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The Ice Crystal-Graupel Collision Charging Mechanism of Thunderstorm Electrification

by

Peter Berdeklis

1998

A thesis submitted in conformity with the requirements for
the degree of Doctor of Philosophy, Graduate Department
of Physics, University of Toronto

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The Ice Crystal-Graupel Collision Charging Mechanism of Thunderstorm Electrification
Peter Berdeklis
Doctor of Philosophy, 1998
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Abstract

The ice crystal-graupel collision charging mechanism, which is considered important in thunderstorm electrification, was studied using a newly developed Triple Interaction Facility. The facility is the first to allow independent control of the solid, liquid and vapor phases of a simulated cloud.

The charge transferred to a riming target by collisions with ice crystals was measured over a wide range of cloud conditions. It was found that the charge transfer was a function of the temperature and the effective liquid water content of the cloud. It was also found that the charge transfer to the target was extremely sensitive to the relative humidity at which the ice crystals were grown. Higher relative humidity always promoted stronger negative charging, while lower humidity led to weaker negative or stronger positive charging. The sensitivity of the charge transfer to relative humidity was greatest at temperatures between -13°C and -18°C , with the strongest negative charging observed near -15°C . Within this temperature range the average charge transferred to the target could be varied from $+5$ fC per ice crystal collision to -25 fC per collision by varying the humidity from ice to water saturation, respectively. In experiments performed at humidities near water saturation and at temperatures near -15°C , charge transfers greater than -60 fC per collision were observed within the first 30 seconds after ice crystals are initiated. It was also found that at high relative humidity the maximum negative charge transferred to the target occurred at a velocity of 5 m s^{-1} and was much weaker at 3, 4 and 6 m s^{-1} . A parameterization of the results is presented for use in numerical thunderstorm models.

Analysis of the data indicates that a competition between negative charge transfer between the ‘liquid-like’ layers of the colliding ice particles and positive charge transfer due to different dislocation densities can provide a consistent physical model of the effect of relative humidity. It is shown that differences in the relative humidity can explain quantitatively the discrepancies between the observations of previous investigators. A new conceptual model of thunderstorm electrification is also presented.

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Chapter One

Introduction

Thunderstorms are one of nature's most spectacular events, and the wonder that they inspire makes them an obvious subject for basic research. Thunderstorms and lightning have, therefore, been the focus of many studies, especially within the last 50 years. Recent field studies have provided a great deal of information about the dynamic and electric structure of thunderstorms, as well as the distribution of charge within clouds, and the conditions required to initiate lightning.

An important goal of thunderstorm research is to determine how thunderstorms become charged, a process known as thunderstorm electrification. Several mechanisms have been suggested. The mechanism of ice crystal-graupel collision charging is the most widely accepted because a large body of quantitative experimental evidence is available to support it. However, after 20 years of laboratory research, the theory is still unable to explain many important field observations, and the physics underlying the charge transfer is still not understood. Furthermore, the results of different research groups do not agree and explanations for the discrepancy are not satisfactory.

The goal of this work is to measure the charge transfer that occurs during ice crystal-graupel collisions, thereby providing quantitative data that can be used in future numerical and theoretical thunderstorm models. To achieve this aim an experimental investigation was performed under conditions that better approximated thunderstorm conditions than previous investigations.

This work begins with a literature survey to provide the context for the research and the experimental results. It is presented in Chapter 2. Previous field and laboratory studies will be discussed, as motivation for the current work, and a conceptual model of thunderstorm electrification will be presented. Chapter 3 introduces the experimental facility that was designed for the current study. The performance of the facility and the experimental technique are described. The results of the investigation are presented in Chapter 4. This includes a detailed analysis of a typical experiment. Parameterizations are given of the observed charge transfer. These parameterizations can be used in future numerical thunderstorm models to test conceptual models of thunderstorm electrification. The results are discussed in Chapter 5. They are compared with previous studies and used as a basis for a new conceptual model of thunderstorm electrification.

Chapter Two

Background

2.1 Introduction

The premise of current theories of thunderstorm electrification is that the large-scale electric structure of a thunderstorm is created by charging mechanisms that act at the scale of cloud particles and by the dynamics of the cloud which redistributes the charge carriers. The large-scale dynamic and electric structure of storms is determined in field experiments. They include *in situ* measurements by aircraft of the size, concentration, type (liquid or solid) and charge carried by individual cloud and precipitation particles. Determining the mechanism by which the particles become charged is the jurisdiction of laboratory studies, in which the evolution of single precipitation particles can be followed under controlled conditions.

The experimental investigation that will be presented here is a laboratory study. Its goal is to better describe one of the mechanisms that may lead to the charging of precipitation particles: the ice crystal-graupel collision charging mechanism, also known as non-inductive precipitation charging. However, to understand the relevance of the experimental results, some familiarity with the electric and dynamic structure of a thunderstorm is required. A brief description of that structure will be given, followed by a survey of previous laboratory studies and a description of the current model of thunderstorm electrification that is based on the results of those studies. Finally, the

limitations of this model and of previous studies are discussed, as the basis and motivation for the current work.

2.2 Thunderstorm Electric Structure

2.2.1 Large Scale Electric Structure

The charge in an average mature thunderstorm is arranged approximately as a vertical tripole (Krehbiel, 1986; Williams, 1989; Takahashi, 1996), shown schematically in Figure 2.1. The three charge centers that make up the tripole are known as the upper positive, the main negative, and the lower positive charge regions.

The upper positive charge region extends from cloud top to just above the main negative charge region. If an anvil has formed at the top of the cloud then the upper

Figure 2.1 The tripole structure of a thunderstorm. The negative charge center is always observed between the -5°C and -20°C altitudes; the positive charge region is always above it. In early electrification, the negative charge center first develops in the updraft-downdraft transition zone, between the -12°C and -20°C levels. The lower positive charge region is not distributed uniformly throughout the cloud, but consists of small pockets of positive charge. After Krehbiel (1986).

positive charge region extends through the anvil. The main negative charge region is 1-3 km deep and always centered between the -5°C and -20°C altitudes (Krehbiel, 1986). Localized pockets of positive charge found beneath the negative charge region, near cloud base, comprise the lower positive charge region. All three regions have charge densities on the order of 0.1 to 10 C km^{-3} . They can extend horizontally over tens of square kilometers (Krehbiel *et al*, 1979) and over hundreds of square kilometers in the case of positively charged anvils, so the total charge in a cloud can vary from a few coulombs to a few hundred coulombs (Krehbiel, 1986).

The tripole charge structure is remarkably consistent for storms observed in a wide variety of geographies and climates. Large, strongly convective summer storms in Florida often have cloud bases as warm as 20°C . Still, the negative charge region is found at the -5°C to -20°C level, which corresponds to an altitude of 6 to 8 km (Krehbiel, 1986). The negative charge region is found in the same temperature range in shallow, and often strongly sheared, electrified winter storms in Japan. These storms have cloud bases a few degrees below the freezing point and the -5°C to -20°C temperature range corresponds to an altitude of 2 to 4 km (Takahashi, 1996). Therefore, it appears that the location of the negative charge region in the cloud is related to the temperature, rather than altitude or pressure.

Krehbiel (1986) provides further support for the constant temperature level of the negative charge region in a study of a Florida storm. As the storm grew vertically over a period of approximately 9 minutes the altitude of the upper positive charge region rose from 10 to 14 km, corresponding to a temperature change of -40°C to -60°C . The rise of

the upper positive charge region matched the rise of the radar detectable cloud top over the same period. However, the negative charge region remained at -15°C (7 km) throughout the growth of the storm.

The strong vertical growth of the storm observed by Krehbiel (1986) suggests that charged cloud particles must have been continuously removed from the negative charge region. Therefore, new negatively charged particles must have been continuously created at that altitude. The negative charge center may, therefore, be an active source of charge in the cloud.

Dye *et al* (1986) presented an *in situ* study of a small Montana thunderstorm, early in its electrical development. They observed that the negative charge center was initially localized within the transition zone between the (side-by-side) updraft and downdraft regions, at altitudes between the -15°C and -20°C levels. In a later study Dye *et al* (1988) again found that the initial negative charge was associated with an updraft-downdraft transition zone, this time at the -12°C level, with the charge confined to a region approximately 500 m across. The confinement of the early negative charge center is consistent with the cellular nature of thunderstorm electrification. In mature multicellular storms charge generation is associated with individual convective cells within the overall storm, each of which lasts approximately 10 to 15 minutes (Krehbiel, 1986).

The updraft-downdraft transition zone is considered a preferred region for rapid growth of precipitation (Dye *et al*, 1976). Dye *et al* (1986) suggested that this region was

also important to the charging because it allowed re-circulation of graupel¹ particles. They also observed that the initial accumulation of negative charge was associated with the high radar reflectivity region of the storm, representing large precipitation particles. This suggests that the electrical development of a thunderstorm is associated with the development of precipitation.

2.2.2 Large Scale Electric Fields

Even in clear air there exists an atmospheric electric field of between -100 and -200 V m^{-1} at the ground, known as the fair-weather field². The field is created by the 300 kV potential difference that exists between the negatively charged ground and the positively charged ionosphere. This potential difference is believed to be maintained by global thunderstorm activity (Reiter, 1992).

Beneath a thunderstorm the electric field is opposite in sign from the fair-weather field, except beneath the positively charged anvil. Before the initial electrification of a storm vertical electric fields measured at the ground are typically less than 1 kV m^{-1} . The initial electrification is not observed until vigorous convective growth of the storm occurs, raising the radar detectable cloud top to above the -20°C altitude. At that time electric fields at the ground rise rapidly to approximately 8 kV m^{-1} and can produce the first lightning stroke within 5 to 10 minutes (Krehbiel, 1986; Dye *et al*, 1988).

¹ Graupel are ice particles, 0.5 to 5 mm diameter and with a density of approximately 0.5 g m^{-3} , that grow by the accretion of cloud droplets (riming).

² The sign given to the fair-weather field is a matter of convention. According to the standard physics convention an upward pointing vector field is positive, therefore the fair-weather field is negative. However, much of the older literature in the field of atmospheric electricity adopted the convention that the fair-weather field is positive (Uman, 1969). Either convention is considered acceptable as long as it is stated clearly which is being used (Reiter, 1992). In the current work the standard physics convention has been adopted.

Electric fields measured at the ground are typically much less than those measured within the cloud, because of a shallow 'blanket' of positive charge associated with corona from the ground (Krehbiel, 1986). From inside a growing thunderstorm Dye *et al* (1988) observed the vertical field rise from less than 1 kV m^{-1} to more than 50 kV m^{-1} in less than 15 minutes during initial electrification. However, this is still much less than the field required for the dielectric breakdown of air, which is 1600 kV m^{-1} at an altitude of 6 km. Marshall *et al* (1995) showed that large-scale electric fields inside a thunderstorm rarely exceed 150 kV m^{-1} before lightning strokes discharge the cloud. This electric field corresponds to the 'breakeven' field of air, at which energetic electrons with energies above 1 MeV can produce an electron avalanche. They suggested that the action of these energetic electrons could initiate lightning at the relatively low electric fields observed.

Sudden reversals in the vertical electric field are often observed beneath a thunderstorm, indicating the presence of positive charge above the observing location (Krehbiel, 1986). These reversals are associated with the arrival of a downdraft and the accompanying 'rain gush', and are known as field excursions associated with precipitation. Balloonborne measurements of lower positive charge centers have also been correlated with the occurrence of field excursions at the ground.

Finally, field reversals are also observed during the dissipating stage of a thunderstorm, associated with the storm's physical collapse (Krehbiel, 1986). At this stage the field is observed to oscillate from positive to negative and back over a period of an hour or more. This reversal is known as the end of storm oscillation (EOSO). Little is

known about the charge structure of a thunderstorm during EOSO or how it changes to produce the field reversals.

2.2.3 Microphysics

To study thunderstorm electrification in the laboratory, the microphysical conditions in the charge generating region of a thunderstorm must be recreated. Unfortunately, such data are scarce because the strong updrafts and downdrafts associated with this region present a danger to aircraft. Thus, much of what little microphysical data is available is restricted to relatively small thunderstorms (Gardiner *et al*, 1985; Dye *et al*, 1986, 1988).

Electric fields in a thunderstorm do not exceed $+100 \text{ V m}^{-1}$ until 5 mm diameter graupel and ice crystal concentrations of at least 10 per litre are observed. Dye *et al* (1986) observed ice crystal concentrations of approximately 30 per litre and a liquid water content (LWC) of 0.5 to 1.0 g m^{-3} in the region of strongest electric field. The concentration of graupel with diameters of at least 1 mm in this region was 1.5 per litre, with 4 mm diameter graupel present in concentrations of 1 m^{-3} . The maximum observed ice crystal concentration and LWC were 64 per litre and 2.5 g m^{-3} , respectively, but these concentrations were not observed in the region of strongest field. Maximum 1 mm diameter graupel concentrations were 2.7 per litre with 5 mm diameter graupel present at 1 m^{-3} . Similar observations were made by Dye *et al* (1988) and Gardiner (1985).

Less than 10% of ice particles were observed to carry a charge of greater than $\pm 5 \text{ pC}$, the minimum detectable charge in all three studies. However, particles carrying detectable charge were observed even at the earliest stages of electrification, when

thunderstorm electric fields were still weak. As the electrical development of the storm progressed, the maximum observed charge carried by ice particles increased in magnitude from ± 10 pC to -50 pC, while the proportion of ice particles carrying detectable charge remained less than 10%.

The microphysical data support the theory that charging is associated with the development of precipitation in the thunderstorm. Furthermore, strong electric fields and lightning discharge have rarely been reported in ice-free clouds, which are often observed in the tropics. Therefore, it appears that thunderstorm electrification is the result of interactions involving ice particles.

2.3 Ice Crystal-Graupel Collision Charging

Several possible mechanisms of thunderstorm electrification have been suggested over the years. Of those mechanisms only three continue to attract support: convective charging (Vonnegut *et al*, 1995); inductive precipitation charging (Mason, 1988); and ice crystal-graupel collision charging, also known as non-inductive precipitation charging (Kuettner *et al*, 1981). Each of these mechanisms can explain some aspect of the observed thunderstorm charge structure but there is still discussion about which mechanism dominates, especially during the initial electrification.

The convective charging mechanism suggests that the charge structure of a thunderstorm is created by the large-scale convective motion in the storm. Cloud and precipitation particles are charged by capturing ions available beneath the cloud and in the surrounding air, and the dynamic structure of the storm acts to distribute the charged

particles appropriately. However, it has not been shown that such motion could occur quickly enough to account for the rapid development of the electric field that is observed. Furthermore, the number of ions available beneath the storm, which provide the charge on the cloud particles, appears to be insufficient to account for the electrification until strong electric fields have already developed in the storm (Saunders, 1995).

Unlike the convective mechanism, the inductive charging mechanism suggests that the electrification of a thunderstorm is the result of collisions between cloud and precipitation particles in the storm. The charge transfer is driven by the electric field of the storm, which polarizes the particles vertically. When smaller particles rebound off of the underside of larger particles, the polarizations create a potential difference that drives the charge transfer with a strength that is proportional to the electric field. However, charged precipitation particles observed at the earliest stages of the electrification are often found to carry much more charge than could be explained by the prevailing electric field (Saunders, 1995).

Most researchers currently support the ice crystal-graupel collision charging mechanism, mainly because of the relatively large body of experimental results that support it. However, contradictory observations by different researchers and inconsistencies between the theory and observations remain unresolved. This situation provides the motivation for the current study. A short description of the history of the ice crystal-graupel collision charging mechanism will highlight the current limitations of the theory.

2.3.1 *Experimental Evidence*

Reynolds *et al* (1957) presented the first observation of ice crystal-graupel collision charging, using an experiment that would become the standard setup for the studies that would follow. Their experimental apparatus is shown schematically in Figure 2.2. The experiment is performed inside a cloud chamber, filled with steam to produce a cloud of supercooled water droplets. The droplet cloud is seeded to produce ice crystals using dry ice (solid CO₂) or an object cooled to liquid nitrogen temperatures. The ice crystals collide with a rime covered target particle that is rotating in the cloud chamber. The charge transferred to the riming target is measured as a current or voltage to ground. Ice crystal concentrations and sizes are determined by collecting ice crystals that fall out of the cloud onto formvar³ covered slides.

Reynolds *et al* (1957) reported two results that are consistent with those of later studies. First, it was found that no significant charge transfer occurred in a cloud composed entirely of supercooled droplets or entirely of ice crystals. Second, the charge transfer to the target was found to be a function of the LWC of the cloud. Reynolds *et al* varied the LWC and observed that the charge transferred to the target changed from negative to positive as the LWC was increased to above 0.6 g m⁻³. The temperature at which these experiments were performed was not specified.

Contrary to future studies, Reynolds *et al* (1957) suggested that the sign of the charge transfer is a function of the relative number concentrations of droplets and ice

³Formvar is a liquid plastic solution. Cloud particles that are collected on a formvar covered slide leave impressions as the liquid plastic hardens. When the cloud particles evaporate the impression left in the plastic can be observed under a microscope. See Section 3.1.2.1.

crystals. They found that at ice crystal concentrations of 10^2 to 10^5 per litre the riming target charged negatively. At ice crystal concentrations of 10^4 to 10^6 per litre the target charged positively.

The work of Reynolds *et al* (1957) was quantified and extended over a wide range of cloud conditions by Takahashi (1978) and Jayaratne *et al* (1983). Both studies found that the charge transfer was primarily a function of LWC and cloud temperature. For a fixed LWC, the sign of the charge transfer changed from positive to negative as the cloud temperature was lowered (see Figure 2.3). The temperature at which this transition occurs is known as the reversal temperature. When the LWC was changed, the reversal temperature changed as well. Jayaratne *et al* found that the sign reversal occurred at

Figure 2.2 The experimental setup used by Reynolds *et al* (1957), Takahashi (1978) and Jayaratne *et al* (1983). The cloud chamber is filled with steam and seeded to produce ice crystals. The charge transferred to the riming target by colliding ice crystals is measured as a current or voltage to ground. Jayaratne *et al* (1983) also performed experiments with a stationary target, where the cloud was drawn past the target.

warmer temperatures as the LWC decreased. Saunders *et al* (1991) found that when the effective LWC (ELWC)⁴ was increased above 1.3 g m^{-3} no reversal temperature existed to temperatures below -30°C .

Jayaratne *et al* (1983) also found that the charge transfer is a function of ice crystal size, target speed and the presence of contaminants in the supercooled water droplets. Increases in ice crystal size and target speed caused the charge transfer to increase exponentially, with exponents 4.9 and 3.2, respectively.

The study of ice crystal size was extended by Keith and Saunders (1990) to ice crystals with diameters up to $800 \mu\text{m}$. They found that the dependence of the charging on ice crystal size is much greater for small ice crystals than for larger ice crystals, with exponents 3.8 for small ice crystals (less than initially reported by Jayaratne *et al*, 1983), 1.9 for ice crystals with diameters between $155 \mu\text{m}$ and $452 \mu\text{m}$, and 0.9 for larger crystals. Despite the weaker dependence on size for larger ice crystals, observed charge transfer magnitudes were much higher than for small crystals. Maximum observed transfers were 90, 120 and 220 fC per collision ($1 \text{ femtoCoulomb} = 10^{-15} \text{ Coulomb}$) for 300, 380 and $800 \mu\text{m}$ ice crystals, respectively. Jayaratne *et al* (1983) reported approximately 1 fC per collision for $100 \mu\text{m}$ ice crystals. It was suggested, therefore, that collisions with large ice crystals would play the dominant role in thunderstorm electrification despite their low concentrations.

⁴The ELWC is the mass of liquid water that would be swept out of a cloud by a target. It is a function of the particle shape and the droplet size distribution of the cloud. See Section 3.1.1.4.

Figure 2.3 The charge transferred to the target per collision, measured by Jayaratne *et al* (1983), as a function of temperature for a speed of 2.9 m s^{-1} and an effective liquid water content of 1 g m^{-3} . After Jayaratne *et al* (1983).

Keith and Saunders (1990) also extended the study of the effect of velocity. They found a different dependence on velocity for positive and negative charge transfers. Positive charge transfers measured at -12°C increased with velocity as $V^{2.5}$ (less than reported by Jayaratne *et al*, 1983). Negative charge transfers measured at -25°C increased as $V^{2.8}$.

Jayaratne *et al* (1983) found that the type and concentration of contaminants in the cloud droplets affected the charging in a complicated manner. Even low concentrations of contaminants changed the sign of the charge transfer observed and increased the charge transfer by an order of magnitude. Maximum charge transfers increased from 1 fC to 10 fC per collision. Similar observations regarding contaminants were made by Reynolds *et al* (1957).

2.3.2 A Model of Thunderstorm Electrification

Jayaratne and Saunders (1984) used the results of Jayaratne *et al* (1983) as the basis for a simple model of thunderstorm electrification, shown schematically in Figure 2.4. At temperatures below the reversal temperature (higher altitudes), graupel and soft hail particles in the cloud acquire negative charge after collisions with ice crystals. Ice crystals are charged positively by the same collisions. At temperatures above the reversal temperature (lower altitudes), the sign of the charge transfer is positive to the graupel.

