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during riming**

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Measurements of electric charge separated during the formation of rime by the accretion of supercooled droplets

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In these experiments, the electric charge carried by single particles ejected from the surface of a graupel particle growing by riming was measured. Simulated graupel pellets were grown by accretion of supercooled water drops, at temperatures ranging from -2 to -10°C in a wind tunnel at air velocities between 5 and 10 m s^{-1} , with the goal of studying the charging of graupel pellets under conditions of secondary ice crystal production (Hallett-Mossop mechanism). The graupel, and induction rings upstream and downstream of the graupel, were connected to electrometers and analyzing circuits of sufficient sensitivity and speed to measure, correlate and display individual charging events. The results suggest that fewer than 1% of the ejected particles carry a measurable electric charge ($>2\text{ fC}$). Further, it was observed that the graupel pellets acquire a positive charge and the average charge of a single splinter ejected is -14 fC . This mechanism of ejection of charged particles seems adequate to account for a positive charge of around 1 pC that individual precipitation particles of mm-size could acquire in the lower part of the cloud, which in turn could contribute to the lower positive charge region of thunderstorms.

1 Introduction

Hallett and Mossop (1974) and Mossop and Hallett (1974) observed that ice splinters were ejected during the formation of rime by the accretion of supercooled droplets on a cylinder of diameter 2.4 mm at 3 m s^{-1} , providing that the air temperature is between -3 and -8°C . In addition, Mossop (1976) showed that the ice splinters are produced only when rime grows in a supercooled cloud containing water droplets with a spectrum that extends beyond $24\text{ }\mu\text{m}$ diameter.

The ejection of the splinters is thought to be produced when the supercooled droplets freeze on impact with the accreted rime. Choullarton et al. (1978, 1980) proposed that under suitable conditions, the freezing droplets form a shell of ice which can shatter

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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. Dong and Hallet (1989) proposed another mechanism; they suggested that the thermal gradients experienced by an accreted droplet can result in thermal shock and shattering of ice during freezing. Regardless of which mechanism is correct, the net effect is to produce a large number of small ice particles that could 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. Evidence that the Hallett-Mossop (H-M) mechanism is an important source of ice particles in clouds has been reported in field measurements by Hallett et al. (1978) in Florida cumuli and Harris-Hobbs and Cooper (1987) in cumuli in Montana, Florida and California.

Despite the evident importance of the H-M mechanism for secondary ice crystal production in clouds, there is only one work in the literature (Hallett and Saunders, 1979) that describes studies of the possibility that the ejected ice fragments were electrically charged and that they can contribute to thunderstorm electrification. Hallett and Saunders (1979) determined in an indirect way the electric charge of the ejected fragments by measuring the charging current to a riming rod moving through a cloud of super-cooled water droplets under conditions of secondary ice crystal production. The measurements were performed at -4°C with air velocities between 1 and 3.5 m s^{-1} . The authors concluded that the magnitude of the charges on the fragments was too small to be able to account for the observed electrification rates in thunderstorms. They observed 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 separated. The results of this study led to subsequent research of graupel charging during ice crystal collisions.

In this study we extend the laboratory experiments performed by Hallett and Saunders (1979) and provide direct measurements of the electric charge carried by single particles ejected from the surface of a simulated graupel growing by riming. The

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experimental evidence shows that the graupel pellets acquire a positive charge and the average charge of single splinter ejected is $-14 \times 10^{-15} \text{C}$. Further, it was observed that fewer than 1% of the ejected particles carry a measurable electric charge. The results obtained are used to evaluate the contribution that the ejection of charged particles can make to cloud electrification, in particular to the lower positive charge region.

2 Experimental

The experiments were carried out by using an open circuit wind tunnel which was assembled in a cold room of height 2.5 m and floor area $2 \times 2 \text{ m}^2$. Figure 1 shows the general layout of the apparatus. The experimental devices used in this work are similar to those used by Avila and Pereyra (2000), Pereyra et al. (2000, 2008), Pereyra and Avila (2002), and Bürgesser et al. (2006).

Riming occurs on a fixed brass cylinder of radius 2 mm (target), which is placed in the measuring tube perpendicular to the airflow. The measuring tube is a cylindrical tube of section 33 mm diameter connected to an air pump which controls the velocity of the droplets past the target.

