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Charge Separation Mechanisms in Clouds

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Abstract Mechanisms of thunderstorm electrification are presented and discussed.

Keywords Charge separation mechanisms: drop break-up, ion charging, convective charge transport, inductive processes · Ice particle mechanisms: Workman–Reynolds freezing potentials, contact potentials, dislocation charges, temperature gradients, melting effects, ice splinter charges, fragmentation effects · Ice crystal/graupel charging: thunderstorm observations, charging requirements, laboratory studies, ice/ice charging mechanism

1 Introduction

Worldwide thunderstorm activity is responsible for maintaining a weak negative charge on the Earth's surface and a corresponding positive charge in the atmosphere. In general, cloud to ground lightning brings negative charge to ground from a negatively charged region in the cloud that also releases positive ions by point discharge in the strong electric field region below a thunderstorm. The resulting global fair weather electric field is around $-120\ \mathrm{V\,m^{-1}}$ at the ground and decreases to zero in the conducting region of the ionosphere. Wilson (1916) assumed a vertical charge dipole within thunderstorms and determined that the charge regions are usually positive above negative: he measured electric field changes at the ground caused by intra-cloud lightning in which the dipole charges neutralised each other. This picture was confirmed with extensive electric field change measurements made by Krehbiel et al. (1979) in New Mexico, in which the locations and values of charges in thunderstorm charge centres were determined. More complicated charge centre distributions have been reported, for example, by Stolzenburg et al. (1998).

The generally accepted concept for the development of the thunderstorm charge dipole is the physical separation of oppositely charged particles within the cloud. Larger cloud particles fall under gravity while smaller particles are transported in the updraught; if these

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particles carry negative and positive charges respectively then the normal charge dipole will result.

2 Charging Mechanisms

2.1 Drop Break-up

Many mechanisms have been proposed to account for the observed charges on cloud particles. Lenard (1892) noted the electrical effects associated with drop break-up near waterfalls. The larger droplets became positively charged while the fragments were negative. This may be explained by the negative electric charge on the surface of water that is carried away on the smaller fragment droplets. Blanchard (1963) observed that bubble bursting over the oceans releases positively charged jet droplets formed from the positive liquid inside the drops; these charged droplets may be carried up into clouds by the local air currents. However, the break-up of individual water droplets in clouds is a rare event, surface tension forces are sufficient to hold all but the largest drops together even in the presence of severe turbulence. Drop break-up may only occur during collisions of two particles, when other, stronger, charging processes may take place leading to an ordered separation of opposite charges in the cloud.

2.2 Ion Charging

Charging processes have been considered involving atmospheric ions produced by cosmic rays and by radioactivity in the ground. The ionisation in a volume of free air over land is around 11 ion pairs per cubic centimetre per second. Thunderstorms themselves produce positive ions in the high field regions below cloud by corona discharge from sharply pointed objects. Lightning bringing negative charge to ground injects large numbers of positive ions into the cloud where the ions may become involved in subsequent cloud particle charging.

Gerdien (1905) used the result that water molecules deposit more readily on negative ions than on positive ions as a process of cloud particle initiation; however, unrealistically high supersaturations of several hundred percent are required to activate droplet growth on ions, as shown by C.T.R. Wilson with his cloud chamber.

Wilson (1929) proposed a mechanism of selective ion capture whereby a cloud particle will be polarised in the pre-existing vertical fair weather electric field and so will carry a positive charge on its lower half and an equal negative charge on its upper half. As it falls, the lower charge attracts negative ions which are captured and lead to the net negative charging of the falling particle. But the process is limited because sufficient build up of negative charge on the particle will lead to the subsequent capture of positive ions. The mechanism may increase the vertical electrical field to about 50 $\mathrm{kV\,m^{-1}}$ before this limiting charge is reached; however, this field strength is inadequate to cause electrical breakdown and is about an order of magnitude lower than the typical maximum field strength measured in thunderstorms. This "influence" mechanism was one of the first to invoke the inductive process of thunderstorm charging, as discussed in Sect. 2.4. Elster and Geitel (1913) used the concept for the charging of polarised drops when smaller droplets rebounded from their underside, thus removing positive charge to be carried up on the smaller droplets while the negative drop fell and strengthened the electric field. However, in stronger fields, coalescence is the likely result of a collision.

2.3 The Convective Mechanism

Wilson proposed a “convective” mechanism involving the movement of charges carried by the natural convection currents in a storm cloud. He suggested that ion capture by cloud particles leads to the initial cloud electrification. This theory was proposed by Grenet (1947) and has been championed by Vonnegut (1953), whose concept is represented in Fig. 1. Positive ions near the ground are attracted to the cloud to be captured by droplets and carried to the cloud top in the updraught. In turn, this positive region attracts negative ions to the cloud that are captured by falling particles whose charge then strengthens the lower negative charge centre. The cycle continues with substantial field intensification.

It is hard to visualise the ordered separation of charge in this process that would lead to an electric field strength capable of initiating lightning. Vonnegut himself made several predictions about the charge distribution in clouds that would be consistent with his theory. He wrote “If this theory is correct, these measurements should disclose large masses of electrically charged air that are some distance from the region of precipitation, a situation that would be unlikely if precipitation were responsible for electrification. Furthermore these measurements should show that in both the positive and negative regions of charge, the greater part of the charge is generally in the form of small charged cloud particles and Aitken nuclei rather than precipitation particles. In some cases one might expect that appreciable electric fields had developed before the precipitation particles had formed.” However, measurements by Reynolds and Brook (1956) showed that rapid electrification of thunderstorms was associated with the development of ice phase precipitation particles. Besides, many measurements of charges on cloud particles in thunderstorms showed them to be significantly charged (e.g., Gaskell and Illingworth 1980).