Ice crystals charged positively by collisions at altitudes above the reversal temperature will rise in the updraft-downdraft transition zone to form the upper positive charge region and the positively charged anvil. Graupel charged negatively by the same collisions will descend relative to the ice crystals because they are heavier. They will be

Figure 2.4 A conceptual model of thunderstorm electrification presented by Jayaratne and Saunders (1984). Rising positive ice crystals create the upper positive charge region. Rising negative ice crystals from below and falling negative graupel from above meet around the level of the reversal temperature to form the negative charge center. If falling negative graupel undergo enough collisions below the reversal temperature they acquire net positive charge and can form a pocket of lower positive charge.

met by rising ice crystals, charged negatively by collisions at altitudes below the reversal temperature. The rising negative ice crystals and falling negative graupel particles meet at altitudes surrounding the reversal temperature to form the negative charge center of the storm. Finally, some of the descending negatively charged graupel may undergo enough collisions below the reversal temperature to change their net charge to positive. In regions of the cloud where conditions favor this sign change, a pocket of lower positive charge will form.

2.3.3 Problems With The Model

Although the qualitative theoretical model of thunderstorm electrification presented by Jayaratne and Saunders (1984) seems plausible, there are several inconsistencies with the experimental results. The first is that the magnitude of the charge transfer observed by Jayaratne *et al* (1983) is insufficient to explain the charge observed on cloud particles. Dye *et al* (1986) performed rough calculations of ice crystal collision probabilities and charge transfers, based on the results of Jayaratne *et al*, and compared the results to the particle charges observed *in situ*. It was found that, having “selected very favorable values for each of the parameters”, concentrations of 100 per litre or more were required to explain the rapid initial development of the electric field (from 100 V m^{-1} to 8 kV m^{-1} in less than 8 minutes) or the presence of graupel with detectable charge at early stages of the electrification.⁵ Ice crystal concentrations of more

⁵ In a later paper, Latham and Dye (1989) repeated the calculations assuming most of the charge transfer came from collisions with larger ice crystals. They found that the number of collisions was sufficient if ice crystals between 300 and 500 μm were assumed to exist in concentrations of 1 to 7 per litre. These concentrations correspond to those observed by Dye *et al* (1986) for this size range. However, Latham and Dye assumed that the dependence of the charge transfer on ice crystal size would be to the exponent 4 for all sizes. Keith and Saunders (1990) later showed that the dependence was approximately to the exponent 2 and 1 for ice crystals of 300 and 500 μm , respectively. Therefore, Latham and Dye over-estimated the charge acquired by graupel.

than 100 per litre may be present in larger thunderstorms. However small thunderstorms, like the one studied by Dye *et al*, have appreciably lower concentrations and cannot provide enough ice crystal-graupel collisions to account for the observed charge.

Another problem is that the qualitative thunderstorm model assumes a rather convenient set of conditions. The schematic shows a reversal temperature of approximately -15°C , which is within the temperature range typically observed for the negative charge center of a thunderstorm. According to the results of Jayaratne *et al* (1983) and Saunders *et al* (1991), this reversal temperature would correspond to an effective LWC of approximately 0.5 g m^{-3} (the corresponding actual cloud LWC would be higher and would depend on the droplet size distribution). However, much higher LWC's are often observed in thunderstorms – large thunderstorms often exceed LWC's of 2 g m^{-3} . If the resulting effective LWC is 1 g m^{-3} the reversal temperature would move to approximately -20°C , as shown in Figure 2.3. For effective LWC's greater than 1.3 g m^{-3} no reversal temperature was observed, and graupel particles were always charged positively by collisions. In this case the model cannot explain the development of the main negative charge center. Instead the expected charge structure would be inverted, with negatively charged ice crystals above a positive charge center. This is never observed.

Figure 2.5 illustrates the greatest difficulty with the ice crystal-graupel collision mechanism. Figure 2.5 (a) shows the sign of the charge transfer versus temperature and effective LWC as observed by Jayaratne *et al* (1983), Saunders *et al* (1991), and other studies from the same group (known as the Manchester group). Figure 2.5 (b) is a similar

diagram of the results of Takahashi (1978). Clearly there is significant disagreement in the reversal temperatures observed in these studies. At 1 g m^{-3} Takahashi reports a reversal temperature of -9°C , compared to approximately -20°C reported by Jayaratne *et al.* Using the different experimental results, numerical models of thunderstorm electrification arrive at significantly different thunderstorm charge structures. Different

Figure 2.5 Charging patterns observed by (a) Jayaratne *et al* (1983) and (b) Takahashi (1978). The lines indicate charge sign reversal temperatures. The charging patterns observed are significantly different, and no satisfactory explanation for the difference has been presented. After (a) Saunders (1993) and (b) Takahashi (1978).

groups using either of the two data sets in their models claim varying degrees of success.

Recently attempts have been made by the Manchester group to recreate the experiments of Takahashi (1978). They suggested that the differences were due to different ways of measuring the cloud LWC in the two experimental setups (Brooks and Saunders, 1995). Takahashi has not accepted this argument, and claims to have recently reproduced his original results (Takahashi, personal communication).

2.4 Proposal For Current Research

The difficulties described clearly indicate the need for an independent investigation. However, the cloud chamber technique used in previous experiments has some serious flaws that must be addressed. Primarily, the conditions in a cloud chamber cannot be controlled well enough to provide reliable data. In particular, ice crystals growing in the chamber do so at the expense of the liquid water in the cloud and sediment out, so the LWC and droplet size distribution are changing continuously. Since changes in the LWC are known to affect the sign and magnitude of the charge transfer, analysis of the charging data is extremely complicated.

Furthermore, the ice crystal concentrations used by Jayaratne *et al* (1983) and the Manchester group are much too high. Jayaratne *et al* quote ice crystal concentrations of 10^6 per litre or more. No estimate of ice crystal concentration is given by Takahashi (1978). However, the method of seeding described (a piece of dry ice placed in the chamber for the duration of the experiment) would suggest very high ice crystal concentrations, perhaps even higher than those of Jayaratne *et al*. High ice crystal

concentrations make very small charge transfers easier to measure because the number of collisions with the riming target are high. However, the conclusion of Reynolds *et al* (1957), that the charging is a function of the ratio of the concentrations of ice crystals and droplets, has largely been ignored. This conclusion suggests that the results of Jayaratne *et al* and Takahashi may not be directly applicable to thunderstorms, since even large storms have much lower ice crystal concentrations (order of 100 per litre or less).

A new study of the ice crystal-graupel collision charging mechanism is warranted if it can provide better control of the simulated cloud conditions and more accurately recreate the microphysical conditions in the charge generating region of a thunderstorm. The strong dependence of the charging on cloud conditions suggests that such an experiment may provide data significantly different from those presented by either the Manchester group or Takahashi. New results should better reflect the charge transfers occurring during ice particle collisions in a thunderstorm. The Triple Interaction Facility, developed for this study and introduced in the following chapter, provides the environment for such an experiment.

Chapter Three

Experimental Facilities and Procedures

3.1 The Triple Interaction Facility

An experimental investigation of ice crystal-graupel collision charging requires a facility capable of producing a cloud composed of ice crystals, supercooled water droplets, water vapor and graupel. The simple cloud chamber design used in previous studies is capable of producing such a cloud. However, as described at the end of Chapter 2, the cloud chamber produces conditions that are constantly changing during an experiment and that often do not represent the environment observed in thunderstorms.

A new facility has been developed for this study that can produce three phase clouds without the limitations of the cloud chamber design. The facility is known as the Triple Interaction Facility, shown schematically in Figure 3.1. The name is taken from a report of a workshop on “The future of laboratory research and facilities for cloud physics and cloud chemistry” (List *et al.*, 1986). The report discusses the need for a facility that could be used to study interactions between three or more precipitation and cloud particle types under well controlled conditions. The current facility is the first reported to meet the prescribed requirements.

The Triple Interaction Facility consists of three distinct parts: the icing physics wind tunnel, the ice crystal growing chamber, and the measuring system. The tunnel and

Triple Interaction Facility (TIF)

Figure 3.1: The Wind Tunnel: [1]Tunnel fan; [2]Heaters; [3]Turning vanes; [4]Pipe to vacuum pump; [5]Cooling element; [6]Access door; [7]Double wall flaps; [8]Perforated wooden board; [9]Tunnel kettle; [10]Ice crystal injection; [11]Water injection; [12]Contraction; [13]Pressure measuring nipples; [14]Measuring section (a: inner wall; b: outer wall). The Ice Crystal Chamber: [15]Ice crystal growing chamber (2.4m x 2.4m x 1.2m); [16]Pipe access door; [17]Steam kettle in insulated can; [18]Chamber fan; [19]Chamber window; [20]Cold room; [21]Connecting pipe (a: ice crystal pipe; b: dry ice pipe).

chamber are used to create the cloud for all triple interaction experiments. The measuring system is experiment specific and can also accommodate investigations that do not involve electrical charging. Each part of the facility will be discussed separately. In the following subsections, all numbers in square brackets refer to the numbers that label Figure 3.1.

3.1.1 The Wind Tunnel

The heart of the Triple Interaction Facility is the University of Toronto's icing physics wind tunnel, first described by List *et al* (1987). The wind tunnel has been used for over a decade to study growth and heat and mass transfer of graupel and hailstones (Zheng and List, 1996; Greenan and List, 1995; Cober and List, 1993; Garcia-Garcia and List, 1986; Lesins and List, 1986). The tunnel has an air tight closed circuit, that can be pumped down to pressures as low as 30 kPa. Tunnel configuration can be adjusted to simulate a wide range of cloud conditions that can be held stable for hours.

For the current experiments the wind tunnel was configured to simulate graupel growth at laboratory pressure (approximately 100 kPa). The configuration was identical to that used in the experiments of Cober (1991), except that somewhat higher velocities were used in the current study.

3.1.1.1 Velocity and Temperature Control

Air flow in the wind tunnel is provided by a centrifugal fan [1]. Air is circulated counterclockwise, as shown by the arrows in Figure 3.1, to create an updraft at the measuring section of the tunnel. Depending on the tunnel configuration and fan speed, air speeds of 0.6 to 30 m s⁻¹ can be achieved in the measuring section. The current

experiment was performed at speeds between 3 and 7 m s⁻¹, which correspond approximately to the fall speeds of 3 mm to 5 mm diameter graupel (Locatelli and Hobbs, 1974; List and Schemenauer, 1971).

Tunnel air speed is determined by measuring the venturi pressure drop of the flow across the contraction [12] below the measuring section. The pressure drop between two nipples [13] mounted flush with the inner tunnel walls is measured using a Setra Systems Model 264 low differential pressure transducer. The voltage output of the transducer is continuously monitored and converted into an air speed by the wind tunnel computer. One hundred velocity measurements are averaged and the average is recorded at a rate of approximately three times per second. A Prandtl tube, accurate to ± 0.1 m s⁻¹, was used to calibrate the tunnel air speed versus voltage output from the pressure transducer. The Prandtl tube was placed in the center of the measuring section during calibration, at the location where the riming target would be placed.

Temperature control is provided by combining the actions of a refrigeration unit [5] and three heaters [2]. The refrigeration unit provides excellent stability at very low temperatures, but at temperatures above -15°C the compressor operates near threshold and has a tendency to cycle on and off. The heaters provide a thermal load above -15°C that the compressor easily and continuously removes, without cycling. The compressor and heater powers can be adjusted independently to provide cloud temperatures ranging from +30°C to -30°C. At temperatures above -6°C the tunnel temperature can be kept constant to within $\pm 0.3^\circ\text{C}$, and to less than $\pm 0.1^\circ\text{C}$ at lower temperatures.

For low to moderate speed graupel experiments, modifications are made to the tunnel to reduce the maximum air speed and improve temperature stability. The double wall flaps [7] are lowered and a perforated wooden board [8] is placed in the air flow. This redirects a large volume of tunnel air from between the inner walls [14a] to the square-ring outer walls [14b] of the measuring section, thus reducing the volume of air passing through the inner measuring section and lowering the air speed. The increased airflow passing through the outer square-ring of the tunnel draws heat away from the walls, providing a very flat temperature profile within the measuring section at low speed.

Tunnel temperature is measured using a Copper-Constantan thermocouple (see Figure 3.3) placed inside the measuring section [14], the output of which is amplified 1000 ($\pm 0.1\%$) times by a linear chopping operational amplifier. The thermocouple is shielded by a small piece of metal attached to the inner measuring section wall. The shield prevents rime ice from collecting on the thermocouple during experiments, which would cause the thermocouple to warm. The cold junction of the thermocouple was kept at room temperature, inside a metal box to minimize temperature fluctuations. The temperature of the cold junction was measured using an electronic temperature sensor ($\pm 0.1^\circ\text{C}$), placed in thermal contact with the thermocouple amplifier. Thermocouple output was adjusted to compensate for the cold junction temperature.

The amplified thermocouple output was calibrated against a thermometer immersed in a mercury bath, cooled to -30°C . Thermocouple and thermometer measurements were recorded every 0.2°C as the mercury bath was warmed to $+40^\circ\text{C}$. A triple point tube was also used to calibrate both the thermocouple and the thermometer.

The range of the thermocouple calibration was -29.0 to 37.7°C , and was accurate to $\pm 0.1^{\circ}\text{C}$.

During experiments, the output of the thermocouple amplifier is measured by an analog-to-digital converter in the wind tunnel computer. As with the velocity data, 100 amplified thermocouple measurements are averaged and recorded three times per second.

3.1.1.2 Liquid Water Content

The droplet cloud in the wind tunnel is provided by spray nozzles [11], located 2 m below (upwind of) the riming target (see Figures 3.3 and 3.4). Depending on the air speed and the type and number of spray nozzles used, the cloud LWC can range from 0.2 to 20 g m^{-3} . For this study a single nozzle was used, providing LWC's of 0.2 to 2.0 g m^{-3} . Distilled, de-ionized water was used in all charging experiments.

The spray nozzle consisted of a Spraying Systems Company #1/8 JJ miniature air atomizing nozzle with a J1050 fluid cap. The spray nozzle is operated by injecting pressurized air and water into opposite ends of a small chamber. The water is ejected through a small hole at the top of the atomizing nozzle while the air is directed through six larger holes surrounding the water hole. The pressurized air disrupts the water stream, causing it to break up into droplets of various sizes. The droplet size distribution is a function of the nozzle air flow rate, while the LWC is a function of the water flow rate.

The air and water flow rates to the nozzle are controlled using a set of calibrated flowmeters. Air flow is controlled using a Matheson #605 flowmeter, water flow is controlled with a Matheson #602 flowmeter.

Inside the tunnel the nozzle assembly is heated to prevent freezing of the water line and nozzle tip. The assembly is wrapped in Nichrome wire, a flexible electric heating element, and insulated to reduce heat transfer to the tunnel air. Nozzle temperature is measured using a thermocouple placed on the nozzle tip, near the point of droplet ejection. The power to the Nichrome wire is adjusted to keep the nozzle temperature at approximately +10°C before an experiment is begun. During an experiment, when water and air flow to the nozzle have been established, the nozzle temperature typically drops below +8°C. Calculations presented by Cober (1991) indicate that droplets have sufficient time to cool to ambient tunnel temperatures before they reach the measuring section, even at measuring section air speeds of 7 m s⁻¹. (Below the contraction the air speed is 1/12 that of the measuring section.)

The LWC of the cloud is calculated from the water flow rate as measured with the flowmeter. By geometric considerations, the LWC can be determined using the equation:

$$W_f = \frac{F}{AV} \quad (3.1)$$

where W_f is the cloud LWC measured in g m⁻³, F is the nozzle water flow rate measured in g s⁻¹, A is the cross sectional area of the measuring section in m², and V is the tunnel air speed in m s⁻¹. The LWC calculated by this method agrees well with the estimated LWC derived from measurements with the 2DCG probe (see Section 3.1.1.3).

Equation 3.1 assumes that the distribution of liquid water in the measuring section is uniform. Cober (1991) showed that droplets are evenly distributed in the measuring section except in a 1 cm boundary layer along the perimeter. He showed that the ice that

collected on the measuring section walls corresponded to the mass of liquid water that would be expected in the boundary layer for an even distribution, so Equation 3.1 can be used without correction.

3.1.1.3 Droplet Size Distribution

The droplet size distribution of the tunnel cloud can be varied by adjusting the air flow rate to the nozzle. Cober (1991) measured the droplet size distribution in the measuring section for five different air flow rates, characterized in Table 3.1 and Figure 3.2. The size distributions were measured using the Magnesium Oxide Technique (May, 1950; Cober, 1987), in which droplets are collected on thin glass strips coated in MgO. Droplet impacts leave craters that are proportional to the droplet diameter. The craters are observed under a microscope to determine the size distribution of the cloud.

The size distributions measured by Cober were verified by observations made in the current study, using a 10 μm resolution Knollenberg 2D Cloud Greyscale Probe (2DCG – described in Section 3.1.3.2 and in detail in Appendix A). The LWC of the cloud is estimated from the partial spectra measured with the 2DCG probe (minimum

Nozzle air flow rate [air] = mm	Mean droplet diameter [d] = μm	Mean volume diameter [d _v] = μm	Median volume diameter [d _m] = μm	Number of droplets measured	Target collection efficiency [%]
83.0 ± 0.5	19.2	21.8	24.6 ± 2.0	1396	34 ± 2
75.0	20.8	24.2	29.6	2485	39
68.0	23.0	27.0	34.8	1169	50
63.0	24.6	29.6	39.8	1296	61
61.0	24.8	30.2	42.0	735	51

Table 3.1 Characteristics of the droplet size distributions used in the experimental series. The three distributions marked in bold were used throughout the experimental series, the others in a few experiments at -16°C (see Figure 4.6). Data was provided by Dr. Stewart G. Cober (personal communication), except for the target collection efficiency which was calibrated in the current study (see Section 3.1.1.4).

Figure 3.2 Droplet size distributions used in charging experiments. Labels indicate the mean droplet size for each distribution (see Table 3.1) Figure (a) shows the number concentration and cumulative mass distribution of the three distributions used in all experimental series. Figure (b) shows the same for the two other distributions used only in a few experiments at -16°C . Spectra were measured in ice crystal free clouds using the Magnesium Oxide technique. Data was provided by Dr. Stewart G. Cober (personal communication).

observable particle size, 35 μm). For each distribution the 2DCG estimated LWC corresponds to the mass distribution expected for the observable portion of the droplet spectrum, to within experimental error ($\pm 10\%$). Droplets larger than those measured by Cober are observed, but in very low concentrations.

3.1.1.4 Effective Liquid Water Content

The effective liquid water content (ELWC) is a measure of the mass of liquid water a target would sweep out of a given cloud. Since the collection efficiency of a droplet is a function of the droplet size and target shape, the ELWC for a given target changes with the droplet size distribution. For a given distribution and target shape the ELWC is equal to the actual LWC multiplied by the target collection efficiency for the distribution.

The collection efficiencies of the droplet size distributions listed in Table 3.1 were calibrated at a velocity of 5.0 m s^{-1} and an actual LWC of 0.99 g m^{-3} . Before each calibration experiment the combined mass of the target and a small metal plate was determined using an electronic balance, accurate to 0.01 mg. The small metal plate was placed in a freezer where it cooled to -35°C .

At the start of each calibration experiment the target was shielded so that rime was not collected until the nozzle system reached equilibrium. When the nozzle water flow rate stabilized the shield was removed and an electronic stopwatch was started. The target collected rime for 3 to 4 minutes before the calibration experiment was stopped, corresponding to a volume swept out by the target of at least 200 litres. To end the experiment the shield was replaced and the stopwatch was halted.

The riming target was removed from the tunnel and placed onto the small metal plate. The cooled plate slowed the melting and evaporation of the rime ice and collected any ice that melted off the target. The target, the plate and the rime ice were weighed together and the total mass was subtracted from the mass of the target and the plate alone. The collection efficiency of the droplet size distribution was calculated by dividing the mass of rime collected by the target by the total mass of liquid water available in the swept out volume of the target. The results of the calibrations are listed in Table 3.1.

3.1.1.5 Electrification of the Droplet Cloud

It is well known that the process of droplet formation by breakup, such as that used by the tunnel spray nozzles, produces droplets that are charged (Iribarne, 1972; Iribarne and Klemes, 1974). The charge develops through a shearing of the charged double layer that exists in a liquid, near the interface with a solid or a gas. The average charge acquired by a droplet is a function of several factors, including liquid conductivity, droplet size, and the time required for completion of the disruption. The charge acquired by any given droplet is random.

Droplet charging is observed in the spray-produced tunnel cloud. Section 4.2 presents a typical experiment which illustrates droplet charging. The droplet charging consists of a high frequency signal and a bias current. The high frequency signal is caused by the high time resolution of the charging measurement (3 measurements per second). In the short time required to complete a single measurement, the spectrum of droplets collected by the riming target may be significantly different from the cloud average. Since each droplet of a given size in the spectrum is carrying a random charge,

the charge transferred to the target in 1/3 of a second will have an appreciable standard deviation. The magnitude of the standard deviation is a function of the droplet size distribution and LWC. Smaller mean droplet sizes or higher LWC's produce a higher standard deviation because more droplets are swept out by the target in a given time. However, the standard deviation for a given droplet size distribution is approximately constant for the duration of a single experiment. Therefore, the high frequency droplet charging can be treated statistically as a measurement error that can be determined for each experiment.