The water droplets used to rime the target were generated by water vapour condensation from a boiler located inside the cold room; the boiler output fed a small cubic cloud chamber of 0.5 m side, whose lower vents allow the mixing of the cloud droplets with cold air. The cloud chamber is connected to the measuring tube through a vertical precooling tower ~ 2 m high allowing the cloud of droplets to reach ambient temperature before entering the working section. Figure 2 shows the histogram of the droplet size distribution used in the measurements; the spectrum extends to $60 \mu\text{m}$ with a mean diameter (d_m) $21 \mu\text{m}$. The cloud droplet spectra were obtained by taking cloud samples with a glass strip of 4 mm width covered with a thin film of 5% formvar solution. Several cloud samples were taken at the position of the target for different temperatures. It can be observed that 30% of the droplets had diameter larger than $24 \mu\text{m}$.

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The effective liquid water content (EW) is defined as the portion of the liquid water content (LWC) captured by the target on account of its collision efficiency, then EW depends on the droplet and collector size and their relative velocity. The effective liquid water content was experimentally determined by weighing the deposit of rime collected on the rod target (Δm) during a given time period (Δt) and then using the equation:

$$EW = \frac{\Delta m}{\Delta t V A} \quad (1)$$

where A is the cross-sectional area of the target, exposed to an air flux of velocity V . The mass of the rime was determined by using a balance with an error of 0.1%.

The air temperature was measured by using a calibrated thermistor placed in the wind tunnel downstream of the target. It was continuously monitored throughout the runs. The rime temperature was raised above the ambient temperature by the release of the latent heat of fusion, this temperature was calculated from EW using the Macklin and Payne (Macklin and Payne, 1967) equation, which considers the balance between the rate at which heat is released by the freezing droplets and is exchanged with the environment by forced convection and sublimation. The effects of the surface roughness on the heat balance equation were considered by using the corrections given by Avila et al. (1999) and Castellano et al. (1999). The speed of the airflow past the target was controlled by adjusting the power to an air pump and was determined by using a Pitot-tube type anemometer with an error of $\pm 0.5 \text{ m s}^{-1}$.

The electronic system for measuring and evaluating single events of charged particles ejected from the rimer was similar to that used by Avila et al. (2003) and consisted of electrometers connected to the target and induction rings placed upstream and downstream of the target, as shown in Fig. 1. The electrometers are sensitive charge amplifiers capable of detecting electric charges larger than 2 fC ($2 \times 10^{-15} \text{ C}$) and sufficiently fast to measure, correlate and display individual charging events. Examples of waveforms caused by an event of charge separation by ejection can be seen in Fig. 2 of Avila et al. (2003). The downstream ring was used to identify charged particles originating at the graupel; thus, a genuine event should show equal charges of opposite

sign on the two interacting particles, both charges were measured and correlated. The experiment was designed to exclude spurious charging events from analysis; in fact, charged particles entering the apparatus could be excluded from analysis by the signal from the upstream induction ring.

3 Results and discussions

According to Hallett and Mossop (1974) and Mossop and Hallett (1974) and subsequent work in this area, splinter production takes place at cloud temperatures between -3°C and -8°C ; for this reason, the range of temperature chosen for the present investigation spans -2°C to -10°C to ensure complete coverage of the range of temperature where the Hallett-Mossop mechanism is active. The liquid water content was also varied in a range which is characteristic of the conditions existing within real clouds.

Saunders and Hosseini (2001) conducted laboratory experiments in which simulated graupel pellets were moved on a rotating frame through a cloud of supercooled droplets to become covered in rime ice. Ejected ice fragments were counted after they grew to visible sizes in the cloud. They studied the effect of rimer velocity on the splinter production in the range $1.5\text{--}12\text{ m s}^{-1}$, and found that the maximum secondary ice particle ejection occurs at 6 m s^{-1} . The current experiments were performed at four different velocities around 6 m s^{-1} (5.3 , 6.8 , 8 and 9.4 m s^{-1}) in order to get the most favorable production of splinters.

Each one of the current experiments lasted 500 s and was started with the target clean of ice. The riming process occurred during the first 300 s and then the cloud droplet supply was cut off while the airflow remained for up to 200 s. Typically, the charged particles ejected from the target appeared after the first minute of starting the riming process and significant charge separation was observed only under riming conditions. Figure 3 shows the production of charged particles from the graupel as a function of time for a cloud temperature of $(-7.6\pm 0.2)^{\circ}\text{C}$, $EW=(0.61\pm 0.09)\text{ g m}^{-3}$ and velocity 6.8 m s^{-1} . Each bar represents the charge on an ejected crystal, which was mainly negative, as can be seen from the figure.

