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Laboratory experiments on the effect of organic acids and metals on the charge transfer during ice crystal – graupel collision under the conditions similar to stratiform clouds

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Abstract

Laboratory investigations have been carried out to find the sign and magnitude of charge transfer during the collision between ice crystals and graupel in the presence of supercooled water droplets inside the cylindrical steel chamber kept inside a walk-in
5 cold room, which can reach a temperature of -30°C . The experiments were performed with pure water (Milli-Q, 18.2 Megohms-cm) and with trace amounts of organic acids and metals at low Rime Accretion Rate ($<0.40\text{ g m}^{-2}\text{ s}^{-1}$) applicable to stratiform clouds in the temperature range -5 to -27°C . The ice crystals and supercooled droplets col-
10 lided with the graupel target at a constant impact velocity of 2.2 m/s. The presence of trace chemicals (organics or metals) saw significant increase in the magnitude of charge transferred per crystal collision to the graupel. These experiments therefore suggest that the presence of trace amounts of organics or metals in cloud water can significantly alter the electrical structure of stratiform clouds.

1 Introduction

15 Many laboratory studies of charge transfer involving ice crystal – graupel collisions in the presence of supercooled water droplets have followed from the pioneering work of Reynolds et al. (1957). They whirled ice spheres representing graupel pellets through a cloud of ice crystals and supercooled water droplets to measure the resulting charge transfer to the sphere and reported high negative charges on rimed graupel at about
20 -25°C . They were the first to note that droplets alone give no charge transfer, but for substantial charge transfer to occur with ice crystals, the presence of supercooled water droplets was essential. They found that contaminants (e.g. sodium chloride smoke) in the cloud in concentrations of about 10^{-4} molar in the rime transferred a negative charge to the rime. This was regardless of the relative amounts of water droplets and
25 ice crystals in the cloud. The maximum diameters attained by the crystals in the course of an experiment were about $100\text{ }\mu\text{m}$.

Brooks et al. (2006) measured rain events occurring between September 1996 and

May 1998 in Wilmington and found that formic and acetic acids are the most abundant organic acids comprising approximately 75% of the total molar quantities. The annual volume weighted concentrations of formic and acetic acids in the rainwater were 9.9 and 7.3 μM respectively.

5 Organic acids contribute significantly to the rainwater acidity in urban areas (Sakugawa et al., 1993; Kawamura et al., 1996; Pena et al., 2002) and 80–90% of the acidity in remote areas of the world (Andreae et al., 1988). Formic and acetic acids dominate precipitation acidity observed in polluted and remote regions of the world (Galloway et al., 1982; Keene and Galloway, 1986; Keene et al., 1983).

10 The combustion of fossil fuels and biomass are sources of organic acids and vehicle exhausts as well as smoke stacks emit these compounds (Kawamura et al., 1985; Talbot et al., 1988). The burning of leaves, hay, and wood also releases formic and acetic acid (Talbot et al., 1988; Hartmann, 1990). Sources of these acids include direct anthropogenic emissions (Kawamura et al., 1985; Grosjean, 1989; Talbot et al., 1988) and biogenic emissions (Andreae et al., 1988; Talbot et al., 1988, 1990; Keene and Galloway, 1986, 1988; Khare et al., 1997; Kumar et al., 1996).

15 Studies by Koulousaris et al. (2009) on precipitation over the Aegean show that Saharan dust storms supply significant amounts of natural Fe to the northern Aegean atmosphere. Anthropogenic emissions were largely responsible for the Cu concentrations in the precipitation. The concentrations of trace metals have been determined in airborne particles (Orlic et al., 1999) and biomass-burning impacted air masses (Muralidharan et al., 2000).

20 Rainwater samples collected during the peak monsoon period over Hyderabad, India show the presence of trace metals (Satyanarayanan et al., 2006). Prospero et al. (2001), Jaffe et al. (2003), Holmes and Miller (2004), have studied long distance transport of metals emitted to the atmosphere from anthropogenic and natural sources. Several studies exist on the concentrations of trace metals in atmospheric particles and rainwater in the western Mediterranean Sea (Chester et al., 1990, 1993; Migon et al., 1991, 1997; Guieu et al., 1993, 1997, 2002; Guerzoni et al., 1997, 1999; Luck and

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Othman, 2002; Sandroni and Migon, 2002). Nriagu and Pacyna (1988) reported increasing amounts of trace metals, particularly heavy metals into the environment as contaminants and pollutants.

5 Workman and Reynolds (1950) found that a large potential difference arises between the liquid and solid phases as dilute aqueous solutions are frozen. The sign and magnitude of the potential difference developed and the quantity of charge separated were functions of the kind and amount of contaminants dissolved in the water.

10 Takahashi's (1978) experiments show that not only the magnitude of the charge separated in ice crystal – graupel collisions but also the sign of the charge strongly depended upon the temperature and cloud liquid water content (CWC). At a temperature of -20°C without ice crystals in the chamber, the rime was slightly electrified negatively for a sodium chloride solution (10^{-4} N) and positively for an ammonium sulphate solution (10^{-4} N). With ice crystals present, a chamber temperature of -20°C and CWC of 1 g m^{-3} , the rime was strongly electrified negatively irrespective of the solution used.

15 Jayaratne et al. (1983) reported that the magnitude and sign of the charge transferred to the riming surface when ice crystals bounce off it in the presence of supercooled water droplets were functions of temperature, crystal dimension, relative velocity, liquid water content (LWC) and the impurity content of the riming target. They found the critical charge sign reversal temperature was about -20°C at a LWC of 1 g m^{-3} with the riming target charging positively at temperatures above the charge reversal temperature and negatively below this temperature. Igniting various materials in the cloud chamber shifted the drop size spectrum to smaller sizes with a maximum droplet diameter less than $10\text{ }\mu\text{m}$ and a modal diameter below $4\text{ }\mu\text{m}$. Introduction of NH_3 gas resulted in a strong positive charging of the graupel when the clouds were seeded to produce ice crystals at all temperatures.

