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6. AUTHOR(S)

Dr. Clive Saunders

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

UMIST
Dept of Pure and Applied Physics
PO Box 88
Manchester M60 1QD, UK

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To: Janet Johnston
Department of the Air Force
Air Force Office of Scientific Research (AFMC)
European Office of Aerospace Research and Development
223/231 Old Marylebone Road
London, NW1 5TH, UK

From: Clive Saunders
Pure and Applied Physics Department
University of Manchester Institute of Science and Technology
Manchester, M60 1QD, UK

Phone 44 61 200 3909
Fax 44 61 200 3941

Email Clive.Saunders@UMIST.AC.UK

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A Laboratory Study into the Charging of Ice in Thunderstorms

Abstract

In the laboratory, the interactions of ice crystals with rime coated targets provides a simulation of the charging of graupel pellets in thunderstorms. The experimental techniques used in previous studies have come under criticism and this study examines the importance of the precise conditions used in the laboratory measurements. The procedure used to initiate ice crystals influences the crystal sizes and concentrations and it was expected that this would, in turn, influence the charge transfers. However the largest problem to be shown up by the present work is in the measurement of the liquid water content in the cloud. Also, questions had been raised about the effect of rime density on the charging, but our studies of the effect on charging of increasing the rime density to its maximum possible value, show no influence of density on the sign of the charge transferred.

Preamble

There has been a lengthy and detailed discussion between two schools of laboratory experimentalists in the field of thunderstorm electrification. Both groups maintain that their own experiments are valid representations of thunderstorm conditions and yet there are important differences between their results.

The intent here is to set up apparatus to test the hypothesis that the precise details of the method of making these charge transfer measurements is crucially important in controlling the results. The first step has been to do experiments that follow exactly the procedures described by each research group. Can both sets of results be duplicated? If so, then the reasons for the discrepancies between the two studies can be examined by changing the cloud conditions in a controlled fashion.

Background to the study.

Reported differences between the results of various laboratory simulations of the charging of riming graupel pellets during ice crystal collisions continue to confuse the issue of thunderstorm charging processes and mechanisms. Theories to account for the nature of the observed charge transfers and their dependence on temperature and liquid water content are still under active debate. Some depend on surface potentials caused by riming while others involve charges on dislocations in the ice structure. A liquid like layer (Baker and Dash, 1992) carrying negative charge on the ice particle surfaces is often invoked - the layer has a thickness that is dependent on surface growth rate. The two interacting surfaces (crystals and graupel) have different growth rates, controlled by droplet accretion rates, local heating and local vapor transfer, leading to the observed charge transfers. These theories are still controversial, for example, even the sign of the charge layer in the liquid like layer is disputed. The mechanism itself is not at issue in the present work; details of possible charging mechanisms are given by Saunders (1992).

There are two recent sets of laboratory experiments involving the study of charge transfer during graupel interactions with smaller ice particles that are the subject of this study, one study was performed in UMIST, Manchester and the other in Hawaii. A list of such experiments including the two under current debate is given below.

A catalogue of charging experiments

A summary of experiments involving ice crystal collisions with a riming target.

Reynolds, Brook and Gourley (1957). 4 mm ice sphere at 8.4 m/s. Liquid Water Content (LWC) 0.25 to 4 g/m^3. Crystals up to 100 μm diameter. At -25°C, rimer charges negatively; with high ice crystal concentration, rimer charges positively.

Church (1966). Similar method to Reynolds, Brook and Gourley. At -15°C with a low LWC, rimer charges negatively. Increase LWC, rimer charges positively.

Magono and Takahashi (1963). Charge transfer dependence on LWC and temperature for ground-up snow particles (500 μm) interacting with a stationary riming target rod at 2 m/s. LWC up to 4 g/m^3. Positive rimer charging at temperatures above -5°C at 2 g/m^3, also positive at lower LWC at lower temperatures. Negative rimer charging elsewhere.

Takahashi (1978). Moving ice covered rods rods at 9 m/s. LWC up to 20 g/m^3. Crystals 10 to 100 μm. Charge transfer dependence on liquid water content and temperature

(See Fig 4). Above -10°C the rime charges positively; at lower temperatures, the rime charges negatively for liquid water contents between 0.2 and 4 g/m^3.

Hallett and Saunders (1979). Moving iced rods at speeds up to 3.5 m/s. Crystals up to 200 μm. Positive charging noted at temperatures above -18°C.

Jayaratne, Saunders and Hallett (1983). Same apparatus as Hallett and Saunders. Dependence of charging on velocity and crystal size. Positive rimer charging above reversal temperature, negative below; reversal temperature dependent on cloud LWC. Increasing LWC moves reversal temperature to lower temperatures.

Jayaratne and Saunders (1985). Importance of droplet sizes noted. With droplets too small to collide efficiently with the target, it charges negatively, while, for the same LWC but with larger droplets that hit the target, it charges positively. Subsequent UMIST work therefore quotes effective liquid water content, EW, rather than LWC.

Keith and Saunders (1990). Extension of the above work (JSH 1983) to larger crystals (800 μm) with moving and stationary targets. Crystal size dependence found. Velocity dependence, for constant rate od rime accretion, determined.

Saunders, Keith and Mitzeva (1991). KS 90 extended with a study of the effect of cloud liquid water content on charging. Equations derived giving values of charge transfer as a function of crystal size, velocity, temperature and Effective Liquid Water Content. Figure 2 shows a schematic of the charge sign regimes.

Saunders and Brooks (1992). Crystal interactions with a riming target experiencing wet growth - no charge transfer when the crystals stick to the target.

The experiments summarised above all use a temperature controlled cloud chamber into which water droplets are introduced to form a cloud. Ice crystals are, usually, artificially nucleated by dry ice or liquid nitrogen and the crystals grow at the expense of the water droplets. Charge transfer is studied when the crystals and droplets collide with a riming target which may be stationary while the cloud is drawn past; alternatively, riming rods, or spheres, move through the cloud on a rotating frame. The riming target is connected to a charge detector and from a knowledge of the ice crystal concentration (and collection efficiency) the charge transfer per crystal collision (or per crystal separation) is determined. All the studies agree that droplets alone produce no charge transfer at interaction speeds of a few meters per second. At speeds above about 10 m/s there is evidence of positive target charging associated with droplet splashing. The studies also agree that a cloud consisting of ice crystals alone gives negligible charge transfer during collisions with an ice target; the presence of both supercooled droplets and ice crystals is required for significant charge transfer.

Experimental techniques

Laboratory simulations of thunderstorm conditions in the UMIST cloud chamber have provided information concerning the charging of ice particles in thunderstorms. The work has also been extended to the study of the charging of aircraft surfaces by the collisions of ice particles. The cloud chamber (Figure 1) is inside a temperature controlled cold room capable of achieving temperatures down to -40°C. Thunderstorm conditions are obtained by introducing water droplets from a boiler or from ultrasonic atomizers. The droplets supercool and the ice phase is then introduced artificially using a thin metal wire pre-cooled to liquid nitrogen temperature. The ice crystals grow in the droplet cloud at the expense of the droplets. By controlling the length of nucleation time and the droplet input rate, clouds having various liquid water contents, drop size distributions, crystal sizes and concentrations may be achieved. Large ice crystals, up to 800 μm diameter, can be grown by levitating the cloud by means of low speed vertical airflow through the cloud chamber.

Charge transfer experiments are carried out when the crystal cloud is drawn past a stationary ice target at 3 m/s. The targets are connected to electrometers so that, with a knowledge of the crystal concentration, the charge transferred per crystal collision may be determined. We have also studied the separation probability following ice crystal collisions so that the charge transfer per crystal separation event can be calculated.

The results of some of these studies are summarised in Figure 2. At a given temperature,

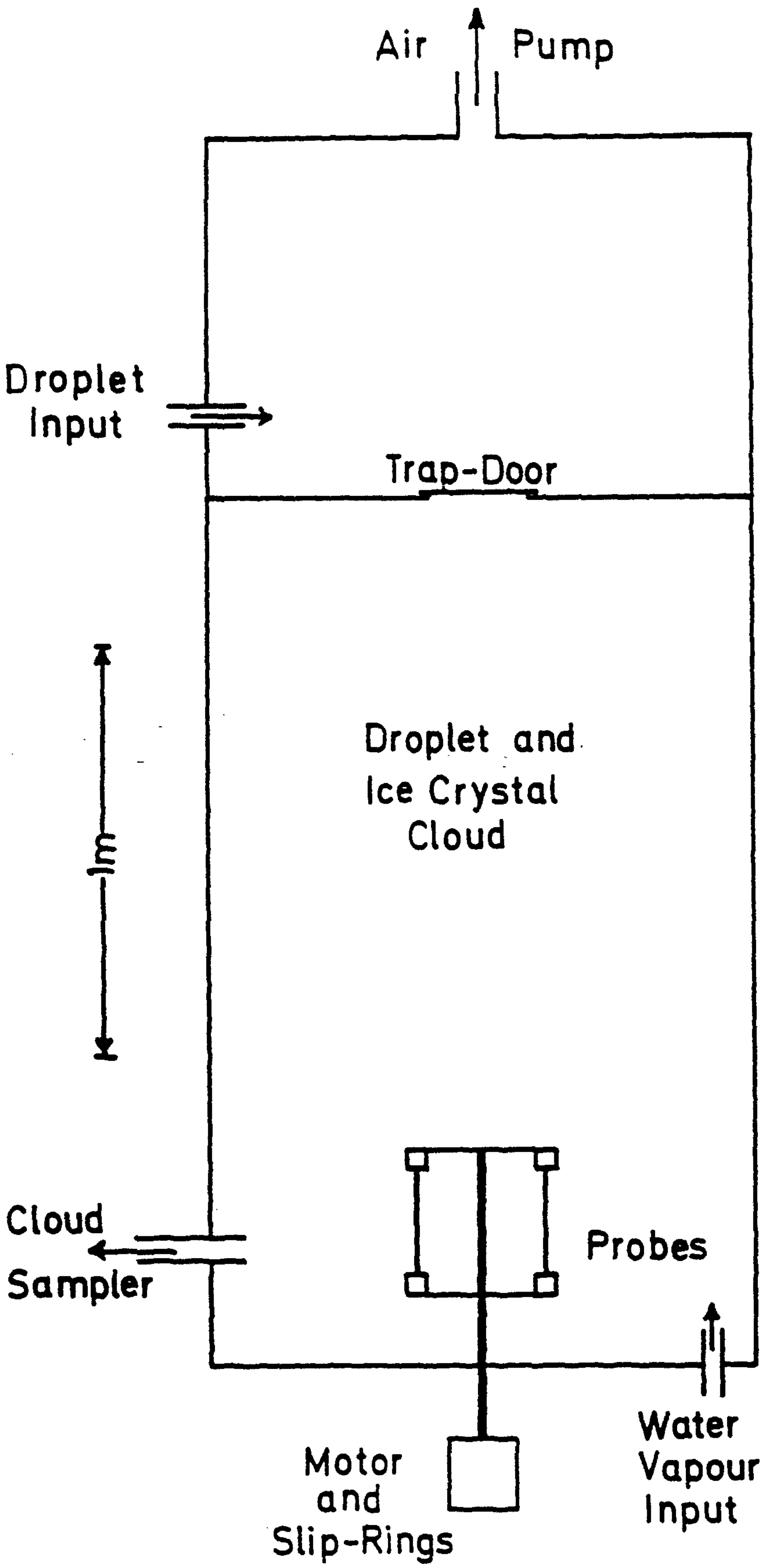


Figure 1. The cold chamber in which thunderstorm conditions are simulated.