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One of the most remarkable results of this study is the low number of charged particles per *mg* of rime accreted found (~ 0.5 particles per *mg* rime) in comparison to the splinter production by the H-M mechanism as determined by several authors. For instance, the number of splinters produced per *mg* of rime accreted measured by Saunders and Hosseini (2001) is at least two order of magnitude larger than the number of charged splinters measured in this work. The most important difference between the present study and the earlier reports is that in the current study the accretion of supercooled droplets was produced on a fixed target, instead of a rotating one as used in previous work. However, there is no reason to assume that this difference in the measurement technique could be the cause of the difference between the results. Likely, most of the ejected particles by the H-M mechanism are too small to carry a detectable electric charge; only a few of them have suitable size to carry a charge on the order of 1 fC. Indeed, Bader et al. (1974) found evidence that sub-micron particles of ice are produced when water droplets freeze on riming, these splinters can be as small as 0.2 μm diameter and he also observed that appreciable numbers of small splinters are produced. The maximum charge that a liquid drop of 0.2 μm diameter may hold before disruption is around 0.5 fC as given by the Rayleigh Limit (Rayleigh, 1882); this magnitude of charge cannot be detected by the amplifier used in this study. Therefore, the results seem to indicate that only a small percentage of splinters produced by the H-M mechanism ($\sim 1\%$) are ejected with electric charge of magnitude above 1 fC.

Table 1 lists the values of the variables used in each run: ambient temperature (T_a), rime temperature (T_r), effective liquid water content (*EW*), velocity (*V*), number of charged particles produced in the run (*N*), total charge (*Q*) acquired by the graupel per minute and the percentage (%) of the ejected particles with negative charge. By considering all the experiments reported in Table 1, it was observed that the 93% of the ejected charged particles were negatively charged.

The results from Table 1 show that for $T_a > -6^\circ\text{C}$ the production of charged particles was substantially lower than that obtained for $T_a < -6^\circ\text{C}$. Secondary ice crystal production by the Hallett-Mossop mechanism has been observed at $T_a > -6^\circ\text{C}$ by many

authors, which confirms that these particles could be uncharged or with undetectable electric charge.

The ice particle production from freezing droplets is governed by the way in which they are accreted and also by the environmental conditions. As can be seen from Table 1, the number of charged particles produced per run was highly variable, indicating that the ejection of charged particles depends on microscopic processes that cannot be completely described by environmental variables such as T_a , EW and V . From the rime temperatures listed in Table 1, it is possible to discern, using the Macklin and Payne (1967) heat balance equation, that the graupel was in the dry growth regime in all experiments.

The results of all the experiments are illustrated in Fig. 4, where the charging rate of the graupel (Q) is plotted as a function of cloud temperature and the rate of rime accretion, $RAR=EW \times V$, which represents the mass of rime accreted on the graupel per unit time and unit surface area (Eq. 1). The error bars indicate the range of values which have been averaged in deriving the plotted points. Figure 4a shows, once again, that the charging rate of the graupel was very low at $T_a > -6^\circ\text{C}$; while for temperatures $-6^\circ\text{C} < T_a < -10^\circ\text{C}$ the charging rate increases as the temperature decreases. Figure 4b seems to indicate that the range $4 \text{ g m}^{-2} \text{ s}^{-1} < RAR < 8 \text{ g m}^{-2} \text{ s}^{-1}$ is the most suitable for the production of charged particles during the riming process.

Figure 5 displays the trend of the total charge acquired by the graupel (Q_t) during each run as a function of the number of charged particles produced in the corresponding run (N). Although an important dispersion of the data is shown in the graph, a linear trend is observed between the total charge and the number of ejected particles. If the total charge is directly proportional to the number of particles ejected, then

$$Q_t = qN \quad (2)$$

where q represents the average charge separated per event, which can be calculated from the slope of the fitted line in Fig. 5, giving a value of 14 fC. Since all the experimental data were used in this graph, the result suggests that q is roughly independent

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of T , EW and V . Therefore, the variations of Q with T_a and RAR observed in Fig. 4 are not consequences of the variations in q but the variations in N and the increase in charge in Fig. 4a is simply indicative of an increase in the number of charged particles ejected at lower temperatures and does not imply that multiplication itself increases at lower temperatures. Foster and Hallet (1982) reported a wider range of temperatures for multiplication, between -3 and -11°C .

