

## A Possible Role of Ice-Forming Nuclei in Rain Formation

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### ABSTRACT

Population of cloud droplets in a cloud may be decreased under some circumstances by their removal (along with other particles) by an ice crystal growing on an ice-forming nucleus. Further condensation of water vapor should take place on remaining cloud droplets, which should consequently grow to larger sizes.

### 1. Introduction

The presence of ice in a cloud is required for the formation of rain by the Bergeron process. Ice crystals can appear in a cloud as droplets freeze, as larger freezing drops splinter, or as growth around ice-forming (sublimation) nuclei. The rate of growth of an ice crystal depends primarily on diffusion of water vapor and to a much lesser extent on the rate of release of the heat of crystallization (Marshall and Langeben, 1954);

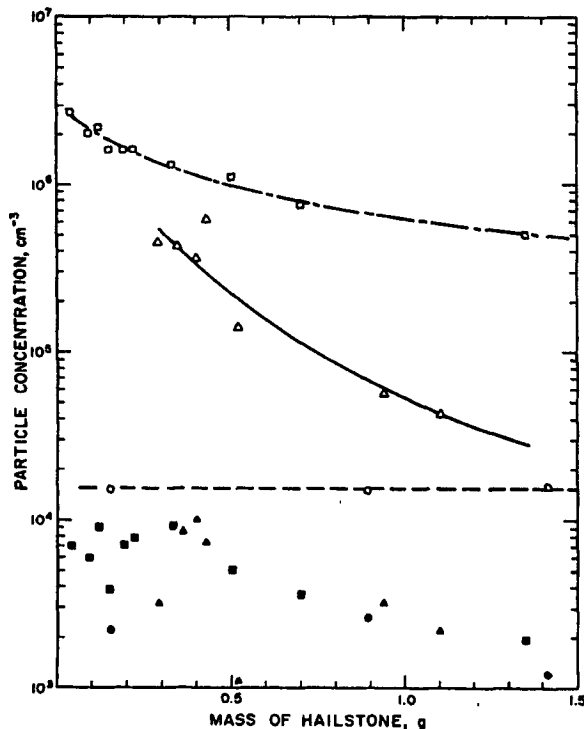


FIG. 1. Concentration of water-insoluble particles in different types of hailstones. Hailstone types: milky (squares), mixed (triangles) and transparent (circles). Particle size range:  $1.5 \leq d < 3$  (open symbols) and  $9 \leq d < 11$  (closed symbols).

during growth of the ice crystal, diffusiophoresis and Stefan flow (Davies, 1967) are operating, and will cause removal of aerosol particles from the gaseous phase.

### 2. Removal of aerosol particles

Let us consider evidence gathered from laboratory and field experiments to assess the degree of removal of aerosol particles from air within a precipitating cloud. Fig. 1 shows the number of water-insoluble particles captured by different types of hydrometeors during different thunderstorms in 1966 in Colorado. The highest concentration of  $1.5\text{--}3\ \mu$  diameter particles was found in milky ice (graupel) collected within a pre-

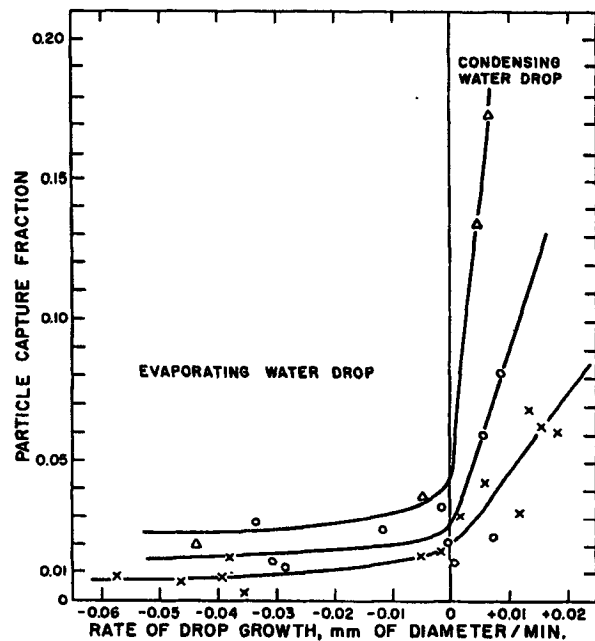


FIG. 2. Capture efficiency of condensing and evaporating water drops for different aerosol particles. Diameter ( $\mu$ ): 1.9 (ZnS, crosses); 0.37 ( $\text{TiO}_2$ , circles); 0.05 (AgI, triangles).

