Further analysis of the effects of supersaturation on graupel charging – modeling study

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ABSTRACT: Charge transfer measurements in laboratory studies of ice crystal interactions with riming graupel show a dependence of the sign of charging on the cloud temperature and effective liquid water content. The charge sign appears to be influenced by the relative vapor diffusional mass growth rates of the ice surfaces, which is controlled by the cloud supersaturation experienced by each ice surface. In particular, the two surfaces may experience different supersaturations from each other if regions of the cloud with differing supersaturations are involved in mixing processes. This study investigates the effects of cloud supersaturation on the details of the charge sign regime. It points to the need for considerations of cloud supersaturation to be included in our understanding of thunderstorm charge generation.

INTRODUCTION

Since Reynolds et al (1957) showed that charge transfer when ice crystals rebound from riming graupel offers a viable thunderstorm charging process, there have been many confirmatory laboratory studies. In particular, Takahashi (1978) and Jayaratne et al (1983) noted a charge transfer regime in which graupel charges positively or negatively depending on the cloud liquid water content and the temperature, the equal and opposite charge sign residing on the separated ice crystals. Thus, the classic thunderstorm dipole may be achieved, when positive ice crystals are carried aloft while the negative graupel falls. Jayaratne et al (1983) pointed out that the opposite, positive, charging of graupel in lower, warmer, cloud regions could account for the lower positive charge, giving agreement with the tripole model discussed by Williams (1989).

Baker et al (1987) suggested that the sign of the charge transfer is controlled by the relative diffusional growth rates of the two interacting ice surfaces, with the faster growing ice surface charging positively. The ice crystals grow by vapor diffusion from the environment while the graupel surface also grows from this far field source but, in addition, grows by deposition of vapor from recently arrived supercooled droplets whose temperature rises to $0^{o}C$ while they freeze on the surface - the temperature difference between the freezing droplets and the colder rime surface drives additional vapor to the graupel. Williams et al (1991) pointed out that riming also heats the rimer surface, which will reduce the depositional vapor growth. Thus the sign of the charge is influenced by several competing diffusional growth rates that vary with accretion rate, temperature and cloud supersaturation.

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Laboratory studies by Saunders et al (1991) and Saunders and Peck (1998) of charge transfer when crystals rebound from riming-graupel, determined a charge sign reversal line on an EW vs Temperature plot, as shown in red in Figure 1. The rimer charges positively at high EW, and negatively at low EW. EW is the effective liquid water content, being the mass collected on the riming target determined by weighing the accreted rime or, as in Keith and Saunders (1990), by the heat rise of a similar target moving through the cloud with the riming target.

Saunders et al (1991) used the data obtained in the Manchester laboratory to generate a series of equations that describe riming graupel and crystal charge exchange, Q femtoCoulombs, for each separating ice crystal as a function of the cloud variables. The charging equation involves the relative graupel/crystal velocity, V, and the size of the crystals, d

$$Q = B\, d^{a}\, V^{b}\, q \quad \text{fC}$$

where the constants B, a and b are functions of crystal size. Charge transfer per ice crystal separation event, q, is obtained from the laboratory experiments obtained with 110 µm diameter ice crystals and an airstream velocity of 3 m s^{-1}. Multiple crystal separation events give rise to a charging current to the ice covered metal riming target connected to an electrometer. Then q is calculated from a knowledge of the crystal concentration in the cloud, determined with a continuous crystal sampler, together with values of the Event Probability, which is a measure of the ice crystal collision and separation probability (Keith and Saunders, 1989), with a typical value of order 0.1.