There are few previous studies which have reported measurements of charge separation for ejection of particles from the surface of graupel growing by accretion of supercooled water droplets. For temperatures between -15°C and -30°C and velocities greater than 17 m s^{-1} , Avila et al. (2003) found that substantial electric charge separation can occur in droplet/graupel collisions. They noted the production of charged fragments with charges ranging from -300 to 500 fC . Indeed, the ranges of temperatures and velocities used in this study are far from the ranges where the H-M mechanism is certainly effective. Hallett and Saunders (1979) measured charge separation under conditions of secondary ice crystal production. Although they concluded that this mechanism is unsuitable to explain the observed electrification rates in thunderstorms, they estimated an approximate charge per ice particle ejected of 50 fC . Although, the magnitude of the charge is larger than the average charge per charged splinter obtained in the current work (14 fC), both are of the same order of magnitude. Likely, this charge has been overestimated in their study given that their estimates include various assumptions and inherent uncertainties, including the number of splinters produced by the charging rod and the ice crystal/graupel collisions produced by the secondary ice particles ejected from the experimental device used in the measurements. The advantage of the current work is that the charge of each single ejected particle was measured, thus the number and the charges of the splinters can be accurately determined.

The physical mechanism of multiplication was largely discussed by Chourlarton et al. (1978), Dong and Hallet (1989) and Saunders and Hosseini (2001). The physical mechanism responsible for the electric charge carried by the particles ejected from

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the graupel growing by riming is still a matter of discussion. Likely, the temperature gradient maintained between a freezing droplet ($\sim 0^{\circ}\text{C}$) and the graupel surface ($\sim T_r$) generates a net electric charge on the freezing droplet due to the different mobilities of H^+ and OH^- in ice (thermoelectric effect – Latham and Mason, 1961) with the colder ice surface becoming positively charged and the warmer freezing droplet negatively charged. Thus, it is possible that charge separation occurs when the freezing droplets fragment and a portion of droplet is ejected from the surface.

Values of the freezing and subsequent cooling times of the water droplets on an ice surface were calculated for various values of the ambient temperature and droplet radius by Macklin and Payne (1967). They estimated that the freezing times of water layers of $20\ \mu\text{m}$ thickness is between 10 and 50 ms for temperatures above -10°C . On the other hand, theory and experiment show that about 10 ms are required for the process of proton migration by temperature gradients (Latham and Stow, 1967). Both times are of the same order of magnitude; therefore, this mechanism, in principle, could be viable to account for the observed charge separation during the ejection process. However, more accurate estimations of these times are required in order to validate the mechanism.

It is important to note that maybe an important number of fragments ejected from the graupel growing by riming could carry electric charge with magnitude under the detection level. However, it is not possible to quantify this production so far.

4 Effects on cloud electrification

In order to evaluate the contribution that the ejection of charged particles can make to cloud electrification, we have estimated that the rimed surface of the artificial graupel capable of ejecting charged particle is approximately $132\ \text{mm}^2$. By taking a charging rate of $50\ \text{fC min}$, then, this mechanism is able to produce approximately a charging rate of $0.4\ \text{fC min mm}^2$ of graupel surface. Thus, soft hail of 1, 2 and 3 mm radius could be charged with rates of 5, 20, $45\ \text{fC min}^{-1}$ respectively; and in 30 min (which

is a characteristic time of residence of particles within the clouds) they could acquire charges of 0.15, 0.6 and 1.4 pC, respectively.

Although the values of the charges that soft hail pellets could acquire via this mechanism are relatively small, they are of the same order of magnitude as many of the charge on precipitation particles of *mm*-size reported in measurements made in real clouds. For instance, Mo et al. (2007) performed aircraft measurements of the electric field, and hydrometeor size and charge in a precipitation shaft beneath the base of a small convective cloud which was electrified. They were able to detect precipitation particles larger than 0.2 mm and charges greater than 0.5 pC and found that 70% of the particles carried charge greater than 0.5 pC, most of the particles carried charge between 0.5 and 10 pC and the maximum charge was 25 pC. They also found that 98% of the charged particles were charged positively.