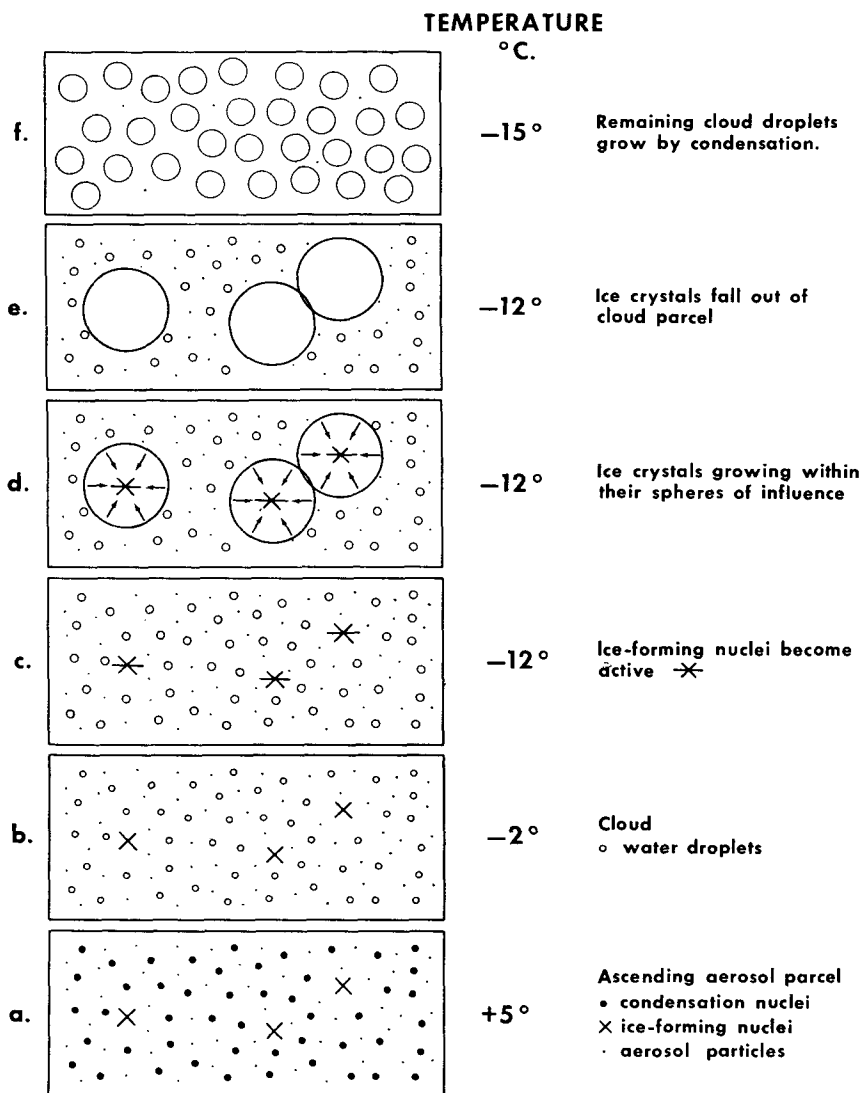


FIG. 3. Growth of cloud droplets according to the present model.

cipitating cloud; the concentration was as high as  $3 \times 10^6$  particles  $\text{gm}^{-1}$ . In contrast, concentrations in clear ice, corresponding to frozen raindrops, were  $1.5 \times 10^4$  particles  $\text{gm}^{-1}$ . The masses of rimed ice crystals and graupel were between  $10^{-4}$  and  $2 \times 10^{-3}$  gm. Therefore, 300–6000 particles could be present per ice agglomerate. Higher concentrations than those present in milky ice were found in snowflakes collected during 1967 snowstorms in Winter Park, Colo. The concentration of 1.5–3  $\mu$  diameter particles ranged from 10 to  $1.2 \times 10^4$  per  $4 \times 10^{-4}$  gm in average rimed snow crystals. This corresponds to  $3 \times 10^7$  particles  $\text{gm}^{-1}$  of ice at the beginning of the storm and  $2.4 \times 10^4$  particles  $\text{gm}^{-1}$  near the end. Concentrations of 300 particles per ice crystal were very common in a snowstorm. It should be realized that not all particles were captured as single particles by phoretic forces. Cloud droplets and particle-rich residues from evaporated drops could be removed by an ice crystal by

other scavenging processes, especially since the evaporating supercooled droplets are in motion in a polarized diffusion field.

It is not known to what degree the removal of 9–11  $\mu$  diameter aerosol particles is caused by phoretic forces. The concentration of these particles in the air was about one-tenth and their concentration in ice about one-hundredth that of 1.5–3  $\mu$  particles, indicating the lack of an enhanced scavenging mechanism. The scattered points for the 9–11  $\mu$  diameter particles show that the removal of these particles is independent of the type of ice phase. It is concluded that 9–11  $\mu$  cloud droplets will not be removed effectively by phoretic forces.

