Solid Precipitation in Super-cooled Cloud : Part1

—Freezing of a Supercooled Water Drop due the Collision of Ice Crystals—

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The freezing-probability of a supercooled water drop, which grows by the capture of cloud droplets, due to the collision of ice crystals formed on ice nuclei was numerically studied in clouds with various conditions. In all cases, the probability increases about 100 times during staying of the drop at cloud top and most of collisions with ice crystals take place when the drop stays near cloud top. The accumulated number of ice crystals colliding with the drop increases with the vertical velocity of air-current and the height of cloud top. The accumulated number of colliding ice crystals is larger in clouds of small water content of cloud droplets than in clouds of large water content after a sufficiently long time, although it tends to increase with the water content in early stage of the growth of the drop. Most favourable water content for the collision is dependent upon the velocity of updraft and the time length. In clouds which are lower than 3000 m in the height of cloud top and which are a few m/s in the velocity of updraft, at most one drop of 10 to 100 supercooled water drops can collide with one ice crystal in 1.0 to 1.5 hour.

1. Introduction

Since Bergeron (1933) suggested that ice phase was required in creating heavy rainfall, his hypothesis has been supported by many meteorologists. However, the true role of atmospheric ice nuclei in the rainfall concerned with ice phase has remained unsolvable. Particularly it is not clarified yet whether or not the freezing of supercooled water drops is closely associated with atmospheric ice nuclei.

In "Project White Top", Koenig (1963) and Braham (1964) observed that the concentrations of ice pellets which existed in the cloud at temperature warmer than -10°C were not related with the concentration of ice nuclei at ground level. They concluded that their observations could be well explained by the measurements of droplet splintering of Mason and Maybank (1960). Further, Koenig (1966) made a study numerically by simulating cloud microphysical processes to test the hypothesis that the drop freezing due to splintering was an important mechanism in cumulus cloud glaciation. He stated that the essential fea-

tures observed in glaciating clouds were simulated if the drop-freezing due to ice splintering mechanism was incorporated in the cloud model.

Meanwhile, in the observation of solid precipitations in the area of the northwest coast of the main island of Japan, Isono et al. (1966) found a close relation between the fallout rate of snow crystals or graupel pellets and the concentration of atmospheric ice nuclei which were active at the temperature of the tops of cloud. They concluded that ice nuclei actually played an important role in the formation of snow crystals and graupel pellets and that their concentration controlled the rate of precipitation, at least, in the case of cold convective clouds.

Thus, the importance of atmospheric ice nuclei in the freezings of supercooled water drops is not made clear yet. This problem will have to be solved quantitatively. In this paper, under the assumption that a supercooled water drop becomes frozen when it collides with an ice crystal associated with an ice nucleus, the freezing probability of the supercooled water drop growing by the

capture of cloud droplets is numerically studied in clouds with various conditions.

The number of ice crystals, colliding with a supercooled water drop as a result of the difference in fall velocities, depends the size of the water drop and the size distribution of ice crystals. The changes with time and height in the size of the drop which, being carried upward by vertical air current, grows mainly due to the capture of cloud droplets, are related with the water content, the size distribution of cloud droplets, the initial size of the drop at cloud base, and the vertical velocity of air current. The size distribution of ice crystals depends on the sizes of ice nuclei, the degree of super-saturation over ice and the vertical velocity of air. Thus, the freezing of the supercooled water drop due to the contact of ice crystals is closely related with many factors mentioned above. For example, the probability of the freezing is perhaps large in a cloud which is associated with the intense updraft because the supercooled drop can continue to be supported in the high layer by air current for a long time where a larger number of ice crystals generally exist. The aim of computations in this paper is to study the effects of those, vertical velocity of air, the water content of cloud droplets, the height of cloud top and the size of a supercooled water drop at the cloud base, on the freezing-probability of the drop (in other words, the number of ice crystals colliding with the drop).

A supercooled water drop containing an ice nucleus can freeze when the drop reaches the temperature at which the ice nucleus becomes active. This probability will also be discussed briefly.

The freezing process originated from ice splintering and spontaneous nucleation are not taken into account here.

2. Governing equations

The computation is carried out under following conditions:

(1) The vertical velocity of air current (w), the water content of cloud droplets (L), the air temperature (T), the height of cloud top above cloud base (H) and the size distributions of cloud droplets and ice crystals all of these, are constant with time and are

horizontally uniform.

(2) Although w and L do not change with height, these values were taken to become zero discontinuously from cloud top to the above layer. The vertical shear of horizontal wind is not considered here.