25 Several researchers have conducted charge transfer experiments with ionic compounds like ammonium sulfate and sodium chloride. Jayaratne et al. (1983) and Jayaratne (1999) used an ultrasonic droplet generator (atomizer) to form a cloud of pure water and weak solution. At a liquid water content of 1 g m^{-3} and crystal size of $50\text{ }\mu\text{m}$

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the current to the rime was negative when the cloud was contaminated with NaCl solution (5×10^{-5} N and 2×10^{-5} N) at all temperatures with an impact speed of 3 m/s. When the concentrations were reduced to about 8×10^{-6} N, the rime acquired a positive charge at -5°C and a negative at -15°C . Jayaratne et al. (1983) and Jayaratne (1999) reported that with cloud contaminated with ammonium sulphate solution (2×10^{-5} N and 2×10^{-4} N) the rime charged positively. Increasing the concentration to 10^{-3} N the rime charged negatively above -11°C and positively below. They also observed that the sign reversal temperature increased with contaminants at concentrations found in cloud water.

Takahashi (1985) also reported positive electrical charges were separated on ice when ammonium sulphate solution (10^{-4} N) was removed from the ice surface at all temperatures. However when NaCl solution (10^{-4} N) was used the separated charges were positive at warmer temperature and negative at temperatures less than -11°C . The electrical sign of the separated charge is the same as that reported in the freezing potential experiments of Workman and Reynolds (1950).

The charge transfers were parameterized by Saunders et al. (1991) in terms of effective liquid water content (EW), ice crystal size and graupel/crystal velocity to use in numerical models of thunderstorm electrification. They showed positive charging of the graupel above the reversal temperature and negative charging below this. In addition, they found positive and negative charging regimes at temperatures below about -20°C and above about -16°C respectively at low values of EW.

Brooks et al. (1997) considered further the importance of the rime accretion rate (RAR) and pointed out that the crystal/graupel velocity, whose important effect on the magnitude of charging was identified in earlier work, has an additional importance through its contribution to the RAR (RAR is the product of EW and the crystal/graupel relative velocity). Thus, they had redrawn the positive and negative charging zones of Saunders et al. (1991) as a function of temperature and RAR. Experiments showed the validity of these conversions. Parameterization of charge transfer as a function of RAR replaced EW.

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Saunders et al. (2006) suggested that the most important influence on charge sign is the relative diffusional growth rate of the two ice surfaces at the moment of impact and that this is affected by an increase in cloud supersaturation experienced by the ice crystals during the cloud mixing process just prior to the collision.

Williams (1995) explained the three general categories of thunderstorms, stratiform precipitation and winter storms, ordinary thunderstorms and the super hailstorm in terms of the electrical, kinematic and microphysical cloud conditions. Stratiform clouds and winter storms are shallow with low liquid water content ($<0.2 \text{ g m}^{-3}$), low updraft speed ($<1 \text{ m s}^{-1}$), ice particle fall speed $1\text{--}3 \text{ m s}^{-1}$ and with a predominant negative dipole (with a layer of negative charge above a layer of positive charge) in the ice region. They also exhibit a prevalence of positive ground flashes.

The objective of the present work is to study charge transfer under conditions similar to those that occur in stratiform clouds (low RAR $<0.40 \text{ g m}^{-2} \text{ s}^{-1}$ and in the temperature range -5 to -27°C) and the early stages of convective clouds by using pure water (Milli-Q) and to see what changes occur when trace amount of organic acids and metals are added to the cloud.

2 Experiments

Experiments were carried out inside a steel chamber kept in a walk-in cold room ($2.4 \times 1.2 \times 2.4 \text{ m}$) which can reach a temperature of -30°C . Figure 1 shows the schematic representation of the experimental apparatus. The cylindrical steel chamber has a capacity of 238 litres. There are two aluminum tubes connected to the side of the chamber, one at 0.14 m from the top and the second one at 0.16 m below this top tube. Suction pumps connected to the tubes draw the cloud from the chamber. The flow speed is calibrated with an accuracy of 1 cm s^{-1} using a constant temperature linearized anemometer (TSI model 1051-2D).

A spherical glass flask (Borosil) of 500 ml was used to heat pure water and other chemical solutions at a controlled rate. This spherical glass flask was kept inside a

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semi-spherical heating mantel, which completely covers the lower half of the flask. The vapor enters the experimental chamber from the neck of the spherical glass flask through a teflon tube of 15 cm length and 2 cm diameter to form a cloud of supercooled water droplets and water vapor. The solution in the flask never exceeded three-quarters
5 of the capacity of the flask. The solution was only bubbling gently. The heating/boiling temperature of the solutions varied from +98 to +99.5 °C. Ice crystal formation in the cloud is initiated by momentarily introducing a rod dipped in liquid nitrogen. A platinum target plate of 18 mm×4 mm kept vertically inside the upper aluminum tube connects to the input of a charge amplifier. Graupel is grown on this platinum target plate.

10 The cloud of supercooled droplets and ice crystals from the steel chamber are drawn past the graupel grown on the platinum target at a constant speed of 2.2 m s⁻¹. Although the updraft speed in stratiform clouds is <1 m s⁻¹ the ice crystal fall velocity is between 1–3 m s⁻¹ (Williams, 1995). The maximum speed that our suction pump could deliver was 2.2 m s⁻¹, so this speed represents both stratiform cloud and the
15 early stages of cumuliform cloud. The target is cleaned and fresh graupel is made before each run. The cloud of supercooled droplets and ice crystals are drawn past the rime target 45 s after seeding. A charge amplifier measures the charge transfer to the graupel during the collisions with ice crystals. A data logger continuously collects the temperature of the cloud and the output of the charge amplifier.

20 Formvar coated slides having the same size as the platinum target plate are placed inside the lower tube, on which ice crystals and droplets are collected at an impact velocity of 2.2 m s⁻¹. These slides are used for the computation of RAR and microphysical analysis. RAR is calculated by using the equation:

$$\text{RAR} = \{(4/3) \times \pi \times r^3 \times N \times \rho\} / (A \times t) \quad (1)$$

25 Where r is the average radius of the supercooled droplets, N is the total number of supercooled droplets, ρ is the density of water, A is the area of the slide, and t is the exposure time of the slide.

3 Chemical analysis

Chemical analysis of the solution before boiling and that of the precipitation particles (cloud droplets/ice crystals) which fell to the bottom of the chamber was done to see if the chemicals in the solution were ending up in the precipitation. As gentle bubbling
5 happens, it is possible that bubble breaking at the surface would put involatile species into the particles through the mechanism that generates sea salt aerosols (Monahan et al., 1986; Woodcock et al., 1963).