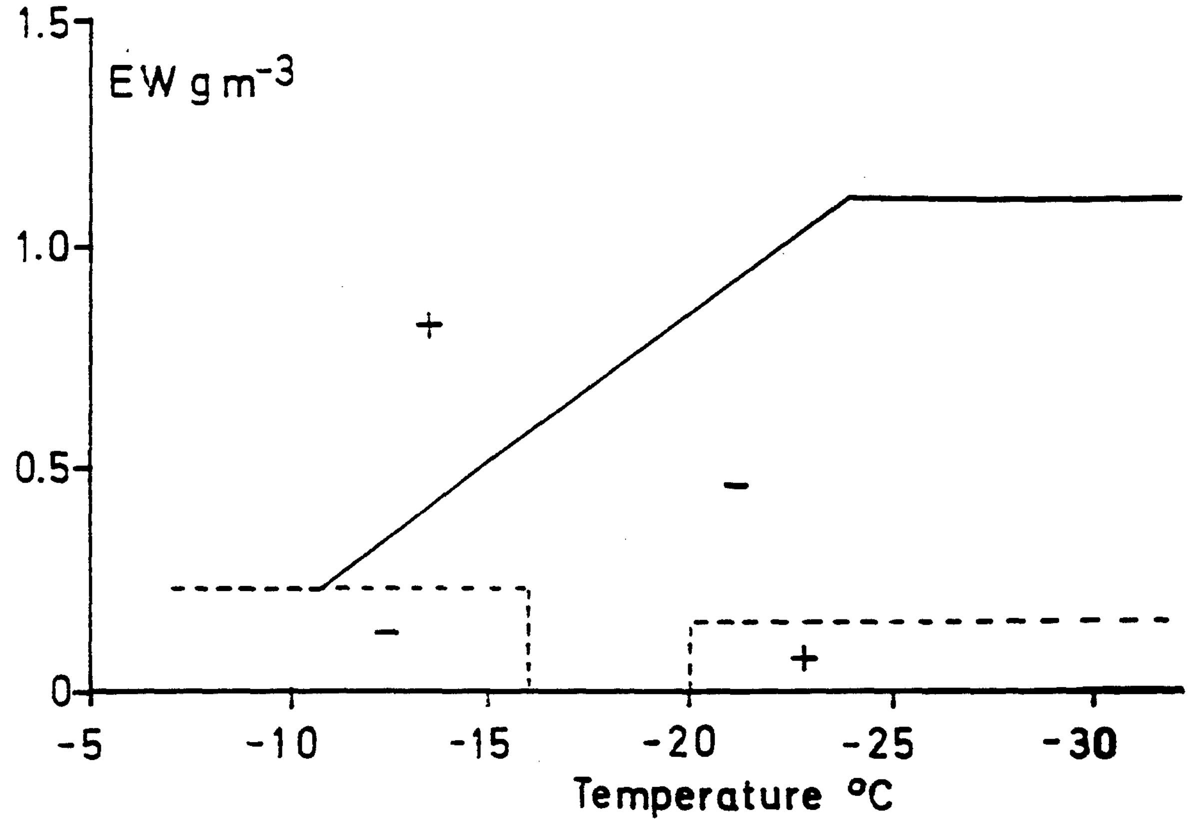


Figure 2. Rimer Charging according to Saunders, Keith and Mitzeva, 1991.

with charge transfer due to ice crystal interactions, increasing the liquid water in the cloud can cause the sign of the charge to a riming target to change sign. Similarly, charge sign reversal occurs at lower temperatures for a given value of cloud water content. Thus the diagonal line in the figure represents "Charge Sign Reversal".

A picture of thunderstorm charging can then be built up relying on the charge reversal effect as shown in Figure 3. Thunderstorms have a vertical charge dipole with positive charge in the upper regions and a negative charge center lower down. Lightning is initiated either within the cloud or from the negative center to ground when the electric field has developed sufficiently to cause breakdown. Successive charge transfer events between ice crystals and riming graupel pellets lead to electric field development. At low temperatures (typically around -15°C), the graupel pellets charge negatively and fall against the updraft to form the lower negative charge center. The positively charged ice crystals are carried aloft to form the upper positive charge center. At higher temperatures (typically above -10°C), the charge generation process is reversed and so negative crystals are carried up to re-inforce the negative charge zone while positive graupel falls to form a lower positive charge center. This lower center possibly acts as a trigger zone for lightning initiation.

The above studies are in agreement that substantial charge transfer occurs when ice crystals rebound from riming graupel. Calculations have shown that the charge is significant to the electrification of thunderstorms. The studies agree that rimers may charge positively or negatively depending on the temperature and cloud liquid water content. In general, increasing the LWC can reverse the negative charging of a rimer to positive.

The other major study is by Takahashi (1978). He moved ice target rods at 9 m/s through a cloud of supercooled water droplets and ice crystals and he obtained the results shown in Figure 4. Differences between the studies are apparent by comparing Figures 2 and 4 and they are not directly attributable to the velocity differences in the two studies - in fact the differences would be even more evident if the UMIST experiments were conducted at 9 m/s. The vertical scale in Figure 4 is given as the liquid water content in the cloud (LWC) while the vertical scale in Figure 2 is the effective liquid water content (EW), which is the water content of the droplets captured by the target. There is a link between EW and LWC; at

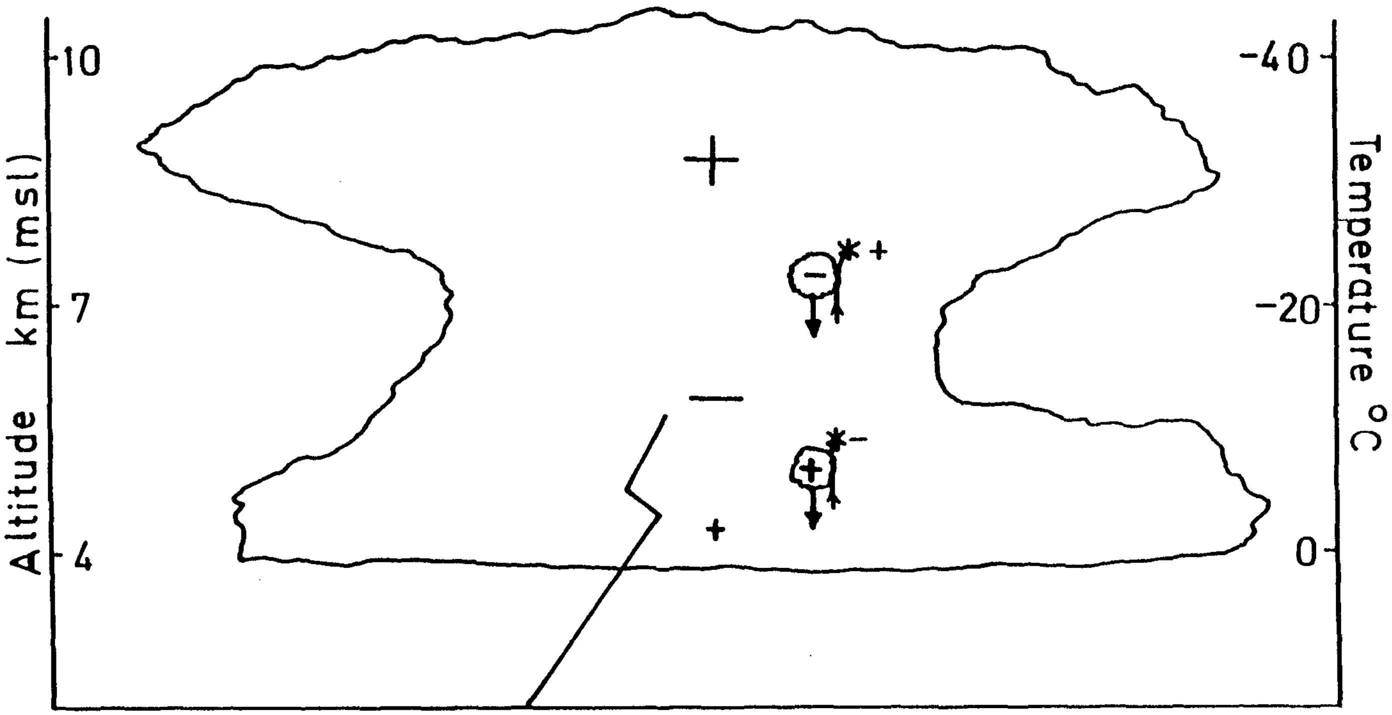


Figure 3. Graupel charging in thunderstorms.