(3) The air temperature at cloud base is 0°C and the lapse rate is $0.7^\circ\text{C}/100\text{m}$. The air in cloud is always saturated over water.

(4) Each ice crystal was formed on an ice nucleus and it grows only by sublimation. Ice splinters were not taken into account.

(5) The radii of a cloud droplet and an ice nucleus are 6μ and 0.5μ , respectively.

(6) All of ice crystals treated in the computation have generally the shape of circular disk. The mass of each ice crystal is proportional to the square of the radius which is defined to be the radius of the sphere containing the crystal. As obtained empirically by Nakaya and Terada (1935), the proportionality coefficient is 1.52×10^{-3} in the case of plane dendrite if the units of the mass and the radius are gr and cm, respectively. This value is adopted as an approximate one and the relationship between the mass and the radius is expressed by

$$m = 1.52 \times 10^{-3} r^2 \quad (1)$$

where m and r are the mass and the radius of an ice crystal, respectively.

Under these assumptions, the growth rate of a supercooled water drop is given by the following familiar equation

$$\begin{aligned} \frac{dR}{dt} &= \frac{1}{4\rho_w} E_L V L \\ \frac{d}{dt} &= (w - V) \frac{d}{dz} \end{aligned} \quad (2)$$

and the number (N) of ice crystals which collide with the drop in unit time is

$$N = \pi R^2 V \int_{r_0}^{r_1} E_c n' dr \quad (3)$$

where the fall velocities of cloud droplets and ice crystals relative to the air are assumed to be nearly zero. Here,

R : the radius of a supercooled water drop.

V : the fall velocity of the water drop relative to the air which is given by the experiment of Gunn and Kinzer (1949).

ρ_w : the density of liquid water.

E_L : the collection efficiency of cloud droplets with radius of 6μ by the water drop with radius of R .

E_C : the collection efficiency of ice crystals with radius of r by the water drop with radius of R .

$n'dr$: the number density of ice crystals with radius of r to $r+dr$.

r_0 : the minimum radius of an ice crystal.

r_1 : the maximum radius of an ice crystal.

It is clear from the assumptions that, if the vertical velocity of the air is given, the radius of an ice crystal at the level of Z is determined by Z and the activation height (Z_c) of an ice nucleus on which the crystal was formed. Therefore, $\int_{r_0}^{r_1} E_C n' dr$ can be rewritten as follows:

$$\int_{r_0}^{r_1} E_C(r, R) \cdot n' dr = \int_{z_0}^Z E_C[r(z_c, z), R] n(z_c) dz_c, \quad (4)$$

where $n(z_c) dz_c$ is the number density of ice nuclei activated in the layer between Z_c and $Z_c + dZ_c$. Z_0 is the height of cloud base.

When the fall velocities of ice crystals relative to the air are neglected, the number of ice crystals in an air parcel at the level of Z equal to the total number of ice nuclei which were activated in the air parcel until the parcel reaches the level of Z . Therefore, the number density of ice crystals at the level of Z is given by $\int_{z_0}^Z n(z_c) dz_c$ which can be directly obtained from the empirical formula of temperature spectrum on the activation of natural ice nuclei (see Fletcher 1962) as follows;

$$\int_{z_0}^Z n(z_c) \cdot dz_c = A \exp \left[-\frac{\partial T}{\partial z} \beta (z - z_0) \right]. \quad (5)$$

Here A and β are parameters to be determined empirically. The value of β was taken to be 0.6 although it is known to vary from 0.4

to 0.8. The value of A is typically about 10^{-5} /litre, though the variations occur within a range several decimal order. Here, 10^{-5} /litre is adopted for convenience sake. These values of A and β give a concentration at -21°C corresponding to about one activated nucleus per litre.

The growth rate of an ice crystal due to sublimation is given by

$$\frac{dm}{dt} = [p - p_e(T_s)] \frac{1}{R_w T} 4\pi C D \quad (6)$$

and

$$L_s \frac{dm}{dt} = 4\pi C K (T_s - T). \quad (7)$$

Here

P : ambient water vapor pressure

$P_e(T_s)$: the saturation water vapor pressure over ice at temperature of T_s

T : ambient temperature

T_s : the surface temperature of an ice crystal

R_w : the gas constant of water vapor

C : the electrostatic capacity of an ice crystal which is taken to be $(2/\pi)r$

D : the diffusion coefficient of water vapor in air

L_s : the latent heat of sublimation

K : the thermal conductivity of air

If it is eliminated from equations (6) and (7) by using the integral of Clausius-Clapeyron's formula,

$$\frac{dm}{dt} = \frac{4\pi C \delta}{f(T)} \quad (8)$$

$$\delta = \frac{\rho_w^s(T) - \rho_i^s(T)}{\rho_i^s(T)}$$

$$f(T) = \frac{J L_s^2}{K R_w T^2} + \frac{1}{D \rho_i^s(T)}$$

can be induced under the above-mentioned assumption that the air in cloud is saturated over water. J is the mechanical equivalent of heat. $\rho_w^s(T)$ and $\rho_i^s(T)$ are saturation water vapor densities over water and ice, respectively.

