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***In Situ* Measurements of Electric Charges on Individual
Ice Crystals by the Application of a Horizontal
Electric Field
(Electrification Mechanisms of Snow Crystals: Part II)**

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Abstract

To measure ice crystal charges directly without any touching, a horizontal electric field was applied in two types of procedures.

In the first procedure, which gives the distribution patterns of the ratio of charge to mass in every exposure for a certain interval, large fractions of ice crystals showed no detectable charges although a part of large and minute ice crystals showed predominantly positive and negative charges respectively.

In the second procedure, which gives the time changes of ice crystal charge with finer time resolution regarding the ice crystals which have relatively large quantities of charges, it was shown that large ice crystals carrying positive charges fell into the early and final stages where the ice crystals grew well, and that on the contrary, numerous and minute ice crystals fell into the middle stage where ice crystals were not supplied with sufficient moisture.

It was concluded and reconfirmed that growing ice crystals have a tendency to carry positive electric charges preferentially and evaporating or slowly growing ice crystals carry negative charges. These properties are also discussed in comparison with some natural phenomena.

1. Introduction

The electrification mechanism of snow crystals so far have not been investigated although it is considered that since snow crystals occupy a major part of winter thunderclouds, their electric charges contribute essentially to the electrical structure of such clouds.

Magono and Orikasa (1966) carried out many simultaneous observations of electric charges of snow crystals and the atmospheric electric potential gradient at the ground surface, and reported that when a steady, light and quiet snowfall occurred, the snow crystals which were almost all unrimed clean crystals, always carried negative electric charges, and there the potential

gradient was maintained positive at several fold values of fair weather values. Kikuchi (1975) reported that rimed snow crystals and flakes carried positive charges and clean snow crystals without riming carried negative charges. According to their observations, it appears that, on the average, snow crystals carried negative charges predominantly, insofar as they were measured at the ground surface.

To investigate the electrification mechanisms of snow particles, Takahashi (1973) performed a laboratory experiment by using bulk ice and reported that the ice surface acquired positive and negative charges in its growing and evaporating processes.

It seems more reasonable to say that an electrification mechanism of snow crystals is essentially the same as that of minute ice crystals produced in the laboratory. Magono and Iwabuchi (1972) measured the electric charges on individual ice crystals in a cold room and reported that although, predominantly, they were found to carry positive charges on the average, ice crystals carried negative charges in the temperature where columnar type ice crystals predominated, and generally the signs of ice crystal charges seemed to have some temperature dependences. Meanwhile, Kikuchi (1973) also showed in Antarctica, that in general, crystals of the warm type (dendrites, sector and plates) exhibited negative charge, while crystals of cold type (columns, side-plane and combination of bullets) exhibited positive charges.

Endoh (1979) measured the electric charges of diamond dust in Inuvik, Canada and reported that the condition of moisture around ice crystals contributed to the signs of electric charge. On the other hand, Endoh (1981) performed some laboratory experiments and reported that similar relations were found to be held between the signs of ice crystal charges and environmental humidity. These experiments, however, seemed to contain a questionable problem regarding their filtration processes which is apt to cause some secondary charge generations. It is desirable that those results should be reconfirmed by different experimental methods.

2. Experimental Methods

The electric charge of ice crystals, fog and cloud droplets have not been measured by electrometric devices, because the quantities of charges are too small to be measured by them. Application of a horizontal electric field to these particles were used for this purpose by Twomey (1956), Magono and Kikuchi (1961), Magono and Iwabuchi (1972) and Endoh (1979). The principle

of these measurements was developed by Wells and Gerke (1919).

Experiments were carried out by two types of procedures.

2.1 Charge to Mass Distribution

To obtain the shape and size of individual ice crystals and their quantities of charges at the same time, an apparatus as shown in Fig. 1 was utilized. When clouds of many droplets were filled in the tower shown in the left half of the figure and cooled to become supercooled droplets, seeding was done by introducing a copper block which was sufficiently cooled by dry ice outside of the tower before hand. Ice crystals produced there grew and fell toward the bottom of the tower and these were measured by an equipment specially designed and shown in detail at the right half of the figure. It has a collimator (A in the figure) of a length of 15 cm in order to avoid the particles which have a large horizontal component of the fall velocity according to the equipment used by Latham and Smith (1970). As ice crystals fall through a slit of 2 mm gaps (B in the figure). They were diffracted by an electric field of DC 2300 Volts per 17 mm gaps, depending on the values of their charges and fell on a slide glass covered by silicon oil.