The bias current is a function of two effects. First, when droplets separate from the nozzle water stream, the stream is left with a residual charge equal and opposite to that acquired by the droplet. However, the stream is still in contact with the nozzle assembly, which is grounded to prevent the buildup of charge. Some of the residual charge on the water stream passes to ground through the nozzle assembly, leaving a net charge on the droplet cloud. Second, the collection efficiency of the riming target is not uniform for all droplet sizes. Smaller droplets have much lower collision probabilities than larger droplets. Since the droplet charging is a function of droplet size, the riming target would acquire a net charge even in a cloud that is net neutral.

The magnitude and sign of the bias current is random, and can vary significantly between experiments performed under identical conditions. However, during a given experiment the bias is constant to within ± 0.5 pA. This is illustrated in the experiment presented in Section 4.2. Therefore, the bias current can be determined in the ice crystal

free phases of a given experiment, and subtracted from the current associated with ice crystal charging.

3.1.1.6 Relative Humidity

The relative humidity in the measuring section is measured by the psychometric method, using a thermocouple placed next to the thermocouple used to measure tunnel temperature. The thermocouple was calibrated in the manner described in Section

3.1.1.1. When both thermocouples are ice free, tunnel temperatures determined by the two thermocouples agree to within $\pm 0.1^{\circ}\text{C}$.

To determine the relative humidity a thin coating of ice is deposited onto the second thermocouple. If the tunnel air is supersaturated or subsaturated with respect to ice, the ice coating on the thermocouple will warm by deposition or cool by sublimation, respectively. The temperature difference between the dry and ice bulb thermocouples is used to determine the tunnel relative humidity with respect to ice (RHI). For an ambient tunnel temperature of -15°C , a difference in temperature of $\pm 0.1^{\circ}\text{C}$ corresponds to $\pm 4.3\%$ RHI.

Below -15°C thermocouple measurements always differ by 0.1°C so relative humidity measurements are not considered accurate below this temperature. At higher temperatures relative humidities in the measuring section are always observed between 100% RHI and 109% RHI. The tunnel relative humidity increases with LWC. Since the cloud droplets act as a source of water vapor, more droplets raise the humidity.

A kettle [9] placed inside the tunnel can be used to produce steam to increase the relative humidity. Under ideal conditions it can be raised beyond 100% with respect to water (RHW). By varying the power to the kettle the relative humidity can be adjusted. In preliminary experiments, humidities as high 140% RHI were measured when the kettle was used. However, the control over the exact relative humidity is poor because the rime ice on the tunnel walls will absorb a large amount of the water vapor. Furthermore, the kettle is a large heat source inside the tunnel, so temperature control is less effective. Typical temperature stability during relative humidity experiments is approximately $\pm 1^\circ\text{C}$. The tunnel kettle was not used in the current study.

3.1.2 The Ice Crystal Chamber

The Triple Interaction Facility was designed to produce a three phase cloud: ice crystals, droplets, and vapor. As is described in Section 3.1.1, the wind tunnel is responsible for generating the droplets and vapor in the cloud. The ice crystal chamber [15] is the source of ice crystals for the facility.

The chamber has dimensions 122 cm x 244 cm x 239 cm (7.1 m^3), and is contained in a large cold room (walk-in freezer) that can be cooled to -25°C . Typically, the chamber is kept at approximately the same temperature as the wind tunnel. However, the temperature of the chamber can be different from that of the tunnel for experiments that study the effect of ice crystal habit. The chamber is connected to the wind tunnel by a 5 m long, 10 cm diameter aluminum pipe [21a] that is surrounded by a second, 15 cm diameter PVC pipe [21b] that is filled with dry ice. The inner pipe enters the tunnel

through one of the access ports [10], 10 cm below the spray nozzle. Inside the chamber a lid is hinged to the inner pipe to control access to the tunnel [16].

Steam is produced using an insulated kettle placed beneath the chamber [17], and enters the chamber through a plastic funnel. The chamber fan [18] is used to maintain the temperature in the chamber before ice crystals are initiated, and to cool and dry the chamber between experiments. Ice crystals are initiated in the chamber by briefly inserting a piece of metal that has been cooled in liquid nitrogen. The ice crystals grow at the expense of the liquid water in the chamber cloud.

Ice crystals are forced into the tunnel by the tunnel vacuum pump [4] which is run continuously and creates a pressure difference between the chamber and the tunnel. Ice crystals are carried to the tunnel in the airflow as soon as they are first observed in the chamber. The travel time from the chamber to the tunnel measuring section is 10 to 15 seconds. With the pump operating at full power ice crystals are observed in the tunnel measuring section for more than 6 minutes before the chamber is depleted. Typical experiments are run for 2 to 4 minutes only to ensure that the ice crystal chamber conditions remain relatively constant.

3.1.2.1 Ice Crystal Habits, Sizes and Concentrations

Ice crystals were collected in the cloud chamber using the formvar replication technique (Takahashi and Fukuta, 1988). Slides coated with liquid formvar were placed on the floor of the cloud chamber. Ice crystals that fall onto the slides leave impressions in the formvar as it dries. The slides are allowed to sit in the cold room for 30 minutes, allowing the ice crystals to evaporate and leaving only the impression of the ice crystal in

the dry formvar. The slides can then be removed and observed under a microscope at room temperature.

The ice crystal habits observed in the chamber vary with temperature in the manner described by Takahashi *et al* (1990). For example, at -8°C plates are the predominant ice crystal habit; at -16°C dendrites are observed. Triangular habits have been observed at all temperatures, in very low concentrations. These ice crystals are likely created because the cloud is seeded with liquid nitrogen. Similar triangular habits have been observed in high cirrus clouds at temperatures below -40°C (Hallett, 1997).

Ice crystals as large as $180\text{ }\mu\text{m}$ have been observed in the chamber, although most are between 50 and $100\text{ }\mu\text{m}$. Ice crystal concentrations in the chamber have not been measured, but visual estimates suggest concentrations of at least 10^4 per litre.

Ice crystal concentrations and sizes in the measuring section are determined using the PMS 2DCG probe, described in Section 3.1.3.2 and in detail in Appendix A. The probe sample volume is located in the center of the measuring section, 30 cm below the riming target. Ice crystals are distinguished from droplets by a simple habit determination algorithm. The resolution of the probe is $10\text{ }\mu\text{m}$, and the algorithm provides a minimum detectable ice crystal size of $40\text{ }\mu\text{m}$.

Figure 4.2(b) gives an example of 10 second average ice crystal concentrations observed in the measuring section in a typical experiment. Ice crystal concentrations of 60 to 300 per litre are typical, with maximum concentrations of 450 per litre. Maximum ice crystal sizes in the measuring section were 80 to $90\text{ }\mu\text{m}$. Ice crystals of all sizes are

present in any 10 second average. However, most ice crystals are 50 to 70 μm in the longest dimension.

3.1.3 The Measuring Section

The measuring section ([14] and Figure 3.3) of the wind tunnel is where target particles are suspended and observations are made. It has a double wall construction with an inner cross section of 17.8 cm x 17.8 cm and a height of 70 cm. The walls of the measuring section are made of aluminum and act as a Faraday cage around the target particle. As a result, very little external electrical noise is observed.

The measuring section instrumentation consisted of: two thermocouples to measure temperature (Section 3.1.1.1) and relative humidity (Section 3.1.1.6); a pressure transducer to measure tunnel pressure and air speed (Section 3.1.1.1); a picoammeter; and a 2DCG probe. Two IBM 386 personal computers were used to collect data from the measuring section. One computer was dedicated to running and recording data from the 2DCG probe, the other collected data from the remaining instruments. The clocks on the two computers are synchronized to within ± 0.5 seconds of each other before the start of each experiment.

3.1.3.1 The Photographic System

The inner and outer walls of the measuring section can be exchanged to allow the use of a variety of camera equipment to provide a photographic record of experiments. Photographs were taken using a Nikon F camera with a 55 mm micro lens and a Nikon M extension tube. In this configuration the camera has a depth-of-field of approximately 8 mm. The camera is mounted on the access door of the measuring section. The access

door has a recessed cavity in which the camera sits. When the access door is closed the inside of the cavity is flush with the inner wall of the measuring section, allowing the camera to be as close to the target particle as possible.

Three strobe lights are used to illuminate cloud particles for visual and photographic observations, a Strobotac 1538-A controlling two Stroboslave 1539-A. The lights shine through glass windows located 45° , 90° , and 135° from the camera mount on the measuring section access door. Cloud droplets could only be observed using the 45° and 135° strobes. Ice crystals were best seen when the 45° and 90° lights were used. When all three lights are used the whole cloud was illuminated, but ice crystals were difficult to observe because of the intensity of the light reflected by the much higher droplet concentration. With only the 45° and 90° lights active very few droplets were

Figure 3.3 (a) Horizontal and (b) vertical cross section of the wind tunnel measuring section: [1]Inner walls; [2]Outer walls; [3]Target particle; [4]Particle support; [5]Teflon collar; [6]Low noise coaxial cable; [7]Access door; [8]Camera; [9]Strobe lights; [10]Strobe windows; [11]Thermocouples; [12]Riming shields; [13]2DCG windows; [14]2DCG laser beam; [15]2DCG object plane; [16]Pressure transducer nipples. Note that the thermocouples [11], the 2DCG windows [13], and the riming shields [12] are actually located on the perpendicular walls, beneath the strobe lights [9].

illuminated and the ice crystals could be easily observed.

3.1.3.2 The 2DCG Probe

A Particle Measuring Systems (PMS) 2-Dimensional Cloud Particle Greyscale (2DCG) probe was used to determine the number concentration and size distribution of wind tunnel cloud particles during experiments. The probe was especially designed for use in laboratory studies (Particle Measuring Systems, 1977). It has a resolution of $10\text{ }\mu\text{m}$, which is significantly better than the $25\text{ }\mu\text{m}$ resolution typical of aircraft probes.

Since the data acquired by the 2DCG probe is critical to the analysis of the charge transfer experiments, significant efforts were made to ensure the proper operation of the probe and correct analysis of probe data. The general operation of the probe, the method of data analysis, and an analysis of error is presented in detail in Appendix A and is summarized here.

Figure A.1 shows a schematic of the optical system of the probe. The 2DCG is designed to take shadow images of particles that pass through a laser beam shining on a series of 60 optical array elements. The horizontal resolution of the optical array is $10\text{ }\mu\text{m}$, providing a total width of $600\text{ }\mu\text{m}$. The probe electronics converts, in parallel, the output from each optical element into 2 bits of digital data that correspond to 4 possible levels of shadow. The shadow levels correspond to 0%, 25%, 50% and 75% shadowed. The probe electronics sample the data at a rate determined by the True Air Speed (TAS) clock. The TAS clock signal is provided by a Wavetek Model 19 digital function generator and is set to provide a vertical resolution that matches the horizontal resolution.

For a tunnel velocity of 5 m s^{-1} the TAS clock frequency is set to 500 kHz, which gives a vertical resolution of $10 \text{ }\mu\text{m}$. Particle images are recorded in the probe hardware memory and downloaded by the probe computer.

Image file sizes ranged from 3 to 25 megabytes in size depending on the droplet size distribution, the LWC and the duration of the experiment. Probe data was analyzed after experiments using custom analysis software. The software identified particle images, applied selection criteria (see below), and estimated particle concentrations and associated errors.

Joe and List (1987) presented a technique for improving the accuracy of 2D greyscale probes. It was shown that the darkness of the shadow in an image could be calibrated against the distance of the object from the optical plane of the probe, as a function of the object size. The criteria was used in the current work to accurately define the sample depth of the 2DCG, in the manner described by Joe and List (1987).

The 2DCG probe was calibrated for image size and for the selection criteria of Joe and List (1987) by analyzing images of glass beads of known size. The selection criteria were applied by the analysis program to reject images that were outside of the defined sample depth of the probe.

3.1.3.3 The Charging Measurements

Figure 3.4 shows the target particle support system and the instrumentation used for charging measurements. The rigid, stationary particle support is made of electrically insulating PVC plastic. Enclosed in the hollow support is a low-noise coaxial cable,

connected to a contact strip. The strip provides both electrical contact and mechanical support to the target particle, which is inserted into a 6 mm hole at the bottom of the support. The support passes through the inner wall of the measuring section through a 5 cm diameter Teflon collar (see Figure 3.3). The collar prevents rime ice from creating a secondary path from the target to ground.

At the other end of the coaxial cable, the target is connected to ground through a 500 M Ω resistor and a Keithley model 485 digital picoammeter. The model 485 is a 4½ digit, $\pm 20,000$ count picoammeter with a resolution of 0.1 pA. It can perform up to 3 readings per second.

The picoammeter was calibrated following the procedure specified in the instrument manual (Keithley Instruments, 1984b). The calibration was performed using a

Figure 3.4 Structural and electrical schematic of a typical target particle and the particle support. The cylindrical brass target (48 mm long, 5 mm diameter) is used to simulate a graupel particle.

Hewlett-Packard 741B DC Standard voltage source that can generate a voltage up to 1 V that is accurate to 20 μV . The manufacturer specified accuracy of the picoammeter is $\pm(0.4\%$ of the reading + 4 counts (0.4 pA)) for the 2 nA scale at which the picoammeter was operated.

The 500 M Ω resistor serves to minimize amplifier noise in the picoammeter (Keithley Instruments, 1984a, 1984b) and to minimize the currents from any thermal EMF's in the system. The low noise coaxial cable and the aluminum walls of the measuring section also act to minimize electrical noise in the system. The observed baseline noise, with the tunnel running at maximum air speed and no cloud in the measuring section, is typically less than ± 0.1 pA, the resolution of the picoammeter. A 0.1 pA current corresponds to a potential difference of 50 mV between the target particle and ground.

The digital output of the picoammeter is collected by the wind tunnel computer, which also provides timing for experiments. Whenever the picoammeter signals that a reading is available the tunnel computer records the picoammeter output and state, the time, and 100 point averages of the temperature, relative humidity, laboratory air pressure, and tunnel velocity. At the end of each experiment comments are recorded and appended to the data file for the experiment.

3.2 Experimental Procedure

Each experiment is performed at fixed ice crystal chamber and tunnel temperatures, air speed, liquid water content, and droplet size distribution. The tunnel

and chamber temperatures are typically set at the start of each day, and all experiments performed that day are at the same temperature.

Prior to an experiment the riming target is cleaned of any ice collected during the previous experiment, so that each experiment begins with the target in the same condition. The ice crystal chamber kettle is turned on, to allow the kettle time to warm and for steam to accumulate in the chamber. The pipe access lid is opened. The 2DCG probe and the tunnel vacuum pump are also started before the experiment begins, and the desired wind tunnel air speed is set.

An experiment is begun by starting the wind tunnel computer, at which time a hand held stopwatch is also started. Ten seconds after the experiment begins, the air flow to the tunnel nozzle is started, set for the desired droplet size distribution. Water flow to the nozzle is started 10 seconds later. The experiment is run for at least 1½ minutes with only a droplet cloud to allow time for the nozzle system to reach equilibrium and to record a baseline for the charging measurements.

When the water flow to the nozzle system has stabilized the experiment is continued in the chamber cold room. When sufficient steam is available in the chamber ice crystals are initiated in the chamber by inserting a nitrogen cooled metallic object. The stopwatch time and the temperature in the ice crystal chamber are recorded. The time is recorded again when ice crystals are first observed in the chamber, as is a qualitative estimate of the density of steam in the chamber (see Section 4.3.3).

The ice crystal phase of the experiment continues for 2 to 4 minutes. During this time fine adjustments are made to the cloud LWC and tunnel air speed. Visual observations of the ice crystal and droplet concentrations are made through the camera access port in the measuring section door, using the appropriate tunnel strobe lights. The temperatures of the nozzle, the ice crystal pipe, and the tunnel cooling element are recorded. The ice crystal phase ends when the pipe access lid is closed, after which the chamber kettle is shut off. The time that the access lid is closed is recorded, as is the chamber temperature.

The droplet cloud is maintained for at least 1 minute after the end of the ice crystal phase. At the end of the experiment the water flow to the nozzle is stopped. The nozzle air flow is shut off at least 30 seconds later. The times that the nozzle water and air flow are stopped are recorded. The tunnel computer and 2DCG probe are stopped no less than 10 seconds after the nozzle air is shut off. After the tunnel computer is stopped, the wind tunnel vacuum pump is turned off.

After each experiment, comments are recorded and appended to the experiment data file. Comments always include the recorded times and temperatures, particular experiment conditions, observations of the target after the experiment, and other general comments and observations.

Chapter Four

Results

4.1 Introduction

The analysis of ice crystal-graupel collision charging experiments involves calculating the average charge transferred to the target particle per ice crystal collision for specified cloud conditions. In the current study cloud conditions spanned temperatures between -5°C and -21°C ; LWC's between 0.25 and 2.0 g m^{-3} ; three droplet size distributions; ice crystal chamber relative humidities between ice and water saturation; and velocities from 3 to 6 m s^{-1} . In total 172 experiments were performed. The data from these experiments are catalogued in Appendix B.

This chapter begins with the presentation of a selected typical experiment that will be analyzed in detail to illustrate the stability of the experimental conditions and general characteristics of the analysis. Next, the results of the entire experimental series will be presented, along with general comments. A complete discussion of the results is reserved for Chapter 5. Possible parameterizations of the data will be offered at the end of this chapter. Note that all errors quoted in the following sections represent one standard deviation from the mean.

4.2 Results of a Single Experiment

Figure 4.1(a) shows the current from the riming target to ground as a function of elapsed time. The corresponding tunnel conditions are shown in Figure 4.2. The experiment was performed with a cloud LWC of $1.43 \pm 0.02 \text{ g m}^{-3}$, at a temperature of

$-21.04 \pm 0.04^{\circ}\text{C}$ and a velocity of $5.29 \pm 0.05 \text{ m s}^{-1}$. The tunnel conditions and errors are calculated from measurements made from the beginning of the analysis baseline (2:10 minutes elapsed time) until the end of the ice phase. The droplet distribution with a mean diameter of $20.8 \mu\text{m}$ was used in this experiment, giving an ELWC of 0.56 g m^{-3} .

As discussed in Section 3.1.1.4, droplets formed by spray nozzles are charged by the disruption process. The current associated with the charged droplets can be seen in Figure 4.1(a) during the interval labeled “Ice Free”. The droplet current includes both a high frequency noise and a baseline bias current. The baseline bias current in this experiment was nearly zero ($0.02 \pm 0.26 \text{ pA}$), however non-zero bias currents of $\pm 2 \text{ pA}$ were typically observed. The highest bias currents observed were between $+6$ and $+7 \text{ pA}$, which occurred in four experiments performed at -5°C using the $19.8 \mu\text{m}$ mean diameter droplet distribution. The time required to reach a stable bias current varied between experiments, usually ranging from 30 seconds to 3 minutes. The bias current of the experiment shown in Figure 4.1(a) stabilized in approximately two minutes. The four experiments that had 6 to 7 pA bias currents were exceptions, taking up to 7 minutes to stabilize. However, regardless of the time required to reach a stable bias current, once achieved the bias current remained constant to within $\pm 0.5 \text{ pA}$ nearly indefinitely. For example, constant bias currents were observed for 7 minutes in the four experiments that had high bias currents. This stability was observed for all cloud conditions.

The high frequency noise is a generally a function of droplet size distribution, with smaller droplets producing more noise. The standard deviation of the droplet charging about the bias current in Figure 4.1(a) is 3.5 pA , which is higher than typical for

Figure 4.1 (a) The charging pattern and (b) the ten second average number concentration of 'not round' images for an experiment performed at -21°C , an ELWC of $0.56 \pm 0.006 \text{ g m}^{-3}$ and a velocity of $5.29 \pm 0.05 \text{ m s}^{-1}$. The $20.8 \text{ }\mu\text{m}$ mean diameter droplet distribution was used. The background current and number concentration were calculated using a baseline from 2:10 to 3:10 minutes. Ice crystals are initiated in the ice crystal chamber at 3:00 minutes, are first observed at 3:10 and are first detected in the measuring section between 3:30 and 3:40 minutes.

Figure 4.2 The tunnel conditions corresponding to the experiment shown in Figure 4.1. The average temperature and velocity in the measuring section for the duration of the baseline and ice phase of the experiment (2:10 to 7:00 minutes) were $-21.04 \pm 0.04^{\circ}\text{C}$ and $5.29 \pm 0.05 \text{ m s}^{-1}$.

this droplet distribution. Typical standard deviations for the 19.2 μm , 20.8 μm and 24.8 μm distributions are 3.0 pA, 1.5 pA and 0.6 pA, respectively. The reason for the higher droplet noise in this experiment is unknown, but both higher and lower than average standard deviations were occasionally observed with all droplet size distributions. In the current experiment the standard error of the estimate of the mean current, determined using a 1 minute baseline from 2:10 to 3:10 minutes (179 measurements), was 0.26 pA.

Figure 4.1(b) shows the ten second average number concentration of 'not round' cloud particle images, larger than 30 μm diameter, recorded by the 2DCG probe. 'Not round' is defined as any image that has maximum and minimum radii that differ by at least 1 pixel. Almost all ice crystals images are classified as 'not round' according to this criteria, but so are a large fraction of droplet images. These droplet images produce the background noise observed in the ice free phase of the experiment. The 'not round' droplet images are caused by three factors: the resolution of the probe; a small difference between the droplet velocity and the probe True Air Speed setting; or droplet trajectories that are not perpendicular to the sampling plane of the probe. The magnitude of the number concentration baseline is proportional to both the cloud LWC and mean droplet diameter. The mean baseline was always calculated over the same time interval that was used to determine the bias current. In this case the baseline concentration was 403 ± 57 per litre. The standard error of the estimate of the mean concentration, determined using 6 ten second averages, was 23.3 per litre.