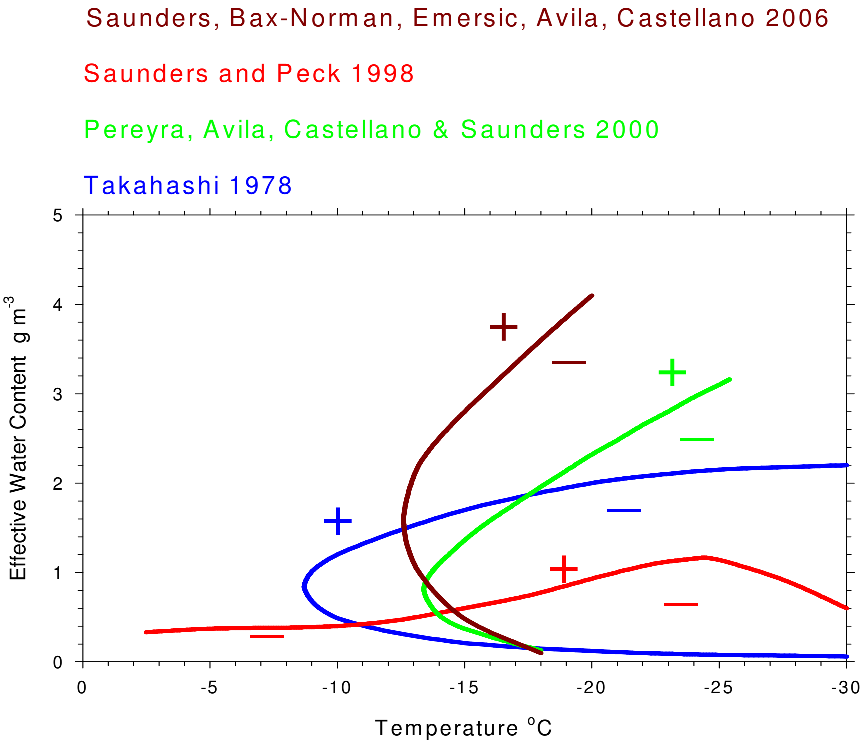


Figure 1 Graupel charge sign reversal lines from various laboratory studies. The Manchester 1991 line is red. The charge sign reversal line obtained by Takahashi (1978) is blue. Subsequent laboratory studies by Pereyra et al (2000) (green) and Saunders et al (2006) (brown) produced other charge sign reversal lines.

Saunders et al (2004) put forward a possible explanation for the differences in the characteristics of the reversal lines based on the different laboratory techniques used in the various studies. The red line results occur when the ice crystals are grown in the same cloud chamber as the droplets used in the charge transfer riming experiments (the "one-cloud" case). The higher EW charge sign reversal lines on Figure 1 occur when the ice crystals are mixed into a cloud of droplets that has not been vapor depleted by growing ice crystals - the crystals are drawn to the target and mixed with droplets just before they encounter the riming target. An example of this "two-cloud" mixing apparatus is shown in Figure 2. In this way, the supersaturations experienced by the crystals and rime at the moment of impact are not necessarily the same. In particular, in this "two-cloud" case, the growing crystals remove water vapor from their local droplet cloud and so reduce the supersaturation. When these crystals are mixed into the undepleted droplet cloud, the crystals experience an increased growth rate that favors their becoming positively charged and the graupel negatively charged, according to Baker et al (1987).