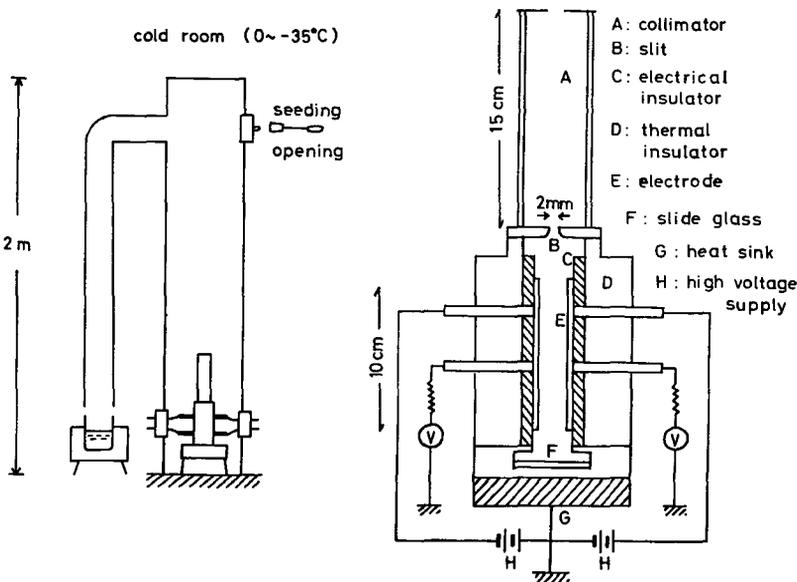


Fig. 1 Experimental apparatus in cold room (left) and measuring part of electric charge of ice crystals by the horizontal electric field (right).

The quantities of electric charges on an ice crystal were calculated from the displacement S from the center of slide glass which was decided by ice crystal fall without fields. The ratio of charge q to mass m of ice particles is described

$$q/m = (gS)/(Eh)$$

where g is the gravitational acceleration, E is an applied field intensity and h is the falling distance through the field. In this case, $E=1500$ V/cm, $h=10$ cm, then

$$q/m = 50S$$

Measurable ranges are limited between -160 and $+160$ with an error of ± 20 in a dimension of e.s.u./gr.

As ice crystals were controlled to fall for 3 or 4 minutes in a series of the cycle, a slide glass was exposed for a minute and displaced slightly every minute. As soon as the ice crystal fall ceased, the slide glass was drawn out and photographed under a microscope.

2.2 Transitional Sign Change

To obtain quick time changes of ice crystal charges in a cycle of their fall and to verify the results of the time dependence of charge described by Endoh (1981), the following experiments were carried out in the laboratory.

The electric charge of individual ice crystal was measured from the diffraction of falling trajectory under a horizontal electric D.C. field with the equipment used by Magono and Iwabuchi (1972). Because the diameter of the cloud chamber of the equipment was small as 15cm, the supplied super-cooled droplets were deposited on the wall of the cylindrical metal chamber. Therefore, water vapour was supplied continuously from above by warming distilled water. Ice crystals were formed by seeding with a cooled copper rod at upper part of the chamber. Since this experiment was focussed on the time changes of ice crystal charges, only the signs of charges on ice crystals which had considerably large quantities and was diffracted strongly, were observed by the naked eye and recorded by an audio tape recorder.

3. Results

All experiments were performed under a temperature around -15°C .