The estimated error of the number concentration for each size bin is calculated for every ten second average. Each estimate includes the standard deviation for the number of particles counted, assuming a Poisson distribution, and estimated errors for the depth-of-field calibration (see Appendix A). The error for the total number concentration is calculated from the error estimates for each size bin using standard error propagation analysis.

The ice phase of the experiment began at 3:00 minutes elapsed time, when ice crystals were initiated in the ice crystal chamber. The temperature in the chamber at the start of the ice phase was -17°C . Ice crystals were first observed in the chamber at 3:10 minutes. Ice crystals are typically observed in the wind tunnel measuring section 10 to 20 seconds after they are observed in the chamber. This is consistent with the first peak in number concentration observed during the ice phase of the experiment (elapsed time 3:30 to 3:40 minutes, concentration 511 ± 67 per litre). The ice phase continued for 4 minutes, after which the chamber access door was closed. However, measurable ice crystal concentrations were observed for less than 2 minutes (3:30 to 4:50 minutes). After the drop in ice crystal concentration the current from the target to ground returns to within ± 0.5 pA of the baseline by 5:30 minutes.

After the ice crystal chamber access door is closed at 7:00 minutes the current from the target to ground returns to a baseline that is 2.4 pA more positive than the initial baseline, until 9:00 minutes when the water flow to the nozzle is stopped. This appears to be caused by ice crystals that are observed in the measuring section after the end of the ice phase. It is believed that these ice crystals are fragments of the rime that collects on

the chamber access door during the ice phase and that break off when the access door is closed. The sign of the change in the baseline is consistent between experiments, with magnitudes between 0.5 and 3 pA. As a result, the post ice phase baseline is never used in the analysis.

For each number concentration measurement made during the ice phase the corresponding ten second average current to ground was calculated, as was the standard error of the estimate of the mean. For each point the charge transfer per ice crystal collision, C , was calculated using the following formula:

$$C = \frac{I}{A V N} \quad (4.1)$$

where I is the average current to ground minus the mean bias current, N is the number concentration minus the mean bias concentration, A is the cross sectional area of the target particle ($2.4 \times 10^{-4} \text{ m}^2$), and V is the average tunnel velocity from the beginning of the baseline until the end of the ice phase. The error for each estimate is calculated using standard error propagation analysis.

Note that no adjustment is made to the estimated charge transfer to account for the collision efficiency of the cylindrical riming target with the ice crystals, which is known to be less than unity for ice crystals smaller than $80 \text{ }\mu\text{m}$ (Keith and Saunders, 1988). Therefore, the actual charge transferred to the target during an ice crystal collision will be systematically greater than suggested by Equation 4.1.

A common feature of experiments performed at intermediate to high ice crystal chamber humidities (see Section 4.3.3) was that charge transfers estimated from data

points within 30 seconds to 1 minute of the beginning of the ice phase were always much more negative than those at later stages. This effect is illustrated in Figure 4.1. The charge transfer per ice crystal collision calculated using data from the ten second intervals beginning at 3:30, 3:50 and 4:00 minutes are -14 ± 10 , -17 ± 7 , and -18 ± 14 fC per ice crystal collision, respectively. The charge transfer calculated at 4:40 minutes is -4.1 ± 2.0 fC per collision. This is caused by the depletion of the water vapor in the ice crystal chamber by the growing ice crystals, which is shown in Section 4.3.3 to favor more positive charging. Despite this behavior, the average charge transfers were calculated using all charge transfer estimates made within the ice phase of an experiment, except where noted (see Section 4.3.3 and Figure 4.10). This leads to a systematic underestimate of the charge transfer at high relative humidities that is not accounted for in the error analysis.

The final estimate of the charge transfer per collision is determined by taking the weighted average of the ten second estimates. Only points with number concentrations near to or greater than the estimated concentration error are included in the average. Lower concentrations have much larger associated charge transfer errors, larger than the estimated charge transfer, and so have little influence on the weighted average. The number of points used for the weighted average varied between experiments from 1 to 17, although 4 to 8 was typical. The standard deviation associated with the weighted average is equal to the square root of the inverse of the sum of the weights (Mandel, 1964). In the current experiment the final estimate was $C = 5.5 \pm 1.9$ fC per collision, using the four data points listed above.

Note that all charge transfer estimates were made using ten second intervals.

Using different intervals in the analysis may lead to somewhat different estimates for the weighted average, since the number of data points per experiment and the associated errors would change. The effect of changing the interval would depend on the interval chosen, so this effect was not been considered in the analysis of experimental error.

4.3 Results of the Experimental Series

The experimental series revealed dependencies of the charge transfer on droplet size distribution, temperature, ELWC, ice crystal chamber relative humidity and velocity. These dependencies will be presented in the following sections. Note that, except where indicated, chamber relative humidities were ‘intermediate’ (between ice and water saturation; see Section 4.3.3) at the time of ice crystal initiation.

The series also revealed no dependency of the charge transfer on the temperatures of the ice crystal chamber or the connecting pipe. The ice crystal chamber temperature was typically within $\pm 3^{\circ}\text{C}$ of the tunnel temperature at the time of ice crystal initiation. However, in some experiments the chamber temperature was as much as 10°C warmer than the tunnel temperature. No measurable effect on the charge transfer was observed when the chamber and tunnel temperatures were not the same. The temperature of the connecting pipe typically ranged from -30°C to -10°C as the dry ice in the cooling pipe evaporated over a period of several experiments. Again, no measurable effect on the charge transfer was observed.

4.3.1 *The Effect of Droplet Size Distribution*

Experiments were performed using cloud LWC's ranging from 0.25 to 2.0 g m^{-3} .

Experiments at fixed LWC were repeated using three droplet size distributions, with mean droplet diameters of 19.2 μm , 20.8 μm and 24.8 μm . Figure 4.3(a) shows the estimated charge transfer per collision as a function of LWC for the three distributions, measured at -21°C . The charge transfer observed at similar LWC's varies with the distribution. For the 24.8 μm and 20.8 μm distributions the maximum charge transfer is observed at a LWC of 1.0 and 1.4 g m^{-3} , respectively. For the 19.2 μm distribution the peak charge transfers occur at 0.5 and 1.5 g m^{-3} .

Figure 4.3(b) shows the same results, this time plotted against the effective LWC for each distribution. The figure shows that the peak charge transfers correspond to an ELWC of approximately 0.55 g m^{-3} . The relationship between the charge transfer and the ELWC holds well throughout the range observed. The exception is the sharp spike observed at an ELWC of 0.16 g m^{-3} , which corresponds to the 19.2 μm distribution peak at 0.48 g m^{-3} LWC. This spike is the result of variations in the ice crystal chamber relative humidity that will be described in Section 4.3.3.

In light of the dependence on droplet size distribution, the following sections present charge transfer measurements as a function of ELWC rather than actual cloud LWC. However, the droplet size distribution used in each experiment will still be indicated on all graphs.

4.3.2 The Effect of Temperature and Effective Liquid Water Content

Figures 4.3(b), 4.4, 4.5 and 4.6 show the charge transfer as a function of ELWC observed at tunnel temperatures of -5°C , -8°C , -11°C , -14°C , -16°C , -18°C and -21°C .

(a)

(b)

Figure 4.3 The effect of droplet size distribution. Figure (a) shows the charge transferred per collision for three droplet size distributions, for liquid water contents (LWC) between 0.25 and 1.9 g m^{-3} . Figure (b) shows the same data plotted against the effective LWC for each distribution. The results are consistent with a dependence on effective LWC rather than actual LWC.

Figure 4.4 The effect of temperature and effective liquid water content. The average charge transferred to the riming target is plotted as a function of ELWC. At temperatures above -10°C positive charge was transferred to the target at all ELWC's.

Figure 4.5 The effect of temperature and effective liquid water content. The sign of the charge transfer to the target changes from positive to negative below -11°C for ELWC's greater than 0.5 g m^{-3} . At -14°C a strong sensitivity to the relative humidity of the ice crystal chamber is observed, accounting for the scatter of points at 0.5 g m^{-3} .

Figure 4.6 The effect of temperature and effective liquid water content. Below -16°C the charge transferred to the target is negative at all ELWC's. At -16°C the large scatter of data points is indicative of a very strong sensitivity to the relative humidity of the ice crystal chamber. The series at -16°C includes two additional droplet distributions. The results of these experiments were consistent with a dependence of the charging on ELWC.

For fixed ELWC the magnitude and sign of the charge transfer is a function of temperature. Above -10°C the charge transferred to the riming target is positive at all ELWC's; below -15°C the charge transfer is always negative. The exact temperature of the sign reversal is a function of the ELWC, ranging from between -8°C and -11°C at 1.0 g m^{-3} to -16°C at 0.1 g m^{-3} . Within the range of ELWC's studied, the reversal temperature seems to warm with increasing ELWC.

The data are generally consistent with a dependence of the charge transfer on ELWC rather than LWC. Notable exceptions are the sharp spike at -16°C , 0.33 g m^{-3} ELWC, and the large variability between experiments at -14°C and -16°C with ELWC's between 0.5 and 0.6 g m^{-3} . As with the spike observed at -21°C , these features are caused by variations in the ice crystal chamber relative humidity.

4.3.3 The Effect of Ice Crystal Chamber Relative Humidity

As mentioned in previous sections, it was observed that the charge transfer to the target was dependent on the relative humidity in the ice crystal chamber. The effect is illustrated in Figure 4.7. It was found that the sign of the charge transfer could be changed by varying the chamber relative humidity, under otherwise identical tunnel conditions.

The Triple Interaction Facility was not equipped to measure the relative humidity in the ice crystal chamber. The chamber humidity was classified qualitatively as low, intermediate or high, depending on the density of steam observed in the chamber at the time of ice crystal initiation. The steam density was determined using a flash light, shone through the chamber window. Low chamber humidity corresponded to 'wisps' of steam,

Figure 4.7 The charge transfer observed when the ice crystal chamber was near ice and water saturation at the time of ice crystal initiation. For all four cases the tunnel temperature is $-14.1 \pm 0.2^\circ\text{C}$, the velocity is $5.26 \pm 0.03 \text{ m s}^{-1}$, and the $20.8 \mu\text{m}$ mean diameter droplet distribution was used.

Figure 4.8 The charging pattern and measuring section relative humidity observed at ice crystal chamber relative humidities near ice saturation. The charge transfer to the target is positive. The average charge transfer per ice crystal collision is $+4.4 \pm 2.3$ fC. Tunnel conditions: $-14.3 \pm 0.05^\circ\text{C}$; 5.28 ± 0.05 m s⁻¹; 0.52 ± 0.005 g m⁻³ ELWC.

Figure 4.9 The charging pattern and measuring section relative humidity observed at ice crystal chamber relative humidities near water saturation. The charge transfer to the target is strongly negative. The average charge transfer per ice crystal collision is -13.1 ± 7.0 fC. Tunnel conditions: $-14.0 \pm 0.01^\circ\text{C}$; $5.29 \pm 0.04 \text{ m s}^{-1}$; $0.51 \pm 0.005 \text{ g m}^{-3}$ ELWC.

Figure 4.10 The charging pattern and measuring section relative humidity observed at intermediate ice crystal chamber relative humidities, between ice and water saturation. The sign of the charge transfer changes during the duration of the experiment. The average charge transfer per ice crystal collision is -2.2 ± 0.3 fC per collision from 3:20 to 5:31 min., $+0.93 \pm 0.71$ fC from 5:31 to 6:11 min., and $+6.2 \pm 2.3$ fC from 6:11 to 7:00 min. Tunnel conditions: $-14.2 \pm 0.01^\circ\text{C}$; $5.23 \pm 0.05 \text{ m s}^{-1}$; ELWC $0.51 \pm 0.005 \text{ g m}^{-3}$.

Figure 4.11 The relative humidity series. All experiments were performed at an effective liquid water content of 0.51 g m^{-3} and a velocity of 5.3 m s^{-1} . The only difference between measurements at the same temperature is the relative humidity of the ice crystal chamber. Higher chamber humidity always promotes charging that is more negative. The ice and water saturation envelopes are calculated from Equations 4.3 and 4.4, respectively.

entrained within mostly clear air. An even distribution of steam throughout the chamber, but with good visibility through the steam, was classified as intermediate humidity. High humidity was reached when the corners of the chamber opposite to the window were no longer visible.

Low ice crystal chamber humidity corresponds to approximately 100% relative humidity with respect to ice. High humidity corresponds to approximately 100% relative humidity with respect to water. Intermediate humidity is between ice and water saturation.

Figure 4.8 shows the charging pattern and measuring section relative humidity for an experiment performed at low ice crystal chamber humidity, near ice saturation, at a temperature of -14°C , ELWC of 0.5 g m^{-3} , and using the $20.8 \mu\text{m}$ distribution. Figure 4.9 shows the same data for an experiment performed under identical tunnel conditions but at high chamber relative humidity, near water saturation. In both cases the measuring section relative humidity is approximately 103% of ice saturation. However, at low chamber humidity weak positive charging is observed, while strong negative charging is observed at high chamber humidity.

Figure 4.10 shows an experiment performed under the same conditions as Figures 4.8 and 4.9, but at intermediate relative humidity, between ice and water saturation. In this case the sign of the charge transfer to the target, with respect to the baseline, changes from positive to negative within the duration of the experiment. The sign change appears to occur as the steam in the chamber is depleted by the growing ice crystals. Similar sign

change charging patterns were observed in two other intermediate relative humidity experiments: again at -14°C , 0.5 g m^{-3} ELWC using the $20.8 \text{ }\mu\text{m}$ distribution and at -15°C , 0.5 g m^{-3} ELWC using the $24.8 \text{ }\mu\text{m}$ distribution.

Figure 4.11 shows the average charge transfer to the target at a constant ELWC as a function of temperature and ice crystal chamber relative humidity. At temperatures below -10°C relative humidities near water saturation always caused a stronger negative charge transfer than humidities near ice saturation. Above -10°C humidities near water saturation weakened the positive charge transfer to nearly zero. In preliminary experiments, not shown in Figure 4.11, negative charging was observed at -5.5°C and -2.5°C . These experiments were performed at an ELWC of 0.7 g m^{-3} and velocities of 4 m s^{-1} and 3 m s^{-1} , respectively. Chamber relative humidities near ice saturation always caused the charge transfer to be more positive.

For each temperature the range of the minimum and maximum average charge transfers, corresponding to the highest and lowest relative humidities, is well defined. The greatest effect of the relative humidity was observed between -13°C and -17°C . At other temperatures charge transfers observed at ice crystal chamber humidities near water saturation and near ice saturation differ by 2 to 8 fC per collision. Between -13°C and -17°C the range is between 20 and 25 fC per collision, and changes in the sign of the transfer were often observed.

As with the experiment described in Section 4.2, estimates of the charge transfer made with data from the first 30 seconds to 1 minute of the ice phase of near water

saturation experiments were always much greater than the experiment average. For the experiment shown in Figure 4.9, estimates from ten second intervals between 5:00 and 5:40 minutes suggest charge transfers of -44 ± 27 fC per ice crystal collision, compared to -13 ± 7 fC per collision averaged over the entire ice phase. Near -16°C maximum negative charge transfers greater than -60 fC per collision were observed near the start of the ice phase. Since these very strong charge transfers are very short lived phenomena, typically lasting much less than 1 minute, these data are not shown in Figure 4.11. They are also not represented in the range of charge transfers described by the 'envelope' curves in Figure 4.11 (see Section 4.4). Only charge transfers averaged over the entire ice phase are shown, except where a sign change during an experiment was observed as in the experiment shown in Figure 4.10.

4.3.4 The Effect of Velocity

Figure 4.12 shows the weighted average of the strongest negative charge transfers achieved at ice crystal chamber relative humidities near water saturation and a temperature of -16°C , for tunnel velocities of 3, 4 and 6 m s^{-1} . The maximum charge transfer at these velocities is much weaker than the maximum charge transfer observed at 5 m s^{-1} . The charge transfers observed are approximately equal to the maximum negative charge transfers observed at temperatures -11°C and -20°C in Figure 4.11. Time constraints did not permit a further investigation of the effect of velocity in this study.

4.4 Parameterizations of the Charge Transfer Per Collision

Figure 4.13(a) shows a surface plot of the charge transfer per ice crystal collision as a function of temperature and ELWC. The surface represents 134 of the 141 data

The Effect of Tunnel Velocity

Figure 4.12 The velocity series. The negative charge transfer observed with the ice crystal chamber at water saturation is plotted as a function of tunnel velocity. The data at 5.3 m s^{-1} was calculated from the intermediate to high relative humidity experiments shown in Figure 4.11. All experiments were performed at an effective liquid water content of $0.50 \pm 0.02 \text{ g m}^{-3}$ and a tunnel temperature of $-16.1 \pm 0.6^\circ\text{C}$. The plotted points represent weighted averages of the charge transfer observed in 2, 3, 8 and 3 experiments performed at 3.1 , 4.1 , 5.3 and 6.1 m s^{-1} , respectively. The negative charge transfer observed at 5.3 m s^{-1} is much stronger than those observed at any other velocity.

points shown in Figures 4.3 through 4.6. Negative charge transfers stronger than -20 fC, observed in nine experiments performed at -16°C, were not included because they represent very high relative humidity experiments. Figure 4.13(b) shows the surface plot of a polynomial fitted to the experimental data. The fitted surface represents the charge transfer under conditions of intermediate ice crystal chamber relative humidity.

In order to parameterize the charge transfer per collision, data were chosen at 5 constant ELWC's: 0.24 ± 0.05 , 0.35 ± 0.05 , 0.55 ± 0.05 , 0.70 ± 0.05 , and $0.98 \pm 0.02 \text{ g m}^{-3}$. Third order polynomials were fitted to the charge transfer as a function of temperature at constant ELWC, using the unweighted least squares method. Since data points at constant temperature and ELWC do not necessarily represent estimates of the charge transfer under identical conditions, because of variations in the ice crystal chamber relative humidity, the weighted least squares method was not used.

The coefficients of the five charge transfer versus temperature fits were then fit to third order polynomials of ELWC, again using the unweighted least squares method. This technique resulted in much better agreement with the data, as measured using the χ^2 statistic, than was achieved using the surface fitting techniques described by Press *et al* (1992). The resulting function of charge transfer to the target, C , in fC versus temperature, T , in °C and ELWC, E , in g m^{-3} is as follows:

$$\begin{aligned}
 C = & (48.16 - 363.7E + 760.7E^2 - 472.7E^3) \\
 & + (17.01 - 132.1E + 257.8E^2 - 153.58E^3)T \\
 & + (1.488 - 11.84E + 22.58E^2 - 13.40E^3)T^2 \\
 & + (0.03762 - 0.3045E + 0.5786E^2 - 0.3460E^3)T^3
 \end{aligned} \tag{4.2}$$

Figure 4.13 Surface plots of (a) the charge transfer observed and (b) the charge transfer predicted by Equation 4.2, as a function of temperature and effective liquid water content.

Figure 4.14 The difference between the observed charge transfers and the model prediction of Equation 4.2. (a) A surface plot of the difference as a function of temperature and effective liquid water content. (b) A frequency plot of the difference for all 134 data points, plotted in 1 fC intervals.

The parameterization is valid for temperatures between -5°C and -21°C , an ELWC between 0.1 g m^{-3} and 1.0 g m^{-3} , and a velocity of 5 m s^{-1} .

It should be noted that the number of significant figures quoted in Equation 4.2 does not indicate the accuracy of the model estimate. Rather, the precision of the parameterization is necessary to account for the range of the ELWC that is modeled, which covers one order of magnitude. The accuracy of the model prediction is indicated by the data presented in Figure 4.14. The difference between the measured charge transfer and the model estimate, as a function of temperature and ELWC, is shown in Figure 4.14(a). The largest errors occur at low ELWC where few data points were available and at -18°C where the model overestimates the negative charge transfer to the target. Large errors also occur at -15°C but these differences are due to the scatter of points caused by the sensitivity to the ice crystal chamber relative humidity. Figure 4.14(b) shows a histogram of the difference between the measured charge transfer and the model estimate for all 134 fitted points. Over 73% of the points are within $\pm 3 \text{ fC}$ of the measured value.

The effect of ice crystal chamber relative humidity was characterized by two curves, shown in Figure 4.11, that model the extremes of the charge transfers observed at the lowest and highest humidities. These curves correspond to chamber relative humidities of 100% with respect to ice and water, respectively. The envelope of charging, C_i , in fC, observed near ice saturation was fit to a third order polynomial function of temperature, T , in $^{\circ}\text{C}$, as follows:

$$C_i = -26.9 - 9.04T - 0.711T^2 - 1.60 \times 10^{-2}T^3 \quad (4.3)$$

For chamber humidities near water saturation the envelope of charging, C_w , in fC, was fit to a fourth order polynomial of temperature:

$$C_w = -23.6 - 3.43T + 0.771T^2 + 0.133T^3 + 4.45 \times 10^{-3}T^4 \quad (4.4)$$

Equations 4.3 and 4.4 are valid for an ELWC of 0.5 g m^{-3} at temperatures between -5°C and -19.5°C and at a velocity of 5 m s^{-1} .