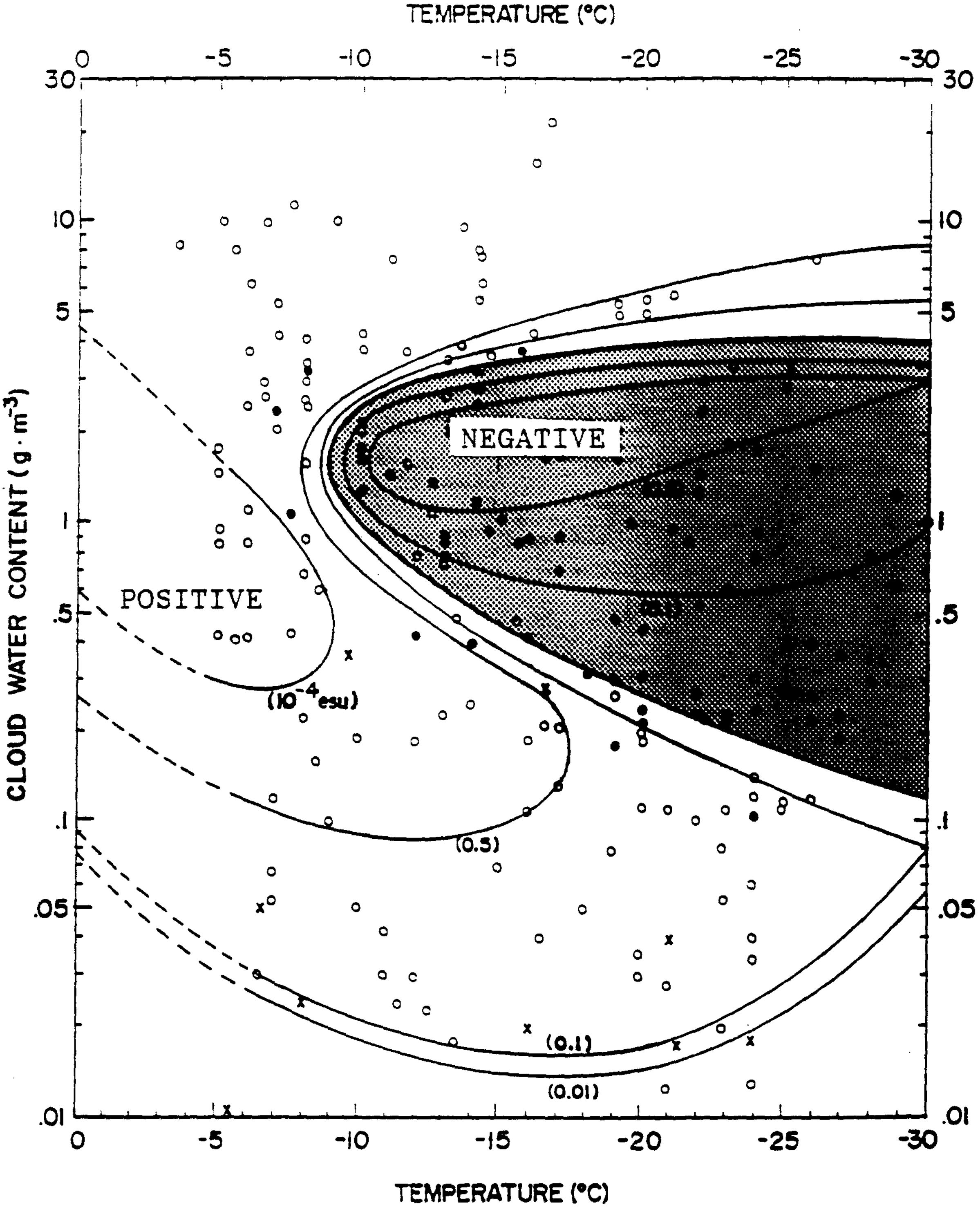


Figure 4. Rimer Charging according to Takahashi, 1978.

high speeds (10 m/s), most droplets are captured by riming (millimeter) targets and so EW = LWC. For small droplets and at lower speeds, such as the fall speed of small graupel pellets (5 m/s), EW may be around 0.5xLWC. This is insufficient to reconcile the two data sets shown in Figures 2 and 4. An important difference between the results is that the negative/positive transition, with increase in LWC, occurs at high LWC values for Figure 4. Figure 4 has LWC values sufficiently high to cause the riming target to experience wet growth, as pointed out by Williams et al (1991), for which condition, according to Saunders and Brooks (1992), there should be no charge transfer because the crystals adhere to the target. In general, the negative charging zone of Figure 4 occurs at much higher LWC values than for Figure 2.

Another important difference between the data sets is that Figure 4 has exclusively positive charging at temperatures above -10°C, whereas Figure 2 has negative rimer charging at low values of LWC at temperatures around -10°C. (Other recent work in UMIST has shown that positive charge to the rimer does occur at LWC values less than 1 g/m^3 at temperatures above -6°C).

It is important that the discrepancies between Figures 2 and 4 should be accounted for because the two sets of data are being used in considerations of charge generation in thunderstorms and obviously, differences in the liquid water contents required for negative charging lead to uncertainty in the validity of the laboratory data.

There have been attempts to account for the large differences in values of liquid water content required for charge sign reversal between the two studies, but they are qualitative in nature and cannot justify the five to tenfold differences in critical LWC values nor the quoted maximum LWC reported in Figure 4 of over 20 g m^{-3}. Several mechanisms accounting for the observed charge transfers have been proposed, and recently a theory specifically based on the results shown in Figure 4 has been put forward. Both sets of data are being included in numerical models used to calculate the rate of electric field growth in thunderstorms. The large differences between the two principal data sets are causing wasted effort and confusion in the development of charge transfer theories and models of field growth and so the present study was initiated with the intention of accounting for the discrepancies, and thus clarifying the situation.

Further considerations of the laboratory techniques

In order to simulate a soft-hail pellet falling through a cloud of supercooled water droplets and ice crystals, the data shown in Figure 4 were obtained with a 3 mm diameter metal target rod mounted on a rotating apparatus so the rod moved through the cloud in the cold chamber at 9 m s^{-1}. The data for Figure 2 were obtained with a 5 mm stationary metal rod target with the cloud drawn past at a speed of 3 m s^{-1}; experiments were also performed at speeds between 1.5 and 25 m s^{-1} while keeping the rate of cloud capture constant by adjusting the cloud water content, with no change in sign of the charge transfer as a function of velocity. The droplet cloud is formed from condensed vapor or by spraying droplets from an atomiser; the droplets rapidly supercool to the cloud chamber temperature.

In all these experiments it is necessary to initiate ice crystals in the cloud by a cloud seeding technique in order to overcome the low concentration of natural ice nuclei. (In clouds in the atmosphere, ice multiplication processes occur that overcome this ice nucleus shortage). Ice crystals were introduced into the laboratory clouds by seeding with solid carbon dioxide, or by briefly introducing a metal wire pre-cooled in liquid nitrogen. The crystals grow rapidly at the expense of the available water vapor, causing the droplets to evaporate; the vapor source is maintained to replenish the cloud.

The Present Studies

In the present experiments, rotating apparatus similar to that used to obtain Figure 4 was constructed and used alongside the stationary target apparatus used for Figure 2, in the same cloud chamber. Both moving and stationary targets were made of 3 mm diameter metal rods; they both experienced crystal and droplet interactions at 9 m s^{-1}. The sign of the charge transferred to the targets during ice crystal interactions was determined by connecting the targets to electrometers. It was anticipated that the two sets of apparatus would give opposite signs of charge transfer for values of LWC of around 1 g m^{-3} at -15°C (see Figures 2 and 4) and so this situation was investigated first. It was noted with some surprise that both targets charged positively under these conditions so long as the targets collected the same amount of deposit, the positive sign being in agreement with Figure 2. The experiments show that target rotation leads to a 10% reduction in the LWC experienced by the target, which is insufficient an error to account for the large differences between Figures

2 and 4.

Evidently the reason for the discrepancies between Figures 2 and 4 is not attributable to any effect of the rotation of the apparatus as we had at one time supposed. Other differences in technique in the conduct of the two original experiments were then examined. For Figure 4, pieces of solid carbon dioxide were used to seed the cloud whereas for Figure 2, a cold wire technique was used; however, it was found that seeding by either technique produced the same charge sign results.

A further difference in the seeding techniques was then tested. For Figure 4, the seeding method involved the continuous nucleation of the cloud, the pieces of solid carbon dioxide remained in the cloud chamber throughout the experiment, while for Figure 2, the droplet cloud was seeded briefly with the cold wire. With the continuous seed method, with either solid carbon dioxide or a small dewar of liquid nitrogen in the chamber, the charge sign detected by the moving and stationary apparatus was the same. The important difference between the two seeding techniques showed up in the determination of the LWC in the cloud by weighing the ice deposits on the two targets. With continuous seeding, the deposits indicated a water content in the cloud up to ten times greater than that being injected from the vapor source.

Details of the Takahashi vs UMIST experiments

A number of notable differences exist between the equipment and procedures used by Takahashi (1978) and Saunders et al (1991). The riming targets used by Takahashi consisted of two identical rods mounted at the ends of an arm that rotated about its midpoint, the rods were connected to an electrometer to allow measurement of the collected charge. Grounded metal cones at the ends of the arm shielded the base of the riming rods, preventing a rime bridge forming between them and the grounded arm. The arm was rotated to give an air speed of 9 m s^{-1} at the rods. The liquid water content was measured by collecting the rime from a small rotating probe situated below the charging targets. The UMIST experiments use a fixed riming rod mounted in a tube through which the cloud is drawn at the required velocity. Saunders et al determined the liquid water content by measuring the temperature rise due to the release of latent heat from freezing droplets on a secondary target, similar to the first and mounted in the second arm of a double tube, and applying the Macklin and Payne (1967) equation giving a link between rimer temperature rise and the rate of accretion of supercooled water droplets.

The equipment used in the present work is mounted in the cloud chamber shown in Figure 1. A tube holding the fixed riming target protrudes into the chamber from one wall and air is drawn through this from an air pump outside the cold room. A pair of rotating targets, modeled after Takahashi's equipment sit in the center of the cloud chamber; all the riming rods are of the same diameter (3 mm) and length (30 mm). A small rotating liquid water content probe, similar to Takahashi's, is mounted on the far side of the chamber. In the bottom corner of the chamber a short tube projects through the chamber wall, this is connected to a suction pump inside the cold room; an aperture in the tube outside the cloud chamber permits a formvar coated glass rod to be exposed to the air flow when the cloud is drawn through the tube at high enough speed to ensure 100% collection of all cloud particles drawn by; droplet and ice crystal spectra may then be obtained from microscopic analysis of the formvar replicas.

The fixed riming rod is connected by a co-axial cable to a charge amplifier situated outside the cold room; one of the rotating rods is connected directly to a similar amplifier mounted on the rotating shaft below the riming rods, the signal is brought out of the cold room via slip rings and a co-axial cable. RC filters remove 50 Hz pick-up from the signals before they are logged to a PC via an analogue to digital converter; samples are taken at 100 Hz and averaged over 1 s before being saved for later viewing and analysis; the output from each amplifier is also displayed continuously throughout the experiment. The second rod on the rotating arm was not electrically connected, and was easily removable in order to weigh the mass of collected rime; the rod in the tube could similarly be removed for weighing.

The liquid water content probe consisted of a 1.5 mm diameter rod, bent to form three sides of a rectangle in a U shape; this was mounted on a small electric motor and rotated in the cloud. The probe was removable for weighing. When determining the collected rime mass the various targets are sealed in pre-weighed plastic bags, thus avoiding the problems associated with scraping rime off targets before weighing and preventing loss by evaporation after removal from the cold room.

The concentration of naturally occurring ice nuclei is very low, in natural clouds ice multiplication processes overcome this shortage and produce many more crystals than could otherwise form; in the laboratory it is necessary to artificially seed the cloud with ice nuclei. Saunders et al (1991) seeded ice crystals by briefly inserting a brass rod, cooled to liquid nitrogen temperature, into the cloud; the ice nuclei introduced by this method grow rapidly at the expense of the cloud droplets, which evaporate to provide vapor for the crystal growth, before falling out of the cloud a few minutes later; the vapor input to the chamber is maintained to replenish the cloud. The crystal and droplet spectra change constantly throughout the run after seeding in this fashion. Takahashi placed a small container of solid carbon dioxide in his chamber, this seeded crystals continuously for a period of about an hour before completely evaporating, and allowed the cloud to reach a steady state before results were taken. In the present series of experiments variations on both seeding methods were used, a wire basket of solid carbon dioxide positioned at the top of the main cloud chamber provided continuous seeding for up to an hour, alternatively a tub of liquid nitrogen could seed for approximately 20 to 30 minutes and produced a greater number of crystals. Brief seeding with a cooled rod could be carried out either in the main cloud chamber or in an upper chamber connected to the main one by a trap door. In the latter case the crystals had time to grow before falling into the lower chamber and their number concentration was reduced since only a fraction of those seeded actually fall through the trap door.