3.1 Charge to Mass Distribution

The obtained pictures of Figs. 2 to 5 show the time changes of the

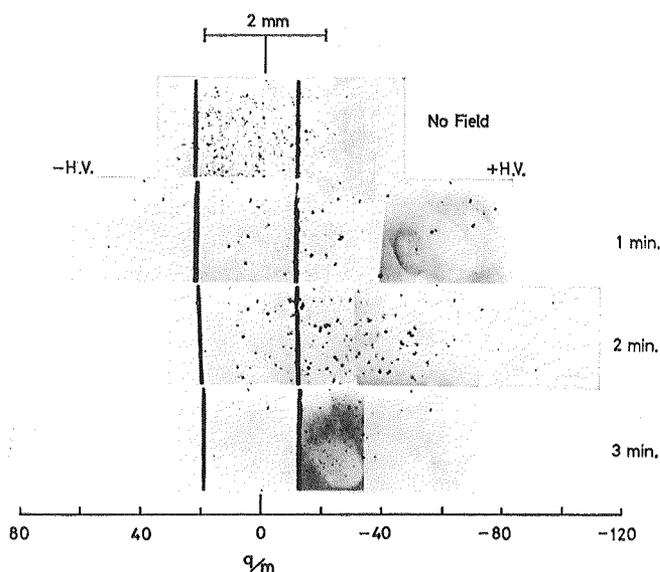


Fig. 2 Transitional distribution of electric charge versus mass of ice crystals.
 abscissa: ratio of charge to mass of ice crystal in e.s.u./gr with polarity.
 ordinate: ending time of time lapse exposure.

distribution of the ratio of charge to mass. In these pictures, the axis of the abscissa indicates the ratio of q/m in a dimension of e.s.u./gr and the left and right sides correspond to the positive and negative signs of electric charge respectively. The time indicated in ordinate axis shows the end time of an exposure for every minute from the time of seeding.

In Fig. 2, on the average, negatively charged ice crystals were shown predominant. Especially during the second period (2 min.), many ice crystals are seen to lean to the right side of negative charge rather better than that for the first period (1 min.). In Fig. 3, the charge distributions lie widely from negative to positive sides. For the second period (2 min.), large ice crystals are seen to distribute widely, but for the fourth period, the size of ice crystals are seen to become smaller and are distributed more closely than that of the second period. In the case of Fig. 4, the slide glass was not drawn throughout the cycle. It is seen that a number of large size crystals fall on the left side of the positive charge and distribute concentratedly and isolatedly far from zero point, while small size crystals fall slightly leaning toward the right side of the negative charge. In Fig. 5, it is shown that ice crystals which fell till the end of the first period (1.5 min.) were predominantly

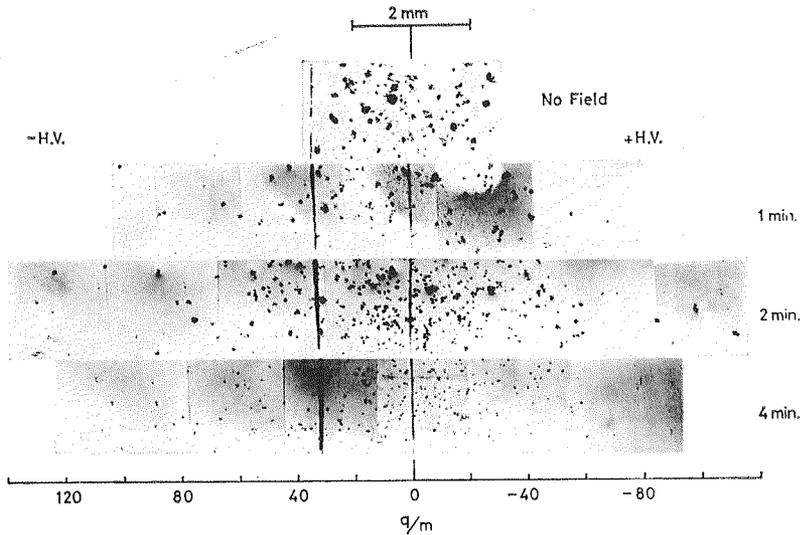


Fig. 3 Transitional distribution of electric charge versus mass of ice crystals.
 abscissa: ratio of charge to mass of ice crystal in e.s.u./gr with polarity.
 ordinate: ending time of time lapse exposure.

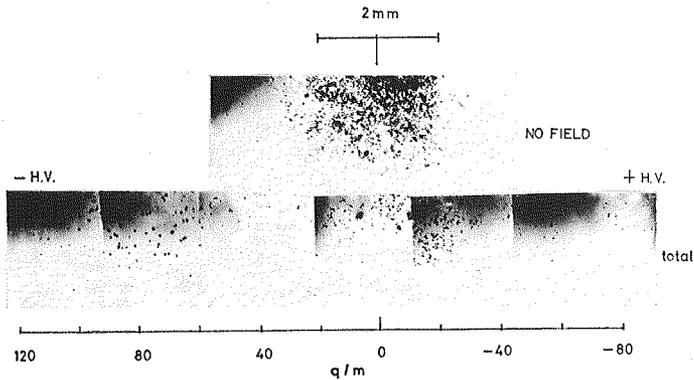


Fig. 4 Total distribution of electric charge versus mass of ice crystals.

distributed on the left side of the positive charge, while ice crystals which fell in the second period (3 min.) were distributed on the right side of the negative charge. Moreover the size of the former and latter are seen as relatively large and small respectively. Throughout the experiments, it is noted that considerable amounts of minute ice crystals are seen to fall near the zero point.