As can be seen in Figure 4.11, Equation 4.3 provides good agreement with the most positive charge transfer observed. Equation 4.4 disagrees slightly with the data, predicting the strongest negative charge transfer at -17°C rather than at -15°C . The negative charge envelope may be better approximated by a discontinuous function that is polynomial below approximately -10°C and zero at temperatures above. However, selecting the correct temperature for the discontinuity would require data between -11°C and -8°C that are not currently available. Therefore, a discontinuous fit to the high humidity charging is not presented in this study.

It should be emphasized that Equation 4.4 is a parameterization of the maximum negative charge transfer averaged over the entire ice phase of a high relative humidity experiment. As discussed in Sections 4.2 and 4.3.3, charge transfers observed during the first 30 seconds to 1 minute of the ice phase, when the humidity in the ice crystal chamber is the highest, are much stronger than the average over the entire ice phase. Therefore, Equation 4.4 should be considered a conservative estimate of the maximum negative charge transfer associated with relative humidities near water saturation.

Chapter Five

Discussion

5.1 The Physics of the Charge Transfer

The dependence of the charge transfer on cloud conditions indicates that the transfer is driven by one or more properties of the colliding particles that change with these conditions. If more than one property drives the transfer then they may act together or in competition. Furthermore, the properties that drive the charge transfer may be different for the riming target and the vapor-grown ice crystals.

In this section an analysis is made of possible changes to the state of the colliding ice particles that could occur as the relative humidity and velocity are varied. These changes are compared to current theories of the physical mechanism that drives the charge transfer. The charging mechanisms presented in this section have been considered in other studies for the last 10 years and are reviewed by Saunders (1993). However, no consistent model of the charge transfer has yet been offered. In this section and in Section 5.2.2 an attempt will be made to present a consistent physical model that matches the current observations in experiments and thunderstorms. Where possible, experimental tests of this model will be suggested.

5.1.1 The Effect of Relative Humidity

Section 4.3.3 presented three experiments, performed under identical tunnel conditions, that differed only in the ice crystal chamber relative humidity. It was shown that the charge transfer to the target was positive at low relative humidity, near ice

saturation, negative at high relative humidity, near water saturation, and that the sign of the charge transfer changed during the experiment at intermediate relative humidity, between ice and water saturation. In all three cases the relative humidity in the tunnel was approximately 103% of ice saturation. The dependence of the charge transfer on the ice crystal chamber humidity, rather than the tunnel humidity, suggests that the ice crystals have some 'memory' of the growing conditions in the chamber. This memory must last at least ten to fifteen seconds, the ice crystal travel time from the chamber to the measuring section.

Ice crystals grown at humidities near water saturation can differ from those grown near ice saturation in five ways: a change in ice crystal habit; higher surface temperature; the development of steps on the surface; higher dislocation density; and an increase in the thickness of the 'liquid-like' layer. These differences will be analyzed to determine the factors which may imprint the chamber conditions onto the ice crystals.

5.1.1.1 A Change in the Ice Crystal Habit

Hanajima (1949) studied the relationship between ice crystal habit, temperature and humidity. He found that, at approximately -16°C , raising the humidity from ice to water saturation changes the observed ice crystal habit from columns to plates to dendrites. Humidity driven transitions from columns to plates were observed at temperatures ranging from -12°C to -20°C . At -15°C the ice crystal habit changes from plates to dendrites with increasing relative humidity.

Changes in ice crystal habit are sudden. For example, dendrites are only observed at -15°C above 107% relative humidity with respect to ice, while plates are observed

below. At -14°C plates are observed at all relative humidities above 102% with respect to ice, columns below. However, the intermediate humidity experiment presented in Section 4.3.3 showed a gradual transition from negative to positive charging as the chamber humidity dropped towards ice saturation. Mixed habits are observed in ice crystals that are grown under changing cloud conditions, but the transition from pristine to mixed habit would still be sudden. The observed charge transfer as a function of chamber humidity (Figure 4.11) is continuous, so it cannot be explained by sudden changes.

5.1.1.2 The Ice Crystal Surface Temperature

Growth by deposition warms the ice crystal through the release of latent heat, and the faster the growth rate the warmer the ice crystal. Near ice saturation the ice crystals grow very slowly and have a temperature near the ambient air temperature. At water saturation the ice crystals grow rapidly and have a surface temperature warmer than ambient. Between ice and water saturation the surface temperature would increase monotonically with humidity, providing a continuous ice crystal memory.

However, the difference in surface temperature would not exist for the required travel time as Cober (1991) showed for droplets. Droplets with $80\text{ }\mu\text{m}$ diameter take less than 0.65 seconds to cool from 10°C to within 0.01°C of -15°C . This is much less than the time required for the droplet to travel from the nozzle to the measuring section. Ice crystals $80\text{ }\mu\text{m}$ in diameter, the largest observed in the tunnel, have much less mass and heat capacity and therefore shorter relaxation times. It is therefore reasonable to assume that the ice crystals in all three experiments have the same surface temperature at the time

of collision with the riming target, and so surface temperature cannot provide the memory for the relative humidity effect.

5.1.1.3 Steps and Dislocations

Steps and dislocations, shown schematically in Figure 5.1, are macroscopic and microscopic changes in the ice crystal structure, respectively. Since steps and dislocations are structural changes they can provide a long-term memory of the growth history of the ice crystal similar to the ice crystal habit. However, unlike the ice crystal habit, the density of dislocations and the height of steps are continuous functions of the humidity.

Steps are thin layers of molecules deposited on the surface of the ice crystal from the vapor. They are 200 Å to 10 µm in height and grow away from the edges of the ice crystal where the relative humidity is highest (Hobbs, 1974). The height and growth rate of steps is a function of the humidity but there are no reports in the literature of charges

Figure 5.1 Schematic diagram of ice crystal (a) steps, (b) dislocation and (c) liquid-like layer. The height of steps, the density of dislocations and the depth of the liquid-like layer all increase monotonically with the growth rate of the ice crystal.

associated with steps. Therefore it is not clear how steps would affect the charge transfer, except perhaps by changing the contact area of the collision.

Hobbs (1974) defines a dislocation as “a line defect in a crystal which disrupts the otherwise ideal arrangement of the atoms or molecules.” Itagaki (1970, 1983) showed that dislocations carry positive charge, between 10^{-12} and $10^{-10} \text{ C m}^{-1}$. Keith and Saunders (1990) noted that the surface density of dislocations was proportional to the growth rate of the ice crystal. They proposed that different dislocation densities on the surface of the riming target and the colliding ice crystals could explain the dependence of the charge transfer on temperature and ELWC.

The charge carriers in ice are protons (H^+ ; Hobbs, 1974). Therefore, a negative charge transfer to the riming target due to differences in the dislocation density would actually be a transfer of protons from the target to the colliding ice crystal. High chamber relative humidity always promotes stronger negative charging of the target. However, a high dislocation density on the ice crystal, caused by rapid growth in the chamber, should promote positive charging of the target because of the excess positive charge available on the colliding crystal. Since it does not, the dislocation density cannot provide an explanation for the dependence on the growing conditions observed, unless there is another competing memory mechanism that provides stronger negative charging. The competing charge transfer may be provided by mass exchange between the liquid-like layers on the ice surfaces.

5.1.1.4 The Liquid-Like Layer of Ice

The liquid-like layer of ice, shown in Figure 5.1(c), is only a few mono-molecular layers thick. It is not considered normal liquid water because it exists in equilibrium with ice on the one side and vapor on the other (Hobbs, 1974). Modern theories of the liquid-like layer show that it is a necessary consequence of the pressure and Gibbs free energy gradients between the crystal lattice and the vapor. Baker and Dash (1989) cite experimental evidence of a liquid-like layer on ice at temperatures as low as -30°C . The thickness of the liquid-like layer is a function of ambient temperature and growth rate, with higher temperatures and faster growth rates creating thicker layers (Baker and Dash, 1989).

An electrical double layer exists at the liquid-vapor surface of the liquid-like layer, similar to the double layer observed at bulk water-vapor interfaces (Iribarne, 1972). The double layer provides an excess of negative ions at the liquid-vapor interface. Baker and Dash (1989) proposed that negative charge transfer between colliding ice particles could be the result of fluid transfer between the liquid-like layers. They argued that when the ice particles come into contact liquid tends to flow from thicker to thinner layers, taking with it excess negative ions from a shearing of the double layer. Since the thickness of the liquid-like layer is proportional to the growth rate of the ice from the vapor, the particle growing more slowly would charge negatively.

Charging through liquid-like layer mass exchange is consistent with the observation that the charge transfer to the target becomes more negative with increasing ice crystal chamber relative humidity. A competition between positive charge transfer

due to dislocations and negative charge transfer due to mass exchange may explain the sign change observed during the intermediate humidity experiment presented in Section 4.3.3. An ice crystal that initially grows rapidly under intermediate to high relative humidity enters the tunnel with a thick liquid-like layer and transfers negative charge to the target. As the humidity in the chamber drops the thickness of the liquid-like layer is reduced but dislocations created during the initial rapid growth remain. When the liquid-like layer effect weakens the dislocation effect becomes dominant and positive charge is transferred to the target.

Note that the target could also charge positively if it was growing more quickly than the colliding ice crystal, by vapor deposition rather than by accretion. In this case the liquid-like layer of the target would be thicker than that of the ice crystal. This positive charge transfer would occur even if a high density of dislocations did not exist on the ice crystals from previous rapid growth. Similarly, a higher density of dislocations on the target particle than on the ice crystal would lead to negative charging of the target. Therefore, competition is not a requirement for charge transfer of either sign.

Although mass exchange between ice particle liquid-like layers can explain the dependence of the negative charge transfer on the chamber relative humidity, it is uncertain that this mechanism can provide a memory to the ice crystal that would last at least 10 to 15 seconds as observed. However, the observed dependence on relative humidity cannot be explained by any other mechanism offered in the literature (contact potentials between rimed and unrimed surfaces, Caranti and Illingworth, 1983; fracture of

rime structures charged due to temperature gradients, Caranti *et al*, 1991), and no other ice crystal properties are known to change with relative humidity.

No estimates have been made of the relaxation time of the liquid-like layer thickness on ice particles growing in changing cloud conditions. If calculations show relaxation times on the order of ten seconds or more, then the liquid-like layer must be considered the dominant charge transfer mechanism in ice crystal-graupel collision charging.

5.1.2 *The Effect of Velocity*

The previous section presents an explanation for the observed effect of relative humidity on the charge transfer between colliding ice particles. What remains to be explained is the unusual dependence of the charge transfer on the relative velocity of the colliding particles.

Figure 4.12 shows the maximum negative charge transfer observed at velocities of 3, 4, 5 and 6 m s⁻¹. All the experiments were performed at -16°C, 0.5 g m⁻³ ELWC and high ice crystal chamber humidities, near water saturation. Much stronger negative charge transfer is observed at 5 m s⁻¹ than at any other velocity. At 3, 4 and 6 m s⁻¹ the maximum negative charge transfer observed was approximately -5 fC per collision, compared to -16 fC per collision at 5.3 m s⁻¹. In all these cases the ice crystals were grown under nearly identical conditions.

Two of the properties of the collision that change with the velocity are the target rime ice density and the contact time of the collision.

At high velocities the momentum of the droplets causes them to spread before they freeze. The spreading droplets fill gaps in the rime structure and increase the density of the rime on the target. Conversely, at low velocities the rime density is lower. The density of the rime also changes with temperature for a constant velocity. At lower temperatures the droplets have less time to spread before they freeze, and so the rime density is lower. If the rime density affects the negative charge transfer then velocities higher and lower than 5 m s^{-1} would have charge transfers similar to higher and lower temperatures, respectively. In fact the charge transfers observed at 3, 4 and 6 m s^{-1} are similar to those observed at -20°C and -12°C in Figure 4.11. However, no current theories of ice particle charge transfer suggest a dependence on the rime density.

The target rime density is also a function of droplet size distribution. Larger droplets take longer to freeze so the rime density increases with mean droplet size. Three droplet size distributions were used in the experimental series. Similar maximum negative charge transfers were observed for all three, which suggests that a change in rime density cannot explain the observed velocity dependence. However, the droplet size distributions used may not produce a sufficient change in rime density to reproduce the effect shown in Figure 4.12 for velocity.

The collision velocity also affects the particle contact time during which the charge transfer takes place. Baker and Dash (1989) suggested that the mass transfer between liquid-like layers is driven by differences in fluid pressures, rather than the kinetic energy of the transfer. Elementary consideration would suggest that longer contact times allow more charge transfer, with the time required to reach equal fluid

pressures as an asymptotic upper limit. This would predict equal or higher charge transfers at lower velocities, but this is not observed. However, longer contact times may allow time for a 'neutralizing current' to develop across the contact point that would partially discharge the initial transfer. This is shown schematically in Figure 5.2.

A neutralizing current is an essential feature of the theory of droplet charging presented by Iribarne (1972). If a neutralizing current develops then the total charge transferred would not be dependent linearly on contact time. Rather, there would be a maximum charging when the difference between the charge transferred by the fluid pressure and by the neutralizing current is maximum. In this case 5 m s^{-1} may be a preferred velocity for negative charge transfer to the target, as is observed in Figure 4.12.

The velocity series presented in Section 4.3.4 is insufficient to distinguish between the rime density and the contact time theories. Future studies may continue this investigation by mapping the maximum negative charge transfer as a function of temperature for a velocity less than 5 m s^{-1} . A maximum in the negative charge transfer may be observed at a temperature warmer than -16°C , where the rime density at lower velocity would equal the rime density at 5 m s^{-1} and -16°C . If a maximum is observed at

Figure 5.2 Schematic diagram of the charge transfer between liquid-like layers. Initially fluid pressure forces negative ions from the electrical double layer of the thicker liquid-like layer to the thinner one. The potential difference developed by the fluid pressure charge transfer generates a current that neutralizes the initial transfer. This mechanism was described by Iribarne (1972) to explain the charging of droplets formed by disruption.

warmer temperatures then the charge transfer is probably a function of the rime density.

If the maximum charge transfer remains near -16°C then the stronger maximum observed at 5 m s^{-1} is likely caused by a neutralizing current in the liquid-like layer at lower velocities, and 5 m s^{-1} is a preferred velocity for negative charge transfer to the target.

5.2 Comparison with Previous Experimental Studies

5.2.1 *The Effect of Relative Humidity*

One of the greatest puzzles in the field of thunderstorm electrification has been the disagreement of the experimental results of Takahashi (1978) and Jayaratne *et al* (1983), discussed in Chapter 2. While both studies found that the charge transfer is a function of the cloud temperature and LWC they differed in most other respects. Jayaratne *et al* and the Manchester group have always maintained that the charge transfer is a function of the ELWC rather than the LWC, and this is supported by the results presented in Section 4.3.1. However, even if the results of Takahashi are adjusted to an estimated ELWC for the droplet size distributions used, the charging pattern observed by Takahashi differs significantly from that observed by Jayaratne *et al*. This disagreement has not yet been satisfactorily explained, and has been a major concern in the field for the last 15 years. The dependence of the charge transfer on the relative humidity may provide an explanation for the different charging patterns observed in these works.

Figure 5.3 shows a comparison of the charge transfers reported by Takahashi (1978), the Manchester group, and Equation 4.2 of this work. The results of the Manchester group were calculated using equations presented in Saunders *et al* (1991) and are adjusted for differences in velocity and ice crystal size according to the power law

Figure 5.3 A comparison of the charging observations of different studies at an effective LWC of 0.5 g m^{-3} . The thin solid lines indicate the envelope of charge transfer observed by varying the ice crystal chamber relative humidity from ice to water saturation. The dashed line is calculated from equations presented in Saunders *et al* (1991) and are adjusted for differences in velocity and mean ice crystal size. The dash-dot lines are taken from contour lines plotted in Figure 8 of Takahashi (1978), by assuming that 1 g m^{-3} LWC equals 0.5 g m^{-3} ELWC. The circles mark the data points taken from his figure. The two Takahashi lines show the results with and without adjustments for the velocity from Saunders *et al* (1991). Note that the unadjusted positive charge transfer of Takahashi (1978) rises to a maximum of 33 fC at -6°C , and is not shown in this figure. The thick solid line is calculated from Equation 4.2.

dependence described in that work. The data for Takahashi (1978) were taken from contour curves plotted on Figure 8 of that work, which is shown schematically in Figure 2.5, assuming that 1 g m^{-3} LWC was equivalent to 0.5 g m^{-3} ELWC. The data for Takahashi (1978) is shown with and without the adjustment for velocity presented by Saunders *et al* (1991). Figure 5.1 also shows the relative humidity envelopes of Figure 4.11, calculated using Equations 4.3 and 4.4.

With the exception of the unadjusted positive charge transfers observed by Takahashi (1978) above -9°C , all the charge transfers fall well within the relative humidity envelopes. It is therefore reasonable to assume that the different charging patterns observed by Takahashi and the Manchester group are purely the result of different cloud chamber relative humidities.

Jayaratne *et al* (1983) describes the experimental procedure of the Manchester group in some detail. In their experiments they define the ‘reference stage’ as the instant at which droplets first reappear in the cloud chamber, after the cloud has been totally depleted of droplets by the growing ice crystals. Estimates of the charge transfer as a function of temperature were made with data recorded at the reference stage because “all other variables were the same”. Since just before the reference stage the chamber cloud had been depleted of droplets, the humidity in the chamber just before the time of the measurement would necessarily be near ice saturation. This is consistent with the charging pattern shown in Figure 5.3, which suggests relative humidities near ice saturation, because of the ice crystal memory of the relative humidity.

The experimental procedure of Takahashi (1978) is not described in sufficient detail to estimate the relative humidity in his experiments. The unadjusted negative charging data shown in Figure 5.3 suggests that the experiments may have been performed at relative humidities near water saturation. However, that similarity may be artificial. The data points shown correspond to an actual LWC of 1 g m^{-3} . This was chosen because the highest reversal temperatures and strongest charging observed by Takahashi (1978) were at a LWC of approximately 1 g m^{-3} . Since the droplet size distribution used in particular experiments is unknown (Takahashi, personal communication), this LWC may or may not correspond to an ELWC of 0.5 g m^{-3} . However, the relative humidity envelopes shown in Figure 5.3 could contain charge transfers observed by Takahashi (1978) for LWC's between 0.35 and 3.5 g m^{-3} .

The effect of relative humidity can also explain the observation of Reynolds *et al* (1957) that the sign of the charge transfer changed from negative to positive when the number concentration of ice crystals was increased to greater than that of the cloud droplets. When the ice crystal concentration is very high they act as a stronger sink for vapor than the droplets are a source. This lowers the cloud chamber humidity towards ice saturation and promotes positive charging. When droplets outnumber ice crystals the humidity rises towards water saturation, promoting negative charging.

In the current experiment the ice crystals are grown in a different environment than the riming target, often at much higher humidities. The studies of Reynolds *et al* (1957), Takahashi (1978) and Jayaratne *et al* (1983) were all cloud chamber experiments, so the ice crystals and riming target were always exposed to the same relative humidities.

Still, the relative humidity appears to affect the charge transfers in previous studies as it does in the current experiment, suggesting that the vapor in the cloud affects the riming target differently than it does the vapor-grown ice crystals. This is not entirely unexpected.

Droplets freezing on the riming target surface would warm the surface much more than vapor deposition would warm the ice crystal surface. Therefore, even under identical cloud conditions the surface states of the riming target and the ice crystals are not the same. Apparently the difference in the surface states is sufficient to preserve the effect of the relative humidity.

5.2.2 The Effect of Velocity

The results of Takahashi (1978) shown in Figure 5.3 may provide further insight into the physics underlying the charge transfer. The negative charge transfers observed by Takahashi (1978) are within the high relative humidity envelope, whether adjusted for velocity or not. The unadjusted positive charge transfers observed are much higher than the maximum positive charge transfers in this work, but they agree well with all three studies when adjusted according to the velocity dependence observed by Keith and Saunders (1990).

The positive charging data suggests strongly that adjustments for the velocity are necessary. However, the velocity series presented in Section 4.3.4 showed that the maximum negative charge transfer was independent of velocity, except at 5 m s^{-1} . This is contrary to the results of Keith and Saunders (1990) who found that the charge transfer

was a strong function of the velocity. They observed that at -12°C positive charge transfers increased as $V^{2.5}$, and negative charge transfers at -25°C increased as $V^{2.8}$.

Two mechanisms were suggested in Section 5.1.2 to explain the observed velocity dependence: changes in the rime density of the target and changing contact times for the charge transfer. Neither theory could be rejected based on the limited experimental series available in this work, but assume for a moment that the velocity dependence is caused by the change in contact time. In this theory it was suggested that the negative charge transferred at low velocities may be limited by neutralizing currents that partially discharge the charge transferred by mass exchange between the colliding liquid-like layers.

It was argued in Section 5.1.1 that positive charge transfer to the target may be the result of a transfer of protons from the ice crystal if a higher density of dislocations are present on the ice crystal. The results of the Manchester group are consistent with chamber humidities near ice saturation at the reference stage. However, the initial relative humidity would be much higher so that the ice crystals would grow rapidly at first, providing a high dislocation density on the crystal but a thin liquid-like layer.