The solution droplet entering the chamber can freeze thus retaining the salt. The solution drop can also evaporate in the chamber leaving a floating aerosol particle. An
10 ice crystal or droplet can capture this particle. Studies show that soluble gases and aerosols are incorporated into the cloud droplets or ice crystals through scavenging mechanisms (e.g. Pruppacher, 1980; Kalina and Puxbaum, 1994; Dixon et al., 1995; Oberholzer et al., 1992; Schwikowski et al., 1994; Baltensperger et al., 1994, 1998). Bell and Saunders (1995) have investigated the scavenging efficiencies of ice crystals
15 of sizes ranging from 35 to 150 μm and found the scavenging efficiencies vary from 0.2 to 0.9 for aerosol particles in the range 4 to 6 μm.

Table 1 gives the properties of the chemicals introduced in the solution. Chemical analysis of the precipitation particles collected in a glass beaker at the bottom of the chamber has been carried out using a Dionex DX 100 Ion Chromatographer and
20 Atomic Absorption Spectrometer (Perkin Elmer AAnalyst 400). About half the strength of the chemicals in the original solution ended up in the precipitation collected at the bottom of the chamber. It is possible that this strength is higher because of the scavenging of aerosols in the chamber by the ice crystals and droplets that form the precipitation. Analysis of the rime collected on the graupel target was not possible as the
25 sample volume was less for carrying out chemical analysis. Table 2 shows the details of the chemical analysis.

4 Experimental errors

The charge-detecting amplifier in the upper tube has a sensitivity of 5.442 ± 0.38 mv/pc.

The mixed phased of cloud was sucked through both the side tubes. Inside the upper tube, ice crystals collided with the target and bounced off; at the same time, a formvar coated microscope slide, the same size as the target, collected the ice crystals and supercooled droplets in the bottom tube. Calibration runs showed that the crystal number concentrations inside the cloud sucked through the upper tube was greater on average by 6.2% (with a modal value of 24%) leading to an overestimate of the charge per crystal.

The RAR was calculated from the number of supercooled droplets collected on the formvar slide in the lower tube and also by weighing the platinum target exposed to the cloud for a given time interval in the upper tube simultaneously. The error in estimating the RAR by the method of counting the droplets during the experiments could be determined. The cloud containing supercooled droplets was drawn past the target in the upper tube for a given time interval. The difference in the weight of the target before and after riming gave the RAR. The RAR obtained by weighing was greater than that calculated from the formvar slides with a mean value of +12.5%. The electronic precision balance (Sartorius Model BT224S) used for weighing the slides had an accuracy of 0.1 mg. RAR for the present experiments was calculated from the number of supercooled droplets collected on the formvar slide in the lower tube and the errors are not incorporated in these calculations.

Uncertainty in terms of percentage error in the measurement of ionic components varied between ± 0.5 and $\pm 3\%$, which are acceptable limits. The cloud temperature variation from the seeding time until the end of the run is less than 2°C .

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5 Results

5.1 Charging with pure water (Milli-Q)

The sign and magnitude of charge transfer during the collision between ice crystals and a fixed graupel target growing by riming was determined. There was no charge transfer when the cloud comprising only droplets was drawn past the target. A cloud comprising ice crystals and supercooled droplets was drawn past the stationary graupel/soft hail target 45 s after the cloud was seeded. The positive and negative charging of the riming graupel as a function of RAR, during the collision with ice crystals with an impact velocity of 2.2 m s^{-1} in the temperature range -7 to -25°C is shown in Fig. 2. The graupel charged positively in the temperature range -7 to -13°C . In the temperature range, -17 to -21°C the graupel charged both positively and negatively. The graupel charge was predominantly positive in the temperature region below -21°C . The maximum and minimum crystal sizes for all the experiments were 163 and $10 \mu\text{m}$ respectively with an average size of about $31 \pm 9 \mu\text{m}$ as observed from the formvar replicas. The droplet sizes ranged from 10 to $75 \mu\text{m}$ with an average size of about $20 \pm 4 \mu\text{m}$.

Figure 3 shows the results of an experimental run conducted with an initial cloud temperature of about -19.5°C . The graupel charged positively initially, reached a maximum, then the charging decreased and reversed sign from positive to negative with time. After reaching a negative maximum, it decreased and became slightly positive before reaching the steady state. The variation of the charge transfer with time is due to the changing cloud conditions. The vapor source was continuous throughout the experiments. The changes in cloud conditions or variations in charge transfer are not identical in all subsequent experiments. The arrows shown in Fig. 3 are the points where formvar coated slides were collected for the computation of RAR and for microphysical analysis. Figure 3 also shows the temperature variations during the experimental run.

Charge per crystal collision transferred to the graupel was calculated from the number of ice crystals collected on the formvar-coated slide at different points during the experimental run and the corresponding output from the charge amplifier. The area

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of the formvar-coated side is the same as that of the amplifier target assuming that the number of crystals hitting the target is the same as that collected on the formvar slide. Figure 4 shows the charge per crystal transferred to the graupel at low RAR ($<0.40 \text{ g m}^{-2} \text{ s}^{-1}$) as a function of temperature. The maximum EW with our present experiment is 0.18 g m^{-3} .

The sign of charge transfer at higher temperatures at 0.2 g m^{-3} agrees with the observations of the Manchester experiments (Saunders et al., 2006). In our experiments, we do not have data at lower temperatures at this EW for more comparisons. The data we have collected at lower temperatures are for very low EW.

5.2 Experiments with the presence of organic acids

5.2.1 Charging with formic acid [HCOOH]

Figure 5 shows the positive and negative graupel charging as a function of RAR during graupel – ice crystal collisions in the presence of supercooled droplets made from a solution ($3.6 \times 10^{-5} \text{ N}$) of formic acid in the temperature range -7 to -27°C . A cloud comprising ice crystals and supercooled droplets was drawn past the stationary target 45 s after the cloud was seeded. When the cloud comprised only droplets, no charge transfer was detected. In the temperature range, -7 to -13°C the graupel charged positively. The graupel charged both positively and negatively in the temperature range -18 to -27°C .

Figure 6 shows the charge per crystal transferred to the graupel at low RAR as a function of temperature. The magnitude of negative charge transferred at lower temperatures is higher than that seen in pure water. Whereas, the magnitude of positive charge transferred at higher temperatures remains the same as that of pure water. The maximum and minimum crystal size diameters were 113 and $10 \mu\text{m}$ respectively with an average size of about $33 \pm 10 \mu\text{m}$ as observed from the formvar replicas for all the experiments. The droplet diameters ranged from 10 to $56 \mu\text{m}$ with an average size of about $18 \pm 5 \mu\text{m}$.

