstorms produce mostly positive lightning strokes to ground. The storms have narrow drop size distributions and low liquid water contents yet the positive lightning originates in regions of high radar reflectivity associated with the presence of large hail. Recent work using instrumented balloons by Marshall and Rust (1991), has revealed an extremely complex thunderstorm charge structure and there are observations of both positive and negative lightning at various stages of the development of tornadoes and mesoscale and supercell storms, (MacGorman and Nielsen, 1991; Hunter et al 1992). There is need here for further studies of thunderstorm charge distributions in order to find the time scale of their development, to locate the charge transfer events and to test charging theories and the validity of laboratory experiments. The use of numerical models provides a tool of growing importance in the achievement of these aims.

Recent numerical models of the electrification of thunderstorms have used storm observations as a template on which to build the electrical development. For example, Dye et al (1986), Latham and Dye (1989) and Norville et al (1991) have used a well studied CCOPE cloud in Montana to test electrification by means of crystal/graupel collisions in a

one dimensional model. Helsdon and Farley (1987) have used the same cloud in a three dimensional dynamical model that includes charging by many processes. Ziegler et al (1986, 1991) have used a New Mexico mountain thunderstorm in one and three dimensional kinematic models and have included laboratory crystal/graupel charge transfer data together with droplet/graupel charging by the inductive mechanism. Although the early models disagreed on the timing of the electrical development and the location of the charge centers, all the model results show that charging by particle collisions is adequate to account for thunderstorm electrification, but many assumptions have been made.

The latest models use the charge transfer data of Keith and Saunders (1990) in order to include the dependence of charge transfer upon ice crystal size. No models yet take detailed account of the collection efficiency of ice crystals for graupel for which some data are available (Keith and Saunders 1989). All the modelers note that the sign of crystal/graupel charging is dependent on temperature and liquid water content but this is taken into account only in a limited way at present. For example, Ziegler et al (1991) have used specific charge sign reversal temperatures of -10°C and -20°C for the storm studied and find that a reversal temperature of -10°C matches the cloud observations.

The first modeling studies to use the formulations of Saunders et al (1991) that include the charge dependence on cloud Liquid Water Content (LWC) are presently being performed by Helsdon and colleagues at the South Dakota School of Mines. They have related the reversal temperature to the specific cloud conditions in a full three dimensional cloud physical electrification model. The first output from this work has shown a considerable problem with Region 3 of Figure 5. At altitude, the liquid water content falls and Region 3 shows the positive charging of graupel under low LWC conditions. This leads to a reversed charge dipole that is not realistic. The laboratory work was re-examined, and further tests were made in order to elucidate this problem. It was felt significant that in the work of Jayaratne et al (1983), Region 3 in Figure 5 was not observed - the rimer charged negatively at low LWC. Furthermore, the results of Takahashi (1978) did show positive rimer charging at low LWC. The dilemma was resolved by reproducing the exact experimental methods of Jayaratne et al in the cold chamber shown in Figure 3, which was the chamber used by Saunders et al. The low LWC positive charging was only observed when water droplets were introduced into the cloud chamber from outside. In the Jayaratne method, a boiler internal to the cold room provides the vapor, whereas in Takahashi and Saunders et al there

is an outside source. The introduction of a saturated air mass into the cloud chamber, together with the saturated, or near saturated conditions inside, lead to mixing and the production of a super-saturated cloud. This causes rapid growth of the riming target, and in agreement with the theory that the faster growing particle charges positively, the target, experiencing a low LWC and growing, charges positively. This is not consistent with the concept that when the LWC is increased, the target collects more water, is bathed in more vapor from the freezing droplets and so charges positively, as described above, because, as shown in Figure 5, increasing the LWC when the target is charging positively causes it to charge negatively. Further work is required to elucidate this conundrum but at first sight it appears that the temperature of the rimer is more important than the vapor supply to the rimer and increasing the temperature, at least initially, decreases the growth rate. At higher values of LWC, the rate of liquid accretion is sufficient to ensure positive charging again. At the heart of this problem is a solution to the long standing debate about the differences between the Manchester data and those of Takahashi.

Several problems with the proposed charge transfer mechanisms have been discussed above. It has been shown how each mechanism fits into the overall scheme of rimer charging as a function of temperatue and LWC, through Figure 5. However, there are severe problems with all the mechanisms excepting the conceptual one of "the faster growing particle charges positive". The revision to Figure 5 discussed in the text above, does not influence the discussions on charging mechanisms, however, it will have to be accounted for in future calculations of the sign of charge transfer to riming graupel.

Further airborne and balloon studies are also needed, with advanced ground based remote sensing techniques, to expand our knowledge of the distribution of charge throughout a thunderstorm, and to determine the characteristics of the particles in those regions of thunderstorms where there is strong evidence that electrification is taking place. In particular, we need to know the surface states of particles. For example, a falling graupel pellet will be colder than its environment and so will be growing, thus causing positive charging to be favored, however, a pellet being carried aloft in an updraft, will be warmer than its surroundings and so is likely to have its growth rate reduced, and this may favor negative charging. These cases in which the rimer is not in thermal equilibrium with its surroundings because of thermal lag have not yet been deliberately simulated in the laboratory studies. Here is an important area for future research.

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This report has been reviewed and is releasable to the National Technical Information Service (NTIS). At NTIS it will be releasable to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication.

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