The transfer of protons between colliding ice particles may be driven by the potential difference between the colliding ice particles, or it may be driven by the kinetic energy of the collision. If the transfer of protons were driven by the potential difference then the dependence on velocity should increase with the contact time, up to an asymptotic limit, like the mass transfer between liquid-like layers. The dependence of the

positive charge transfer on $V^{2.5}$ suggests a dependence on the kinetic energy of the collision, perhaps enhanced by the potential difference.

Keith and Saunders (1990) also observe an exponential dependence of the negative charge transfer on velocity at -25°C . Baker and Dash (1989) showed that the thickness of the liquid-layer decreases with lower temperature, and that the layer is only observed above approximately -30°C . Therefore, the charge transfer between liquid-like layers would be weak at -25°C . This is consistent with the small range of charge transfers associated with changes in relative humidity near -20°C — approximately 2 fC per collision (Figure 4.11). Therefore, it is more likely that the negative charge transfer comes from a higher dislocation density on the riming target, caused by rapidly freezing droplets, so protons are being transferred to the ice crystal. In this case the dependence of the positive charge transfer on $V^{2.8}$ again suggests a dependence on the kinetic energy of the collision, now possibly enhanced by weak liquid-like layer charging as well.

Clearly further investigation into the effect of collision velocity is required. The above analysis suggests that the velocity dependence of the charge transfer is different when liquid-like layer charge transfer dominates compared to when dislocation charging dominates. An experimental test of this suggestion could be performed by measuring the velocity dependence of the charge transfer at low chamber relative humidities, at temperatures near -16°C where the liquid-like layer effect is strong. If the analysis is correct, a velocity dependence of approximately $V^{2.5}$ should be observed.

5.3 A Conceptual Model of Thunderstorm Electrification

The dependence of the charge transfer on relative humidity provides a consistent picture of the microphysics that drives the ice crystal-graupel collision charging and explains discrepancies between the results of previous investigations. It also provides insight into the electrification of a thunderstorm.

It has been shown that the effect of relative humidity is much greater than the effect of temperature or LWC. Therefore, to explain the charge structure of a storm regions of high, low and intermediate humidities in the thundercloud should be identified. However, data on the relative humidity in thunderstorms is not currently available. Therefore, in order to develop a model of thunderstorm electrification based on the current results, a conceptual model of the relative humidity in different regions of a thunderstorm is presented.

The region of the thundercloud most likely to have relative humidities near water saturation is between cloud base and the -4°C level, where rising air from below condenses into cloud droplets and where very few ice crystals are present. Relative humidities near ice saturation would be observed near cloud top where entrained air and high ice particle concentrations dry the cloud. Low relative humidities may also be observed near cloud base in the regions where downdrafts and/or precipitation shafts lower cold, dry air from above.

In most of the cloud, droplets and ice crystals co-exist for long periods of time, so the steady-state relative humidity of the air would depend on the relative number concentration of droplets and ice crystals. Ice crystal concentrations observed in

thunderstorms are of the order of 10 to 100 per litre. Therefore intermediate relative humidities are expected, between ice and water saturation. In general, the relative humidity would rise with lower ice crystal concentration, but lower concentrations also reduce the average number of ice crystal-graupel collisions.

Unlike the rest of the cloud, the regions within and surrounding the updraft may have very high relative humidities. The updraft carries warm, moist air from the cloud base to mid-altitudes very rapidly. As the rising air cools adiabatically, the same mass of vapor will increase the relative humidity of the air to near water saturation.

This simple model of the relative humidity in a thunderstorm together with the observations of this study can provide a conceptual model of the electrification of a thunderstorm. The model, shown schematically in Figure 5.4, may explain several observations of natural thunderstorms discussed in Section 2.1, which are not explained by the conceptual model of Jayaratne and Saunders (1984) that was presented in Section 2.3.2.

At the early stages of electrification the negative charge center is confined to a small region near the updraft-downdraft transition zone, at altitudes between the -12°C and -20°C levels (Dye *et al*, 1986, 1988). Also, strong electric fields are not measured until 5 mm graupel are observed in the cloud. When 5 mm graupel are observed the electric field grows rapidly, from 100 V m^{-1} to 8 kV m^{-1} in 8 minutes (Dye *et al*, 1986).

The importance of the updraft region to the electrification could be explained by the effect of the relative humidity on the charge transfer. Since the humidity near the

updraft may be near water saturation, ice crystals that rise in the updraft or that are found in regions surrounding the updraft would grow rapidly in the humid environment. This would 'prime' the ice crystals to provide strong negative charge transfers to colliding graupel. Figure 4.11 shows that the greatest sensitivity to the relative humidity occurs at temperatures between -13°C and -18°C , with the strongest negative charge transfers occurring near -15°C . This agrees well with the -12°C to -20°C altitude of the first observed negative charge center.

The role of the updraft as a vapor source also agrees with observations by Williams and Rutledge (1990). They found that the highest lightning rates observed during an Australian monsoon season corresponded to periods with the highest convective available potential energy (CAPE). Higher CAPE creates stronger updrafts that could raise more moist air from the cloud base, and this seems to be associated with stronger electrification.

The presence of 5 mm graupel in the cloud before the development of strong electric fields is consistent with the velocity dependence observed at relative humidities near water saturation, shown in Figure 4.12. The terminal velocity of 5 mm graupel is approximately 5 m s^{-1} at pressures corresponding to cloud altitudes (List and Schemenauer, 1971). Figure 4.12 shows that 5 m s^{-1} is a preferred velocity for strong negative charge transfer at -16°C , a temperature that corresponds to the altitude of the early negative charge center. Smaller graupel have lower fall speeds, and much lower maximum negative charge transfers are observed at lower speeds. Therefore, rapid early

electrification may only begin when 5 mm graupel are present in the high humidity region surrounding the updraft, because that is when the strongest charge transfers would occur.

In mature storms the presence of the negative charge center at altitudes between the -5°C and -20°C level can be explained as a static process, rather than the dynamic process described by Jayaratne and Saunders (1984). The temperature and ELWC series presented in Section 4.3.2 showed reversal temperatures between -10°C and -14°C for all ELWC's at intermediate relative humidities. This is typically much warmer than the reversal temperatures reported by the Manchester group and corresponds approximately to the temperature at the bottom of the negative charge center. This is consistent with the

New Thunderstorm Electrification Model

Figure 5.4 A new conceptual model of thunderstorm electrification. The main negative charge center develops 'in place' because observed reversal temperatures are near the bottom of the negative charge center. Very strong negative charging is observed around the updraft because rising warm moist air increases the local humidity. Positive charge centers develop at the bottom of updrafts or precipitation shafts where dry air from above negates the high humidity effect.

negative charge center being generated approximately 'in place', rather than by advecting negatively charged particles to the observed altitude as suggested in previous studies.

Finally, unlike the negative charge center, the lower positive charge region is not distributed evenly throughout the cloud but consists of small pockets of positive charge. This may also be explained by the relative humidity series presented in Section 4.3.3, which showed that relative humidities near water saturation weaken the positive charge transfer observed at temperatures warmer than the reversal temperature. If the relative humidity is high at low altitudes, pockets of strong positive charge may only develop in regions that have been dried by downdrafts and/or precipitation shafts. This could explain the local reversal of the electric field below the storm that is associated with the arrival of downdrafts and the corresponding rain gush (Krehbiel, 1986).

This conceptual model offers explanations for several important phenomena, but clearly the assumptions of the model must be verified. In particular, the conceptual model of the relative humidity in a thundercloud must be tested. Future field studies must attempt to measure the relative humidity in various regions of a cloud, and numerical models must include this data to accurately recreate the electrification of a thunderstorm.

Chapter Six

Summary and Conclusions

The current study has provided important insight into the fundamental questions of thunderstorm electrification, both at the scale of the cloud particles and the thunderstorm as a whole. In addition, the results of this study may offer an explanation for the differing observations of previous studies, which have been a long standing concern of the field.

The experiments of this work were performed using a newly developed facility for the study of precipitation particles. The Triple Interaction Facility is the first capable of providing independent control of the solid, liquid and vapor phases of a simulated cloud. The value of such control is demonstrated by the current study.

The experiments revealed several properties of the charge transfer that occurs between colliding ice particles.

- 1) The dependence of the charge transfer on the effective liquid water content of the cloud, rather than the actual liquid water content, was verified.
- 2) This study is the first to observe a dependence of the charge transfer on relative humidity. It was found that increasing the relative humidity at which the ice crystals are grown always favors stronger negative charging, while lowering the relative humidity promotes weaker negative or stronger positive charging. The sensitivity to the relative humidity was strongest at temperatures between -13°C and -18°C . Within this

temperature range the charge transfer averaged over the entire experiment could be varied from +5 fC to -25 fC per ice crystal collision by changing the humidity in the ice crystal chamber from ice to water saturation. This is larger than the range of charge transfers observed by varying the temperature and ELWC. At other temperatures the average charge transfer could be varied by 2 to 8 fC per collision. At humidities near water saturation, charge transfers observed within the first 30 seconds to 1 minute after ice crystals were initiated in the chamber were much stronger than the average over the whole experiment. Within this interval negative charge transfers greater than -60 fC per collision were observed near -15°C, compared to a maximum of -26 fC per collision for the experiment average. At -21°C charge transfers of -13 fC per collision were observed within this interval, compared to a maximum of -6.6 fC averaged over the entire experiment.

3) It was found that at intermediate relative humidity, between ice and water saturation, the sign reversal temperature for the charge transfer warms with increasing ELWC. For ELWC's of 0.4 to 0.8 g m⁻³ the reversal temperature was observed at -11°C. At lower ELWC the reversal temperature was between -11°C and -16°C, and between -9°C and -11°C at higher ELWC. However, it was shown that by varying the ice crystal chamber humidity from ice to water saturation the reversal temperature at a constant ELWC of 0.5 g m⁻³ could be shifted from -17°C to -10°C, respectively. Therefore, the reversal temperature can only be defined at both constant ELWC and constant relative humidity.

4) Finally, it was found that much stronger high relative humidity charging occurs at a velocity of 5 m s^{-1} than at lower and higher velocities. At velocities of 3, 4 and 6 m s^{-1} the maximum negative charge transfers observed were approximately -5 fC per ice crystal collision, at a temperature of -16°C . At 5 m s^{-1} the maximum transfer at -16°C increased to -16 fC per collision.

These observations were analyzed to develop a consistent model of the physical mechanisms that drive the charge transfer. The observed charge transfer could be explained as a competition between two charge transfer mechanisms: negative charge transfer between liquid-like layers on the surfaces of the colliding ice particles and positive charge transfer due to different dislocation densities. Theoretical calculations necessary to support this hypothesis were identified.

The dependence of the charge transfer on the relative humidity was offered as an explanation for the difference between the results of previous investigations. It was shown quantitatively that the range of charge transfers observed by varying the relative humidity could easily envelope the different charge transfers observed in other studies.

The velocity series was analyzed and two possible mechanisms were offered: a dependence on target rime density or a neutralizing current across the attachment point of liquid-like layers. Neither mechanism could be rejected based on the data available, but an experimental investigation that could distinguish between the two was suggested.

The velocity series was also compared to the results of previous studies that showed a significantly different dependence of the charge transfer on the velocity. The

different observations are consistent with the theory of two competing charge mechanisms if each has a different velocity dependence. An experimental test of this theory was suggested.

Finally, a model of thunderstorm electrification was offered, based on a conceptual model of the relative humidity in different regions of a thunderstorm. The relative humidity model provides consistent qualitative agreement between field observations and the current laboratory experiments. Variations in the relative humidity could explain several localized phenomena observed in thunderstorms, including the early electrification observed in the updraft region of the storm and the localization of the lower positive charge center.

The assumptions of this model await verification. The results of the laboratory experiments have been parameterized and are available for use in numerical simulations that can test this thunderstorm model.

Appendix A

The 2DCG Probe

The 2D series of optical array probes were introduced by Knollenberg (1976), and are built by Particle Measuring Systems of Boulder, Colorado. The instruments are designed to take shadow graph images of cloud and precipitation particles aboard research aircraft. The images provide particle size, shape and concentration data. Knollenberg 2D optical array probes have been used in cloud physics for over 20 years, and are considered standard instrumentation in field studies. As a result, many works have been published focusing on the limitations of these probes and techniques to improve the quality of the acquired data (Heymsfield and Parrish, 1979; Heymsfield and Baumgardner, 1985; Joe and List, 1987; Korolev *et al*, 1996).

A 2D Cloud particle Grey scale (2DCG) probe was used to determine the number concentrations and size distributions of wind tunnel cloud particles during experiments. The probe was especially designed for use in laboratory studies (Particle Measuring Systems, 1977). It has a resolution of 10 μm , which is significantly better than the 25 μm resolution typical of aircraft probes. The general operation of the probe, the method of data analysis, and an error analysis will be presented in this chapter.

A.1 Probe Operation

The 2DCG probe is composed of three parts: the optical system; the on-board electronics; and the probe computer. The probe optical system, shown schematically in Figure A.1, consists of a Helium-Neon laser operating at a wavelength of 632.8 nm, a

Figure A.1: Schematic diagram of 2DCG probe optical system. The dashed line indicates the focal plane of the probe optics, approximately 34 cm in front (to the left) of the first optic. Tunnel airflow is out of the page.

system of three focusing and magnifying lenses, and a 60 element photodiode array. The photodiodes are circular and have a center-to-center spacing of $200\ \mu\text{m}$. The probe magnifying lens provides a magnification of 20 times, which gives the photodiodes a total optical width of $600\ \mu\text{m}$ at a resolution of $10\ \mu\text{m}$. Two focusing lenses place the focal plane of the probe at approximately 34 cm in front of the first lens.

The output of the photodiodes is amplified and recorded by the probe's on-board electronics. The rate at which the photodiodes are sampled, known as the slice rate, is determined by the True Air Speed (TAS) clock. The TAS clock pulse is provided by a Wavetek Model 19 digital function generator that is set to provide a vertical resolution of $10\ \mu\text{m}$ at a given air speed. For a tunnel velocity of $5.3\ \text{m s}^{-1}$ the Wavetek was set to output a $530 \pm 0.5\ \text{kHz}$ TTL pulse.

At each clock pulse the photodiode array is sampled in parallel. Photodiode output is converted into a two-bit digital pixel that represents one of four levels of shadowing, corresponding to 0%, 25%, 50% and 75% shadowed. These shadow levels are known as

no shadow, MIN, MID and MAX, respectively (Particle Measuring Systems, 1977). The probe electronics begin recording the output of the photodiode array when an element of the array is shadowed, and continue to record until the entire array is unshadowed or until the probe memory is full (512 slices).

A single sample of the entire photodiode array is known as a slice. Probe images are made up of successive slices recorded from the first partially shadowed slice to the first unshadowed slice. As an image is recorded the probe electronics test selected image criteria to determine image length, maximum shadow level and if the end elements of the array are unshadowed. In the current experiment the selection criteria used were: 2 slices minimum image length; at least one pixel shadowed at the MAX level; and end elements unshadowed. An image was accepted only if it met these criteria.

If an image is accepted by the on-board electronics the recording of slices is halted and the probe computer is signaled. The signal line to the probe computer also controls the gating of an AMD Am9513 system timing controller (Advanced Micro Devices, 1990). When the signal line goes high it stops a clock on the Am9513 and the computer downloads the image from the probe. The computer restarts the probe by lowering the signal line, which also restarts the clock. In this way the clock measures the elapsed 'active' time of the probe, during which the probe is available to accept particle images. The clock has a resolution of 25 μ sec. With each probe image the computer records the elapsed active time, date, time of day, image size, and TAS setting.

The probe laser shines into the tunnel through a set of anti-reflection coated optical windows, 30 cm below the riming target (see Figure 3.4). The probe and the laser rest on a translational table that is mounted on a building wall rather than on the tunnel itself. This eliminates vibrations from the tunnel that could affect the quality of probe images. The translational table is positioned so that the focal plane of the probe is in the center of the measuring section, aligned approximately with the target.

A.2 Determination of the Probe Sample Volume

A consistent problem in the analysis of 2D probe data is defining the sample volume of the probe. The height of the volume is determined by multiplying the True Air Speed by the elapsed probe active time, and the width is determined by the number of elements and the resolution of the optical array. However, the depth of the volume is less well defined. The sharpness of focus, and therefore the level of shadowing, for a particle a given distance away from the focal plane is a function of the size of the object. At sufficiently large distances smaller objects will not shadow the array enough to meet the probe selection criteria. Furthermore, as the distance from the focal plane increases the particle image degrades and particles are incorrectly sized.

Joe and List (1987) showed that the distance from the focal plane of an object could be related to the level of shadow in the grey scale image of the object. They defined the RMAX ratio as the number of image pixels shadowed at the MAX level divided by the total pixels shadowed, and showed that the distance of an object of given size from the focal plane could be calibrated against the RMAX ratio of its image. Once

calibrated, this ratio could be used to reject images that correspond to objects outside of a specified sample depth.

The RMAX criteria was used in the current study to define the sample depth of the 2DCG. The distance of objects from the focal plane was calibrated against the RMAX ratio in the manner described by Joe and List (1987). Small glass beads of known size were dropped through the probe laser beam at various distances from the focal plane and the RMAX ratio of the images was determined. The beads were manufactured by Precision Glass Beads and had sizes of $42 \pm 5 \mu\text{m}$, $125 \pm 15 \mu\text{m}$, $240 \pm 15 \mu\text{m}$ and $400 \pm 15 \mu\text{m}$. The beads were dropped through a hypodermic needle mounted on a vernier translational mount. The translational mount could be positioned to $\pm 0.005 \text{ mm}$. The

Figure A.2 The calibration of the RMAX ratio and the image diameter for $125 \pm 15 \mu\text{m}$ diameter glass beads. The value of the vernier scale reading is arbitrary – only differences were used. The object plane is at the point of maximum RMAX ratio. The RMAX threshold for this size particle was chosen as 0.37, which corresponds to a sample depth of 9.1 cm. The threshold is indicated by the solid vertical lines.

diameter of the hypodermic needle was 0.23 mm for the 42 μm beads and 0.68 mm for the three larger sizes. Figure A.2 shows the RMAX and size calibration data for the 125 μm beads.

Probe images were analyzed using custom analysis software. The software identified objects in the particle image, determined the maximum dimension of the image, applied the RMAX criteria and classified the images as 'round' or 'not round' (see Section 4.2). Accepted images observed in a 10 second interval were counted as a function of size. The 10 second sampled volume was calculated for each size bin by multiplying the elapsed active time, the TAS, the sample depth for the particle size and the effective⁶ width of the optical array.

The 10 second average estimate of the concentration for each size bin was determined by dividing the number of images observed by the volume sampled for that size bin during the interval. The total number concentration was calculated as the sum of the individual size bins. Errors for each size bin estimate and for the total number concentration were determined using standard error analysis techniques, by assuming that the interarrival time of the cloud particles obeys a Poisson distribution (see below).

A.3 Probability Theory of Probe Data Analysis

To determine the cloud particle number concentration and size spectra from the raw 2DCG probe data the 'dead' time of the probe must be considered. The dead time is

⁶ Images were rejected by the probe on-board electronics if either element at the end of the optical array was shadowed. Therefore, the effective width of the optical array is less than the actual width by twice the object radius, since one radius is the minimum distance that its center can be from the end of the array for the object image to be accepted.

the time that the probe is busy processing and downloading the current particle image. Particles that arrive at the probe during the dead time will not be registered and the estimate of the concentration will be systematically lower.

There are two formally correct methods of analysis that treat the probe dead time, but these methods often lead to significantly different estimates of the number concentration. Both methods assume that the distribution of the droplets in the cloud is Poisson, an assumption based on the spatial distribution argument (Parratt, 1961). The first method is to count the number of particles of each size bin observed during a fixed time interval. The mean arrival rate of the particles is estimated by dividing the total number of particles observed by the total observation time. In this method, corrections must be made to the estimate of the number concentration to account for the dead time. The second method (Joe and List, 1987) assumes that the probe is restarted after every image arrival, as if no other images had been observed. It can be shown (the Waiting Time Paradox; Feller, 1968, 1971) that for a Poisson process the distribution of waiting times is the same as the distribution of particle interarrival times. Therefore the average of the waiting times provides an unbiased estimate of the particle interarrival times, and the inverse of the average interarrival time is the estimate of the interarrival rate.

When all particles are considered identical and no particles are rejected, these two methods are simply reciprocals of one another and the predicted number concentrations are identical. However, when particles are rejected because they do not meet the RMAX rejection criteria or when accepted particles are divided into different size bins, the average waiting time method leads to significantly higher concentrations of all particle

sizes than predicted by the counting method. Since both methods provide mathematically correct estimates of the particle concentration and both methods are currently used by different research groups, a mathematical analysis was performed of both methods to determine which is more precise.

A.3.1 The Average Waiting Time Method

The distribution of interarrival times for a Poisson process is described by,

$$G(t) = P\{T > t\} = e^{-\lambda t} \quad (\text{A.1})$$

where the distribution function $G(t)$ gives the probability that the epoch⁷ of the particle arrival, T , will be greater than t (Feller, 1971). If a particle arrival has not occurred before an epoch t , then the conditional probability that it will arrive a later epoch $t_1 = t + t_0$ is given by the distribution function,

$$H(t) = \frac{P\{T > t_1\}}{P\{T > t\}} = \frac{e^{-\lambda(t+t_0)}}{e^{-\lambda t}} = e^{-\lambda t_0} \quad (\text{A.2})$$

which is the same as $G(t_0)$. This describes mathematically a general property of exponential distributions, like the Poisson distribution, that the underlying process has no ‘memory’ of past events. It is this property that explains the classical Waiting Time Paradox described by Feller (1971).