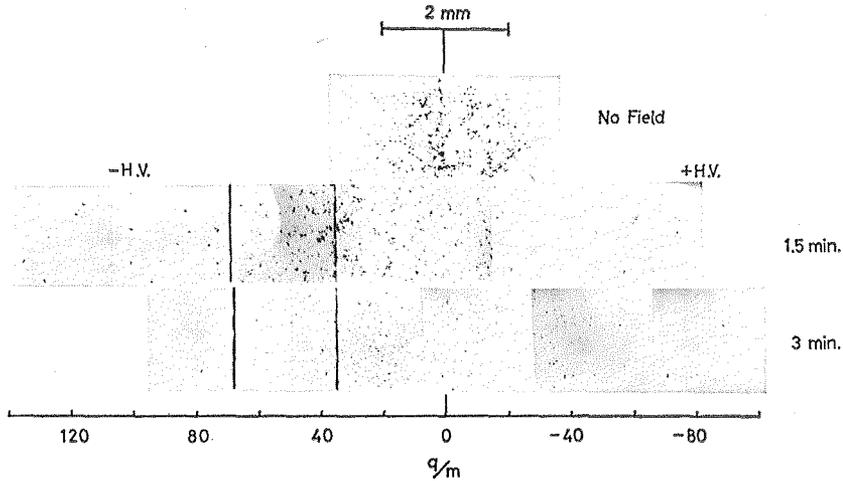


Fig. 5 Transitional distribution of electric charge versus mass of ice crystals.
 abscissa: ratio of charge to mass of ice crystal in e.s.u./gr with polarity.
 ordinate: ending time of time lapse exposure.

3.2 Transitional Sign Change

Since the cloud chamber of the equipment used for obtaining fast time changes of ice crystal charges was relatively small, a deposition loss of supplied droplets to the wall was considerably large. Therefore water vapour was continuously supplied. Accordingly the relations between the existences of supercooled droplets and ice crystals are considered to be different from the previous experiments, where water vapour was not supplied after the time of seeding. To examine this, an impactor method was utilized. Since this method requires a considerably long time for sampling and recording, the experiments were carried out using deliberately the previous larger tower under a situation of over-seeding so that ice crystals were maintained in such way to fall for a long time together with a continuous supply of water vapour. The obtained results are shown in Fig. 6. Solid circles with solid lines and triangles with broken lines indicate the number concentrations of supercooled droplets and ice crystals respectively. The abscissa and ordinate show the time elapsed in minutes and concentration of the particles in a dimension of number per cm^3 respectively. It is seen in the figure that at the beginning, ice crystals alone existed in a concentration of about 10 particles per cm^3 , immediately after a seeding as shown by arrow marks, numerous ice crystals appeared and supercooled droplets began abruptly to decrease,

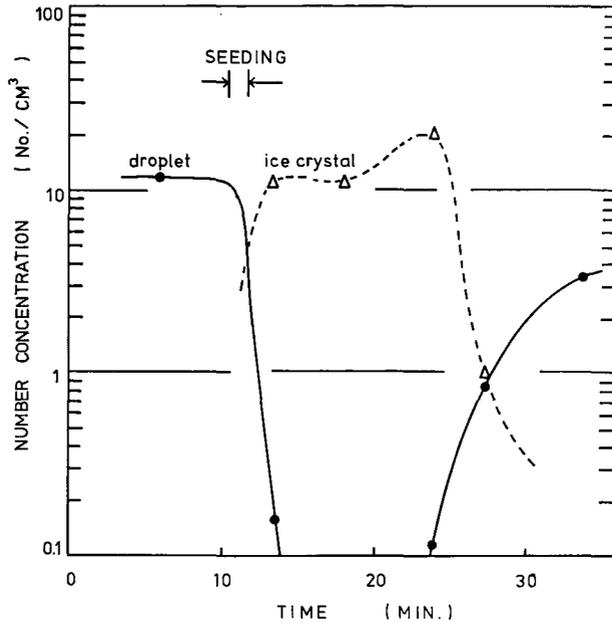


Fig. 6 Time variation of relation in concentration of supercooled droplets and ice crystals after seeding under successive supply of steam.

and soon disappeared. In contrast, ice crystals were maintained in the same concentration of that of initial supercooled droplets for about 10 minutes arising from over-seeding, but after a short lapse of time began to decrease and finally disappeared after which the droplets increased again.