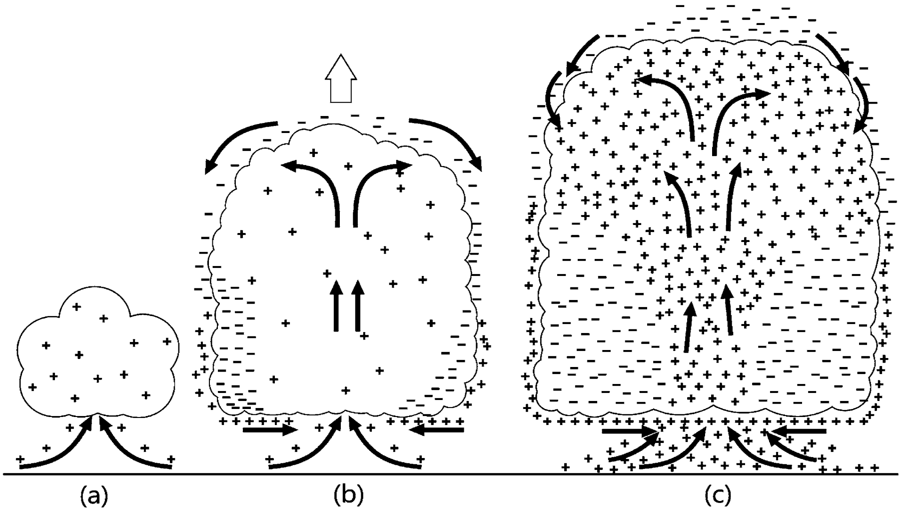


Fig. 1 The convective charging mechanism. (**a**) Positive space charge ingested into cloud. (**b**) A negative screening layer forms on the cloud particles on the outside boundary, which moves down the sides toward the cloud base. Additional positive charge is further ingested at the base, and further negative charge flows to the upper cloud boundary to replace the loss of the screening layer that flowed to the cloud base along the sides. (**c**) The lower accumulation of negative charge increases the electric field strength to a magnitude large enough to generate positive corona from ground objects. The corona becomes an additional source of positive charge that feeds into the cloud. (Emersic 2006)

Masuelli et al. (1997) used a numerical model of the cloud charging involved in the convective process and found an inadequate rate of field development. More recently, Helsdon et al. (2002) examined the convective charging hypothesis using a three-dimensional storm electrification model of a small, weak storm and a larger, severe, storm. With a full treatment of small ions, including attachment to hydrometeors, the inclusion of field-dependent surface point discharge, and the components of the Maxwell current, the results from both storm simulations indicated disorganised, weak electrical structures during the mature and dissipating stages. Furthermore, currents within the storm were dissipative and the cloud acted as a barrier to the external conduction current when convective-only charging was considered. However, since the convective charging hypothesis, by itself, is unable to produce significant charging or strong electric fields in their simulated clouds, they concluded that it is not a viable mechanism for thunderstorm electrification.

2.4 Inductive Charging

The inductive process, shown in Fig. 2, relies on the pre-existing vertical electric field to induce charges so that particle rebounds can separate charge and strengthen the field. Initially, the field may be due to the downward directed fair weather field ($-\underline{\mathbf{E}}$) resulting from positive charges in the atmosphere with a negative ground surface below. The interacting cloud particles have sufficiently high an electrical conductivity that there is time for the induced charges to form in the particles in response to the external electric field. In other planetary atmospheres with materials other than water ice, the particle conductivity and electrical relaxation time needs to be considered both for response to a changing external field and the time required for charge transfer. Collisions of water droplets often leads to coalescence, so the most likely situation in which the inductive process may act in clouds is for rebounding ice/ice or, possibly, ice/water collisions. A smaller cloud particle rebounds from the underside of a larger ice particle in the existing vertical electric field; it removes charge and is carried around the larger particle in the upward moving airstream—gravitational separation then occurs with the larger particle falling while the oppositely charged smaller particle is carried aloft thus strengthening the electric field. But the process does not always work like this; Saunders and Al-Said (1976) showed that when pairs of larger drops collided they partially coalesced, swung around each other and separated induced charge in a way that reduced the ambient field, as shown in Fig. 3.

Experimental studies with ice/ice collisions by Illingworth and Caranti (1985) showed that the charge transfer was limited by the purity of naturally occurring ice. Ice has an

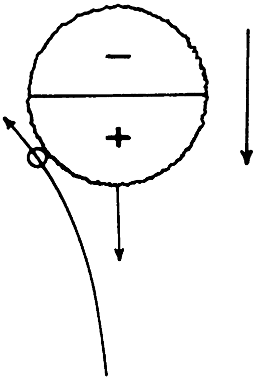


Fig. 2 An uncharged graupel pellet in the environmental vertical electric field. The field induces positive and negative charges as shown. A rebounding cloud particle removes positive charge leaving the graupel negatively charged. The negatively charged graupel falls while the positively charged cloud particle is carried aloft so that the environmental electric field $\underline{\mathbf{E}}$ is strengthened

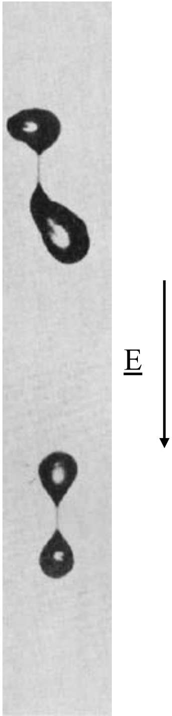


Fig. 3 Collisions between nearly equal sized water drops result in particle separations in the direction of the vertical environmental electric field that are in a direction to dissipate the field. (Al-Said 1977)

electrical conductivity high enough to allow the induced charges to form, but low enough that in the brief collision time, there is insufficient time for a complete transfer of charge. They found that when the ice was doped to increase its electrical conductivity, the theoretical value of induced charge transfer appropriate to two conducting particles was achieved. For this reason the inductive process involving ice/ice collisions has not been considered a viable mechanism for thunderstorm electrification.

Mason (1988) made a convincing case from thunderstorm observations and numerical modelling of the inductive process that it may be a viable mechanism for the case of water droplets rebounding from the underside of ice pellets. Brooks and Saunders (1994) carried out studies of this process in a laboratory cloud chamber in which an ice coated sphere fell through a cloud of supercooled water droplets in a vertical electric field. They showed that measurable and significant charge transfer was achieved when the droplets rebounded off riming graupel pellets, thus reviving the inductive mechanism. This process may help account for observations in thunderstorms of regions of cloud particles that have acquired their charges very rapidly in later stages of storm development when substantial electric fields are already present.

Despite this recent turn around in the fortunes of the inductive process, it does have to overcome a severe problem: observations in the early electrification period of thunderstorms by Christian et al. (1980) showed charges on graupel (small hail pellets) larger than could have been generated in the maximum electric field strength measured in thunderstorms. These results were obtained from an airborne instrument in a New Mexico thunderstorm in which a cloud particle imager and charge induction device gave corresponding values of charge and graupel size for individual precipitation particles. They concluded that cloud particle charges producing the first lightning stroke are unlikely to be due to the inductive process. Other processes must be responsible for the production of the observed charges in the time available.