The average waiting time method exploits the lack of memory of the particle arrival process to determine the number concentration from the probe data. According to Equation A.2 the waiting time between the probe restart and the next particle arrival must

⁷ In probability theory points on the time axis are called epochs, to distinguish them from interarrival times which are intervals on the time axis.

have the same distribution as the particle interarrival time, so they must also have the same expected value. For a Poisson distribution the mean is the maximum likelihood estimator of the expected value (Parratt, 1961), so the mean of the waiting times gives an unbiased estimate of the particle interarrival time.

A.3.2 The Counting Method

In the counting method the number of particles observed in a time interval is divided by the total observation time to estimate the interarrival rate of the particles. However, during the probe dead time particles may arrive which are not counted, leading to an underestimate of the interarrival rate. The estimated rate must therefore be adjusted to account for the lost particles. The average waiting time method requires no such adjustment since the waiting time of each particle is measured without dead time. Therefore, a proper error analysis of the counting method requires an analysis of the correction for particles lost.

After Feller (1971), we consider a delayed terminating renewal process. The renewal process is the arrival of images to the probe. The process has a mean renewal (interarrival) rate of λ . Particles of size D arrive in the probe sample volume at a mean rate of $\alpha \neq \lambda$. A particle is lost if it arrives within a time τ of the arrival of an image. The renewal process terminates when a particle is lost.

The distribution of image interarrival times is,

$$F(t) = \begin{cases} 1 - e^{-\lambda(t-\tau)}, & t > \tau \\ 0, & t \leq \tau \end{cases} \quad (\text{A.3})$$

The probability that a particle of size D arrives within a time τ after the image is

$q = 1 - e^{-\alpha\tau}$, independent of the time t in Equation A.3. Therefore, the defective distribution of the terminating renewal process is,

$$\begin{aligned} L(t) &= (1 - q)F(t) \\ &= e^{-\alpha\tau} (1 - e^{-\lambda(t-\tau)}) . \\ L(\infty) &= L_{\infty} = e^{-\alpha\tau} \end{aligned} \quad (A.4)$$

If M is the epoch of termination then $Z(t) = P\{M \leq t\}$ satisfies the renewal equation,

$$Z(t) = 1 - L_{\infty} + \int_0^t Z(t-y)L\{dy\} . \quad (A.5)$$

The expected value of $Z(t)$ is given by,

$$E_z = \frac{E_L}{1 - L_{\infty}} = \frac{1}{1 - e^{-\alpha\tau}} e^{-\alpha\tau} \left(\frac{1}{\lambda} + \tau \right) = \frac{1 + \lambda\tau}{\lambda(e^{\alpha\tau} - 1)} . \quad (A.6)$$

This is the expected lifetime of the terminating renewal process, that is, the average time between lost counts. The rate of particle count loss is the inverse.

The expected number of observed particles, size D, arrivals is equal to the expected arrivals minus the expected lost arrivals, divided by the time interval to give a rate.

$$\begin{aligned} \alpha' &= \alpha - \frac{\lambda}{(1 + \lambda\tau)} (e^{\alpha\tau} - 1), \quad \alpha' \equiv \text{observed rate} \\ &\equiv \alpha - \frac{\lambda}{(1 + \lambda\tau)} (\alpha\tau + \frac{1}{2}(\alpha\tau)^2 + \dots) \end{aligned} \quad (A.7)$$

For high particle arrival rates or long dead times higher order terms must be considered, but this is not necessary for the conditions typically observed in the wind tunnel.

Therefore, by assuming that $1/\lambda \gg \tau$, i.e. that the average interarrival time is much longer than the dead time, and taking only the first order terms in $\alpha\tau$, Equation A.5 can be easily solved for α :

$$\begin{aligned}
 \alpha &= \frac{N}{t} = \frac{\alpha'}{(1 - \lambda\tau)}, & N &\equiv \text{particles arriving} \\
 & & &\text{in time } t \\
 &= \frac{N'/t}{(1 - \lambda\tau)}, & N' &\equiv \text{particles observed} \\
 & & &\text{in time } t \\
 &= \frac{N'}{t - t\lambda\tau}, & t\lambda &\equiv \text{expected number of} \\
 & & &\text{images in time } t \\
 &= \frac{N'}{t'}, & t\lambda\tau &\equiv \text{total dead time in time } t
 \end{aligned} \tag{A.8}$$

where t' is the total time t minus the total dead time, that is, the total probe active time.

Note that when $\alpha' = \lambda$ the first equation in A.6 is the accepted correction for Geiger counter dead times (Parratt, 1961).

A.3.3 Comparative Error Analysis

Both methods of analysis give formally correct estimates of the particle interarrival times. To determine which method is more precise the error associated with each method was analyzed. In the following analysis it is assumed that all times are measured without error.

In the average waiting time method the time from the probe 'restart' until the arrival of a particle is averaged to give an estimate of the particle interarrival time. If N

particles of size D are observed in an elapsed probe active time T , then the estimate of the particle rate is given by the inverse of the average, $\bar{t}_a$, of the N waiting times, t_n .

$$\begin{aligned}
 \bar{t}_a &= \frac{\sum_{n=0}^N t_n}{N} \pm \frac{\bar{t}_a}{\sqrt{N}} \\
 &= \frac{T_a}{N} \pm \frac{\bar{t}_a}{\sqrt{N}} \\
 \alpha_a &= \frac{1}{\bar{t}_a} \pm \frac{1}{\sqrt{N} \bar{t}_a} \\
 &= \frac{1}{\bar{t}_a} \pm \frac{\sqrt{N}}{T_a}
 \end{aligned} \tag{A.9}$$

where T_a is the total time spent ‘waiting’ for particles of size D . The error in the average interarrival time is the standard error of the mean associated with a Poisson interval (Parratt, 1961) and the error in the arrival rate is calculated using standard error propagation.

To estimate the arrival rate using the counting method the number of particles observed, N , is divided by total probe active elapsed time, T ,

$$\bar{\alpha}_c = \frac{N}{T} \pm \frac{\sqrt{N}}{T} \tag{A.10}$$

where $\sqrt{N}$ is the standard deviation associated with a count of N in a Poisson process (Parratt, 1961).

In both analysis methods the error is inversely proportional to the total observing time. However, in the average interarrival time method the total observing time, T_a , is the time spent observing particles of size D only. If all the observed particles are size D and all observations are accepted then $T_a \equiv T$ and the methods are equivalent. If particles are

classified into size bins and recorded particle images are rejected based on selection criteria then T_a is always smaller than T and the error associated with the average interarrival time method is always larger.

Empirically, approximately 75% of particle images recorded by the 2DCG are rejected based on the RMAX selection criteria. Furthermore, the range of cloud particle sizes observed in the wind tunnel spans 12 size bins. Based only on the ratio of accepted to rejected images, concentrations calculated using the average interarrival time method would have associated errors approximately 3 times those of the counting method. Therefore, the counting method was used in the current study to determine the cloud particle concentration and size spectra.

Appendix B

Summary of Experimental Data

The following tables list the relevant data from the experimental series described in Chapter 4. Reported errors indicate one standard deviation from the mean. The errors quoted for the ice crystal concentrations indicate the standard deviation of the concentrations used, rather than the error of the concentration estimates. The number of points indicate the number of points used in the weighted average that produced the estimate of the charge transfer per collision. The mean droplet diameter

-5°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.16 ± 0.00	19.2	0.47 ± 0.01	-5.02 ± 0.23	5.34 ± 0.06	-8	58 ± 24	7	7.99 ± 2.16
0.19 ± 0.00	20.8	0.49 ± 0.00	-5.01 ± 0.13	5.32 ± 0.04	-7	72 ± 28	8	1.70 ± 1.17
0.25 ± 0.01	24.8	0.49 ± 0.00	-5.08 ± 0.09	5.31 ± 0.04	-8	160 ± 78	3	2.60 ± 1.28
0.34 ± 0.00	19.2	0.98 ± 0.01	-5.04 ± 0.17	5.31 ± 0.05	-8	69 ± 21	4	5.86 ± 2.74
0.38 ± 0.00	20.8	0.98 ± 0.01	-5.01 ± 0.11	5.28 ± 0.05	-7	89 ± 26	3	10.38 ± 5.06
0.38 ± 0.00	20.8	0.98 ± 0.01	-5.29 ± 0.16	5.31 ± 0.05	-7	112 ± 33	7	2.79 ± 1.03
0.50 ± 0.01	19.2	1.46 ± 0.02	-4.97 ± 0.04	5.31 ± 0.07	-7.5	114 ± 42	6	4.87 ± 1.82
0.50 ± 0.02	24.8	0.99 ± 0.01	-4.90 ± 0.15	5.28 ± 0.03	-8	169 ± 63	9	2.60 ± 0.63
0.50 ± 0.02	24.8	0.99 ± 0.01	-5.17 ± 0.12	5.28 ± 0.04	-9.5	226 ± 71	8	10.78 ± 2.32
0.51 ± 0.01	20.8	1.32 ± 0.01	-5.33 ± 0.05	5.33 ± 0.05	-8	113 ± 30	4	3.18 ± 1.34

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.52 ± 0.01	20.8	1.33 ± 0.01	-4.91 ± 0.05	5.27 ± 0.04	-7	119 ± 30	4	5.75 ± 2.60
0.65 ± 0.01	20.8	1.68 ± 0.02	-5.11 ± 0.08	5.32 ± 0.05	-7	144 ± 50	7	6.45 ± 1.54
0.67 ± 0.01	19.2	1.96 ± 0.03	-5.15 ± 0.14	5.30 ± 0.07	-8.5	96 ± 7	3	10.43 ± 3.94
0.74 ± 0.03	24.8	1.47 ± 0.01	-4.72 ± 0.24	5.30 ± 0.03	-8	288 ± 92	11	3.98 ± 0.68
0.76 ± 0.01	20.8	1.96 ± 0.02	-5.01 ± 0.12	5.29 ± 0.05	-8	171 ± 59	4	2.88 ± 1.29
0.75 ± 0.03	24.8	1.47 ± 0.01	-4.99 ± 0.09	5.29 ± 0.04	-8	210 ± 65	5	8.64 ± 2.98
0.99 ± 0.04	24.8	1.96 ± 0.01	-5.06 ± 0.14	5.29 ± 0.04	-8	270 ± 136	6	2.44 ± 0.63

-8°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.19 ± 0.00	20.8	0.49 ± 0.00	-8.44 ± 0.08	5.33 ± 0.05	-11	74 ± 22	4	1.14 ± 1.03
0.38 ± 0.00	20.8	0.98 ± 0.01	-7.91 ± 0.06	5.33 ± 0.05	-10	51 ± 22	6	5.12 ± 3.88
0.49 ± 0.01	19.2	1.44 ± 0.02	-7.97 ± 0.06	5.26 ± 0.06	-10	64 ± 9	4	9.79 ± 4.52
0.50 ± 0.02	24.8	0.99 ± 0.01	-8.11 ± 0.07	5.27 ± 0.03	-11	189 ± 99	7	6.86 ± 1.51
0.73 ± 0.03	24.8	1.45 ± 0.01	-8.04 ± 0.08	5.37 ± 0.04	-10.5	135 ± 55	7	2.50 ± 1.10
0.74 ± 0.03	24.8	1.46 ± 0.01	-7.81 ± 0.06	5.32 ± 0.03	-10	223 ± 110	9	2.87 ± 0.57
0.97 ± 0.03	24.8	1.92 ± 0.01	-7.77 ± 0.05	5.40 ± 0.03	-10	166 ± 90	3	2.57 ± 1.35

-11°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.10 ± 0.00	20.8	0.25 ± 0.00	-11.38 ± 0.04	5.30 ± 0.04	-12	36 ± 27	5	3.10 ± 2.37
0.17 ± 0.00	19.2	0.49 ± 0.00	-11.23 ± 0.05	5.29 ± 0.04	-12	34 ± 31	4	2.34 ± 3.71

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.19 ± 0.00	20.8	0.49 ± 0.01	-11.35 ± 0.04	5.26 ± 0.04	-12	62 ± 44	11	-2.72 ± 1.43
0.25 ± 0.01	24.8	0.50 ± 0.00	-11.49 ± 0.05	5.37 ± 0.03	-10	126 ± 63	2	1.89 ± 0.78
0.34 ± 0.00	19.2	0.99 ± 0.01	-11.25 ± 0.05	5.28 ± 0.03	-12	98 ± 40	2	-1.06 ± 1.96
0.35 ± 0.01	24.8	0.68 ± 0.00	-11.11 ± 0.06	5.31 ± 0.04	-11.5	124 ± 72	17	1.40 ± 6.07
0.35 ± 0.01	24.8	0.68 ± 0.00	-11.16 ± 0.07	5.26 ± 0.03	-12.5	151 ± 76	3	2.27 ± 0.78
0.35 ± 0.01	24.8	0.68 ± 0.00	-11.65 ± 0.12	5.25 ± 0.04	-10	144 ± 72	9	1.59 ± 0.51
0.35 ± 0.01	24.8	0.68 ± 0.00	-12.86 ± 0.09	5.32 ± 0.03	-12.5	185 ± 72	14	1.05 ± 0.30
0.36 ± 0.01	24.8	0.71 ± 0.00	-11.22 ± 0.07	5.21 ± 0.04	-10	156 ± 89	5	5.28 ± 0.27
0.39 ± 0.00	20.8	1.00 ± 0.01	-10.44 ± 0.07	5.22 ± 0.04	-11	83 ± 53	2	-0.11 ± 3.91
0.39 ± 0.00	20.8	1.00 ± 0.01	-10.96 ± 0.10	5.23 ± 0.05	-11.5	88 ± 53	6	1.02 ± 1.78
0.47 ± 0.00	20.8	1.20 ± 0.01	-10.49 ± 0.09	5.25 ± 0.04	-12	68 ± 55	2	-6.63 ± 8.68
0.51 ± 0.00	19.2	1.47 ± 0.01	-11.54 ± 0.04	5.28 ± 0.04	-11	34 ± 42	2	-8.70 ± 11.34
0.50 ± 0.02	24.8	0.99 ± 0.00	-11.46 ± 0.08	5.28 ± 0.03	-12.5	164 ± 72	8	-0.16 ± 0.21
0.50 ± 0.02	24.8	0.98 ± 0.00	-11.83 ± 0.05	5.32 ± 0.04	-11.5	172 ± 67	8	0.03 ± 0.25
0.57 ± 0.01	20.8	1.47 ± 0.02	-11.22 ± 0.10	5.28 ± 0.05	-12	99 ± 64	3	-0.54 ± 2.23
0.57 ± 0.01	20.8	1.48 ± 0.01	-11.43 ± 0.09	5.26 ± 0.04	-11	106 ± 61	10	3.06 ± 1.83
0.68 ± 0.00	19.2	1.97 ± 0.01	-11.23 ± 0.06	5.26 ± 0.03	-11	138 ± 58	2	-2.49 ± 2.44
0.74 ± 0.03	24.8	1.47 ± 0.00	-12.91 ± 0.04	5.29 ± 0.04	-12	180 ± 108	5	-0.65 ± 0.42
0.77 ± 0.01	20.8	1.97 ± 0.01	-11.03 ± 0.05	5.26 ± 0.03	-12	97 ± 70	4	0.29 ± 2.82
0.99 ± 0.04	24.8	1.96 ± 0.00	-11.62 ± 0.08	5.28 ± 0.03	-11.5	222 ± 115	3	-2.67 ± 1.55

-14°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.25 ± 0.01	24.8	0.49 ± 0.00	-14.97 ± 0.10	5.31 ± 0.04	-15	114 ± 34	6	0.52 ± 0.31
0.34 ± 0.00	19.2	0.99 ± 0.01	-14.66 ± 0.09	5.30 ± 0.05	-15	98 ± 34	6	-0.15 ± 1.29
0.35 ± 0.01	24.8	0.70 ± 0.01	-14.63 ± 0.07	5.32 ± 0.04	-15	181 ± 70	3	1.91 ± 0.68
0.51 ± 0.01	19.2	1.47 ± 0.02	-13.72 ± 0.09	5.28 ± 0.07	-15	97 ± 43	8	6.01 ± 1.77
0.50 ± 0.02	24.8	0.98 ± 0.01	-13.98 ± 0.10	5.32 ± 0.03	-13.5	211 ± 56	5	-3.07 ± 0.80
0.51 ± 0.01	20.8	1.31 ± 0.01	-14.19 ± 0.05	5.28 ± 0.04	-16	85 ± 13	5	-3.75 ± 1.56
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.23 ± 0.05	-16	146 ± 70	12	-2.24 ± 0.34 *
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.23 ± 0.05	-16	80 ± 20	3	0.93 ± 0.71 *
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.23 ± 0.05	-16	94 ± 26	3	6.19 ± 2.29 *
0.51 ± 0.01	19.2	1.47 ± 0.02	-14.32 ± 0.05	5.29 ± 0.06	-15	103 ± 39	11	2.76 ± 1.27
0.50 ± 0.02	24.8	0.98 ± 0.01	-14.66 ± 0.09	5.32 ± 0.03	-14	147 ± 39	4	-1.96 ± 0.96
0.52 ± 0.01	20.8	1.34 ± 0.01	-14.08 ± 0.04	5.25 ± 0.04	-17	87 ± 39	3	0.57 ± 0.61 *
0.52 ± 0.01	20.8	1.34 ± 0.01	-14.08 ± 0.04	5.25 ± 0.04	-17	87 ± 30	3	4.48 ± 1.76 *
0.52 ± 0.01	20.8	1.33 ± 0.01	-14.33 ± 0.05	5.28 ± 0.05	-16	96 ± 21	5	4.38 ± 2.25
0.65 ± 0.01	19.2	1.88 ± 0.02	-14.65 ± 0.11	5.25 ± 0.04	-9	78 ± 12	2	-13.58 ± 10.70
0.67 ± 0.01	19.2	1.95 ± 0.03	-13.86 ± 0.05	5.31 ± 0.09	-14	121 ± 47	6	-1.17 ± 1.52
0.74 ± 0.03	24.8	1.47 ± 0.01	-14.17 ± 0.05	5.30 ± 0.04	-15	266 ± 119	12	-0.96 ± 0.21

* Indicates multiple data points taken from single experiments in which the charge transfer is observed to change sign during the ice phase. See Section 4.3.3 and Figure 4.10.