Williams (1989) proposed that the electrical structure of thunderstorms can be represented as a vertical tripole consisting of three charge regions, an upper positive charge region, a midlevel negative charge region, and a lower positive charge region. The lower positive charge region plays a key role to enhance the electric field at the bottom of the main negative charge region, thus important lightning activity can be induced between negative and positive charge regions located in middle and lower parts of a thunderstorm respectively. Nag and Rakov (2009) suggested that the lower positive charge region could help the discharge of a negatively charged leader propagating downward; also the presence of a large amount of lower positive charge may avoid the occurrence of negative CG discharges.

The mechanism of ejection of charged particles could, in principle, explain the positive charge smaller than 5 pC acquired by individual precipitation particles of mm-size in the lower part of the cloud. By assuming that the undetected charged particles ejected from the graupel (charges <1 fC) are also negatively charged due to the temperature difference between the freezing droplet and the rime surface, they could contribute to the positive charge acquired by the graupel growing by riming. However, we cannot quantify this contribution so far.

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In addition, charges larger than around 5 pC found on the precipitation particles could also be acquired by collisions with other ice particles (ice crystals or other graupel pellets) via the non-inductive mechanism (Takahashi, 1978; Saunders et al., 1991, 1999, 2001; Saunders et al., 2006; Avila et al., 1995, 1996, 1998; Pereyra et al., 2000, 2008, etc.).

An important observation is that the sign of the charge acquired by the rime was positive in agreement with the sign of the charged precipitation particles found in the lower region of clouds. Bateman et al. (1999) suggested that the charge of the lower positive charge region should be produced almost entirely by precipitation particles. Thus, we can conclude that the current results suggest that the mechanism of ejection of charged particles under conditions of secondary ice crystal production is a viable mechanism to contribute to the formation of the lower positive charge region.

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Table 1. Values of the variables in each run: ambient temperature (T_a), rime temperature (T_r), effective liquid water content (EW), velocity (V), number of charged particles produced in the run (N), total charge (Q) acquired by the graupel per minute and the percentage (%) of the ejected particles with negative charge.

T_a (°)	T_r (°)	EW (g m ⁻³)	V (m s ⁻¹)	N	Q (fC min ⁻¹)	% (-)
-2.0±0.1	-1.2	0.59±0.07	8.0±0.5	0	0	
-2.1±0.2	-1.3	0.60±0.08	6.8±0.5	0	0	
-2.2±0.3	-1.7	0.33±0.04	8.0±0.5	0	0	
-2.4±0.2	-1.8	0.36±0.04	9.4±0.5	0	0	
-3.4±0.5	-2.1	1.03±0.1	6.8±0.5	0	0	
-3.7±0.5	-2.9	0.59±0.1	6.8±0.5	1	26	100
-3.8±0.5	-2.9	0.61±0.1	6.8±0.5	0	0	
-4.0±0.2	-3.5	0.29±0.1	6.8±0.5	0	0	
-4.3±0.3	-2.6	1.3±0.2	6.8±0.5	0	0	
-4.3±0.4	-2.4	1.3±0.2	9.4±0.5	0	0	
-4.4±0.8	-3.2	0.75±0.09	9.4±0.5	1	7	100
-4.6±0.3	-3.5	0.9±0.2	5.3±0.5	0	0	
-4.8±0.8	-3.8	0.63±0.09	8.0±0.5	3	11	100
-4.8±0.9	-4.1	0.43±0.07	5.3±0.5	0	0	
-5.0±0.3	-4.1	0.57±0.08	6.8±0.5	1	7	100
-5.0±0.4	-4.2	0.42±0.05	9.4±0.5	0	0	
-5.1±0.2	-4.6	0.23±0.03	8.0±0.5	0	0	
-5.2±0.8	-4.8	0.17±0.02	6.8±0.5	0	0	
-5.3±0.1	-4.9	0.18±0.03	5.3±0.5	0	0	
-6.0±0.7	-4.5	1.08±0.1	6.8±0.5	6	21	100
-6.2±0.5	-4.9	0.81±0.1	6.8±0.5	7	56	86
-6.2±0.5	-4.9	0.87±0.1	6.8±0.5	4	13	100
-6.3±0.4	-5.2	0.69±0.1	6.8±0.5	20	71	100

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Table 1. Continued.