### 3. Laboratory studies

Fig. 2 shows the results of a laboratory experiment on the capture of electrically neutral aerosol particles

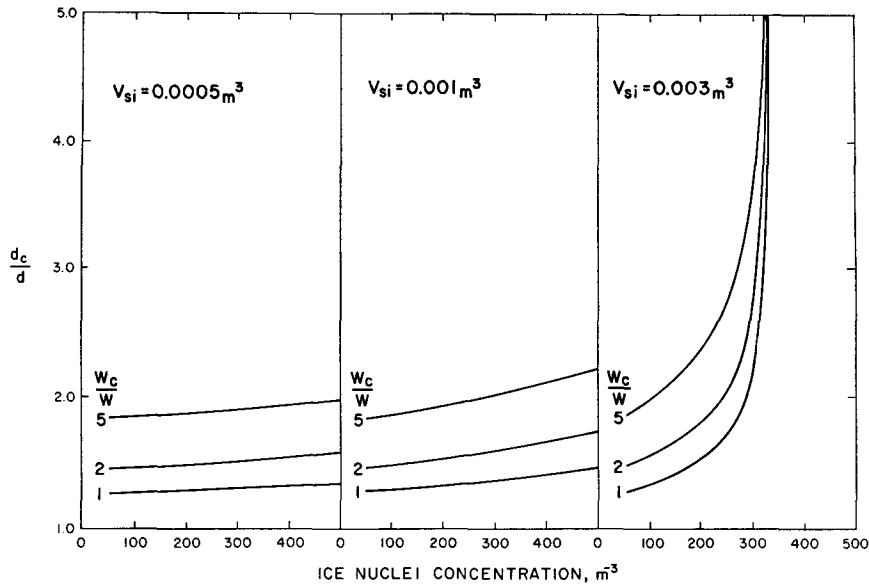


FIG. 4. Change in size of cloud droplets as a function of ice nuclei concentration, liquid water content, and volume of the sphere of influence.

by condensing and evaporating water drops. Water drops were mounted on the end of a gold wire and their size-history followed with a low-power, long-focus microscope fitted with a calibrated filar micrometer. Drop growth was controlled by cooling the drop, humidifying the air, and lining the vessel wall with damp blotting paper. Evaporation of the drop was accomplished by heating it with a nichrome wire micro-heater. Experiments with zinc sulfide particles of  $1.9 \mu$  mean-volume diameter are reported by Rosinski *et al.*, (1963). Most of the experiments with titanium dioxide ( $0.37 \mu$ ) and silver iodide ( $0.05 \mu$ ) particles are reported elsewhere (Rosinski, 1961). The capture efficiency is the ratio of the number of particles captured by a water drop to the number of particles passing through the same cross-sectional area. The rate of removal of aerosol particles by phoretic forces around an ice crystal within a cloud should be larger than the rate observed for a water drop under similar conditions. Ice crystal growth takes place in an environment corresponding to supersaturation with respect to ice; therefore, supercooled cloud droplets in the vicinity of an ice crystal evaporate, creating conditions favorable for rapid growth and strong phoretic effect (Vittori and Prodi, 1967).

#### 4. Theoretical analyses

Let us consider now a slowly ascending parcel of aerosol (Fig. 3a). Condensation of water takes place first on condensation nuclei and then on cloud droplets. In a natural cloud, a spectrum of drop sizes results. However, in this discussion let us assume the result to be a monodispersed cloud (Fig. 3b). Ice-forming nuclei active at temperature  $T$  ( $T = -12^\circ\text{C}$ , Fig. 3c) originate ice crystals, which will grow at this temperature and,

because of the assumed slow ascent, will first use condensable water vapor and then vapor of evaporating cloud droplets as vapor sources for growth. During this growth, all small aerosol particles (including inert particles, condensation nuclei left behind after complete evaporation of cloud droplets, ice-forming nuclei active at lower temperatures, and cloud droplets) are removed by phoretic and other scavenging mechanisms from a volume around each growing ice crystal. This volume is referred to as the "sphere of influence" (Fig. 3d). A similar, but much smaller, sphere of influence exists around each condensing cloud droplet.

Ice crystals grow to a certain size and fall out of the cloud parcel (Fig. 3e). Upon further ascent of the parcel, condensation of water vapor takes place on the remaining cloud droplets (Fig. 3f), distributed within the cloud by turbulence. The liquid water content balance for one cubic meter of cloud after the action of the ice nuclei can be expressed as

$$C(1 - N_T V_{si}) \frac{\pi}{6} d_c^3 \rho = (1 - N_T V_{si}) W + W_c, \quad (1)$$

where

- $C$  = concentration of cloud droplets ( $\text{m}^{-3}$ ),
- $N_T$  = concentration of ice nuclei, active at temperature  $T$  ( $\text{m}^{-3}$ ),
- $V_{si}$  = volume of the sphere of influence ( $\text{m}^3$ ),
- $d_c$  = diameter of cloud droplets after condensation (cm),
- $\rho$  = density of water ( $\text{gm cm}^{-3}$ ),
- $W$  = initial liquid water content ( $\text{gm m}^{-3}$ ), and
- $W_c$  = condensed liquid water ( $\text{gm m}^{-3}$ ).