-16°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.10 ± 0.00	20.8	0.25 ± 0.00	-16.34 ± 0.06	5.22 ± 0.06	-10	40 ± 7	5	0.05 ± 1.18
0.13 ± 0.03	24.8	0.25 ± 0.05	-15.92 ± 0.56	5.24 ± 0.08	-10	134 ± 30	3	-0.66 ± 0.19
0.17 ± 0.00	19.2	0.49 ± 0.00	-15.70 ± 0.07	5.26 ± 0.04	-12	47 ± 18	6	-2.86 ± 1.86
0.17 ± 0.00	19.2	0.49 ± 0.00	-16.14 ± 0.07	5.26 ± 0.03	-10	53 ± 16	2	-0.23 ± 3.05
0.19 ± 0.00	20.8	0.49 ± 0.00	-15.39 ± 0.05	5.26 ± 0.04	-13	76 ± 19	7	-3.67 ± 1.63
0.24 ± 0.00	19.2	0.69 ± 0.01	-15.81 ± 0.06	5.24 ± 0.04	-11	52 ± 3	2	-3.12 ± 7.00
0.25 ± 0.01	24.8	0.50 ± 0.00	-16.46 ± 0.09	5.22 ± 0.03	-12	116 ± 35	7	-3.37 ± 0.75
0.32 ± 0.01	19.2	0.93 ± 0.04	-15.45 ± 0.07	5.40 ± 0.24	-8	60 ± 17	5	-18.06 ± 8.32
0.33 ± 0.00	19.2	0.97 ± 0.01	-16.12 ± 0.14	5.22 ± 0.04	-12	58 ± 7	6	-11.87 ± 7.94
0.33 ± 0.00	19.2	0.96 ± 0.01	-16.25 ± 0.06	5.23 ± 0.05	-16	68 ± 22	8	-8.29 ± 1.93
0.33 ± 0.01	19.2	0.96 ± 0.03	-15.60 ± 0.05	5.25 ± 0.15	-12	63 ± 15	10	-27.78 ± 6.10
0.34 ± 0.00	19.2	1.00 ± 0.01	-15.25 ± 0.06	5.04 ± 0.03	-5	64 ± 29	7	-27.88 ± 7.06
0.35 ± 0.01	24.8	0.69 ± 0.00	-16.03 ± 0.07	5.26 ± 0.03	-11	105 ± 22	5	-5.44 ± 2.37
0.35 ± 0.01	19.2	1.01 ± 0.02	-16.22 ± 0.04	5.28 ± 0.05	-16	96 ± 41	13	-3.16 ± 0.51
0.37 ± 0.00	20.8	0.97 ± 0.01	-15.73 ± 0.05	5.22 ± 0.06	-10	90 ± 9	2	-4.18 ± 3.06
0.38 ± 0.00	20.8	0.98 ± 0.01	-15.31 ± 0.09	5.22 ± 0.06	-13.5	103 ± 24	6	-4.49 ± 1.85
0.38 ± 0.00	20.8	0.98 ± 0.01	-15.90 ± 0.12	5.22 ± 0.06	-12	89 ± 8	6	-5.97 ± 2.93
0.42 ± 0.01	24.8	0.84 ± 0.01	-15.63 ± 0.08	5.26 ± 0.04	-13	79 ± 3	2	-5.03 ± 4.40
0.48 ± 0.02	24.8	0.95 ± 0.01	-16.15 ± 0.07	5.28 ± 0.03	-12.5	148 ± 27	7	-17.59 ± 4.91
0.50 ± 0.00	23.0	1.00 ± 0.01	-16.66 ± 0.06	5.21 ± 0.04	-10	124 ± 62	2	-9.45 ± 5.46
0.49 ± 0.01	23.0	0.98 ± 0.01	-16.64 ± 0.10	5.24 ± 0.06	-13	157 ± 55	6	-6.90 ± 1.68

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.51 ± 0.00	19.2	1.48 ± 0.01	-15.59 ± 0.04	5.24 ± 0.04	-12	47 ± 23	2	-27.60 ± 21.00
0.50 ± 0.01	19.2	1.45 ± 0.02	-16.09 ± 0.07	5.35 ± 0.07	-13	70 ± 23	7	-11.97 ± 3.53
0.49 ± 0.02	24.8	0.97 ± 0.01	-16.15 ± 0.07	5.20 ± 0.03	-12	158 ± 56	2	-15.10 ± 6.03
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.12 ± 0.09	5.29 ± 0.04	-12	132 ± 25	6	-5.94 ± 1.95
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.29 ± 0.04	5.28 ± 0.03	-12	47 ± 33	2	-9.29 ± 14.61
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.26 ± 0.14	5.27 ± 0.04	-14	265 ± 94	3	-1.77 ± 0.66
0.52 ± 0.02	24.8	1.03 ± 0.01	-16.29 ± 0.08	5.34 ± 0.04	-14.5	184 ± 68	7	-3.29 ± 0.75
0.55 ± 0.00	20.8	1.41 ± 0.01	-15.30 ± 0.04	5.35 ± 0.04	-16	129 ± 39	11	-0.86 ± 0.71
0.54 ± 0.01	20.8	1.38 ± 0.02	-15.57 ± 0.06	5.25 ± 0.05	-10	55 ± 21	2	-47.73 ± 42.69
0.55 ± 0.00	20.8	1.41 ± 0.01	-15.84 ± 0.05	5.27 ± 0.04	-12	126 ± 43	10	-2.75 ± 0.85
0.55 ± 0.01	20.8	1.43 ± 0.02	-15.52 ± 0.06	5.45 ± 0.05	-11.5	89 ± 21	10	-26.37 ± 5.76
0.55 ± 0.01	20.8	1.42 ± 0.01	-15.73 ± 0.12	5.34 ± 0.05	-16	74 ± 12	3	-5.19 ± 3.65
0.55 ± 0.01	20.8	1.42 ± 0.02	-16.03 ± 0.07	5.32 ± 0.05	-16	59 ± 10	2	-14.08 ± 11.41
0.57 ± 0.00	20.8	1.48 ± 0.01	-15.81 ± 0.06	5.27 ± 0.04	-10	58 ± 10	2	-47.44 ± 35.64
0.57 ± 0.01	20.8	1.47 ± 0.01	-15.75 ± 0.05	5.30 ± 0.05	-9.5	82 ± 14	2	-26.51 ± 15.04
0.59 ± 0.01	24.6	0.97 ± 0.01	-15.30 ± 0.05	5.21 ± 0.03	-11	104 ± 13	5	-13.67 ± 4.80
0.59 ± 0.01	24.6	0.97 ± 0.01	-15.38 ± 0.04	5.19 ± 0.04	-12	131 ± 58	6	-5.77 ± 1.34
0.59 ± 0.01	24.6	0.96 ± 0.01	-16.12 ± 0.09	5.24 ± 0.05	-12	100 ± 21	5	-7.56 ± 3.20
0.59 ± 0.01	24.6	0.97 ± 0.01	-16.15 ± 0.06	5.21 ± 0.03	-9	189 ± 130	7	-4.30 ± 0.82
0.60 ± 0.01	24.6	0.99 ± 0.01	-15.11 ± 0.05	5.10 ± 0.03	-14.5	133 ± 35	8	-3.22 ± 0.81
0.60 ± 0.02	24.8	1.18 ± 0.01	-16.14 ± 0.03	5.33 ± 0.04	-11	211 ± 65	7	-1.96 ± 0.50
0.61 ± 0.01	24.6	1.00 ± 0.01	-17.29 ± 0.06	5.20 ± 0.04	-13	108 ± 26	8	-8.70 ± 2.34

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.66 ± 0.01	19.2	1.93 ± 0.02	-15.78 ± 0.04	5.37 ± 0.05	-14	92 ± 25	10	-2.19 ± 1.90
0.70 ± 0.03	24.8	1.37 ± 0.04	-15.83 ± 0.06	5.12 ± 0.14	-11	203 ± 51	11	-7.52 ± 1.52
0.74 ± 0.01	20.8	1.92 ± 0.01	-16.06 ± 0.03	5.31 ± 0.03	-14	260 ± 88	1	-15.59 ± 5.82
0.75 ± 0.03	24.8	1.48 ± 0.01	-16.25 ± 0.21	5.24 ± 0.03	-10	178 ± 54	4	-4.74 ± 1.85
0.99 ± 0.04	24.8	1.96 ± 0.02	-16.28 ± 0.04	5.29 ± 0.03	-16	146 ± 28	6	-12.67 ± 5.13
1.00 ± 0.04	24.8	1.98 ± 0.02	-15.70 ± 0.07	5.24 ± 0.04	-14	174 ± 27	4	-11.95 ± 4.40

-18°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (mm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.17 ± 0.00	19.2	0.49 ± 0.00	-17.89 ± 0.03	5.27 ± 0.05	-18	65 ± 16	7	-4.45 ± 1.22
0.17 ± 0.01	24.8	0.33 ± 0.00	-18.40 ± 0.03	5.33 ± 0.03	-16.5	116 ± 44	10	-0.56 ± 0.19
0.34 ± 0.00	19.2	0.99 ± 0.01	-18.25 ± 0.06	5.26 ± 0.05	-15	129 ± 45	4	-2.03 ± 1.17
0.37 ± 0.03	24.8	0.70 ± 0.01	-18.33 ± 0.11	5.34 ± 0.04	-14	155 ± 66	10	-3.34 ± 0.56
0.52 ± 0.04	24.8	0.98 ± 0.01	-18.23 ± 0.03	5.32 ± 0.03	-16.5	131 ± 46	4	-2.89 ± 0.90
0.75 ± 0.01	20.8	1.94 ± 0.02	-18.22 ± 0.04	5.28 ± 0.04	-17	144 ± 38	3	-4.10 ± 2.33
1.02 ± 0.08	24.8	1.94 ± 0.01	-18.26 ± 0.06	5.30 ± 0.03	-17	192 ± 47	3	-3.32 ± 1.56

-21°C Series

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.09 ± 0.00	19.2	0.25 ± 0.00	-20.77 ± 0.04	5.26 ± 0.03	-17	33 ± 7	5	0.16 ± 0.63
0.12 ± 0.00	24.8	0.25 ± 0.00	-21.12 ± 0.03	5.31 ± 0.03	-16.5	72 ± 15	8	-0.85 ± 0.28
0.13 ± 0.00	19.2	0.37 ± 0.00	-20.47 ± 0.05	5.31 ± 0.03	-17	61 ± 20	3	1.41 ± 0.97

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.16 ± 0.00	19.2	0.48 ± 0.00	-21.01 ± 0.04	5.31 ± 0.04	-16	74 ± 26	14	-5.98 ± 0.91
0.16 ± 0.00	19.2	0.48 ± 0.00	-21.39 ± 0.03	5.30 ± 0.05	-17	50 ± 14	4	-11.72 ± 4.36
0.17 ± 0.00	19.2	0.49 ± 0.00	-20.66 ± 0.04	5.31 ± 0.03	-15.5	45 ± 1	2	-8.98 ± 5.71
0.17 ± 0.01	24.8	0.33 ± 0.00	-21.17 ± 0.03	5.37 ± 0.03	-18.5	72 ± 21	6	-0.32 ± 0.29
0.19 ± 0.00	20.8	0.49 ± 0.00	-20.62 ± 0.05	5.33 ± 0.03	-17	70 ± 24	6	-0.35 ± 0.96
0.19 ± 0.00	20.8	0.48 ± 0.01	-21.33 ± 0.04	5.31 ± 0.04	-17.5	58 ± 28	2	-3.29 ± 2.73
0.24 ± 0.00	19.2	0.68 ± 0.01	-21.24 ± 0.06	5.31 ± 0.04	-18	66 ± 25	11	-1.65 ± 0.87
0.25 ± 0.01	24.8	0.49 ± 0.00	-20.93 ± 0.19	5.27 ± 0.04	-19	148 ± 95	9	-0.92 ± 0.57
0.27 ± 0.00	20.8	0.68 ± 0.01	-21.21 ± 0.05	5.31 ± 0.04	-16.5	93 ± 23	5	-1.49 ± 1.12
0.34 ± 0.00	19.2	1.00 ± 0.01	-19.95 ± 0.04	5.25 ± 0.04	-16	111 ± 41	12	-1.19 ± 1.03
0.39 ± 0.00	20.8	0.99 ± 0.01	-20.43 ± 0.05	5.25 ± 0.05	-17.5	72 ± 18	7	-2.48 ± 1.53
0.46 ± 0.00	20.8	1.19 ± 0.01	-21.31 ± 0.05	5.29 ± 0.05	-18	128 ± 18	13	-0.62 ± 0.64
0.51 ± 0.00	19.2	1.47 ± 0.01	-21.29 ± 0.03	5.29 ± 0.03	-16	266 ± 76	8	-4.13 ± 0.88
0.50 ± 0.02	24.8	0.99 ± 0.01	-20.03 ± 0.04	5.29 ± 0.03	-16	273 ± 105	9	-6.55 ± 1.15
0.55 ± 0.01	20.8	1.43 ± 0.02	-21.04 ± 0.04	5.29 ± 0.05	-17	154 ± 36	4	-5.52 ± 1.89
0.60 ± 0.02	24.8	1.18 ± 0.01	-20.73 ± 0.04	5.34 ± 0.02	-17	91 ± 79	1	-2.48 ± 2.31
0.66 ± 0.01	19.2	1.92 ± 0.01	-20.20 ± 0.05	5.25 ± 0.04	-15	102 ± 37	9	-2.33 ± 1.18
0.74 ± 0.01	20.8	1.90 ± 0.02	-20.29 ± 0.45	5.30 ± 0.04	-16.5	128 ± 43	3	-4.11 ± 3.21
0.74 ± 0.03	24.8	1.47 ± 0.01	-21.20 ± 0.05	5.30 ± 0.03	-19	349 ± 96	9	-1.17 ± 0.21
0.97 ± 0.03	24.8	1.92 ± 0.01	-20.39 ± 0.06	5.30 ± 0.03	-17	78 ± 7	5	-0.38 ± 1.73

Ice Crystal Chamber Relative Humidity Series

Low (Near Ice Saturation) to Intermediate Relative Humidity

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.50 ± 0.02	24.8	0.99 ± 0.01	-5.04 ± 0.13	5.28 ± 0.04	N/A	197 ± 67	17	3.15 ± 0.60 †‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-8.11 ± 0.07	5.27 ± 0.03	-11	189 ± 99	7	6.86 ± 1.51 ‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-11.46 ± 0.08	5.28 ± 0.03	-12.5	164 ± 72	8	-0.16 ± 0.21
0.50 ± 0.02	24.8	0.99 ± 0.01	-11.83 ± 0.05	5.32 ± 0.04	-11.5	172 ± 67	8	0.03 ± 0.24 ‡
0.51 ± 0.01	19.2	1.47 ± 0.02	-13.72 ± 0.09	5.28 ± 0.07	-15	97 ± 43	8	6.01 ± 1.77 ‡
0.50 ± 0.02	24.8	0.98 ± 0.01	-13.98 ± 0.10	5.32 ± 0.03	-13.5	211 ± 56	5	-3.07 ± 0.80 ‡
0.52 ± 0.00	20.8	1.34 ± 0.01	-14.08 ± 0.04	5.25 ± 0.04	-17	87 ± 39	3	0.57 ± 0.61 ‡ *
0.52 ± 0.00	20.8	1.34 ± 0.01	-14.08 ± 0.04	5.25 ± 0.04	-17	87 ± 30	3	4.48 ± 1.76 ‡ *
0.51 ± 0.00	20.8	1.31 ± 0.01	-14.19 ± 0.05	5.28 ± 0.04	-16	85 ± 13	5	-3.75 ± 1.56 ‡
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.28 ± 0.04	-16	146 ± 70	12	-2.24 ± 0.34 ‡ *
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.28 ± 0.04	-16	80 ± 20	3	0.93 ± 0.71 ‡ *
0.51 ± 0.01	20.8	1.32 ± 0.01	-14.20 ± 0.09	5.28 ± 0.04	-16	94 ± 26	3	6.19 ± 2.29 ‡ *
0.51 ± 0.01	19.2	1.47 ± 0.02	-14.32 ± 0.05	5.29 ± 0.06	-15	103 ± 39	11	2.76 ± 1.27 ‡
0.52 ± 0.01	20.8	1.33 ± 0.01	-14.33 ± 0.05	5.28 ± 0.05	-14	96 ± 21	5	4.38 ± 2.25 ‡
0.50 ± 0.02	24.8	0.98 ± 0.01	-14.66 ± 0.09	5.32 ± 0.03	-14	147 ± 39	4	-1.96 ± 0.96 ‡
0.55 ± 0.00	20.8	1.41 ± 0.01	-15.30 ± 0.04	5.35 ± 0.04	-16	129 ± 39	11	-0.86 ± 0.71 ‡
0.55 ± 0.01	20.8	1.42 ± 0.01	-15.73 ± 0.12	5.34 ± 0.05	-16	74 ± 12	3	-5.19 ± 3.65 ‡
0.55 ± 0.00	20.8	1.41 ± 0.01	-15.84 ± 0.05	5.27 ± 0.04	-12	126 ± 43	10	-2.75 ± 0.85 ‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.12 ± 0.09	5.29 ± 0.04	-12	132 ± 25	5	-5.94 ± 1.95

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.52 ± 0.02	24.8	1.03 ± 0.01	-16.29 ± 0.08	5.34 ± 0.04	-14.5	184 ± 68	7	-3.29 ± 0.75 ‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.26 ± 0.14	5.27 ± 0.04	-14	233 ± 86	3	1.15 ± 0.43 *
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.26 ± 0.14	5.27 ± 0.04	-14	220 ± 86	3	-0.37 ± 0.31 *
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.26 ± 0.14	5.27 ± 0.04	-14	265 ± 94	3	-1.20 ± 0.40 *
0.51 ± 0.02	24.8	0.97 ± 0.01	-19.76 ± 0.04	5.37 ± 0.04	-16	160 ± 89	3	-2.10 ± 0.79
0.52 ± 0.02	24.8	0.99 ± 0.01	-20.03 ± 0.04	5.29 ± 0.03	-16	273 ± 105	9	-6.55 ± 1.15 ‡
0.51 ± 0.02	24.8	0.98 ± 0.01	-20.06 ± 0.09	5.35 ± 0.03	-16	187 ± 59	11	-2.11 ± 0.40

* Indicates multiple data points taken from single experiments in which the charge transfer is observed to change sign during the ice phase. See Section 4.3.3 and Figure 4.10.

† Data is a weighted average of multiple experiments.

‡ Data are repeated in corresponding temperature series.

Intermediate to High (Near Water Saturation) Relative Humidity

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.48 ± 0.02	24.8	0.95 ± 0.01	-5.30 ± 0.20	5.32 ± 0.03	-5	220 ± 68	7	0.56 ± 0.18
0.50 ± 0.02	24.8	0.98 ± 0.01	-8.32 ± 0.05	5.33 ± 0.03	-10	182 ± 86	4	4.51 ± 1.25
0.50 ± 0.02	24.8	0.98 ± 0.01	-8.35 ± 0.05	5.34 ± 0.03	-10	160 ± 53	7	0.59 ± 0.26
0.50 ± 0.02	24.8	0.98 ± 0.01	-11.25 ± 0.04	5.31 ± 0.03	-10	146 ± 74	6	-6.74 ± 3.28 ‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-11.40 ± 0.14	5.27 ± 0.03	-9	216 ± 79	6	-3.58 ± 0.77
0.50 ± 0.02	24.8	0.99 ± 0.01	-11.72 ± 0.06	5.27 ± 0.03	-9	189 ± 53	5	-2.27 ± 0.87
0.51 ± 0.00	20.8	1.31 ± 0.01	-13.96 ± 0.05	5.29 ± 0.04	-15	55 ± 20	5	-13.06 ± 7.01 ‡
0.51 ± 0.00	20.8	1.33 ± 0.01	-14.11 ± 0.07	5.22 ± 0.04	-12	123 ± 27	11	-14.10 ± 3.07 ‡
0.55 ± 0.01	20.8	1.43 ± 0.02	-15.52 ± 0.06	5.45 ± 0.05	-11.5	89 ± 21	10	-26.37 ± 5.76 ‡
0.51	19.2	1.48	-15.59	5.24	-12	47	2	-27.60

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
± 0.00		± 0.01	± 0.04	± 0.04		± 23		± 21.00 ‡
0.54 ± 0.01	20.8	1.38 ± 0.02	-15.57 ± 0.06	5.25 ± 0.05	-10	55 ± 21	2	-47.73 ± 42.69 ‡
0.55 ± 0.01	20.8	1.42 ± 0.02	-16.03 ± 0.07	5.32 ± 0.05	-16	59 ± 10	2	-14.08 ± 11.41 ‡
0.50 ± 0.01	19.2	1.45 ± 0.02	-16.09 ± 0.07	5.35 ± 0.07	-13	70 ± 23	7	-11.97 ± 3.53 ‡
0.48 ± 0.01	24.8	0.95 ± 0.01	-16.15 ± 0.07	5.28 ± 0.03	-12.5	148 ± 27	7	-17.59 ± 4.91
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.29 ± 0.04	5.28 ± 0.03	-12	47 ± 33	2	-9.29 ± 14.61
0.49 ± 0.02	24.8	0.97 ± 0.01	-16.83 ± 0.06	5.20 ± 0.03	-12	158 ± 56	2	-15.10 ± 6.03 ‡
0.50 ± 0.02	24.8	0.99 ± 0.01	-19.41 ± 0.10	5.29 ± 0.03	-13	145 ± 27	8	-6.43 ± 1.64
0.50 ± 0.02	24.8	0.98 ± 0.01	-19.86 ± 0.13	5.32 ± 0.03	-13	173 ± 55	11	-5.16 ± 0.96

‡ Data are repeated in corresponding temperature series.

Velocity Series

Velocity data shown in Figure 4.12 for 5.3 m s⁻¹ is calculated from -16°C Series and Ice Crystal Chamber Relative Humidity Series, and is not repeated here. Except were noted, all experiments were performed with the ice crystal chamber near water saturation.

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.47 ± 0.02	24.8	0.94 ± 0.01	-16.03 ± 0.05	3.07 ± 0.03	-15	277 ± 96	12	-6.00 ± 0.76
0.50 ± 0.02	24.8	0.99 ± 0.01	-16.00 ± 0.03	3.06 ± 0.04	-16.5	293 ± 119	13	-0.71 ± 0.12*
0.45 ± 0.02	24.8	0.89 ± 0.02	-16.02 ± 0.04	3.07 ± 0.04	-16	490 ± 227	20	-1.21 ± 0.08*
0.50 ± 0.02	24.8	0.98 ± 0.01	-15.99 ± 0.03	3.08 ± 0.04	-17	61 ± 14	6	-2.11 ± 2.43
0.50 ± 0.02	24.8	0.98 ± 0.02	-16.04 ± 0.02	4.12 ± 0.02	-16	149 ± 31	12	-3.71 ± 0.81
0.50 ± 0.02	24.8	0.98 ± 0.01	-16.02 ± 0.03	4.13 ± 0.02	-13	177 ± 120	2	-6.20 ± 2.50
0.50 ± 0.02	24.8	0.98 ± 0.01	-16.34 ± 0.07	4.14 ± 0.02	-15	170 ± 61	8	-6.15 ± 1.46

ELWC (g m ⁻³)	Mean Droplet Diam. (μm)	LWC (g m ⁻³)	Temp. (°C)	Velocity (m s ⁻¹)	Chamber Temp. (°C) ± 0.5 °C	Ice Crystal Conc. (l ⁻¹)	No. of Points	Charge Transfer (fC per collision)
0.50 ± 0.02	24.8	0.98 ± 0.01	-16.31 ± 0.10	6.12 ± 0.03	-15	139 ± 76	4	-4.92 ± 1.62
0.50 ± 0.02	24.8	0.98 ± 0.01	-16.21 ± 0.09	6.13 ± 0.04	-13	153 ± 70	12	-4.18 ± 0.79
0.50 ± 0.02	24.8	0.98 ± 0.01	-16.30 ± 0.07	6.15 ± 0.04	-14	109 ± 44	11	-5.60 ± 1.33

* Experiments were performed at low to intermediate chamber relative humidity, and are not included in Figure 4.12.

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