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5.2.2 Charging with Acetic Acid [CH₃ COOH]

The graupel charging sign with a solution ($2.7 \times 10^{-5} \text{ N}$) of acetic acid as a function of temperature and RAR are shown in Fig. 7. The graupel charged both positively and negatively in the temperature region -5 to -8°C . In the temperature range, -9 to -20°C the graupel charged positively. Below -21°C only negative charge was transferred to the graupel.

The charge per crystal collision transferred to the graupel at low RAR as a function of temperature is shown in Fig. 8. As in the case of formic acid, the acetic acid also gave higher negative charging at lower temperature while the magnitude of positive charging remained at the same level as pure water at higher temperatures. The maximum and minimum crystal sizes were 100 and $10 \mu\text{m}$ respectively with an average size of about $27 \pm 5 \mu\text{m}$ as observed from the formvar replicas collected for all the experiments. The droplet diameter ranged from 10 to $50 \mu\text{m}$ with an average size of about $20 \pm 4 \mu\text{m}$.

5.3 Experiments with the presence of metals

5.3.1 Charging with Ferrous Sulphate [FeSO₄ 7H₂O]

Figure 9 shows the sign of graupel charging with a solution ($5 \times 10^{-5} \text{ N}$) of pure water and ferrous sulphate as a function of temperature and RAR. The graupel charged positively in the temperature region -6 to -12°C . In the temperature region -16 to -24°C the graupel charged both positively and negatively. The charge per crystal transferred to the graupel at low RAR as a function of temperature is shown in Fig. 10. At higher temperatures the positive charging is larger compared to that of water. The maximum and minimum crystal sizes were 138 and $10 \mu\text{m}$ respectively with an average size of about $37 \pm 8 \mu\text{m}$. The droplet diameters ranged from 10 to $50 \mu\text{m}$ with an average size of about $20 \pm 5 \mu\text{m}$.

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5.3.2 Charging with Cupric Sulphate [CuSO₄5H₂O]

Graupel charging sign with solution (5×10^{-5} N) of pure water and cupric sulphate as a function of temperature and RAR are shown in Fig. 11. The graupel charged positively in the temperature range -7 to -19 °C. In the temperature range, -20 to -25 °C the graupel charged both positively and negatively. The charge per crystal collision transferred to the graupel at low RAR as a function of temperature is shown in Fig. 12. The maximum and minimum crystal sizes were 100 and 10 μm respectively with an average size of about 27 ± 8 μm . The droplet diameter ranged from 10 to 50 μm with an average size of about 22 ± 4 μm . With trace amount of metals in the cloud water the positive and negative charging trends at all temperatures were the same as that of pure water. However, the magnitude of positive charge per crystal collision transferred to the graupel was twice that compared to water at higher temperature (≥ -12 °C). At lower temperatures (< -15 °C) the magnitude of positive charge transferred to the graupel per crystal collision was almost the same as that of pure water, but the magnitude of negative charge was almost double that of pure water.

All the results reported were reproducible. Table 3 shows the crystal and droplet size diameters for pure water and chemical solutions.

6 Discussion

Chemical analysis shows that there is dominant presence of organic compounds or metals in rainwater samples collected at various places. Our present experimental results with solution of pure water and organic acids (formic and acetic) showed that both formic and acetic acids produced a large amount of negative graupel charging at lower temperatures as compared to that of pure water. The trend and magnitude of positive charging at higher temperatures was the same as that of pure water. The charging experiments with a solution of metals (ferrous sulphate and cupric sulphate) showed that the positive and negative charging trends at all temperatures were the same as that

of pure water. However, the magnitude of positive charge per crystal collision transferred to the graupel was twice that compared to water at higher temperature. At lower temperatures the magnitude of positive charge transferred to the graupel per crystal collision was almost the same as that of pure water, but the magnitude of negative charge was almost double that of pure water.

Rime gets positively charged when temperatures are warmer than -10 °C (Magono and Takahashi, 1963; Takahashi, 1978; Hallett and Saunders, 1979; Gaskell and Illingworth, 1980; Jayaratne et al., 1983). Takahashi (1985) found that a positive electric charge separates on ice irrespective of the chemical impurities found in liquid water when the liquid water is removed from the ice surface at temperatures warmer than -10 °C. Takahashi (1983) attributed the negative electrification of rime at low temperatures to defect formation during ice fracture. The defects in ice provide a mechanism by which molecular dipoles may change their orientation (Hobbs, 1974).

The activation energy required to dissociate H^+ and OH^- ions in water is lower than that needed in ice and the mobility of the H^+ ion in ice is almost one order of magnitude higher than that of the OH^- ion in ice. Takahashi (1985) proposed that H^+ ions could only migrate from liquid water to ice when liquid water exists on an ice surface.

Our present results are consistent with the observations made by Takahashi (1985). Graupel gets charged negatively only at low temperatures for pure water and for clouds having trace amounts of metals and organics. The only exception is acetic acid where slight negative charge is observed at higher temperatures.

In the case of organics, the magnitude of positive charge separated is about the same as that of pure water. Only one H^+ ion is dissociated from both formic acid and acetic acid, which may explain why the magnitude of positive charge is the same as that of pure water. At lower temperatures, higher negative charge is transferred as compared to water. This may be because the negative ion in water is OH^- while that for formic acid it is COOH^- and for acetic acid it is CH_2COOH^- . The defects on the ice crystals or the higher negative charge transfer may be influenced because of the carbon associated with the negative ion of the organic compound.

Our results with cupric sulfate and ferrous sulfate show that the magnitude of both the positive and negative charge transferred to graupel is higher compared to that of water. There is no shift in the temperature regime with the sign of charge transfer. The positive ions involved in the metals are respectively Cu^{2+} and Fe^{2+} . In this case, the number of ions involved is higher as compared to H^+ in case of water. The conductivity of metals is several orders higher than that of pure water and could be the explanation for the observed higher magnitude of positive charge transfer. The electrical conductivity of copper is better than that of iron. This could be one of the possible reasons for higher positive charge transfer in the case of cupric sulfate as compared to ferrous sulfate. It may be noted that at 20°C the solubility of cupric sulfate is $31.6\text{ g}/100\text{ ml}$ (Material Safety Data Sheet) while that of ferrous sulfate is $25.6\text{ g}/100\text{ ml}$ (Material Safety Data Sheet). At lower temperatures, a marginally higher negative charging as compared to water is observed. In this case the negative ion is that of SO_4^{2-} as compared to the OH^- ion in the case of pure water. Although the ionic dissociation is more as compared to that of water, the increase in magnitude of negative charge transfer is not as high as seen for organics at lower temperatures.