3 Particle Charging Involving the Ice Phase

Observations in thunderstorms have shown that strong electrification follows the development of ice particles. Reynolds and Brook (1956) noted a rapid increase in radar reflected intensity from a storm in which the electric field was approaching breakdown leading to lightning. Illingworth and Lees (1992) used radar to observe the position of lightning and precipitation in a UK summer thunderstorm and confirmed that lightning is co-located with the maximum precipitation radar echo; they concluded that the presence of graupel is required for lightning to occur. Most mechanisms considered today involve cloud ice in the charging process.

3.1 Workman–Reynolds Freezing Potentials

When supercooled water droplets are captured by a falling ice pellet, the water freezes. Workman and Reynolds (1950) measured a freezing potential across the ice/water interface during the freezing process. They suggested that this may lead to charge separation by the shedding of charged liquid water due to splashing during the collision process. In a laboratory study in which the freezing potential was measured as a function of time after the water/ice collision, Caranti and Illingworth (1983) found that the potential developed very slowly. In fact it turns out that the full potential develops in bulk ice in a time longer than the freezing time of the captured small supercooled cloud droplets. Thus the process cannot account for significant charge transfer in thunderstorms.

3.2 Contact Potential

Caranti and Illingworth (1980) and Caranti et al. (1985) noted that an ice particle developed a surface contact potential when it accreted supercooled droplets that froze on its surface. The surface charge was negative, and if considered as a “contact potential” (despite the fact that ice is a proton conductor), the negative potential could account for the negative charging of the colliding ice surface having the larger negative contact potential. However, this process could only account for the negative charging of accreting graupel during ice crystal collisions; besides, laboratory studies have shown that the charge sign is controlled by temperature and water accretion rate. Furthermore, it has been observed that ice surfaces growing by vapour diffusion charge positively and sublimating ice surfaces charge negatively during rebounding collisions with smaller ice particles (as discussed below), but when ice surfaces were caused to grow or sublimate by cooling or heating, their contact potential was not affected. So, the contact potential mechanism has been discounted from further consideration in the terrestrial atmosphere.

3.3 Dislocation Mechanism

Keith and Saunders (1990) suggested that charge transfer in ice/ice collisions may be associated with charges on dislocations in the ice lattice. They reported that dislocations carry a positive charge and that during a collision between an ice crystal and a graupel pellet this charged material may be transferred. They calculated that for a typical number of dislocations per unit area of $5 \times 10^9\ \mathrm{m}^{-2}$, with a charge per unit length of $6 \times 10^{-11}\ \mathrm{C\,m}^{-1}$, (determined by X-ray methods) the charge available on a typical collision area of $55 \times 55\ \mu\mathrm{m}^2$ is +50 fC, which is of the observed order of magnitude from laboratory studies of charge transfer. Dislocation concentration depends on the ice growth rate, and as discussed elsewhere, crystals and graupel grow at different rates as a function of cloud conditions so positive or negative charge transfers may occur. However, in ice, there are mobile ions of both signs which are free to move under local electric fields such as would be set up by a charged dislocation. So, any mass transferred during a collision is likely to consist of the dislocation together with surrounding oppositely charged material. However, in other planetary atmospheres where the colliding particles may consist of materials that develop charged dislocations but may not have mobile charges available, this process may be viable.

3.4 Temperature Gradients in Ice

Latham and Mason (1961), working in the laboratory, studied charge transfer during impacts of ice crystals on an ice sphere representing a falling graupel pellet. They noted that a temperature difference between the particles led to charge transfer, such that the warmer ice particle lost positive charge. They required the graupel to be warmed by collection of supercooled droplets in the cloud chamber (riming), however their simulations did not include riming itself—they relied on artificial heating of the ice surface. They attributed this result to the higher mobility of positive ions in ice compared with negative ions: during contact the positive ions are able to move away from the warmer ice surface leaving it negatively charged. They developed a numerical model, however, the actual charges measured in the laboratory were considerably in excess of the theoretical predictions. Later, Marshall et al. (1978), Gaskell and Illingworth (1980) and Jayaratne et al. (1983) showed in laboratory studies that charge transfers could be obtained in the opposite direction to the direction of the temperature gradient between colliding ice particles. Later it was realised that particle growth or sublimation rates control the sign of charge transfer and naturally, these rates are temperature dependent as well as being influenced by the local cloud supersaturation.

3.5 Melting Effects

Despite graupel usually being charged negatively in the lower charge region of thunderstorm dipoles, rainfall measured below cloud is often positively charged. Dinger and Gunn (1946) proposed a charge transfer process associated with melting. Drake (1968) noted that convection in a melting ice sphere produced negatively charged droplets ejected from bursting air bubbles at the surface. The sign of charge and the conditions under which the charges were separated were highly dependent on the impurities in the melting ice. The positive charge on the melted drops may help account for the lower positive charge region in thunderstorms and for the positive charge on precipitation. The capture of positive ions below cloud will also contribute to the drop charge.

3.6 Ice Splinter Charging During Hallett–Mossop Ice Multiplication

Ice splintering has had a long history of possible involvement in charging. Latham and Mason (1961) noted that ice splinters created during the freezing of supercooled droplets (riming) on a larger ice surface were charged. Hallett and Saunders (1979) studied the charges on ice splinters produced during the Hallett and Mossop (1974) ice multiplication process. The Hallett–Mossop mechanism is an important source of ice particles in clouds, particularly in regions where the temperature is between −3°C and −8°C. The process involves the accretion of supercooled water droplets by falling ice pellets, ice crystals or graupel. The supercooled liquid water immediately freezes and if the conditions are suitable, the freezing droplets form a shell of ice which can shatter under the high stresses involved caused by the effects of expansion upon freezing. The ice surface may break up into fragments, or a spicule of material may be formed through which liquid is ejected that rapidly freezes. The net effect is to produce a large number of small ice particles that grow very rapidly by the diffusion of local water vapour and form ice crystals that can in turn accrete smaller droplets thus continuing the multiplication process.