T_a (°C)	T_r (°C)	EW (gm ⁻³)	V (ms ⁻¹)	N	Q (fC min ⁻¹)	% (-)
-6.7±0.5	-5.7	0.57±0.1	6.8±0.5	5	25	100
-6.9±0.4	-6.2	0.35±0.1	6.8±0.5	5	6	80
-6.9±0.6	-5.1	1.2±0.2	8.0±0.5	1	4	100
-7.1±0.5	-5.6	1.0±0.1	6.8±0.5	1	3	100
-7.1±0.5	-5.8	0.9±0.1	5.3±0.5	0	0	
-7.1±1.2	-5.7	0.8±0.1	9.4±0.5	2	11	100
-7.5±0.3	-6.3	0.7±0.1	8.0±0.5	5	27	100
-7.6±0.2	-6.5	0.61±0.09	6.8±0.5	32	102	97
-7.8±0.2	-6.9	0.55±0.09	5.3±0.5	1	5	100
-7.8±0.5	-6.9	0.40±0.05	8.0±0.5	17	58	100
-7.9±0.3	-7.0	0.41±0.05	9.4±0.5	8	28	88
-8.1±0.1	-7.4	0.28±0.04	6.8±0.5	11	30	100
-8.2±0.1	-7.6	0.23±0.04	5.3±0.5	5	9	80
-8.3±0.4	-6.4	1.3±0.1	6.8±0.5	20	38	70
-8.5±0.4	-7.3	0.7±0.1	6.8±0.5	11	97	91
-8.9±0.3	-8.3	0.24±0.1	6.8±0.5	6	32	84
-9.0±0.9	-7.1	1.2±0.2	8.0±0.5	6	10	67
-9.4±0.4	-7.9	0.9±0.1	6.8±0.5	19	57	85
-9.4±0.9	-8.2	0.62±0.07	9.4±0.5	17	59	88
-9.6±0.3	-8.0	1.1±0.2	5.3±0.5	10	31	100
-9.6±1.4	-8.0	0.9±0.1	8.0±0.5	15	96	100
-9.7±0.2	-8.5	0.64±0.09	6.8±0.5	39	123	95
-9.9±0.2	-9.1	0.40±0.06	5.3±0.5	10	34	100

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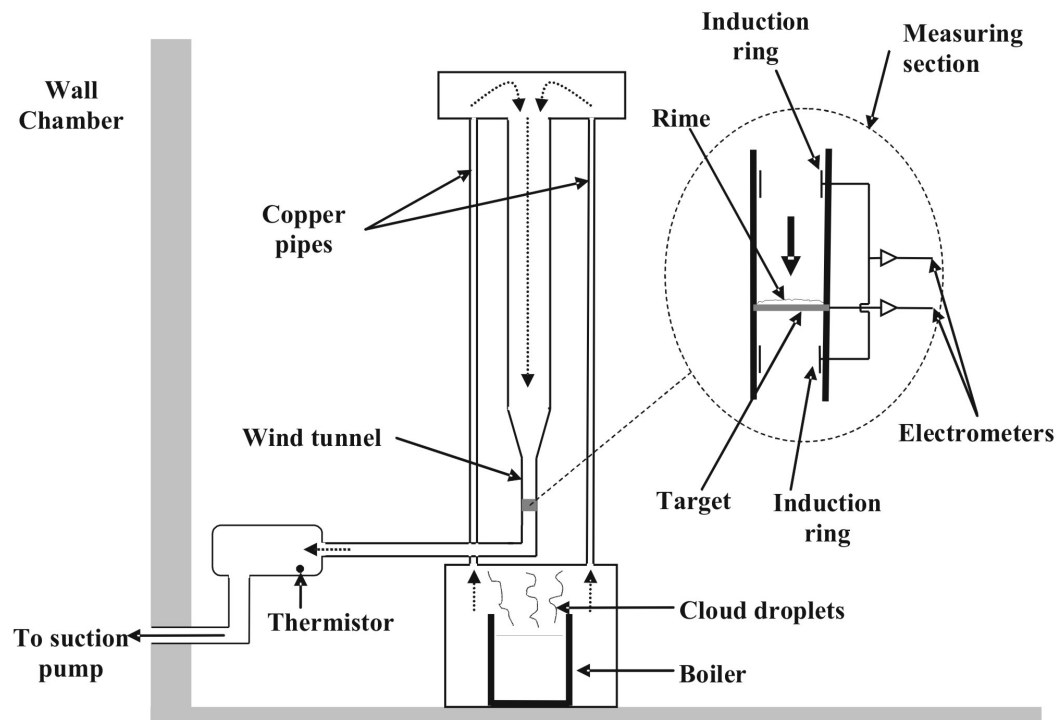


Fig. 1. General layout of the cloud chamber and the associated equipment used in this study.