7 Conclusions

Our experiments on charge transfer in cloud conditions and impact velocity applicable to stratiform and early stages of cumliform clouds show that the sign and magnitude of charge transferred is modified significantly by the presence of trace chemicals (metals or organics) in the cloud water. The magnitude of negative charge transferred to graupel per crystal collision at lower temperatures was five times that of water when trace amounts of organic acids were present in the cloud water. In the presence of metals in cloud water the magnitude of positive charge transferred per crystal collision at higher temperature was almost double and negative charge transfer at lower temperatures was twice as that of pure water. It is therefore possible that the presence of these chemicals in cloud water can significantly modify the polarity and electrical

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strength of stratiform clouds. In order to extend these results to cumulonimbus clouds the experiments need to be conducted at higher cloud liquid water content and impact velocities.

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Table 1. Chemical properties of compounds investigated.*

Name	Formula	Molecular weight (g/mol)	Density (g/cc)
Formic Acid	HCOOH	46.03	1.2
Acetic Acid	CH ₃ COOH	60.05	1.05
Ferrous Sulphate	FeSO ₄ 7H ₂ O	278.02	1.90
Cupric Sulphate	CuSO ₄ 5H ₂ O	249.68	2.286 ^a

* (Material safety Data Sheet, J. T. Bakers, ^aAlfa Aesar Data Book)

Table 2. Results of chemical analysis.

Samples	Chemical components (ppm)										
	F	Cl	NO ₃	SO ₄	NH ₄	Na	K	Ca	Mg	Fe	Cu
Mill-Q Before boiling	0.001	0.010	0.063	0.001	0.261	0.128	0.044	-0.222	0.190	0.012	0.021
Mill-Q Precipitation (droplets/ice crystals)	0.001	0.075	0.020	0.059	0.189	0.123	0.176	0.360	0.033	0.023	0.042
FeSO ₄ 7H ₂ O Solution before boiling	0.000	0.137	0.184	6.104	0.394	0.313	0.292	0.238	0.012	5.024	0.112
FeSO ₄ 7H ₂ O Precipitation (droplets/ice crystals)	0.000	0.283	0.208	3.362	0.208	0.338	0.395	0.278	0.020	2.157	0.050
CuSO ₄ 5H ₂ O Solution before boiling	0.197	0.096	0.000	4.648	0.176	0.443	0.173	0.112	0.210	0.042	4.052
CuSO ₄ 5H ₂ O Precipitation (droplets/ice crystals)	0.000	0.192	0.000	2.185	0.486	0.085	0.417	0.282	0.023	0.112	1.981

Table 3. Crystal and droplet size diameters for pure water and chemical solutions.

Solutions	Crystal diameter (µm)			Droplet diameter (µm)		
	Max	Min	Avg	Max	Min	Avg
Milli-Q	163	10	31±9	75	10	20±4
Formic Acid	113	10	33±10	56	10	18±5
Acetic Acid	100	10	27±5	50	10	20±4
Ferrous Sulphate	138	10	37±8	50	10	20±5
Cupric Sulphate	100	10	27±8	50	10	22±4

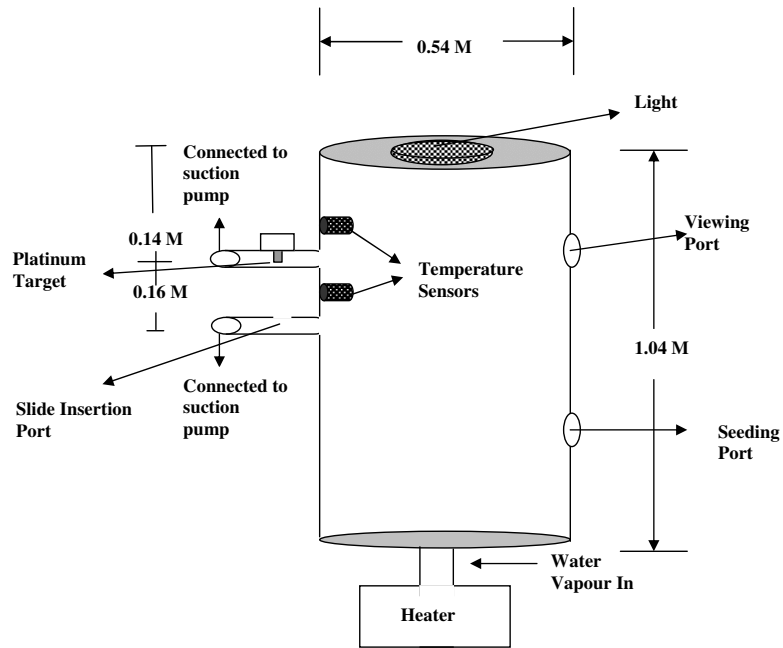


Fig. 1. Schematic representation of experimental apparatus.

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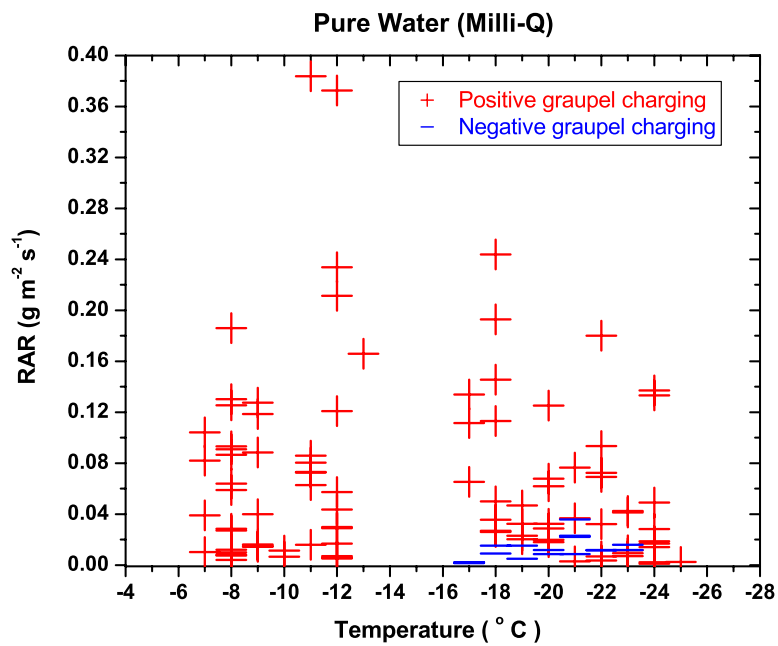


Fig. 2. The positive and negative graupel charging as a function of temperature and RAR for pure water.