Hallett and Saunders (1979) investigated this multiplication process with a view to the possibility that the ejected ice fragments were electrically charged and so could contribute to thunderstorm electrification. They found that the growing ice pellet charged positively with a negative charge on an ejected ice fragment of order -10^{-16} C. Interestingly, when the vapour supply used to grow the cloud droplets was turned off, the charge sign reversed; under these conditions the ice pellet would have started to sublimate. The authors concluded that the sign of the charge transfer depends on the physical state of the rime ice surface and its vapour pressure excess or deficit relative to the environment. This turned out to be a far reaching conclusion, as will be seen below, however the magnitude of the charges on the fragments is too small to be able to account for the observed electrification rates in thunderstorms. They did note that in the presence of liquid cloud, the ice crystals grew rapidly and when these larger crystals collided with a riming ice surface, then substantial charges were transferred. This work led to extensive studies of charging during ice crystal collisions with riming graupel, as discussed in Sect. 4 below.

3.7 Fragmentation Effects

The surface of ice is in an environment in clouds in which it grows by vapour diffusion from the environment. It may, therefore, become covered in frost, which can break off, or be knocked off during collisions with other cloud particles. Takahashi (1978) invoked crystal breakup for his observed charging during crystal collisions with riming graupel. In droplet/graupel collisions at speeds between 10 and 30 $\mathrm{m\,s^{-1}}$, Avila et al. (2003) noted the production of charged fragments. Such speeds are high for terrestrial thunderstorms but may be relevant in other planetary conditions.

Charge transfer associated with surface growth or sublimation has been noted by everyone who has worked in the area of collisional ice charging in clouds. The relative growth rate theory discussed below, has its origins in the often observed result that growing ice surfaces charge positively, while sublimating ice surfaces charge negatively, as noted by Findeisen (1940), Findeisen and Findeisen (1943), Buser and Aufdermaur (1977), Marshall et al. (1978), Gaskell and Illingworth (1980), Jayaratne et al. (1983), Rydock and Williams (1991), Caranti et al. (1991), Dong and Hallett (1992), Saunders et al. (1993), Scavuzzo et al. (1995) and Mason and Dash (2000). Most of these experiments were carried out by artificially cooling or heating an ice surface while its surface was removed by collisions or by fragmentation, thus the laboratory simulations are not representative of the conditions in real clouds. For example, an ice surface grows by diffusion in a real cloud causing the ice surface to be heated by the release of the latent heat of condensation. However, in laboratory simulations, in order to ensure that the ice surface grows, it is artificially cooled. Correspondingly, a sublimating ice surface in clouds loses mass because the surrounding environment is at a lower vapour pressure and it is cooled by the latent heat of sublimation; however, in laboratory simulations the ice would be heated to ensure sublimation. Thus these simulation experiments do not provide realistic cloud or ice surface conditions.

These differences have to be considered when relating the laboratory results to nature—however, the observation that ice surfaces growing by diffusion charge positively and sublimating ice surfaces charge negatively is consistent with observations of the sign of charge on fragments breaking off a growing or sublimating ice surface and provide an important clue to the mechanism of charge transfer when ice crystals rebound from ice particles that are not only growing or sublimating, but are also accreting supercooled water droplets. Various theories have been proposed for these observations, the most recent being a theory relying on the relative diffusional growth rates of two briefly interacting ice particles, as discussed below.

4 Ice Crystal/Graupel Charging

4.1 Background

The beginnings of this non-inductive particle charging mechanism (so-called because the presence of an electric field plays no role in the mechanism) came when Reynolds et al. (1957) measured in the laboratory the charge transferred to riming graupel when ice crystals rebounded and removed the equal and opposite charge. The process is shown in Fig. 4. They reported the negative charging of graupel that could account for the region of negatively charged graupel in thunderstorms. The positively charged ice crystals would be carried aloft to form the upper charge region of the dipole. Takahashi (1978), in similar experiments, showed that the riming graupel charge sign could be negative or positive by crystal rebounds depending on cloud temperature and liquid water content.

Work in this research area started in the Manchester laboratory in 1978 with a search for charged fragments created during the Hallett–Mossop ice multiplication process, as discussed above. It soon became apparent that, although the small ice particles ejected during the process were charged, the charges were too small to be of significance to thunderstorm electrification processes. However, it was noted that after time had been allowed in the cloud chamber for the ice crystals to grow to tens of micrometres in diameter, the charge transfer was considerably increased. It was also noted with interest that under some cloud conditions

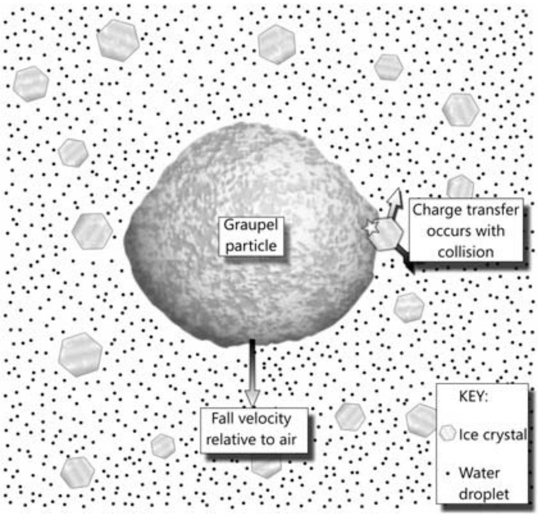


Fig. 4 The charging a graupel particle (soft-hail pellet) during droplet accretion (riming) and ice crystal rebounding collisions. (Emersic 2006)

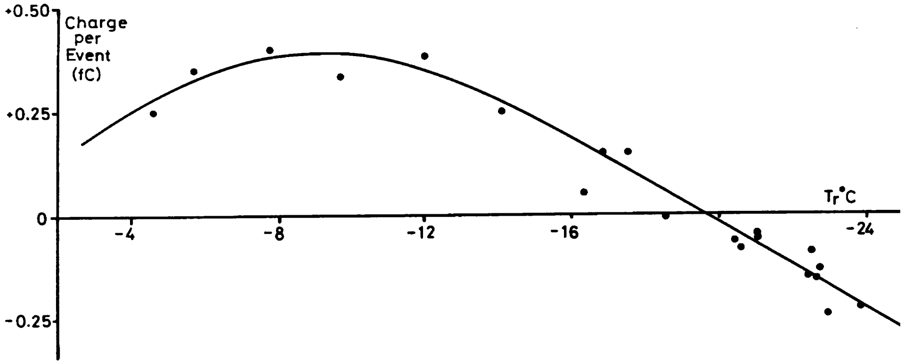
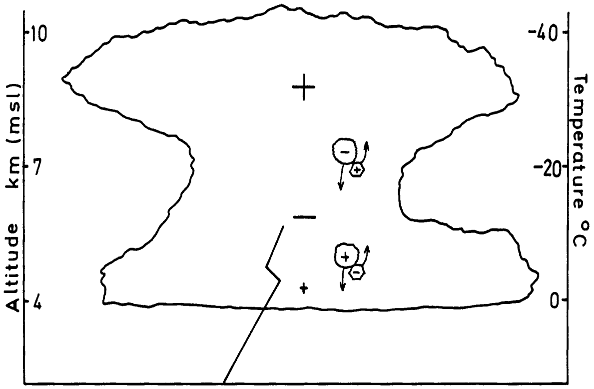