Experiments and results

A preliminary experiment suggested that the rotating target might be depleting the cloud in its immediate vicinity. In order to test this rigorously, the total liquid water content calculated from the rime accreted on each target in an ice free cloud was plotted against the true liquid water content as measured by a Gerber Instruments Particle Volume Monitor (PVM) probe. Mean collection efficiencies of each target for cloud droplets were calculated using the results of Langmuir and Blodgett (1944-45) for the droplet distribution obtained by analysis of formvar slides and shown in Figure 5. The fixed and rotating targets operated at 9 m s^{-1}, and the rotating LWC probe at approximately 3.9 m s^{-1}; the cloud temperature was maintained between -11 and -15 °C. The comparison is shown in Figure 6.

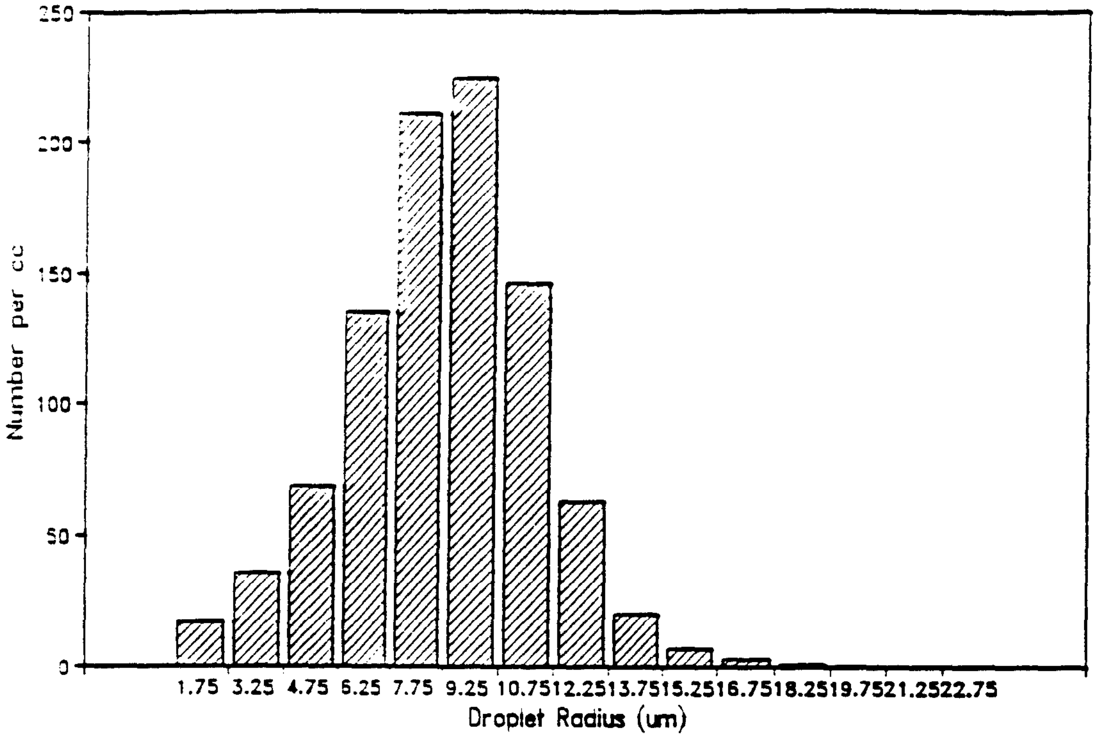


Figure 5. The unseeded droplet spectrum.

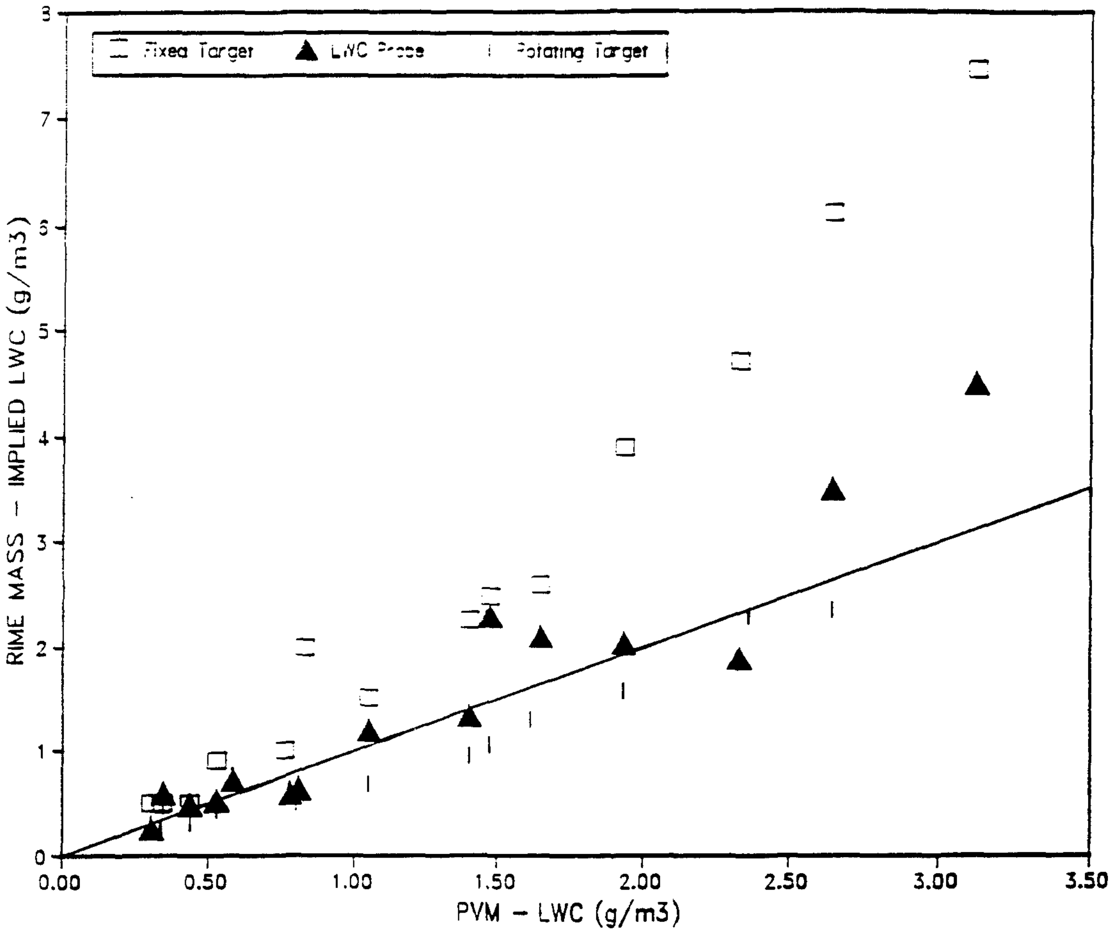


Figure 6. Liquid Water Contents in an unseeded cloud estimated from the mass of ice on the targets and LWC probe, plotted against the true LWC.

The rotating target did not appear to deplete the local cloud to the extent previously suspected, but underestimated the liquid water content by just 11%. The small rotating LWC probe overestimated the liquid water content at higher LWC values and had the greatest scatter in the results due to the difficulty of using this device. The fixed riming target overestimated the liquid water content by a factor of 2.1 probably due to the nature of the flow pattern when the cloud is drawn into the tube in which the target is mounted; the flow is certainly not laminar since the riming rod is set back only 25 mm from the mouth of the tube. The problem was avoided by Saunders et al (1991) who measured their liquid water content on the riming target itself, so their results are still valid for the liquid water content experienced by the target.

It was clear that the two targets saw very different liquid water contents under the same ambient conditions, however, there seemed no reason to suspect that charging results should not be valid provided that the liquid water content experienced by each target was determined.

Two similar sets of experiments were carried out in which the cloud was seeded continuously over a wide range of temperatures and liquid water contents; the first used solid carbon dioxide to seed the cloud with ice crystals, the second used liquid nitrogen. In both cases the crystals were seeded at the top of the lower cloud chamber, all other experimental procedures were common to both sets of experiments. Prior to each experiment, time was allowed for the cloud to become established and reach thermal equilibrium. After the introduction of the seeding substance a further 15 minutes were allowed for the crystal and drop spectra to settle down. In order to prevent the growth of ice crystals on the targets during this time, they were stored outside the cloud chamber until everything else was ready, they were then quickly fixed in position and a further 3 to 5 minute wait allowed for the cloud to settle after the disturbance caused by the opening of the chamber door. The rotating target and the airflow through the fixed tube were started at the same time and run for a timed period, typically 1 to 2 minutes, occasionally as little as 30 seconds, depending on the liquid water content; short periods were used to prevent the excessive build up of rime at high liquid water contents while long periods allowed a more accurate determination of the mass of rime collected at very low liquid water contents. The charge sign results of these experiments are shown in Figures 7 and 8. Total liquid water contents are calculated on the basis of the mass of rime accreted on the targets together with the mean collection efficiencies of the targets for the measured droplet spectrum during the continuously seeded experiments, Figure 9, and can only be considered approximate. It was not possible to determine the liquid water content and drop spectrum with a PVM or Forward Scattering Spectrometer Probe since both instruments use optical scattering techniques which are upset by the presence of ice crystals. Errors due to uncertainties in the air speed are of the order of 10%, errors resulting from changes in collection areas and efficiencies due to the build up of rime are unknown but will probably tend to lead to overestimation of the liquid water content, these were minimised by reducing riming times to prevent excessive rime build up.

The results show negative charging for low liquid water contents at all temperatures, while positive charging is achieved at higher liquid water contents, with the continuous nitrogen seeded case requiring higher values than for the continuous CO_2 seeded experiments. There is little difference in the charge transfer results between the fixed and rotating targets, although the latter requires a slightly higher liquid water content to achieve positive charging due to its lower droplet collection rate, as noted earlier. The measured liquid water content values cover a similar range to those of Takahashi. It is notable that there are values several times higher than those obtained in the tests with the PVM, and over double the known maximum of 4 g m^{-3} being introduced into the cloud chamber. Other experiments showed that liquid water contents determined for the small rotating LWC probe by weighing the rime collected and correcting for droplet collision efficiencies (the method used by Takahashi), are higher still. Data from the CO_2 seeded experiments during which the miniature LWC probe was in use are shown in Table 1 together with the corresponding values from the two charging targets and the estimated maximum possible liquid water content available from the boiler based upon PVM measurements made at the same boiler powers in a droplet only cloud at -15 °C. The actual liquid water content in the cloud should be significantly less than the maximum possible values quoted in the table because the growing ice crystals deplete the droplets.