The change with height in the radius of an ice crystal is determined by equations (1) and (8) and

$$\frac{d}{dt} = w \cdot \frac{d}{dz} \quad (9)$$

Collection efficiencies E_L and E_c in equations (2) and (3) are computed from the efficiency E_0 given by Langmuir (1951). The sizes of collected droplets or crystals are taken into account as follows;

$$\frac{E_L}{E_c} = \left(E_0^{1/2} + \frac{r}{R} \right)^2 \quad (10)$$

$$E_0 = E_0(\alpha, R_e) \quad (11)$$

where α and R_e are a measure of the inertia of the droplet or the crystal and the Reynolds number of the supercooled water drop, which are given by

$$\alpha = \frac{2\rho r_{dc}^2 V}{9\eta R} \quad (12)$$

$$R_e = \frac{2\rho_a R V}{\eta} \quad (13)$$

Here,

ρ : the density of the droplet which is taken to be 1 g/cm³ or the density of the crystal which is taken to be 0.1 g/cm³.

r_{dc} : the radius of the droplet or the crystal.

ρ_a : the density of air.

η : the viscosity of air which is taken to be 1.71×10^{-4} g/cm/s.

Strictly speaking, the collection efficiency given by Langmuir is not applicable to the collision of ice crystals with the water drop, because the shape of an ice crystal is not a sphere. However, the drag force of air on the ice crystal, the form of which is approximated by a circular disk, is nearly equal to that on a sphere containing the disk if the crystal is small enough, because the former is $16\eta r_{dc} V$ and the latter is $6\pi\eta r_{dc} V$ (Lamb, 1945) as far as Stoke's law holds good for airflow around the both. Therefore, the formula (11) may be roughly applicable to the ice crystals.

3. Results of numerical computations and discussions

In this section, results of numerical com-

putations of many cases are discussed to clarify the effects of each parameter on the collision of ice crystals with the supercooled water drop. In computations, the drop with initial radius of R_0 at cloud base is carried upward by vertical air current with velocity of w , growing due to the capture of cloud droplets. The drop continues to stay at cloud top until its fall velocity relative to air becomes larger than vertical velocity of the air. The conditions of computations are given by different combinations of the values of three parameters (w , L and H). The values of parameters adopted in computations are shown in Table 1.

Table 1. The values of parameters adopted in computations.

Ro (mm)	0.02	0.05	0.1	
W (m/s)	1	2	3	
L (g/m³)	0.5	1	2	
H (m)	1000	2000	4000	6000

(a) The growth of a water drop due to the capture of cloud droplets and its trajectory. The size of the drop has a great effect on the number of ice crystals colliding with it. First the change in its radius and trajectory

Fig. 1. The growth of a water drop whose initial radius is 0.02 mm, 0.05 mm or 0.1 mm due to the capture of cloud droplets. Dotted, solid and chain lines represent the cases of 2 g/m³, 1 g/m³ and 0.5 g/m³ in the water content of cloud droplets, respectively.

Fig. 2. The trajectories of water drops with initial radius of $20\ \mu$ in various clouds. Figs. a, b and c correspond to clouds of $0.5\ \text{g/m}^3$, $1\ \text{g/m}^3$ and $2\ \text{g/m}^3$ in the water content of cloud droplets. Dotted, solid and chain lines represent the cases of $3\ \text{m/s}$, $2\ \text{m/s}$ and $1\ \text{m/s}$ in the vertical velocity at air current, respectively. White and black points mean that the drops reach and leave the cloud tops at those times, respectively.

are briefly studied on the relation with w , L and H .

As indicated in Fig. 1, the radius of the drop is expressed as dependent upon time, initial radius and the water content of cloud droplets, but independent of its height, the vertical velocity of air and the height of cloud top, because the falling velocity of the drop and the collection efficiency of cloud droplets in equation (2) are determined only by the radius of the drop. The growth rate is proportional to the water content of cloud droplets.