All results obtained with a small cloud chamber by means of continuous vapour supply are shown in Fig. 7. The abscissa indicates the time. The position of many short spikes indicates the time when a strongly charged ice crystal was observed and its direction of upward and downward shows the sign of electric charge as positive or negative respectively. All experiments were done in almost the same manner, but the duration of ice crystal fall differs from case to case. On the average, in the early and final stage, and in the middle stage, the predominant charge of ice crystals were positive and negative respectively. They showed two reversal times t_1 , t_2 when the sign of charge changed from one to the other as shown in Table 1. In the first reversal time, the sign changed from positive to negative, and in the second reversal time, the sign changed from negative to positive again. A most representative example is

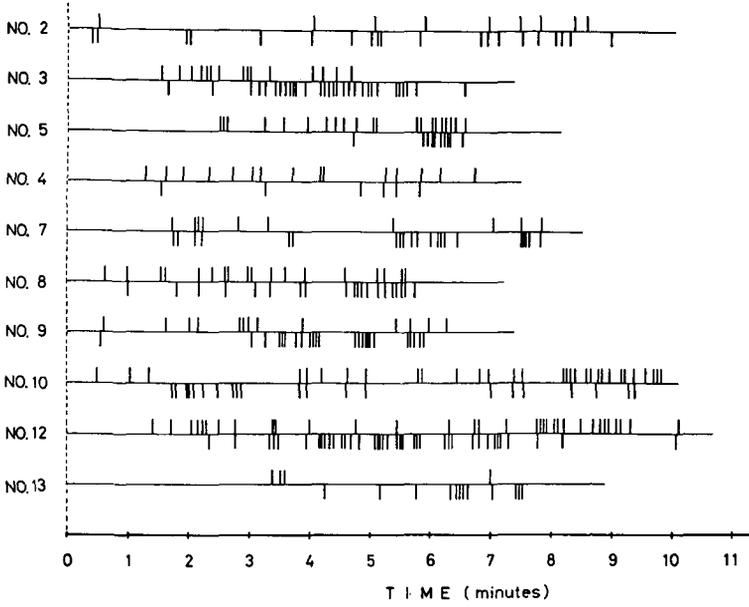


Fig. 7 Detailed records of observed time and polarity of electric charge of ice crystals.

Table 1 Frequencies of positively and negatively charged ice crystals with transition over two reversal times. (after Fig. 7.)

Exp. No.	Positive/Negative			Sum
	t_1	t_2		
3	10/ 2	5/ 28	0/0	15/ 30
4	14/ 1	5/ 8	2/1	21/ 10
5	10/ 2	3/ 4	2/0	15/ 6
7	6/ 4	3/ 17	1/0	10/ 21
8	12/ 6	6/ 14	0/0	18/ 20
9	8/ 2	4/ 22	2/0	14/ 24
10	3/ 0	12/ 18	16/4	31/ 22
12	10/ 5	7/ 31	16/3	33/ 39
13	3/ 0	1/ 12	0/0	4/ 12
Sum	76/22	39/154	39/8	161/179

seen in experiment No. 12. In all cases, the experimental period may be divided into three stages by the two reversal times. The frequency of positive and negative charge of ice crystals covering all cases were summarized in Table 1 and averaged respectively for each stage and shown in Fig. 8.