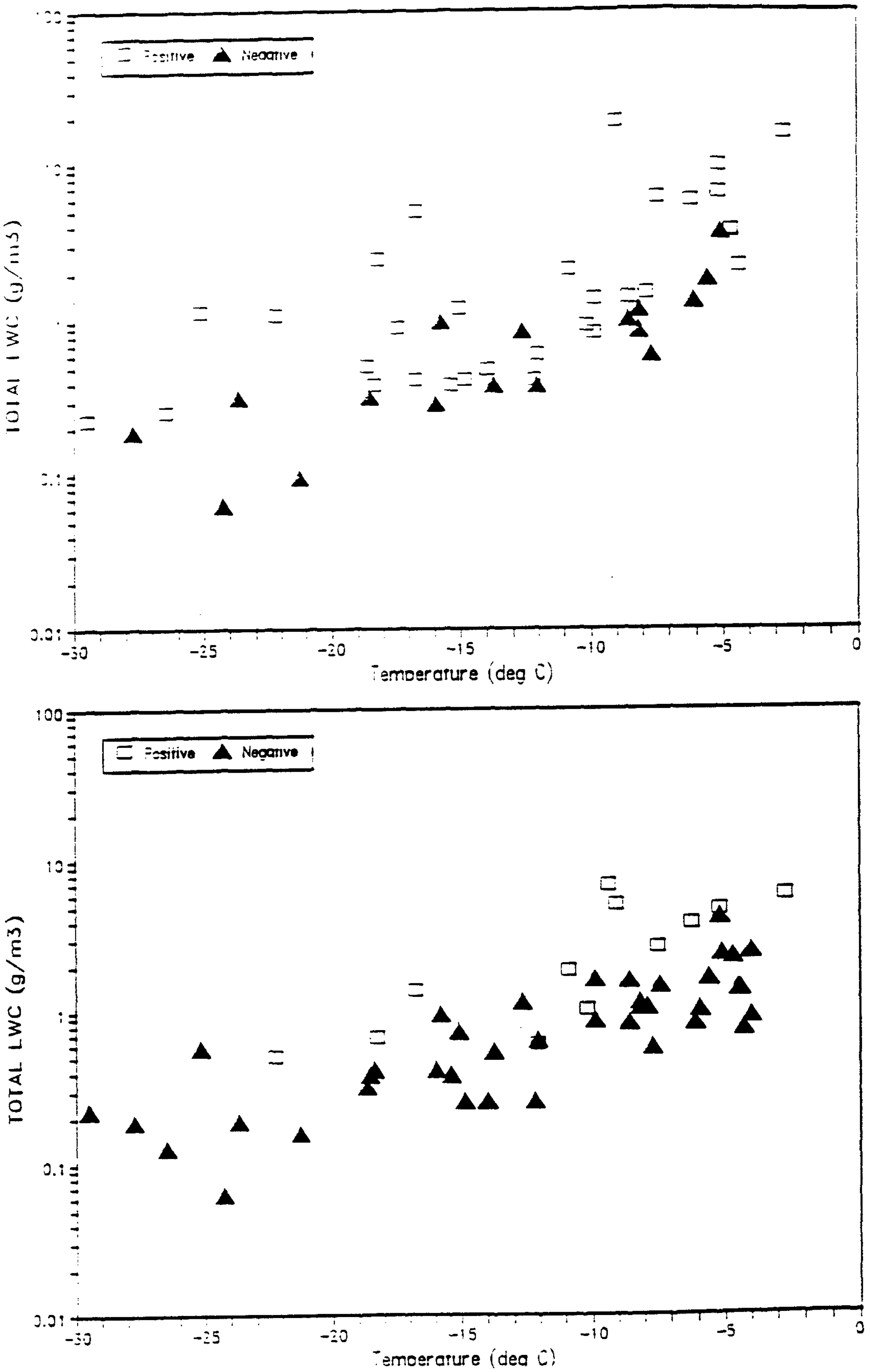


Figure 7. Charge sign with temperature and LWC, CO_2 continuous seed.

Top - Fixed Target; Bottom - Rotating Target.

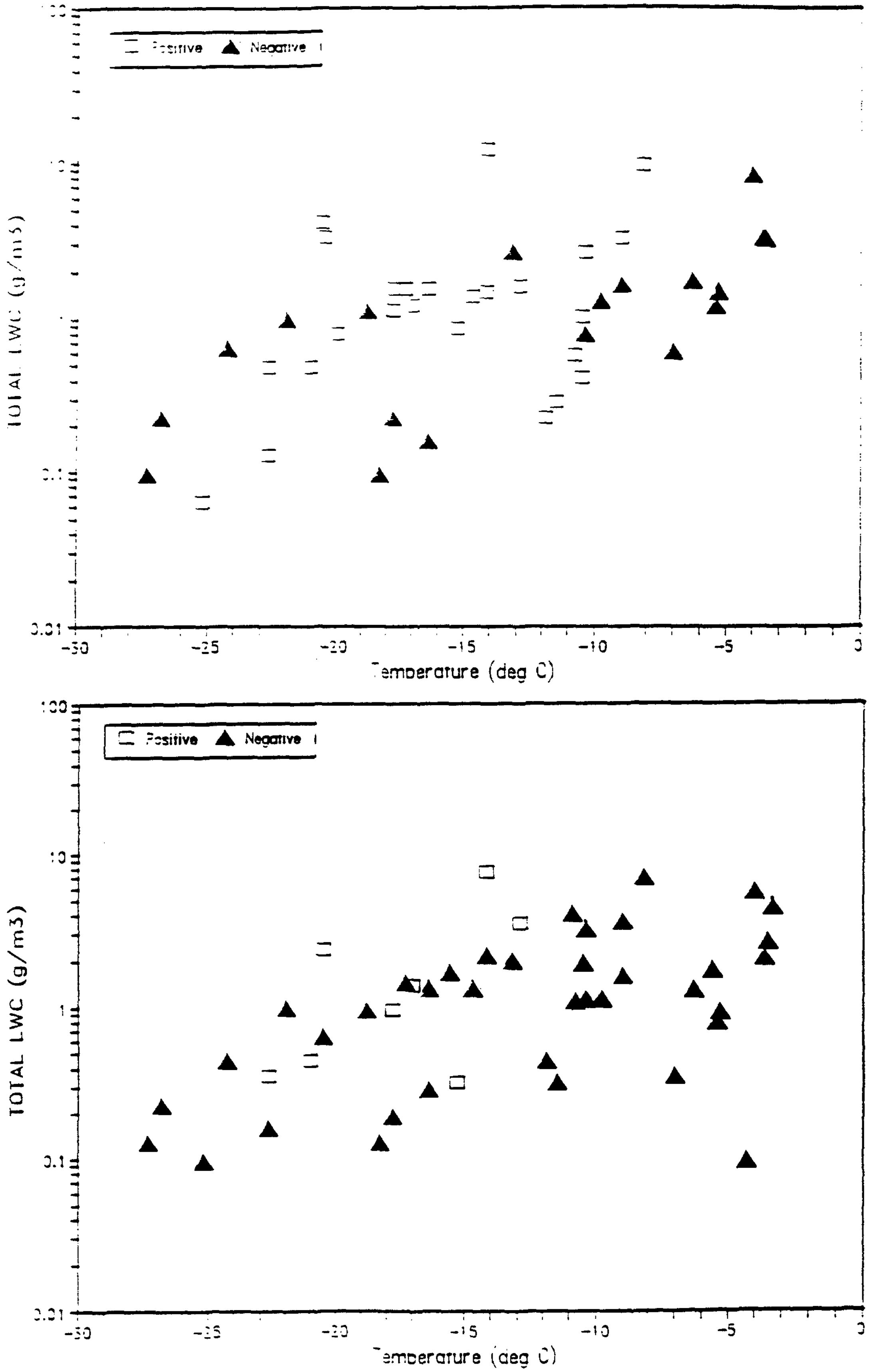


Figure 8. Charge sign with temperature and LWC, Continuous N_2 seed. Top - Fixed target; Bottom - Rotating Target.

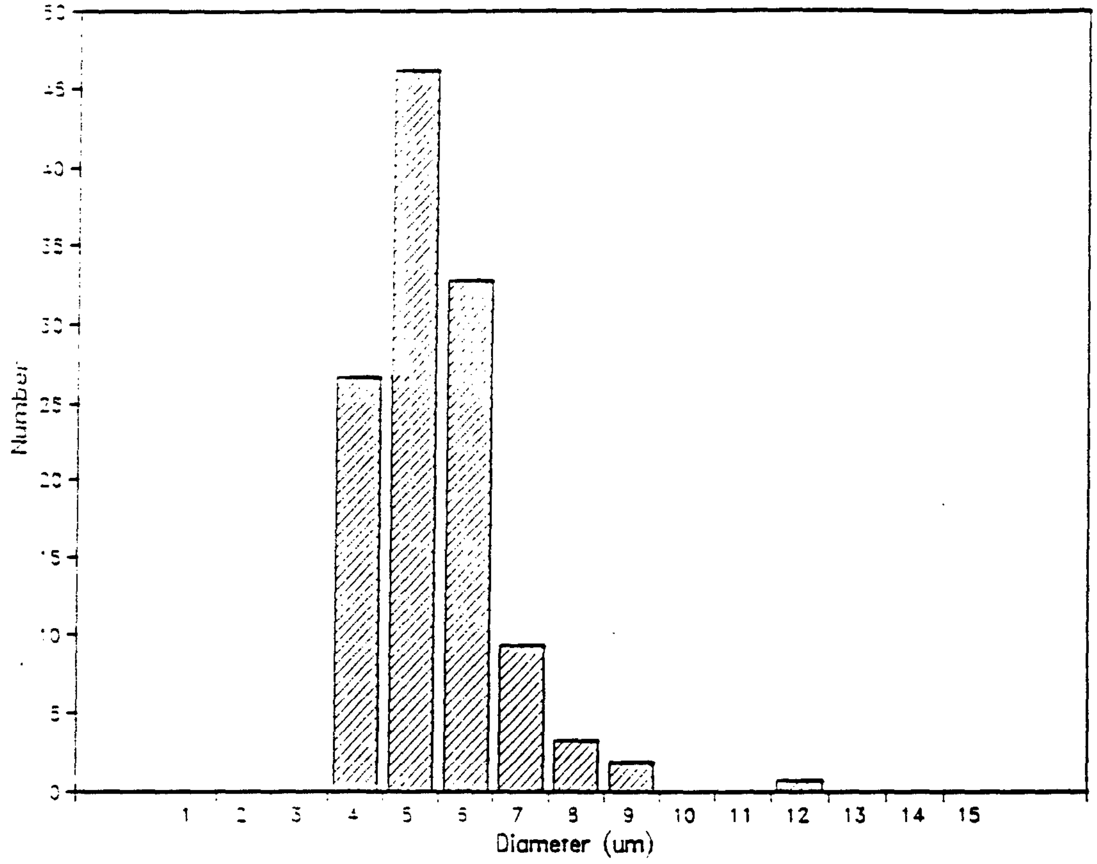


Figure 9. Droplet spectrum in continuous CO_2 seeded cloud at -7°C.

SMALL PROBE	FIXED TARGET	ROTATING TARGET	MAXIMUM POSSIBLE
4.1 (3.3)	0.8 (0.4)	0.7 (0.8)	0.3
2.8 (2.3)	0.3 (0.14)	0.3 (0.33)	0.3
6.3 (5.1)	1.2 (0.6)	1.2 (1.3)	0.4
7.1 (5.8)	2.9 (1.4)	1.8 (2.0)	0.4
6.6 (5.4)	1.5 (0.7)	1.7 (1.9)	0.4
17.5 (14.2)	0.4 (0.2)	-	1.4
8.8 (7.1)	1.8 (0.9)	2.4 (2.7)	1.6
13.3 (10.8)	-	7.6 (8.5)	1.6
8.2 (6.7)	1.2 (0.6)	1.5 (1.7)	2.6
5.8 (4.7)	1.2 (0.6)	0.6 (0.7)	2.6

Table 1 - Measured liquid water contents (g m^{-3}) from collected rime during CO_2 seeded experiments and estimated maximum possible true LWCs. Values in parentheses include compensation for the target collection calibrations from Figure 6.