Fig. 5 The charge transferred to a riming graupel pellet by a separating ice crystal following a collision, at constant cloud water content. (Jayaratne 1981)

Fig. 6 The charging of a thunderstorm leading to the tripole charge distribution. Graupel pellets charge negatively at low temperatures and positively at higher temperatures



a riming ice surface could become charged positively, whereas Reynolds et al. (1957) had concentrated on their observation of negative charging of graupel, a result they used to account for the negative charge region in thunderstorms. A more detailed study by Jayaratne et al. (1983) showed that the charge sign could be positive or negative as a function of the cloud temperature and liquid water content; Fig. 5 shows graupel charge sign reversal at −20°C from experiments at a particular water content. Lower water contents moved the reversal point to higher temperatures favouring negative graupel charging in the cloud. The temperature dependence of the charge transfer helps account for the sign of graupel charging required to account for the usual dipole structure of a thunderstorm reported by Wilson (1916, 1929), and the often observed tripole structure noted by Williams (1989). Figure 6 shows a storm with a tripole charge structure: graupel/crystal charge separation events at higher altitudes and lower temperatures lead to negative graupel that falls under gravity while the positive ice crystals are carried aloft; at lower altitudes and higher temperatures, the charge transfer reverses sign leading to the often observed lower positive charge centre, while the negatively charged ice crystals are carried up to reinforce the negative charge centre. The lower positive charge centre has been detected in thunderstorms and is thought to help the initiation of cloud to ground lightning strokes from the lower region of negative charge.

4.2 Thunderstorm Charge Observations

The work on charge transfer mechanisms follows from an understanding of the cloud physical and electrical properties of thunderstorms. There have been many studies of thunderstorm properties, by remote sensing techniques as well as by research flights through storms. Krehbiel (1986) reported on studies in New Mexico in which the location of charge centres was identified by multiple-station analysis of the electric field change associated with intracloud lightning. They noted that during the course of a storm, the lower negative charge region maintained a steady altitude around 7 km corresponding to a temperature around −15°C. Meanwhile, the upper positive charge centres were observed to be carried up at 8 m s^{-1}, a rate that corresponded to the updraught speed in the cloud. Krehbiel (1986) also compared data on charge centre location in storms in other locations. Thunderstorms in New Mexico, in Florida, and winter storms over the Sea of Japan all possessed a negatively charged region located around the −15°C level despite the considerable differences in the dynamics and characteristics of these various storms.

In a series of multiple aircraft penetrations through thunderstorms in Montana, Dye et al. (1986) reported on simultaneous measurements of cloud parameters and electrical properties. They noted that increases in electric field strength occurred in regions containing a mix of liquid water and of ice particles. Ice crystals and graupel pellets were identified by airborne laser probes carried on aircraft flying in regions of strong electric field. They also reported that electrification appeared to be occurring at the interface between the updraught and downdraught regions of the clouds.

These observations point strongly to a precipitation based charging process of thunderstorm electrification and they strengthened the growing conviction that ice crystals rebounding from riming graupel in the presence of supercooled liquid water is a requirement of the charge transfer process leading to electric field development and lightning.

4.3 Thunderstorm Charging Requirements

In order to ascertain whether laboratory measured charge transfer values are adequate to account for thunderstorm electrification, Mason (1953) used thunderstorm observations to put forward some basic requirements of a viable theory of charge generation.

1) Time available for electric field generation is 30 minutes.
2) Charge generation produces 20 to 30 Coulombs per flash.
3) Charge separation occurs between the 0°C and −40°C levels in a region of radius 2 km.
4) The main negative charge centre is between the −5°C and −25°C levels depending on the cloud physics, the main positive centre is a few kilometres above the negative centre. The lower positive charge is close to the 0°C level.
5) Electric field development is associated with the development of precipitation in the form of soft hail (graupel).
6) The first lightning occurs within 12 to 20 minutes of the first radar detection of large particles.
7) Charge theories/mechanisms must generate 5 to 30 $\mathrm{C\,km^{-3}}$ leading to a charge generation rate of order 1 $\mathrm{C\,km^{-3}\,min^{-1}}$.

From these basic requirements, Mason determined the magnitude of the charge transfer events needed to account for the observed rates of electrification. Falling graupel pellets, radius R, number density n_h, collide with ice crystals, number density n_i.

The number of collisions/m^3/sec $= \mathrm{d}N/\mathrm{d}t = E\pi R^2 n_h n_i V$ where V is the relative velocity between graupel and crystals and E is the crystal/graupel collision efficiency.

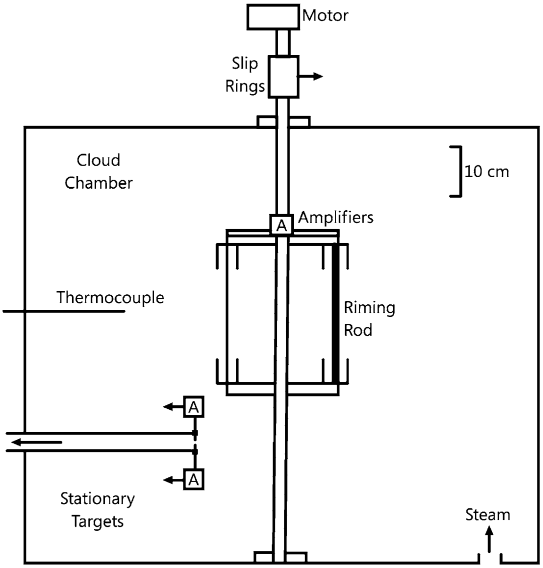


Fig. 7 Cold room cloud chamber and charge transfer apparatus. (Jayaratne 1981)

Graupel precipitation rate, $p = (4/3)\pi R^3 \rho n_h V$ where $\rho =$ the density of a graupel pellet. Each collision produces charge transfer q. To account for observations, the rate of production of charge per unit volume of cloud: $\mathrm{d}Q/\mathrm{d}t = q(\mathrm{d}N/\mathrm{d}t) = (3/4)q(En_\mathrm{i}p/R\rho) = 1$ C km^{-3} min^{-1}.