As the size distribution of ice crystals in model cloud is dependent only on the height and w , the trajectory of the drop gives an important effect on the probability of collision. For example, a large drop which stays for a long time in high level has a high probability to be collided with by ice crystals. Fig. 2 shows the trajectories of drops with initial radius of 20μ in clouds with various conditions. Figures (a), (b) and (c) correspond to clouds in which the water content of cloud droplets are 0.5 g/m^3 , 1 g/m^3 and 2 g/m^3 , respectively. Chain, solid and dotted lines represent clouds in which the vertical velocity of air current are 1 m/s , 2 m/s and 3 m/s , respectively. White or black points mean the height and the time at which the drop reaches or leaves cloud top in each cloud, respectively. Thick horizontal lines in each figure indicate the staying of the drop at the cloud top, 1000 m , 2000 m , 4000 m or 6000 m in height. As the radius of the drop is dependent only upon time for given R_0 and L , the time when the drop leaves cloud top and the radius at that time are independent of the height of cloud top if the vertical velocity of air current is the same. It is shown in the figures that, with stronger updraft, drops can reach the cloud top sooner and stay there longer than with weaker updraft. However, if the water content of cloud droplets is larger, the drop leaves the cloud top sooner, otherwise it cannot even reach the top due to its rapid growth, whereas with small L , it can be carried up to higher level and can stay at the cloud top for long time on account of the growth rate.

(b) The probability of the collision of ice crystals with a supercooled water drop.

Fig. 3 shows the change with time in the number of ice crystals which collide with the drop of 20μ in initial radius in unit time in clouds with various conditions. Ordinate and abscissa are the number of ice crystals colliding in unit time and the time after the drop left cloud base, respectively. Dotted, solid and chain lines represent the values for clouds with 2 g/m^3 , 1 g/m^3 and 0.5 g/m^3 in the water content of cloud droplets. Figs. (a) and (b) correspond to clouds with 2000 m and 4000 m in the height of cloud top. Figures beside curves indicate the vertical velocity of air current in unit of m/s . White and black points mean the same as those in Fig. 2.

In each case, the probability of collision increases about 100 times during the stay of the drop at the cloud top; that is, it increases from $10^{-6}/\text{s}$ to $10^{-4}/\text{s}$ when the cloud top is 2000 m in height and from $10^{-3}/\text{s}$ to $10^{-1}/\text{s}$ in case of 4000 m in height. It is to be noted that the maximum of probability is found little after the drop left the cloud top and its value is almost independent of the water content of cloud droplets. It is determined by the height of cloud top (about $10^{-4}/\text{s}$ in 2000 m and $10^{-1}/\text{s}$ in 4000 m) and the vertical velocity of the air current. Although maximum value is not dependent upon the water content, the time when it is found is inversely proportional to the water content. After the drop left the cloud top, the probability decreases rapidly with time.

It can be said that most of the collisions occur during the stay of the drop at the cloud top. In the clouds with stronger air current, the probability increases much sooner than in the clouds with weaker air current, because the drop can reach the cloud top sooner. Maximum value is also larger, because the drop cannot leave the cloud top until its size becomes larger in the former than in the latter.

In the clouds with the cloud top lower than 2000 m or higher than 4000 m , the change in the probability is similar, although the drop cannot reach the top in some clouds with the higher top (especially in the clouds with large water content).

In actual cloud, the probability may be between the values in the cases with the water content of cloud droplets of 0.5 g/m^3

Fig. 3. The probability of the collision of ice crystals with a water drop. Figs. a and b correspond to the cloud tops, 2000 m and 4000 m in height, respectively. Dotted, solid and chain lines represent the cases of 2 g/m³, 1 g/m³ and 0.5 g/m³ in the water content of cloud droplets, respectively. Figures beside the curves mean the vertical velocity of air current in unit of m/s. White or black points mean that the drop reaches or leaves cloud top at that time, respectively.

and those in the cases of 1 g/m^3 .

(c) The effects of the velocity of updraft on the collision of ice crystals.

The effects of each parameter on the collision will be explained from the view point of the accumulated number of ice crystals colliding with the drop.

The effects of the velocity of updraft are shown in Fig. 4. The ordinate indicates the accumulated number of ice crystals colliding with the drop during 0.5 hr., 1 hr. or 1.5 hr. and the abscissa indicates the vertical velocity. Dotted, solid and chain lines mean the cases of 2 g/m^3 , 1 g/m^3 and 0.5 g/m^3 in the water content of cloud droplets. Figures on the right side of curves represent the height of cloud top. White circles mean that the drop already fell out of the cloud then and the values at these points indicate terminal accumulated numbers.