Most of the calculated liquid water contents are greater than the expected values, many are greater than the maximum possible values, some by an order of magnitude or more, even when the results of the target collection calibrations against a PVM in an ice free cloud are taken into account (the bracketed values); however, they are in the range of values obtained by Takahashi. The figures show that the largest problem occurs for low LWC values. The reason for the overestimation, here and in Takahashi's work, was revealed by an examination of the droplet and crystal spectra in the cloud.

Formvar replicas of cloud particles were made on 4 mm diameter glass rods, briefly exposed to an air flow of approximately 9 m s^{-1}. Rods were used rather than the usual microscope slides because the collection efficiencies of cloud particles on cylinders are better known than those on flat plates, thus increasing the accuracy with which the collected particle spectra may be extrapolated back to the cloud; also the collection efficiency for small particles by 4 mm rods is higher than that with relatively large (25 mm wide) slides. The constant monitoring required by the rotating target meant that replicas could not be made during the same period as charge measurements, but were made immediately before or after. The particle replicas were counted and sized under a microscope. Droplet and crystal spectra for a typical continuous CO_2 seeded run, corrected for the collision efficiencies on the rod (droplets from Langmuir and Blodgett (1944-45), crystals from Keith and Saunders (1989)), are shown in Figures 9 and 10. It should be emphasised that the times for which different rods, and indeed different parts of a given rod, were exposed are not well known and vary considerably, typically between 0.1 and 0.5 s, so it was not possible to determine absolute concentrations of particles, only the shape of the distribution. Drop and ice spectra taken from the same rod may be compared for relative number concentrations. The formvar replicas showed many more crystals than droplets, and after correcting for the collision efficiency of the various particles on the glass rod it was found that crystals outnumbered droplets in the cloud by 50%. It is known that not all the crystals that impact on a rime coated target rebound and separate charge, many of them stick to the rime. The only experimental measurements of ice crystal sticking efficiencies were made by Keith and Saunders (1989) from which it is estimated that the average sticking efficiency for the current crystal spectrum is in the range 0.1 to 0.5. Using approximate values for the volume of the crystals (hexagonal plates are assumed to have a thickness of one third their diameter, rods to be of square cross-section, equal to one third of their length), and assuming a mass density for crystals of 0.5 g cm^{-3}, the fractional contribution to the mass of rime collected on the targets by droplets and crystals was calculated for a range of crystal sticking efficiencies (Table 2). The results indicate that the "rime" collected on the targets actually consists predominantly of crystals. By neglecting the collection of crystals, liquid water contents calculated from the "rime" mass are overestimated. Similar results apply to the experiments with continuous seeding using liquid nitrogen.

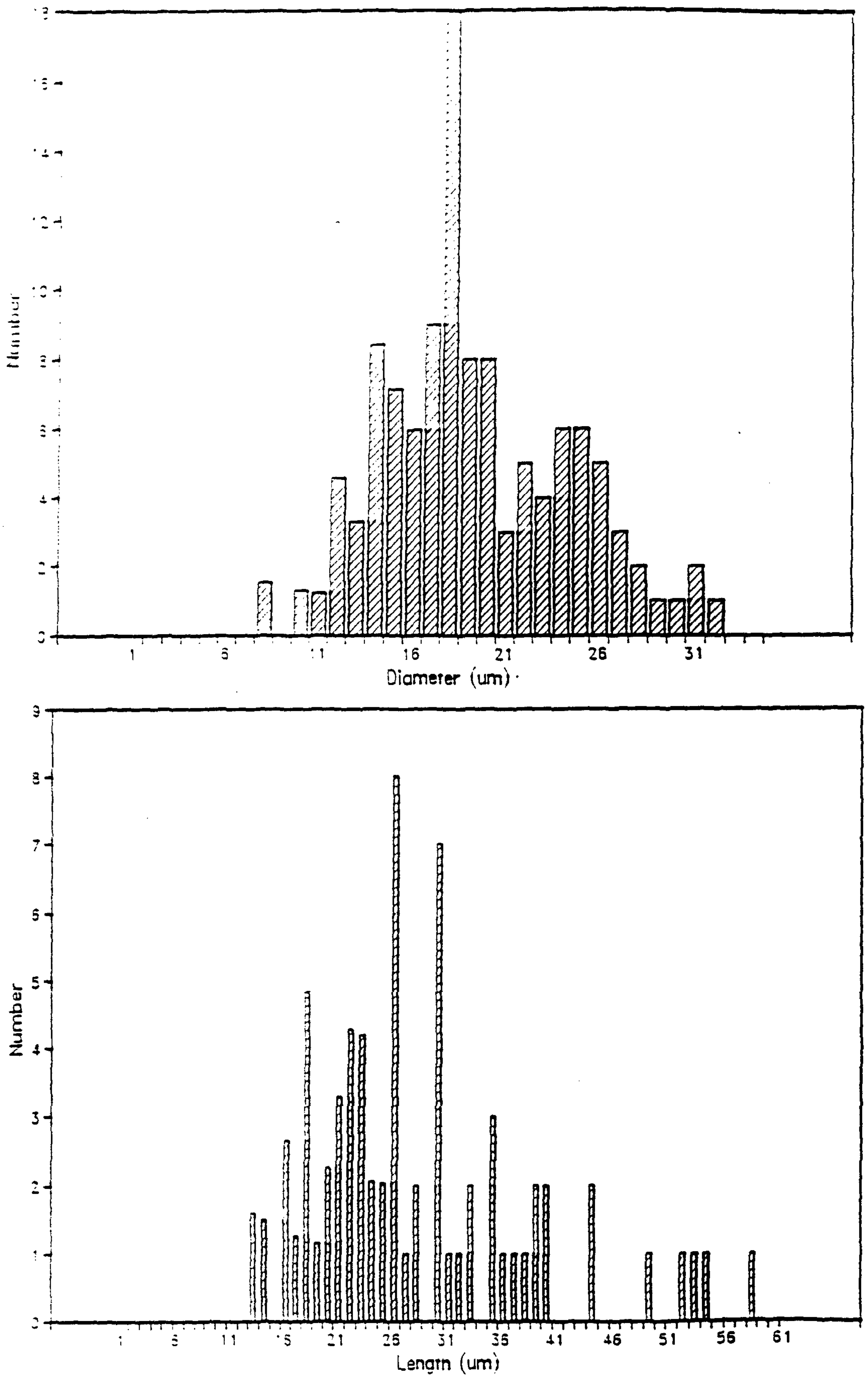


Figure 10. Crystal spectra in continuous CO_2 seeded clouds.
Top - Plate crystals; Bottom - Column crystals.

Crystal Sticking Efficiency	% of Rime Mass from Droplets	
	Charging Targets	LWC Probe
0.1	29.7	27.0
0.2	17.2	15.6
0.3	12.1	11.0
0.4	9.3	8.5
0.5	7.6	6.9
1	3.9	3.6

Table 2 - Calculated percentage of rime mass due to droplets.

Discussion of the charging experiments

It is apparent from Table 2 that the larger part of the rime mass collected on the targets is in fact made up of ice crystals and not water droplets. The reason for Takahashi's overestimation of liquid water contents is now obvious since he assumes that the collected rime consists entirely of droplets. It might be argued that this problem would be reduced in Takahashi's measurements because his droplet spectra show a broader range of sizes, and peak at a diameter of about 10 μm, approximately double that of the present work; however, these spectra were measured in ice free clouds (private communication) and cannot be considered representative of the droplet distribution during his experiments. The precise degree to which the liquid water content has been overestimated is impossible to determine since the sticking efficiencies of crystals are not well known and the relative droplet and crystal distributions in Takahashi's cloud are unknown. Figures 11 and 12 show the ratio of LWC determined by the probe method (including the droplet collection efficiency correction) to the maximum LWC in the cloud before seeding, taken from Table 1. The figures show that the most important problem occurs at low LWC values, below a probe LWC of around 5 g/m^3; at higher values, the LWC present is sufficient to swamp the mass of crystals on the targets.

In the light of the present work, Takahashi's charging zones in Figure 4 must be moved to lower liquid water contents by factors of between two and six; the former is the minimum

Figure 11. Ratio of corrected LWC on probe to total LWC in unseeded cloud against total LWC in unseeded cloud.