For a typical precipitation rate, $p = 5$ cm hr^{-1}. Graupel density $= 0.5$ g cm^{-3}. $R = 2$ mm. $n_i = 0.1$ cc^{-1} for crystals > 80 µm.

So, $q = (4/3)(\mathrm{d}Q/\mathrm{d}t)/(En_\mathrm{i}p/R\rho) = 1.6 \times 10^{-14}$ C per collision, if $E = 1$ and every ice crystal collision results in a rebound.

4.4 Laboratory Studies of Thunderstorm Charging Processes

There has been a long series of measurements in the Atmospheric Science Laboratory in Manchester where large cold chambers permit the growth of clouds to simulate atmospheric processes. Extensive thunderstorm charge transfer experiments have been performed in which the magnitude and sign of the charge transfer during ice crystal rebounds from riming graupel have been determined as a function of cloud temperature, graupel temperature, cloud water content, cloud droplet size distribution, impurity content of the graupel, ice crystal size, relative velocity between the colliding particles and their collision and separation probabilities. The most recent work points to the importance of ice particle diffusional growth rates controlled by the supersaturation in the cloud.

Figure 7 is a typical laboratory cloud chamber located inside a cold room. A cloud of water droplets condenses from the continuous water vapour input. The droplets supercool to the ambient temperature. Ice crystals are initiated by introducing a fine wire cooled to liquid nitrogen temperature and grow from the available water vapour. Figure 8 shows a three minute sequence of crystal growth from the available water vapour and cloud droplets, whose number and concentration decline and recovery during the run track the variations in cloud supersaturation. Metal rods, typically of 5 mm diameter, are moved through the cloud on a rotating frame; droplets accrete on the rod to form rime ice simulating the growth of a graupel pellet. Ice crystals strike the rod and if they rebound, charge is separated. The

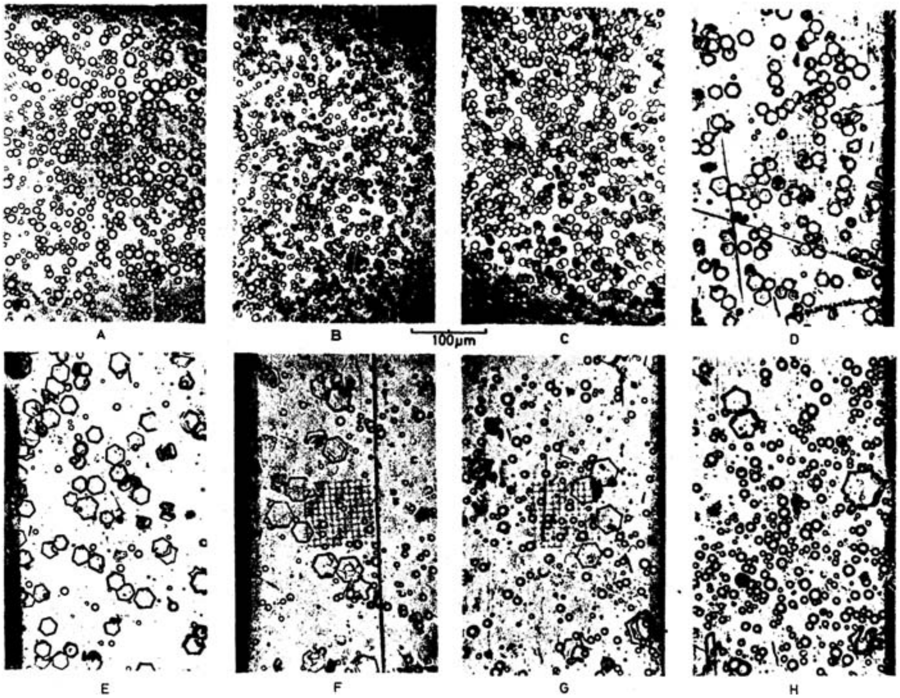


Fig. 8 A time sequence of cloud conditions over a period of three minutes from crystal nucleation. The droplets and cloud vapour provide water vapour for crystal growth. When the crystals have grown, they fall out of the cloud and the continuous vapour supply re-establishes the water droplet cloud. (Jayaratne 1981)

riming target rod is connected via slip rings to an electrometer so that the total charge due to many crystal charge transfer events is measured. Alternatively a stationary target is used while the cloud is drawn past using a suction pump. For the same velocity, between 2 and 9 $\mathrm{m\,s^{-1}}$, representing the fall speed of a graupel pellet, both stationary and moving targets give similar results.

Figure 9 represents a study of the effect of velocity on charge transfer for positive and negative graupel charging. The dependence of charge transfer on ice crystal size is shown in Fig. 10. These results (Keith and Saunders 1990) confirm that the magnitude of charge transfer determined in these laboratory studies is adequate to account for thunderstorm electrification according to the analysis of Mason (1953). Droplet size also influences the charge transfer as shown by Avila et al. (1999).