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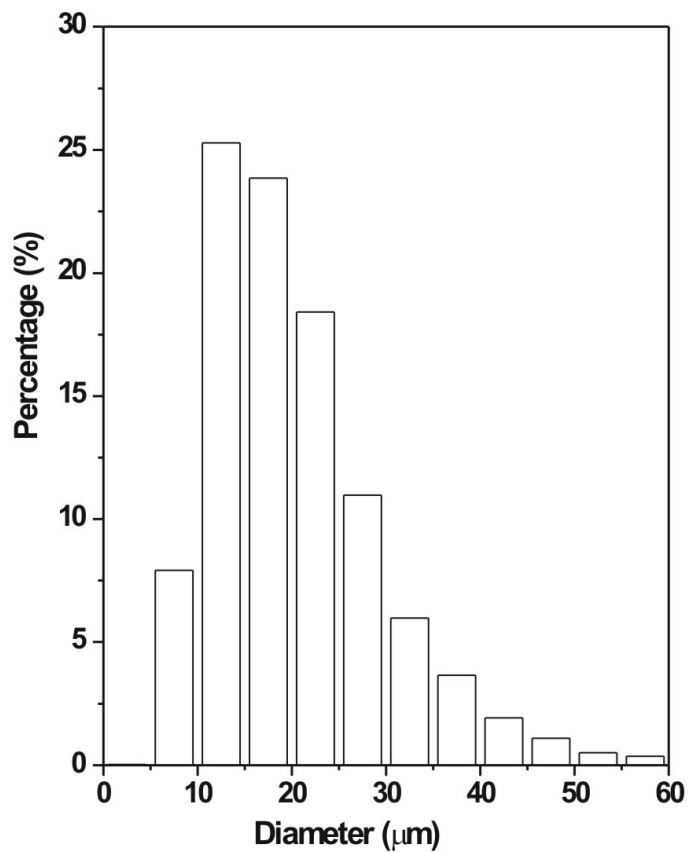


Fig. 2. Cloud droplet size distribution used in the experiments. The mean diameter is 21 μm .

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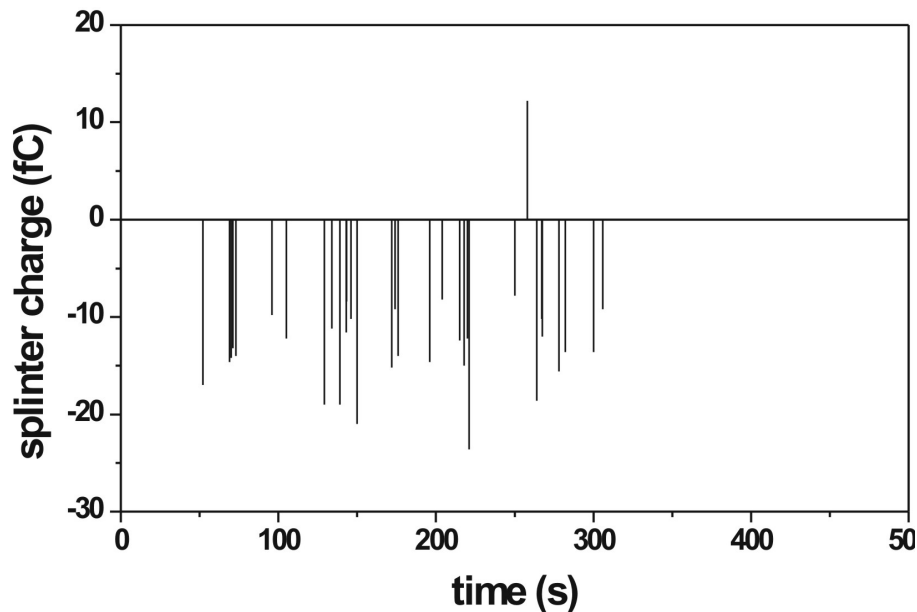


Fig. 3. Production of charged fragments during riming for a cloud temperature of $(-7.6 \pm 0.2)^\circ\text{C}$, $EW = (0.61 \pm 0.09) \text{ g m}^{-3}$ and velocity 6.8 m s^{-1} . Each bar represents the charge of one individual event.