270

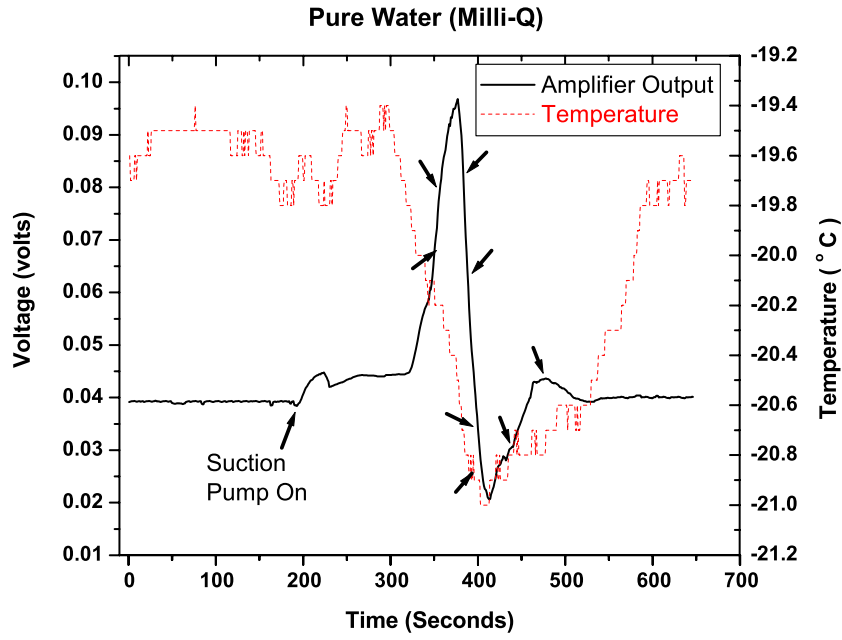


Fig. 3. Experimental runs showing the output of the charge amplifier for pure water with an impact velocity of 2.2 m/s.

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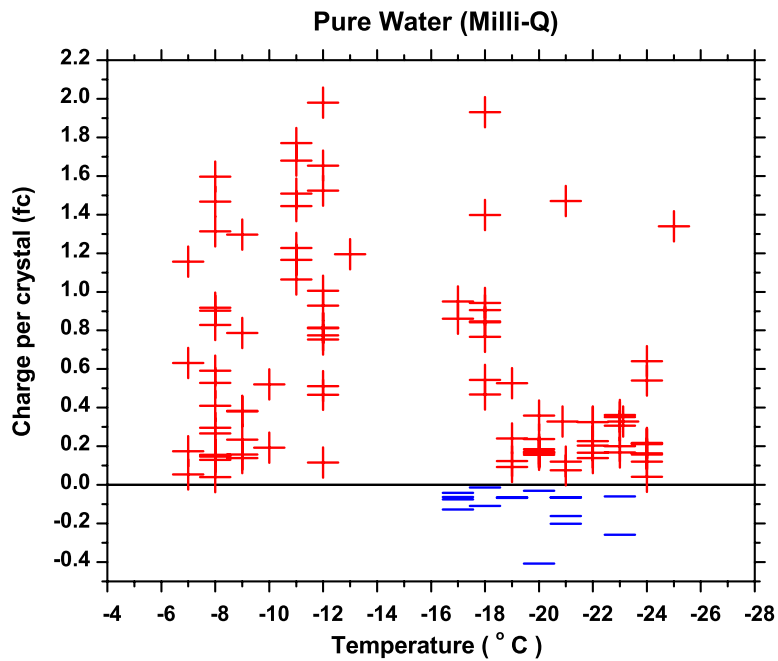


Fig. 4. Ice crystals interacting with graupel made from pure water as a function of temperature at low RAR ($<0.40 \text{ g m}^{-2} \text{ s}^{-1}$).

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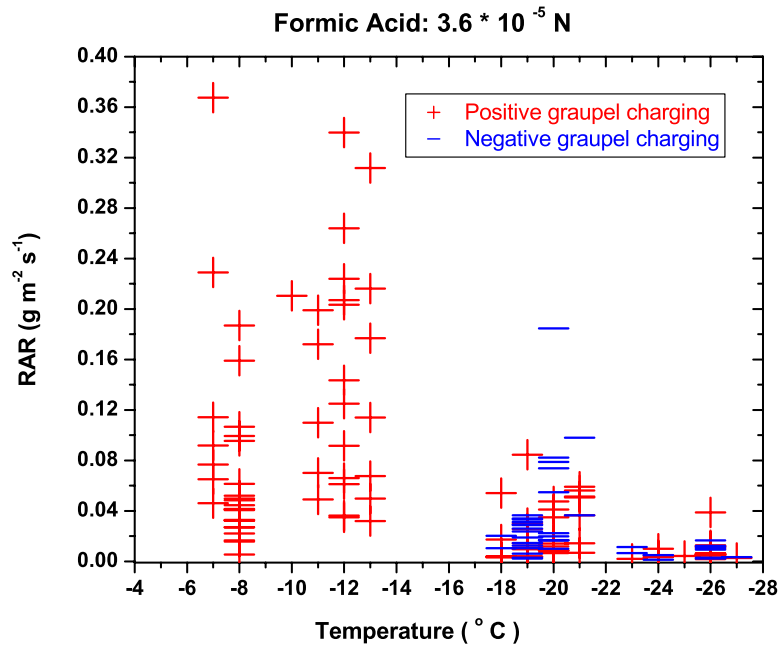


Fig. 5. The positive and negative graupel charging as a function of temperature and RAR for formic acid solution of 3.6×10^{-5} N.