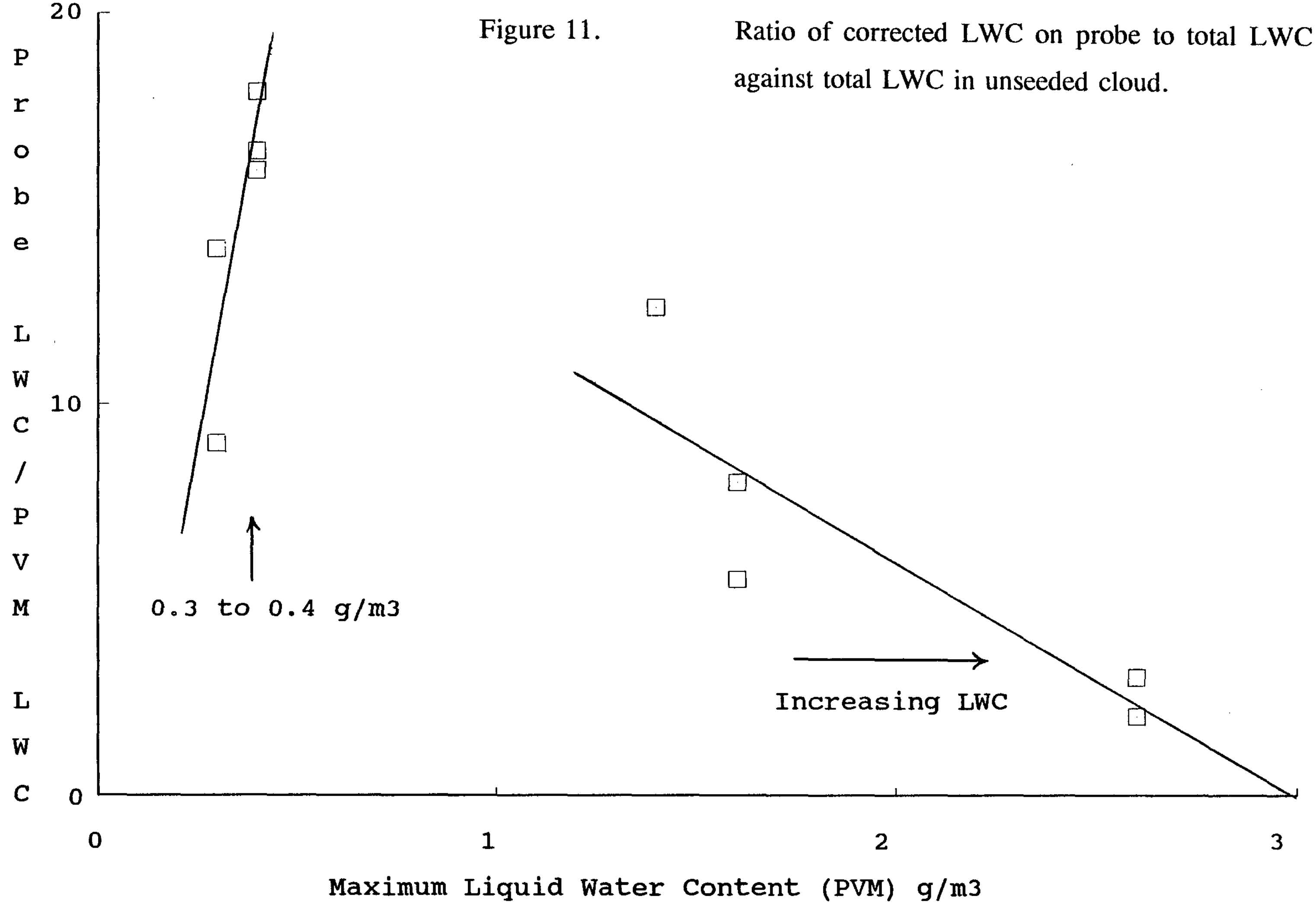
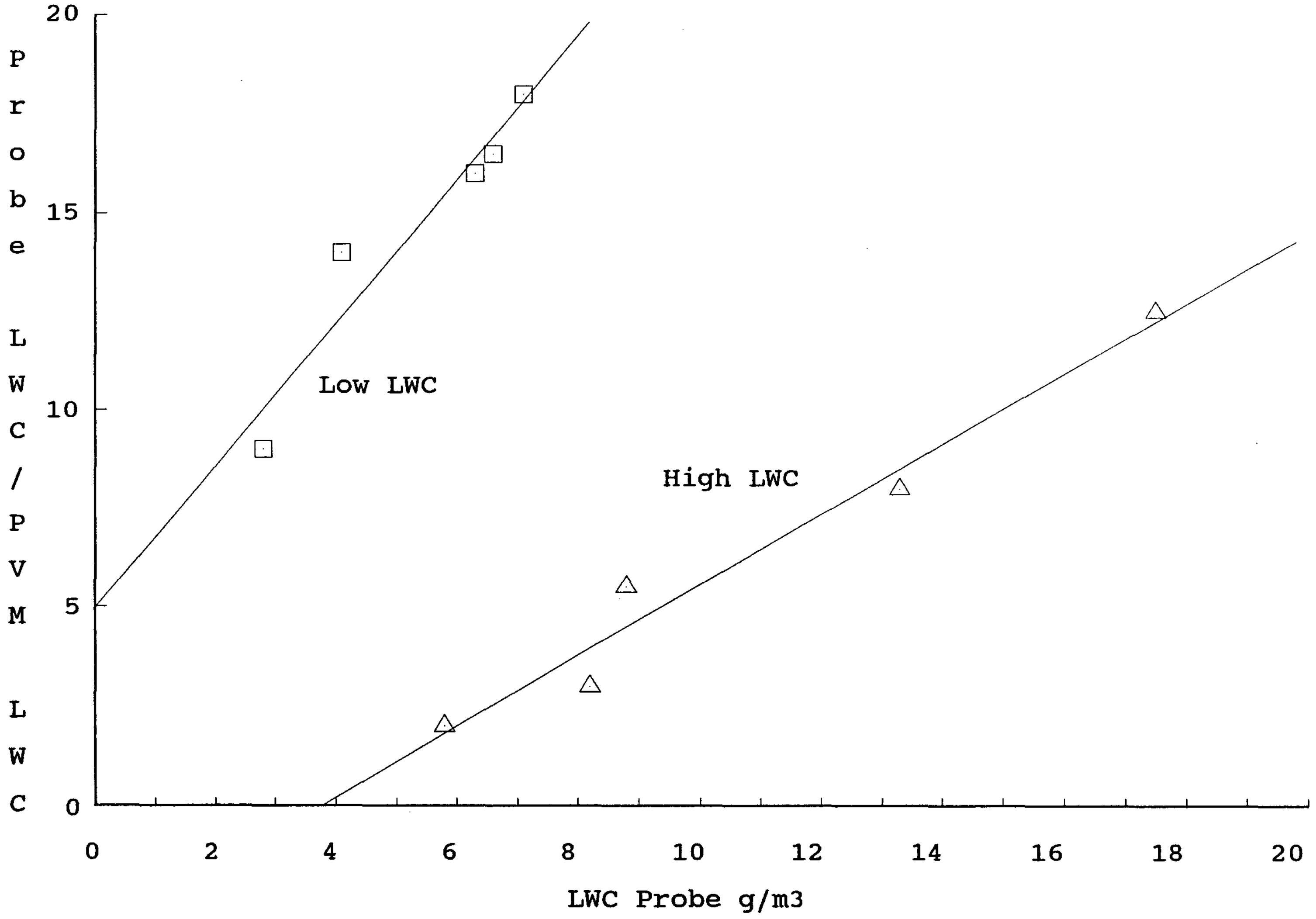


Figure 12. Ratio of corrected LWC on probe to total LWC in unseeded cloud against corrected LWC on probe.



likely error, the latter brings the upper boundary into agreement with the results of Saunders et al (1991) after a correction for the effect of the different velocities at which the experiments were conducted and is within the limits of possible error suggested by the present work.

The collection of crystals by the rimer has serious implications for all studies which make use of the mass of collected rime to determine the liquid water content as in Figures 7 and 8. The degree to which measurements are distorted will depend on a number of factors: the relative number concentrations of droplets and crystals in the cloud, the air velocity and target size, the size of the crystals, and to some extent the temperature. Keith and Saunders found that the sticking efficiency of crystals decreased for crystals greater than 100 μm in size, and did not vary significantly with temperature below about -5 °C, but increased towards unity above this temperature. The sticking efficiency was found to decrease with increasing velocity, which may explain the observation in this work that the LWC probe, rotating at about 4 m s^{-1}, gave a greater overestimation of the liquid water content than either of the riming rods, operating at 9 m s^{-1}. The small differences between the CO_2 and nitrogen seeded case is probably due purely to differences in ice crystal concentrations and hence to the amount of resulting distortion of the calculated liquid water contents. The large scatter in results near to charge reversal is probably also due principally to variations in the ratio of crystals and droplets collected on the targets.

The second major question regarding the results of Takahashi (1978) is his lack of any negative charging at temperatures above -10°C, while Saunders et al (1991) found negative charging at low liquid water contents for all temperatures above -20 °C; in the present work negative charging has also been found at low liquid water contents for all temperatures. The present work found no positive charging at low liquid water contents at temperatures above -10 °C with the Takahashi type rotating targets; however, there was some evidence of weak positive charging, presumably by droplet shedding, in a droplet only cloud. The entry of water vapour and droplets into the experimental chambers may also have some bearing on this problem; in the UMIST experiments water vapour is produced by a boiler situated outside the cloud chamber, and enters the chamber at one corner, away from the riming apparatus. Takahashi precooled environmental air from outside the coldroom so that droplets condensed from the natural water vapor in the air; the cloud was then drawn into the experimental chamber immediately below the riming targets. The air velocity at the inlet is quoted as 1 m s^{-1}, and since the total height of the chamber was only 1.5 m, there would have been a constant strong supply of water vapor to the target, in addition to collected droplets. Baker et al. (1987) suggested that, in a collision between ice particles, the faster growing particle charges positively, if this is so then a strong supply of vapor to Takahashi's target might help explain positive charging at low liquid water contents. A proper resolution of this problem must await further, more detailed, studies.

Studies of rime density as a function of cloud seeding technique.

It is conceivable that ice crystal interactions with a riming target may be affected by the nature of the rime surface - in particular, by the rime density. For this reason, as part of the investigation into possible causes of the differences between the charge transfer results of Takahashi and Saunders, measurements of rime density on the target in the unseeded cloud case were compared with densities measured in the two cases of brief and continuous cloud seeding.

The experiments were carried out in a temperature controlled freezer in which a cloud of water droplets was introduced from a boiler where they rapidly supercooled to the environmental temperature. The liquid water content could be controlled by altering the boiler heater power. The riming targets were made of 0.5, 1.5 or 2 mm diameter wire bent into wide U shapes with vertical arms of length 5 cm and horizontal separation of 12 cm. The rods could be rotated about the vertical axis through the centre of the U at speeds between 1.5 and 7.5 m/s, which are appropriate to the fall speeds of riming graupel in thunderstorms. The speed was monitored by measuring the frequency of closure of a reed switch caused by the passing of a small magnet placed on the rotating shaft. The density of the rime collected on the targets after their rotation through the cloud was measured by weighing the rime deposit after taking its physical dimensions in order to determine its volume.

Figure 13 shows the measured rime density against velocity for the unseeded cloud case. The error in the measurement of rime density is large at 25% but assessment of the size of the irregular rime deposit is not straightforward. The figure shows a general increase in density with velocity. Earlier work by Macklin (1960) had shown a simple relationship betwen rime density, the velocity of the target, the droplet size and temperature:

$$\text{Density} = 0.11 \times (-rV/T)^{0.76}$$

The slope of the line in Figure 13 is $V^{0.755}$ and given constant droplet size and temperature, the experiments appear consistent with the work of Macklin.

The effect of seeding the cloud in the freezer is shown in Figure 14 where the upper line of points was obtained by continuously seeding the cloud by the Takahashi method, while the lower line is for the UMIST method.

These results came as a surprise. It was anticipated that the highest density would be obtained with an unseeded cloud of supercooled water droplets and that the effect of seeding the cloud would be to introduce ice crystals to the chamber that would lower the droplet sizes and produce low density rime. Furthermore, the UMIST brief seed technique, which depleted the droplet cloud less than the Takahashi long seed was expected to reduce the density less than the long seed method. In fact, the highest densities (0.5 g/m^3) were obtained at 7.5 m/s by the long seed method, the unseeded cloud gave 0.35 g/m^3 while the brief seed gave a density of 0.17 g/m^3.

In these experiments, both the long and short seed methods use up the available water vapor and deplete the droplet cloud, so most of the collected material consists of ice crystals. A brief seed provides a smaller number of larger crystals giving a low density deposit while a continuous seed gives a larger number of smaller crystals that are able to pack closely on the target to give a high density deposit. This would account for Takahashi not noticing that his deposit consisted predominantly of ice crystals - as a high density deposit it had the same appearance as a rime deposit made up of water droplets.