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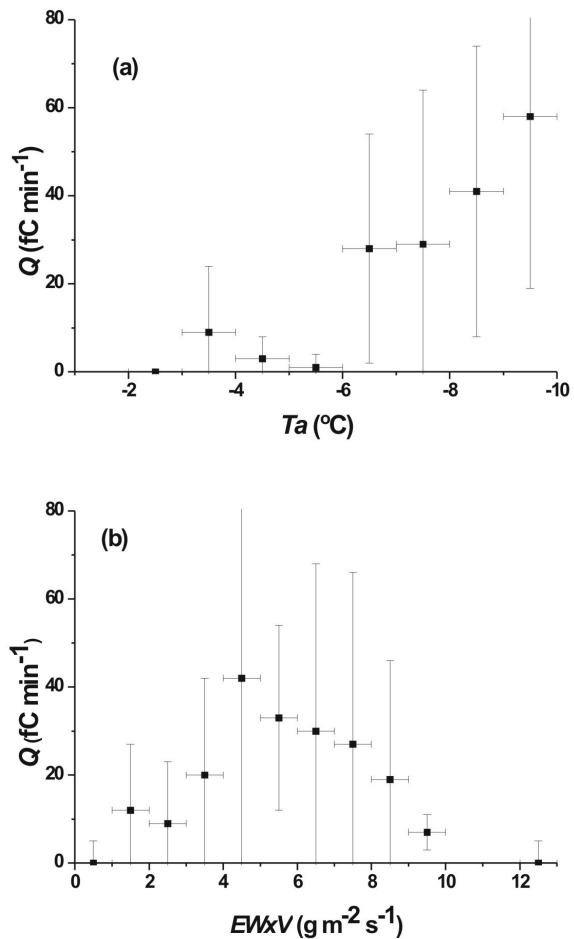


Fig. 4. Plots of the charging rate of the graupel (Q) as a function of cloud temperature and the rate of rime accretion.

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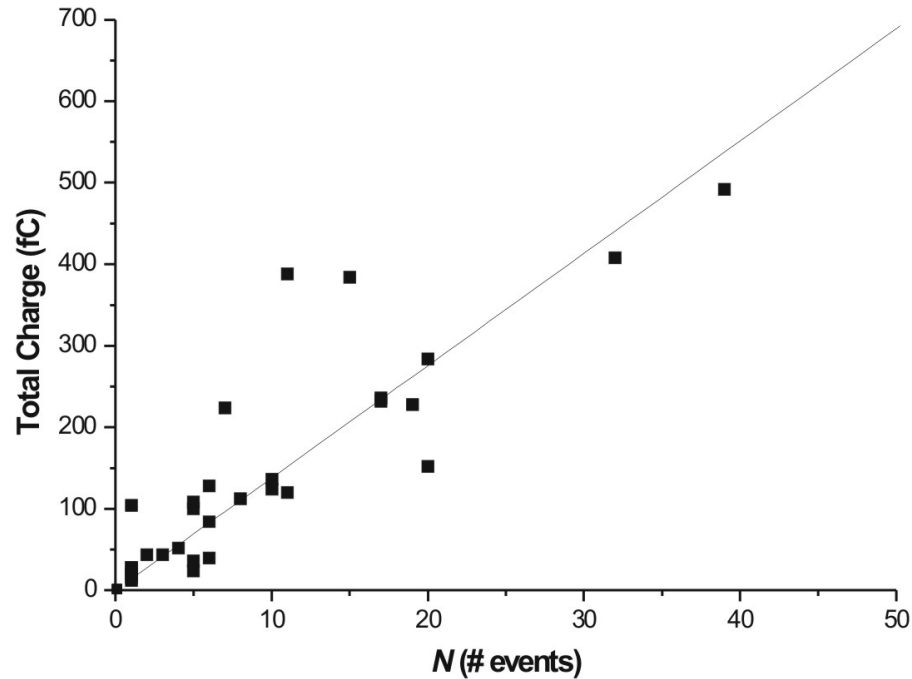


Fig. 5. Plot of the total charge acquired by the graupel (Q_t) as a function of the number of charged particles produced (N). The solid line represents the linear fit of the data.

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