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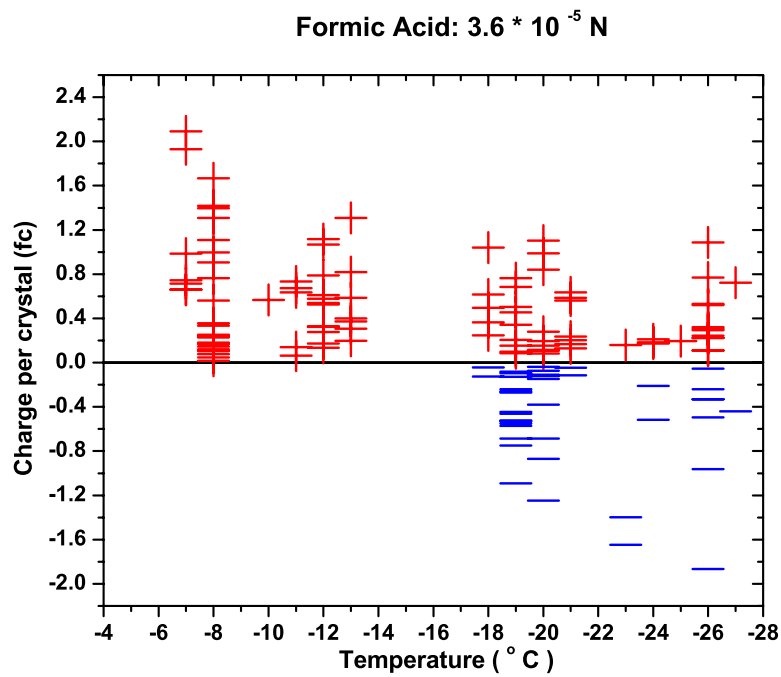


Fig. 6. Ice crystals interacting with graupel made from formic acid solution of 3.6×10^{-5} N as a function of temperature at low RAR ($< 0.40 \text{ g m}^{-2} \text{ s}^{-1}$).

274

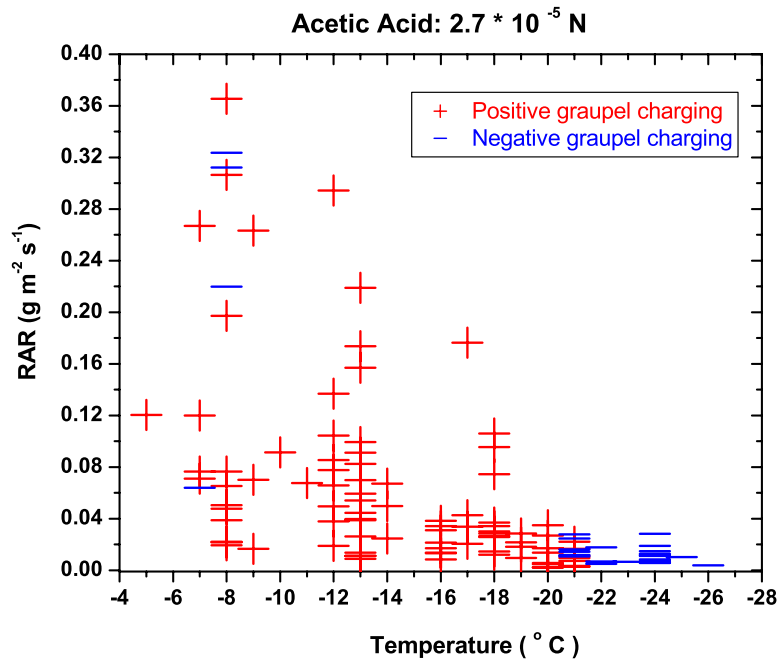


Fig. 7. The positive and negative graupel charging as a function of temperature and RAR for acetic acid solution of 2.7×10^{-5} N.

275

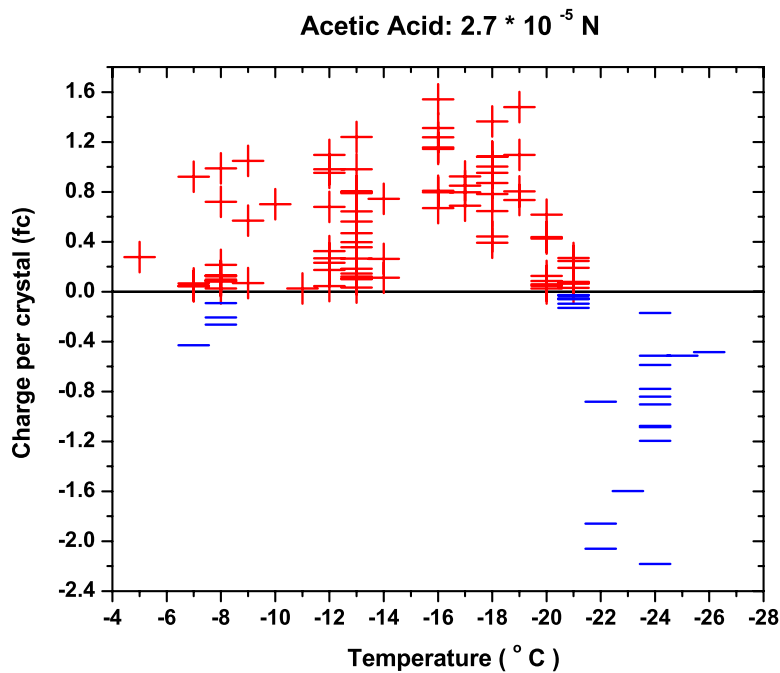


Fig. 8. Ice crystals interacting with graupel made from acetic acid solution of 2.7×10^{-5} N as a function of temperature at low RAR ($< 0.40 \text{ g m}^{-2} \text{ s}^{-1}$).

276

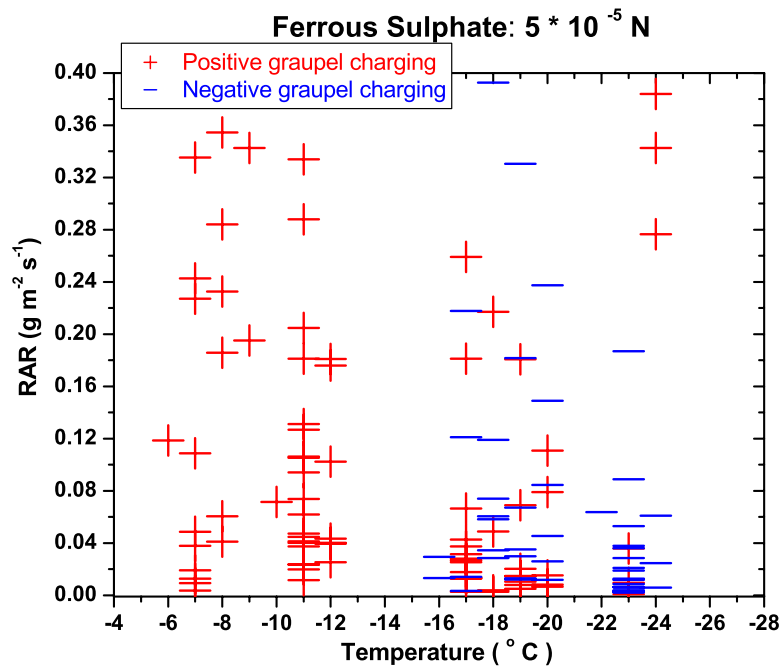


Fig. 9. The positive and negative graupel charging as a function of temperature and RAR for ferrous sulphate solution of 5×10^{-5} N.