The effect of rime density on charging

Given the above results, that there is a difference in the rime density on the riming targets in the brief seed and long seed case, a further study was made to see whether the density could affect the charge transfer. It is possible that the Takahashi data obtained at very high

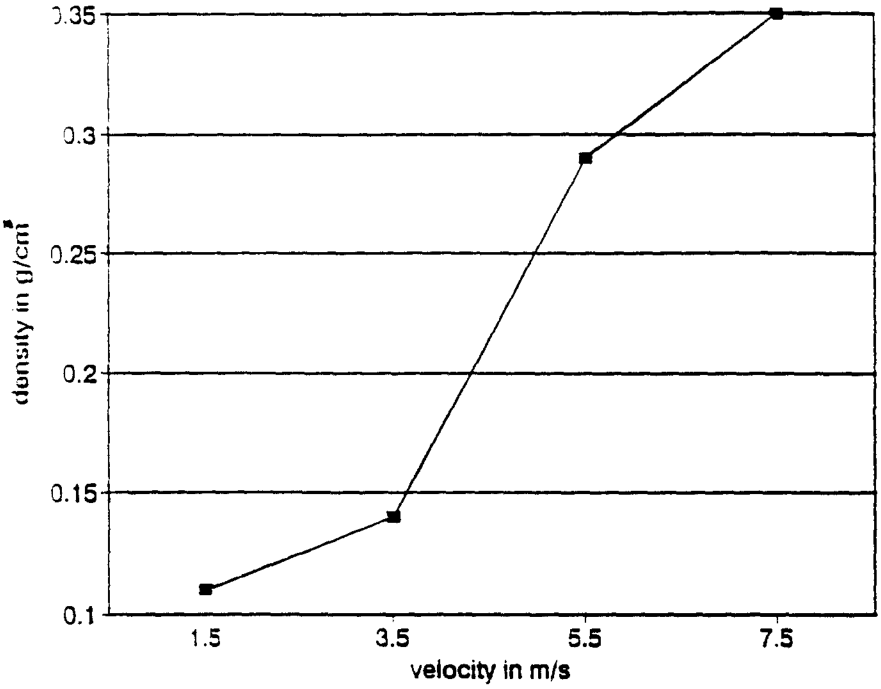


Figure 13. Rime density for unseeded cloud against velocity.

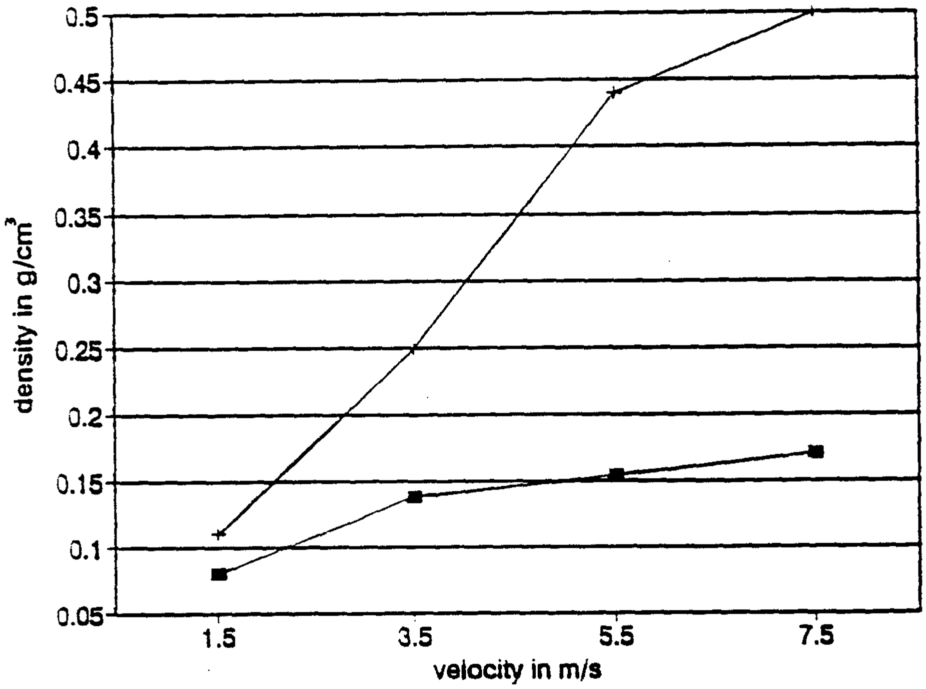


Figure 14. Rime density for seeded cloud case - upper line, continuous seeding (Takahashi); Lower line, UMIST, brief seed method.

liquid water contents could be associated with high rime density whereas this regime could not be obtained with the low density rime obtained with the brief seed method.

A stationary riming target was set up and the current transferred to it while ice crystals impacted upon it could be monitored. High speeds of 20 m/s were used in order to ensure high rime density. During the experiment, the rime surface temperature was measured with a thermistor bead and the density was calculated from the Macklin equation given above for the cloud conditions used. For example, the mean droplet radius in the cloud was 4 μm and at a surface temperature of -6°C the rime density reached 0.9 g/m^3.

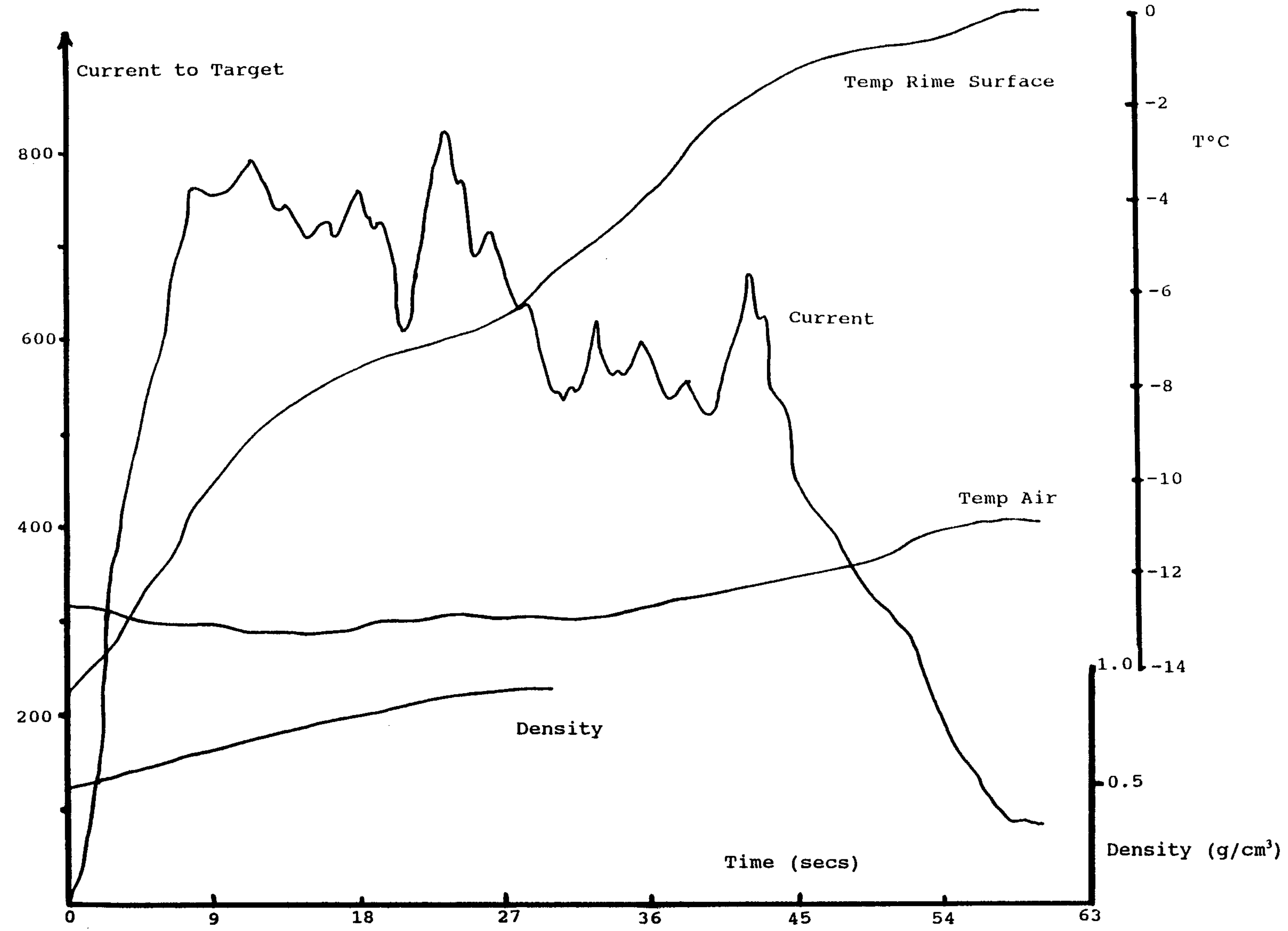
Figure 15 shows the charging current to the target, the temperature of the air and of the target together with the calculated rime density. The maximum density is of course unity.

The question has been raised in discussions with others in the field as to whether the UMIST brief seed technique could actually achieve high rime densities. If it could not, then positive charging at high rime densities could not been observed in the UMIST experiments. Figure 15 shows that high densities can be achieved and that at these high densities, positive charging is observed.

Furthermore, as the surface temperature rises to 0°C during the experiment, the charge transfer decreases dramatically. This is in line with our previous work showing no charge transfer during wet growth conditions when the surface of the rimer cannot remove the heat being released by the arriving droplets fast enough and its surface temperature rises to 0°C. A further consequence of the heat rise of the rimer is that its surface reaches a stage of net sublimation; Williams et al (1991) postulate that this surface condition is associated with the negative charging of the rimer: this is obviously not the case as shown by the continued positive charging of the rimer from low temperatures up to those approaching the wet growth of the surface.

Figure 15.

The current to a riming target, the rime and air temperatures and the density of the rime all plotted against time.



Conclusions

The charge transfer results are the same for the Takahashi type apparatus and for the UMIST apparatus when used in the same cloud. The results are not sensitive to the cloud nucleation technique used nor to differences in the way that the two sets of charge transfer experiments were conducted. It appears that the charge sign reversal at a higher LWC noted by Takahashi than by UMIST, is in fact due to an overestimate of the LWC by Takahashi.

The differences between the liquid water content values quoted by Takahashi (1978) and those of Saunders et al (1991), have been found to be due to the method of determining liquid water content. Takahashi measured the quantity of rime collected on a rotating probe, and assumed that all the rime was due to the collection of droplets. It has been shown that a large proportion of the "rime" is in fact made up of collected ice crystals; by neglecting these, Takahashi greatly overestimated the liquid water content of his cloud. Saunders et al used the heat balance of their rimer to determine the quantity of water collected from the latent heat released by freezing droplets; this method is insensitive to the collection of ice crystals. The magnitude of Takahashi's overestimation is due to the method of seeding; continuous seeding with CO_2 results in the production of a very large number of crystals which greatly deplete the droplets in the cloud. Brief seeding with a cooled rod, as used by Saunders et al, produces fewer crystals and reduced droplet depletion, hence the contribution of ice crystals to the collected rime mass is not significant.

Conditions experienced by the fixed and rotating targets used by Saunders et al and Takahashi, can differ markedly from each other and from the ambient conditions within the same cloud. This highlights the problems involved in comparing different sets of charging results, since nominally identical conditions may in fact differ considerably. The finding may help explain existing discrepancies between various data sets from laboratory charging experiments. Future experiments must be designed and monitored in such a way that their conditions are reproducible by other workers with different equipment. Given the sensitivity of charging behavour to the ambient conditions it is important that the results of field studies are fed back to the laboratory and that experimental studies try to match the conditions found in natural clouds as closely as possible.

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