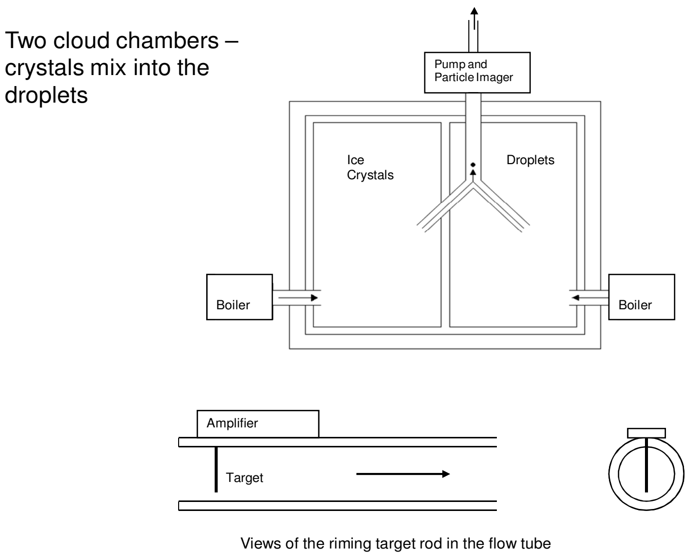


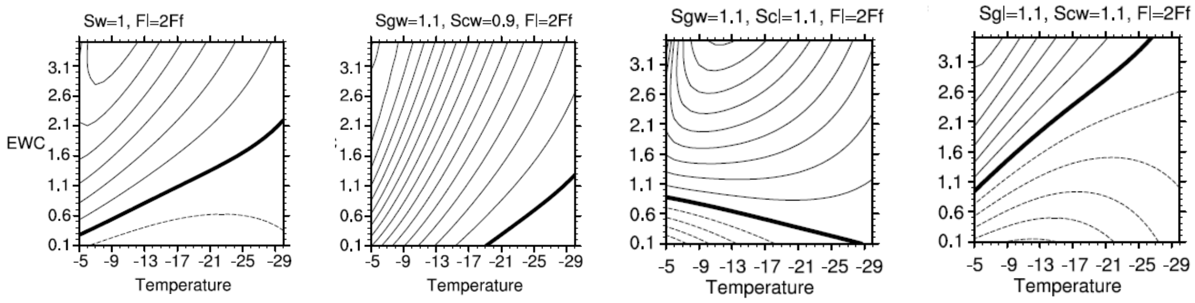
Figure 2 Laboratory cloud chamber for ice crystal collisions with a riming target following cloud mixing. The ice crystals grow in their own cloud of supercooled droplets and deplete the available vapor. The separate droplet cloud is not depleted by vapor loss to ice crystals.

The results are qualitatively explicable in terms of the relative growth rates of the interacting ice surfaces - at high cloud supersaturations, the ice crystals grow faster than the graupel surface and so charge positively, with the rimer charging negatively according to the Relative Growth Rate hypothesis. This mixing effect pushes the charge sign reversal line to higher values of EW. High temperature positive rimer charging in the mixing cloud cases, see Figure 1, is caused by high droplet cloud supersaturation causing rapid rimer growth. On the other hand, Saunders et al (2006) and Emersic and Saunders (2010) showed that by reducing the vapor content to the crystal cloud, then mixing the crystals into the droplet cloud, the crystals experienced a sufficiently high enhanced growth rate to cause negative rimer charging at temperatures as high as -5^{o}C.

The objective of the present work is to use the relative growth rate model to determine values of supersaturation that correspond to the various charge sign regions on an EW/T plot. The plots below in Figure 3 show the "one-cloud" case together with three "two-cloud" scenarios that correspond to a positive rimer charge region at high temperature, a positive region at high EW, and a predominantly negative zone.

ANALYSIS

Mitzeva et al (2005) and Tsenova et al (2009) calculated the diffusional growth rates of the interacting graupel surface and ice crystals for a range of cloud conditions and supersaturations from which they determined the relative diffusional growth rates of the ice surfaces. The relevant equations are given in the Appendix. They showed that for a water droplet cloud at water saturation, that ensured ice supersaturation so both ice surfaces grew from the vapor, the accreting graupel charged positively or negatively according to the values of cloud temperature and EW. The charge sign reversal line on an EW/T plot has some similar characteristics to the reversal line obtained in the laboratory work of Saunders et al (1991) and is the first graph shown in Figure 3, corresponding to the red line in Figure 1. Other cloud supersaturations give rise to various results on an EW/T plot. Figure 3 shows some of these model results where the charge sign reversal lines correspond to various cloud conditions in laboratory experiments. The first figure is for the results obtained in Manchester in which the ice crystals and droplets are in the same cloud as the riming target - so called "one cloud" experiments. The other three figures are for the mixing case, "two cloud" experiments where the rimer and the crystals experience different supersaturations; Sg for graupel, Sc for ice crystals. w and i refer to the saturation ratio with respect to water or ice. Fl is the local, rimer surface, ventilation coefficient, while Ff is the far field ventilation coefficient.



a) "One-cloud" b) "Two-cloud" c) "Two-cloud" d) "Two-cloud"

Positive at high T and low EW

Positive at high EW

Negative

Figure 3 Charge sign reversal lines for various graupel and crystal supersaturations. Positive rimers, continuous lines. Negative rimers, dashed lines.