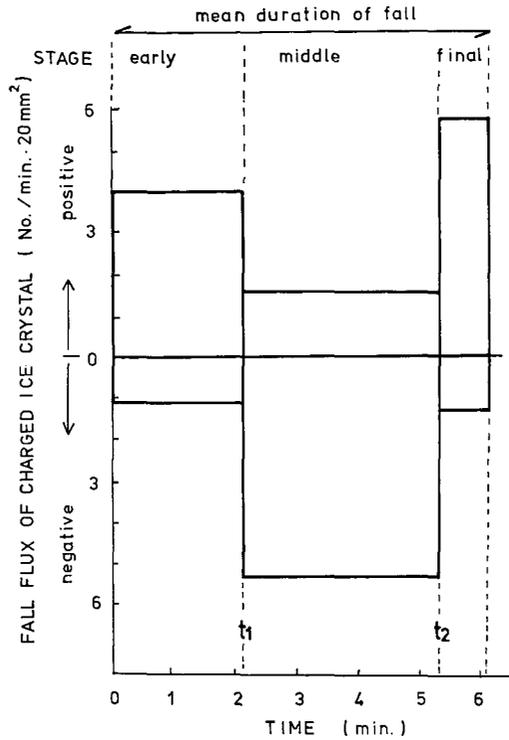


Fig. 8 Prevailing sign of electric charge of ice crystals in transition normalized with two reversal times.

In the early and final stages of the average duration of ice crystal fall, the ratio of positive to negative charge frequency were 3.4:1 and 4.9:1 respectively, where the ice crystal charge was positive predominantly, and in the middle stage, the ratio was 1:3.4, where the crystal charge was negative predominantly. In these experiments, vapour was supplied continuously and ice crystal fall was begun by seeding and stopped after settling. In the early and final stages, ice crystals were observed to fall fast and the size of them are considered to be larger than that in the middle stage. In the middle stage, ice crystals were observed to fall slowly but frequently, and size and number are considered to be smaller and more abundant respectively as compared to that in the early and final stages.

4. Considerations

In the case of time changes of charge distribution as shown in Fig. 5, it is seen that large ice crystal carried positive charges in the early stage and small ice crystals carried negative charges in late stage. These results are considered to be in good agreement with those reported by Endoh (1981). In other case, such a good agreement could not be found. It is considered to be caused by the dull time resolution in these experiments. However it can be found that large and small particles fell in the early and late stages respectively, and carried predominantly positive and negative charges respectively, even though these were held partially or incompletely. Furthermore, in every case, most of ice crystals were found to be distributed around the zero point. These tendencies can be seen in the experimental results reported by Magono and Iwabuchi (1972). They reported that ice crystals whose charge could not be detected occupied 50–80% of all crystals. It is considered to be caused due to an effect of the recombination between charged ice crystals and surrounding charge carriers with opposite sign produced in charge separations.

In the experiments for obtaining a sharper time resolution, although only the signs of large quantities of charges on ice crystals were recorded ignoring the small ones, a tendency of time changes was seen in every case and summarized into Fig. 8. It may be described as follows. In the early and final stages and the middle stage, ice crystals were found to predominantly carry positive and negative charges respectively in almost all cases, and the size of ice crystals could be estimated as relatively large and small in the former and latter cases respectively according to the difference of their falling speed. These difference may be attributed to moisture conditions. In these experiments, water vapour was supplied continuously. The moisture condition in the early and final stages, and the middle stage of these three stages may be considered the correspond to the early and late stages in clearly mentioned experiments and the former experiment carried by Endoh (1981). According to Fig. 6, the fate of ice crystal situation in a cycle is described as follows.

As the cloud chamber filled with steam clouds, which are cooled and immediately become supercooled droplets, seeding is done at upper part of the chamber. As seeding is always made in the state of local over-seeding, the seeds are concentrated partially at the beginning. As they gradually spread, and the front of the seeds touches dense supercooled droplets beneath them; then they grow abruptly and begin to fall to the bottom. On their down way, they scavenge many supercooled droplets completely and become

larger and larger. This is considered to correspond to the early stage of the cycle, where relatively large ice crystals were found. Since succeeding ice crystals cannot entrap any supercooled droplets and grow anymore, their size and falling speed are relatively small, although the number density is high. These ice crystals are considered to correspond to that found in the middle stage of the cycle. Finally small residual fraction of ice crystals may be considered to be overtaken by freshly introduced supercooled droplets and can gain sufficient moisture again and grow quickly. These crystals are considered to correspond to that in the final stage.

According to the above-mentioned situation of moisture and signs of ice crystal charges, while it may be considered that a growing ice crystal carries preferentially positive electric charges, as it grows slowly or begins to evaporate, it has a tendency to carry negative charges.