4.5 The Thunderstorm Charging Mechanism

A thunderstorm charging mechanism based on vapour deposition rate, first proposed by Baker et al. (1987), has been successful in helping to account for differences between the results from various laboratory studies. The concept follows on from the result described earlier that, during collisions leading to the removal of some surface mass from the larger particle, fast growing ice surfaces charge positively and conversely, sublimating surfaces charge negatively. Baker et al., suggested that an additional variable comes into play when two ice surfaces having different vapour diffusional growth rates come into brief contact, namely the surface state of the smaller particle in the collision process. They suggested that

Fig. 9 Charge transfers when ice crystals rebound from riming graupel, as a function of velocity, for the positive and negative charging regimes. (Keith 1987)

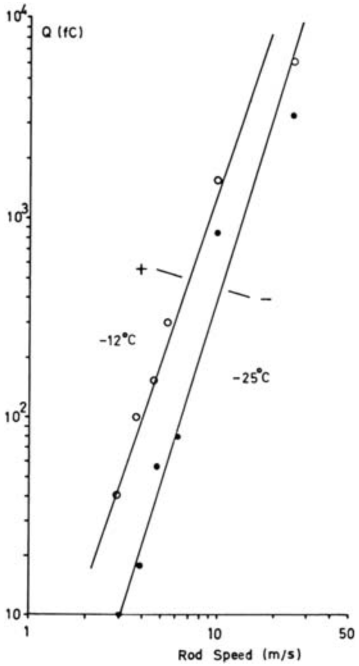
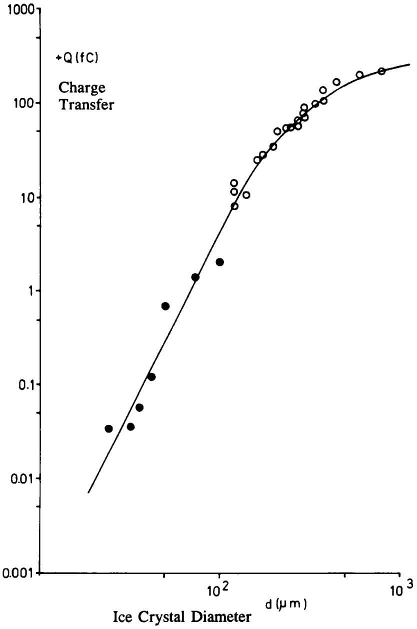


Fig. 10 Charge transfer to a riming target as a function of ice crystal size. (Keith 1987)



the *relative* diffusional growth rates of the interacting ice particle surfaces was the factor that controls the sign of charge transfer. The charge transfer follows the rule that the ice surface that grows faster by vapour diffusion charges positively during ice crystal/graupel

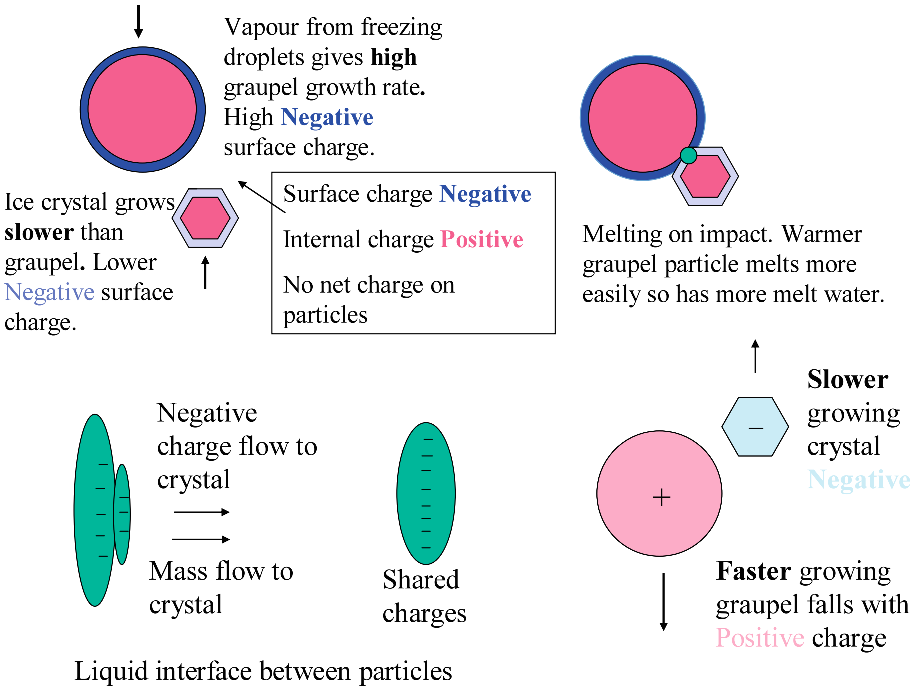


Fig. 11 A graupel pellet, warmed by rime accretion collides with a colder, faster growing, ice crystal. Charges and mass are shared in the melt water. The graupel becomes negatively charged while the crystal carries positive charge

rebounding collisions. This concept has stood the test of time, and has been shown to be consistent with the results obtained in various laboratories.

The theory was developed further by Dash et al. (2001)—-the faster growing ice surface has more negative surface charge available for transfer, and hence charges positively. According to Dash et al., rapid vapour deposition to an ice surface produces disordered growth, with ionic defects at vapour and grain boundary interfaces; faster growth leads to higher charge densities. The OH^- ions are held in position by their hydrogen bonds, but positive ions are able to diffuse away from the surface into the bulk ice, leading to a negative surface potential. Two colliding ice surfaces tend to equalise their surface charges so that the faster grown surface loses negative charge. The collisional impact melts a local volume on each surface with the warmer graupel providing more mass than the colder crystal. The charge exchange takes place in the melt water whereby the melted masses and associated charges are shared between the separating ice particles. In this way, the negative charge shared leads to positive charging of the faster growing ice surface. However, the liquid mass transfer is in the opposite direction to the transfer of negative charge, as noted by Mason and Dash (2000). The equalisation of charges occurs on a time scale of microseconds, which is much less than the estimated 0.1 ms contact time. This provides insufficient time for the deeper protons in the ice to react during the available contact time. Figure 11 represents the case of a faster growing ice crystal rebounding from a slower growing graupel pellet, so that the charge and mass exchange during the collision results in the graupel becoming negatively charged.