DISCUSSION

Charge transfer reversal lines were determined for a wide range of supersaturation values for crystals and graupel and the results were presented by Mitzeva et al (2005). The general conclusion from those studies is that increasing the ice crystal supersaturation leads to more negative graupel; that increasing the graupel supersaturation favors positive graupel and increasing both crystal and graupel supersaturations leads to more negative graupel. All these results are in accord with the Relative Growth Rate hypothesis that the faster growing ice surface charges positively. In the latter case, the small crystals will grow faster than the larger graupel.

Some supersaturation values have been selected here in order to illuminate the importance of supersaturation to particle charging in thunderstorms. In the "one-cloud" case, shown in Fig 3a, where the saturation ratio with respect to water is one (Sw=1) for both the rimer and the crystal surfaces, the calculated reversal line has some similarity to the results of experiments in which the crystals and droplets coexist in the same cloud. (The red line in Figure 1) At lower temperatures the line deviates from experiment indicating that the saturation ratio in the laboratory cloud was greater than 1. The assumption that Sw is constant across the EW/T space is clearly a simplification.

In the three figures for simulated "two-cloud" cases, the reversal lines are highly dependent on the assumed saturation values in the cloud. The values of saturation have been chosen to help identify the cloud conditions in the laboratory that give rise to the results shown in Figure 1. Figure 3b shows positive rimers at high temperatures and low EW corresponding to all the "two-cloud" cases in Figure 1. Both rimer and crystals are supersaturated with respect to ice, so are growing, but the graupel experiences the higher supersaturation, so grows faster and charges positively. This result indicates that in the low growth regimes at high temperature and low EW, the mixing process accelerates graupel growth rather more than crystal growth. The "one-cloud" and "two-cloud" results are in agreement that rimers charge positively at high temperatures, however there is disagreement about the sign of charging at very low EW values, as can be seen in Figure 1. This low EW region of EW/T space needs further laboratory investigation as it may indicate that the "two-cloud" case involves an unrealistically higher graupel growth rate than would be experienced in the saturation conditions in real clouds at very low values of EW.

In Figure 3c the increased crystal growth leads to negative rimer charging at low EW while at high EW the rimer is positive. The reversal line is at lower EW than in Figure 1 showing how mixing favors positive rimers under these conditions. In Figure 3d, the negative zone is extensive due to the crystals, with higher supersaturation, growing faster than the rimer. Here, mixing has enhanced crystal growth.

CONCLUSION

These results will help identify supersaturation conditions in thunderstorms necessary for positive or negative graupel. It is clear that when ice crystals and droplets are well mixed and experience similar supersaturations, the "one-cloud" experiments provide a valid simulation of real clouds. Regions in clouds where ice crystals and riming graupel experience different growth conditions may lead to charge transfers whose sign is highly dependent on local cloud supersaturation values.

APPENDIX

1. *Diffusional growth rate of graupel from the far field* (growth rate of graupel particle from diffusion of water vapor from the surrounding environment) – (Pruppacher and Klett, 1978):

$$\frac{dM_g}{dt}(f) = 4\pi C D_v F_f \left(\rho_a(T_a) - \rho_i(T_g)\right)$$

C – graupel shape factor assumed to be spherical, and $C = R$,

D_v – vapor diffusion coefficient,

F_f – ventilation coefficient for the far field, and

$$F_f = \begin{cases} 1 + 0.108(\mathrm{Sc}^{1/3}\,\mathrm{Re}^{1/2})^2 & \text{if } \mathrm{Sc}^{1/3}\mathrm{Re}^{1/2} < 1.4 \\ 0.78 + 0.308(\mathrm{Sc}^{1/3}\,\mathrm{Re}^{1/2})^2 & \text{if } \mathrm{Sc}^{1/3}\mathrm{Re}^{1/2} \geq 1.4 \end{cases}$$

Re, Sc and Pr – Reynolds, Schmidt and Prandtl numbers

$\mathrm{Re} = \frac{2R_g.V}{\gamma_a}$, $Sc = \frac{\gamma_a}{D_v\sigma_a}$, $\mathrm{Pr} = \frac{\gamma_a}{K}$, where σ_a, γ_a, and K are respectively air density, viscosity, and thermal conductivity.