5. Discussions

An experiment in a similar condition to the present work was carried out by Magono and Iwabuchi (1972). They measured electric charges on several ice crystals in a cycle and repeated it in many cycles. They showed that in general, ice crystal charges predominate negatively. In the middle stages as shown in Fig. 8, ice crystal charges predominated negatively, the duration time was longer than other stages and number density of ice crystals was higher than those. Therefore, a probability of sampling in that stage may be considered to be high for such random sampling procedures. Furthermore, they showed that columnar type crystals had a tendency of carrying positive charges. It may be considered that since the falling speed of columnar type crystals are generally high, measurement tend to be done in the final stage where the ice crystal charge predominate positively.

Some experiments above a charge generation of a fixed bulk ice of frost were done by Findeisen (1940), Findeisen and Findeisen (1943) and Takahashi (1973). Findeisen and Findeisen (1943) showed that growing frosts on metal in high humidity carried positive electric charges and ejected some splinters, and also carried negative charges as they began to evaporate. These results seems to be in agreement with present results, but the temperature of their experiments was too low to run a comparison. Takahashi (1973) reported that ice surface was positively and negatively charged in their growing and evaporating courses. This is in good agreement with the present results except for the difference in size. Hallett and Saunders (1979) measured the electric

charge of a rimer in association with secondary ice production. In their experiment, the rimer carried positive charges as long as steam was supplied, and as the steam was stopped it carried negative charges. It may be considered that some common mechanisms of charge generation probably exist throughout the above-mentioned experiments which are in agreement with in the relation between humidity conditions and signs of ice charges.

The electric charge of individual ice crystal in winter fog (diamond dust) was measured by Endoh (1979) at Inuvik, Canada. It was observed that at an early stage of a time series of fog in the form of a wave pattern arrived, the electric charges of ice crystals were predominantly positive and the concentration and size of ice crystals were relatively high and small respectively, and that, in contrast, in the late stage, electric charges of ice crystals were predominantly negative and the concentration and size of ice crystals were relatively low and large respectively.

The relation between the sign of electric charge of ice crystals and its moisture conditions deduced from the difference of growing stage of ice fog seems to be in good agreement with the results of the present experiment, although the conditions of air temperature were different.

Regarding some observational results of electric charge distribution in winter thunderclouds of Japan, positive charges were frequently observed in the lower half of the cloud depth, above freezing level where moisture was considered to be relatively abundant and precipitation was considered to be only solid particles. On the other hand, at the upper part of cloud and beneath the cloud base, negative charges were often observed, where the moisture condition was considered to be subsaturated to ice, and the snow crystals seemed not to grow or to begin to evaporate.

From the results of many early observations of snow crystal charge on the earth surface, it seems appropriate to consider that clean snow crystals without riming often carried negative charge because they were destined to fall through the subsaturated layer beneath the cloud base, and measured in the subsaturated air on the earth surface.

6. Conclusions

Electric charges of ice crystals produced in the laboratory were measured successfully without direct touching by means of application of a horizontal electric field in two kinds of procedures.

In the experiment for obtaining transitional charge distribution, it may

be concluded separately that large and small ice crystals fell in the early and late stage respectively and carried predominantly positive and negative charge respectively. Most of the ice crystals did not carry any detectable charge. This seems to be due to an effect of recombination loss during their free fall.

In the experiment, where water vapour was supplied continuously, it was found that large ice crystals carrying positive charges fell in the early and final stages, and numerous minute ice crystals carrying negative charges fell in the middle stage. These differences may be considered to be attributable to moisture conditions. The moisture conditions in the early and final stages, and the late stage of these three stages may be considered to correspond to the early and late stages in the former experiment respectively.

Since the difference of ice crystal size may be considered to depend on the condition of moisture surrounding ice crystals, it may be concluded that ice crystals in the growing stage preferentially carry positive electric charges, on the other hand, ice crystals in the evaporating or slowly growing stages have a tendency to carry preferentially negative charges.

Such properties of electrification of ice crystals were reconfirmed in ice fog and also found in winter thunderclouds. Especially beneath and around a cloud, negative charges are estimated to predominate due to the evaporation. It may be considered to be the reason why the electric charge of snow crystals which were measured at the ground surface, were predominantly negative on the average. Accordingly, it may be concluded that since no riming processes were detected and were not considered in this experiment, the results of this experiment coincide and support that of the former experiment by Endoh (1981).

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