The diameter of droplets before appearance of the ice phase in a cloud is

$$d = \left[ \frac{W}{0.524C\rho} \right]^{1/3}. \quad (2)$$

The diameter of cloud droplets after ice crystals have ceased growing is, from (1)

$$d_c = \left[ \frac{(1 - N_T V_{si})W + W_c}{0.524C(1 - N_T V_{si})\rho} \right]^{1/3}, \quad (3)$$

and the change in diameter of cloud droplets,

$$\frac{d_c}{d} = \left[ 1 + \frac{W_c}{(1 - N_T V_{si})W} \right]^{1/3}. \quad (4)$$

This is given in Fig. 4 for three different values of the sphere of influence.

The unknown volume of  $V_{si}$  can be estimated from the field experiment. The measured concentration of 1.3–3  $\mu$  aerosol particles near some clouds was approximately  $10^5 \text{ m}^{-3}$ , and the concentration is assumed to be similar initially in the clouds. Assuming 300 aerosol particles were captured by one ice crystal formed around one ice nucleus, the sphere of influence is equal to  $300 \times 10^{-5} \text{ m}^3 = 3 \times 10^{-3} \text{ m}^3$ .

Let us consider as an example a cloud with  $1 \text{ gm m}^{-3}$  liquid water content distributed over  $2.4 \times 10^8$  cloud droplets of 20  $\mu$  diameter. For an ice-forming nuclei concentration of  $300 \text{ m}^{-3}$ , and if  $V_{si} = 3 \times 10^{-3} \text{ m}^3$ , cloud droplets will grow to 44  $\mu$  diameter upon condensation of  $1 \text{ gm m}^{-3}$  of water vapor. The concentration of the remaining cloud droplets is  $(2.4 \times 10^8) - (3 \times 10^{-3} \times 2.4 \times 10^8 \times 300) = 2 \times 10^7 \text{ m}^{-3}$ .

In reality, cloud droplets are not uniform. Smaller ones evaporate preferentially; therefore, formation of droplets larger than those calculated should take place. On the other hand, not all cloud droplets within the sphere of influence will be destroyed, so the concentration of cloud droplets will be larger than calculated and the droplets will not be as large as calculated. The ice agglomerate in the example above could grow to  $3 \times 10^{-3} \text{ gm}$  by using all available cloud droplets in its sphere of influence. The terminal velocity of graupel of this size would be approximately  $2.2 \text{ m sec}^{-1}$ . Before a graupel reached this size, it would settle through different parcels of cloud and, therefore, its sphere of influence would express the integrated effect of its entire lifetime.

When the concentration of ice nuclei is low, the fraction of the total population of aerosol particles and cloud droplets removed by ice crystals will be so small that turbulent diffusion will restore original conditions.

When ice nuclei concentration is high, many ice crystals will form, all droplets will evaporate rapidly, and scavenged particles will be distributed among a large number of crystals.

Ice-forming nuclei active at a lower temperature than  $T$  are, of course, usually present in the clouds. Some of them will be removed (at temperature  $T$ ) by growing ice crystals; the concentration of those remaining depends on their temperature-concentration spectrum. The concentration of ice-forming nuclei active at a lower temperature than  $T$  should be as low as possible to eliminate the formation of new cloud particles and to minimize the increase of the cloud particle population after the scavenging action of ice-forming nuclei at temperature  $T$ . If this concept is correct, ice-forming nuclei active at low temperatures are of no importance in this mechanism of rain formation. The equation indicates that there is a critical value for  $N V_{si}$ . It seems that the sphere of influence changes much less than the concentration of ice-forming nuclei, and the concentration  $N$  therefore governs the growth of cloud droplets for a given  $W_c/W$  ratio.

It should be pointed out that velocity of updrafts and turbulence within a cloud, microturbulence within the sphere of influence, entrainment of moist air, and temperature distribution within an ascending cloud should be taken into account in evaluating formation of large cloud droplets in the presence of the ice-forming nuclei. Calculation of cloud droplet growth by collection (stochastic process) should be combined with calculation of diffusional growth for clouds containing ice nuclei. If the concept presented here is correct, ice nuclei within a cloud create conditions for diffusional growth of cloud droplets, and may originate rain under favorable circumstances. Two problems to be resolved are: 1) whether and under what circumstances this takes place in natural clouds, and 2) whether it is possible to induce this process of rain formation artificially.

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