$\rho_a(T_a)$ - cloud equilibrium vapor density with respect to water,

$\rho_i(T_g)$ - equilibrium vapor density over graupel surface,

T_g is the spherical graupel surface temperature, from the theoretically obtained Macklin and Payne (1967) relation between T_g and cloud effective water content *EW*, temperature T_a and spherical graupel velocity *V*:

$$EWV\frac{\left[L_f + c_w(T_a - T_0) + c_i(T_0 - T_g)\right]}{4} = \chi \mathrm{Re}^{0.5}\frac{\left[\mathrm{Pr}^{1/3}K(T_g - T_a) + Sc^{1/3}D_v L_v\left(\rho_i(T_g) - \rho_a(T_a)\right)\right]}{2R_g},$$

where L_f and L_v are respectively latent heat of freezing and vaporization, c_w and c_i – heat capacities of liquid water and ice. The numerical factor χ depends on the turbulence in the air stream and on the roughness of graupel surface (Macklin and Payne, 1967). It is determined based on Avila et al., 1998, for spherical graupel:

$\chi = 0.6 + 0.83\exp\left(-5.17\times10^{-7} r_d^4 V\right)$, r_d – droplet size.

2. *Diffusional growth rate of graupel from the local field* (growth rate from diffusion of water vapor from the freezing supercooled cloud droplets on the graupel surface).

The expression used for the diffusional growth rate of graupel from the local field is derived from the expression for the total mass lost by vapor transfer during the freezing of a droplet in Baker et al (1987):

$$\frac{dM}{dt} = 3 f_l \pi D_v a(\rho_0 - \rho_i)\tau_f \text{ , } a=2^{1/3} r_d$$

To determine the growth rate of the spherical graupel due to all frozen supercooled cloud droplets on its surface, the above expression is multiplied by the number of colliding graupel surface droplets with time, $N = \frac{3 S_g V.EW}{4\sigma_w a^3}$.

Thus, the growth rate of graupel from diffusion of water vapor from the freezing of cloud droplets on its surface is:

$$\frac{dM_g}{dt}(l) = 3\pi F_l D_v \left(r_d \sqrt[3]{2}\right)\left(\rho_a(T_0) - \rho_i(T_g)\right)\left(\frac{3S_g V.EW}{4\pi\sigma_w r_d^3}\right)\tau_f \text{ ,}$$

where S_g is the graupel projected area, σ_w is water density, τ_f is the freezing time of a droplet on graupel surface, F_l is the ventilation coefficient for the local field and is evaluated from the engineering chemistry relation between the heat transfer coefficient f_h and the Nusselt (Nu) number. Nu is determined as in Incoprera and Dewitt 1996:

$$F_l = \chi \mathrm{Re}^{0.6} \mathrm{Pr}^{1/3}$$

The freezing time τ_f is determined from Pruppacher and Klett (1978):

$$\left(2\pi.\left(L_v D_v F_l\left(\rho_w(T_0) - \rho_w(T_a)\right) - F_l K T_a\right)\right)\tau_f - \left(T_g K_i r_d \sqrt{\frac{\pi c_i \sigma_g}{K_i}}\right)\sqrt{\tau_f} - \frac{2\pi}{3} r_d{}^2 \sigma_w L_m c_w T_a = 0$$

where K_i is the thermal conductivity of ice, L_m – the latent heat of melting.

3. *Diffusional growth rate of spherical ice crystals* (Pruppacher and Klett, 1978):

$$\frac{dM_{cr}}{dt} = F_c \frac{4\pi R_{cr}(S_i - 1)}{\frac{L_s^2}{K R_v T_a^2} + \frac{R_v T_a}{E_{icr} D_v}}$$

In this work, as ice crystals are relatively small, it is accepted that their ventilation coefficient $F_c = 1$. L_s is the latent heat of sublimation, E_{icr} – the equilibrium vapor pressure over the ice crystal surface, S_i is the saturation ratio of water vapor with respect to ice in the environment, $S_i = \frac{E_w}{E_{icr}(T_a)}$.

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