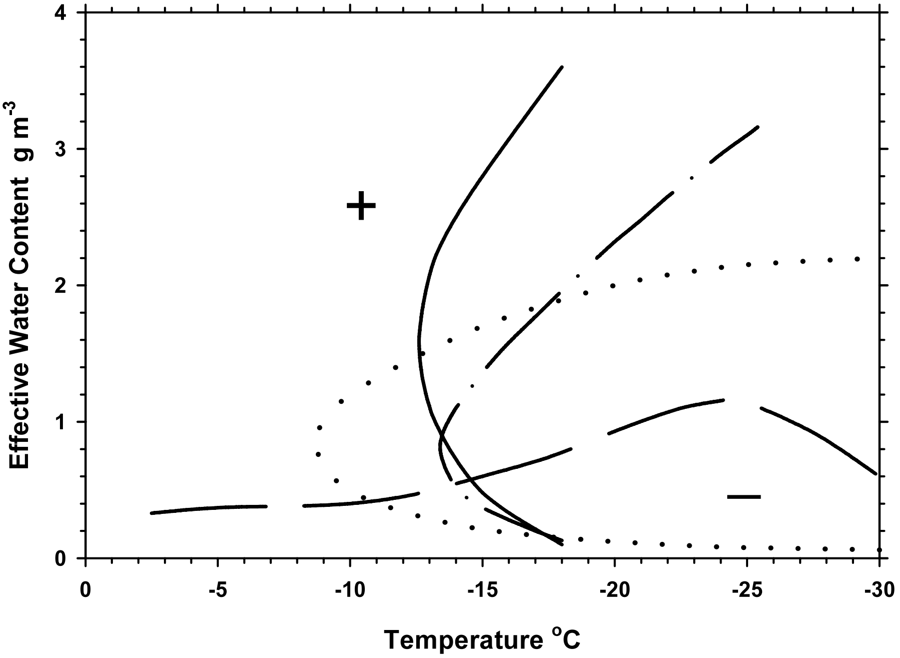
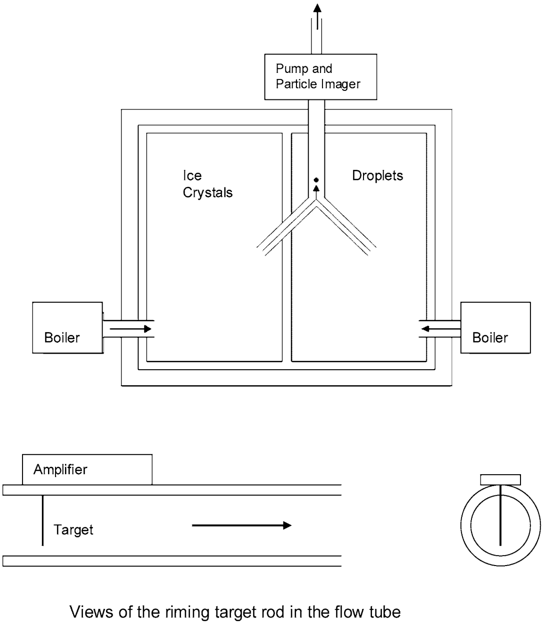


Fig. 12 Rimer charge sign boundaries from various laboratory studies. — Saunders et al. (2006), — · — Pereyra et al. (2000), · · · · · · Takahashi (1978), — — Saunders and Peck (1998)

Figure 12 shows laboratory results from Saunders et al. (2006) together with those from Pereyra et al. (2000), Takahashi (1978) and Saunders and Peck (1998). Plotted on these graphs are the critical values of temperature and cloud effective water content for which the charge transfer during crystal/graupel collisions is zero. The effective water content, EW, is made up of that portion of the cloud water droplet spectrum that by virtue of the droplet size, graupel velocity and the appropriate collision efficiency, strike the graupel pellet. The lowest of the charge sign reversal lines shown in the figure was obtained in experiments in the Manchester laboratory with cloud chamber apparatus essentially similar to that in Fig. 7 (Saunders and Peck 1998). The differences between this line and the charge sign reversal line obtained by Takahashi (1978) are clear and prompted many laboratory studies aimed at resolving the reasons for the differences. The laboratory studies of Pereyra et al. (2000) provided the breakthrough—they mixed ice crystals into their droplet cloud just before the mixed cloud impacted on the riming target. The studies of Saunders and Peck (1998) however, were performed in a chamber in which the ice crystals grew in the same chamber as the droplets. The importance of the cloud conditions for particle growth was investigated in studies by Saunders et al. (2006) whose data line is shown in Fig. 12. Figure 13 shows their apparatus in which the ice crystals grew in a cloud of supercooled droplets, while the droplets that provided most of the rime ice to the target were grown separately. The two clouds were mixed briefly on their way to the target.

The relative growth rate hypothesis can help to account for the results that led to the range of results in Fig. 12, and in particular is consistent with the apparently conflicting results represented by the highest and lowest of the charge sign reversal lines, both sets being obtained in the same laboratory and cloud chamber in Manchester. According to the hypothesis, in order to strengthen the negative charging of a graupel pellet, the impacting

Fig. 13 The cloud chambers and target in which the ice crystals and supercooled droplets are grown in separate chambers. (Bax-Norman 2004)



ice crystals need to grow faster than the graupel ice surface. This is achieved in the case where the two clouds are mixed just before impact, because the crystals growing in a droplet cloud reduced the supersaturation in the chamber, while the other chamber containing the supercooled water droplets is at water saturation. On mixing, the ice crystals experience a higher value of supersaturation and so increase their growth rate leading to the high charge sign reversal line. In the case where the crystals grow in the same droplet cloud, their growth rate is appropriate to a mixed cloud and there is no surge of increased supersaturation when they approach the target. This leads to the lowest of the reversal lines shown on Fig. 12.

These results have relevance to thunderstorm conditions where mixing between cloud parcels having different histories will result in faster, or slower, growth of ice crystals. In other cloud situations, there is longer term stability of conditions and the low EW charge sign reversal line will apply.

Given that ice particles grow in supersaturated conditions, such as in cirrus clouds experiencing an updraught, charge transfer will occur during collisions between non-riming ice particles growing at different rates. The relative growth rate hypothesis predicts that the faster growing ice surface will charge positively. Laboratory measurements have confirmed substantial charge transfer in ice/ice collisions in the absence of supercooled droplets. This will be important in other planetary atmospheres where the liquid phase may not be present.

5 Concluding Remarks

The processes of thunderstorm electrification outlined here have all been considered, in their time, as contenders for the dominant mechanism by which terrestrial thunderstorms become sufficiently charged to produce lightning discharges. All the mechanisms result in charges being carried on cloud particles. The two processes generally acknowledged to be the most likely candidates are the process by which ice particles, growing at different diffusional rates, collide and share charges such that the particle growing fastest charges positively, and the inductive mechanism that relies on the pre-existing electric field to produce induced charges in uncharged particles that may be transferred during collisions.

The reason that these processes are viable is connected with the electrical relaxation time of the earth's atmosphere and the particles' dielectric and physical properties that permit charge transfers in the available contact time and allow the charges to remain on the particles long enough for regions of high electric fields to develop in the cloud volume. In other planetary atmospheres similar considerations must be made in order to identify likely charging processes leading to particle separation under the local gravity. All the processes considered here may well be active in other atmospheres.

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