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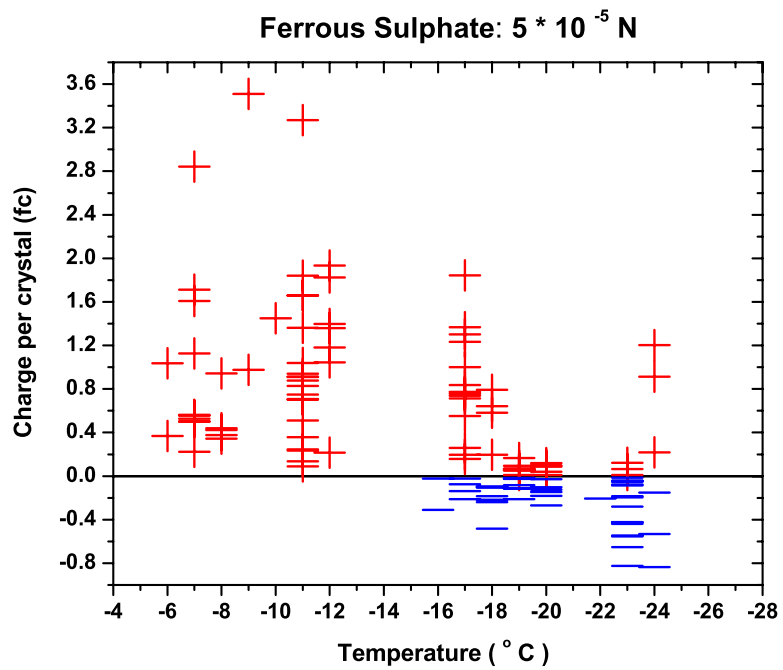


Fig. 10. Ice crystals interacting with graupel made from ferrous sulphate solution of 5×10^{-5} N as a function of temperature at low RAR ($< 0.40 \text{ g m}^{-2} \text{ s}^{-1}$).

278

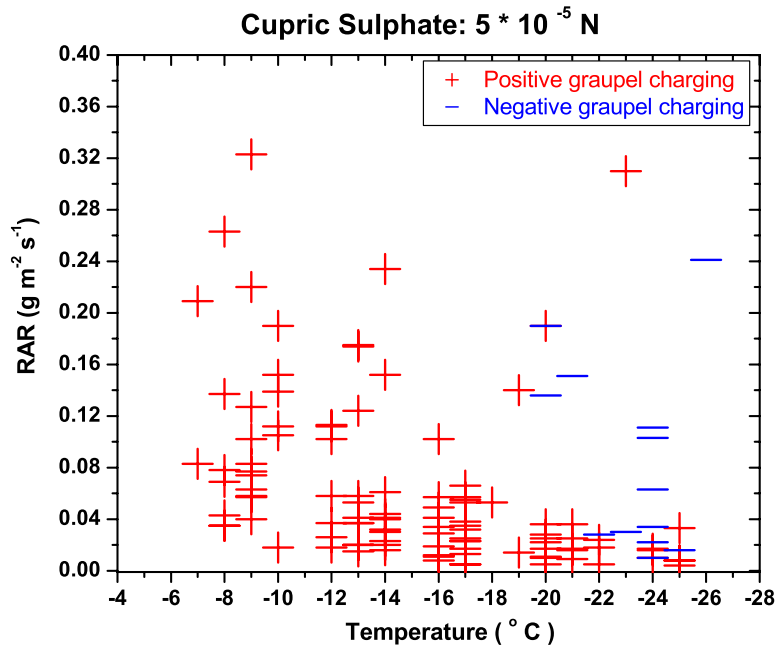


Fig. 11. The positive and negative graupel charging as a function of temperature and RAR for cupric sulphate solution of 5×10^{-5} N.

279

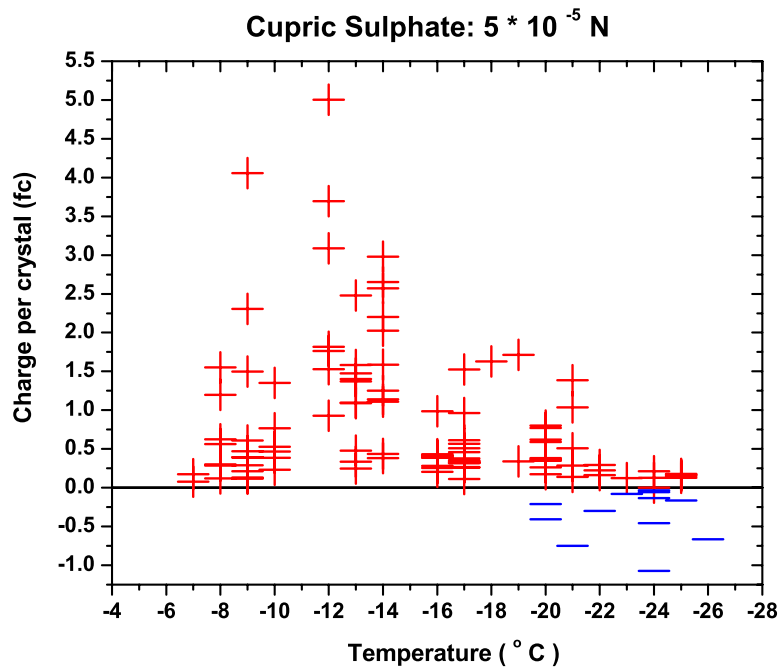


Fig. 12. Ice crystals interacting with graupel made from cupric sulphate solution of 5×10^{-5} N as a function of temperature at low RAR ($< 0.40 \text{ g m}^{-2} \text{ s}^{-1}$).

280