The increase in the accumulated numbers with the increase in the vertical velocity can be found in all cases. It is more remarkable in the cases of the higher cloud top. For

example, in the cloud whose top is 6000 m, the value in the case of 3 m/s is 10^6 to 10^7 times as large as that in the case of 1 m/s . This can be accounted for by the fact that if the drop stays long in such a high level, it is likely to be collided with by many ice crystals since their number density is large there. However, the quantitative results of these computations cannot be directly applied to the real cloud. Because some ice crystals are so large in high level that the assumptions concerned with collection efficiency and the fall velocities of ice crystal are unreasonable.

(d) The effects of the water content of cloud droplets on the collision of ice crystals.

The effects of the water content is shown in Fig. 5. The ordinate indicates the same as that in Fig. 4. Dotted, solid and chain lines show the cases of 3 m/s , 2 m/s and 1 m/s in the vertical velocity, respectively. Figures on the right side of curves represent the height of the cloud top.

After 0.5 hour, it can be seen that the accumulated number is a little larger in the case

Fig. 4. The effects of the vertical velocity of air current on the accumulated number of ice crystals colliding with a water drop within 0.5 hr, 1 hr or 1.5 hr. Dotted, solid and chain lines represent the cases of 2 g/m^3 , 1 g/m^3 and 0.5 g/m^3 in the water content of cloud droplets. Figures on the right-hand side of curves indicate the height of cloud top. White points mean that the drop already fell out of cloud during that time.

Fig. 6. The effects of the height of cloud top on the accumulated number of ice crystals colliding with a water drop. Dotted, solid and chain lines represent the clouds of 2 g/m^3 , 1 g/m^3 and 0.5 g/m^3 in the water content of cloud droplets, respectively. Figures on the right-hand side of curves mean the vertical velocity of air current. The meaning of white points is same as that in Fig. 4.

Fig. 5. The effects of the water content of cloud droplets on the accumulated number of ice crystals colliding with a water drop. Dotted, solid and chain lines represent the clouds of 3 m/s , 2 m/s and 1 m/s in the vertical velocity of air current, respectively. The meanings of figures on the right-hand side of curves and white points are same as those in Fig. 4.

with larger water content. This tendency appears more remarkably in the clouds of stronger vertical current than in the clouds of weaker one. After 1 hour water drops in all clouds with the water content of 2 g/m^3 falls out of clouds. Although the tendency that the accumulated number of ice crystals colliding with the drop is larger in the clouds with larger water content remains in some cases, maximum accumulated numbers can be found in the clouds with medium water content (that is, 1 g/m^3) in other cases. This is due to the fact that since water drops in the clouds with larger water content grow more rapidly and fall out of clouds sooner, these drops cannot collide with so many ice crystals. After 1.5 hour, maximum numbers in the cases with updraft of 1 m/s are found in the clouds of 0.5 g/m^3 in the water content,

although maximum numbers in the cases of 2 m/s or 3 m/s are in the clouds of 1 g/m^3 .

Thus, the drop growing in the cloud with smaller water content cannot collide with so many ice crystals in early stage of its growth. However, the accumulated number of ice crystals colliding with the drop becomes larger after sufficiently long time than that in the cloud with large water content, because the drop falls out of the latter cloud sooner on account of its more rapid growth. The accumulated number of ice crystals colliding for 1 hour in the cloud with updraft of 1 m/s to 3 m/s is the most in the case of about 1 g/m^3 . This most favourable water content is dependent mainly upon the velocity of updraft and the time after the drop left cloud base.

(e) The effects of the height of cloud top on the collision of ice crystals.

Fig. 7. The effects of the initial radius of a water drop on the accumulated number of ice crystals colliding with the drop. Solid and dotted lines represent the clouds of 1 g/m^3 and 0.5 g/m^3 in the water content of cloud droplets, respectively. Figures beside curves mean the vertical velocities of air current in unit of m/s . The height of cloud top is 2000 m.

The remarkable effects of the height of cloud top on the collision is shown in Fig. 6. Dotted, solid and chain lines indicate the clouds in which the water content of cloud droplets is 2 g/m^3 , 1 g/m^3 and 0.5 g/m^3 , respectively. Figures on the right hand side of the curves represent the vertical velocity of air current. White circles mean the same as those in Figs. 6 and 7. It is seen from the figure that the increase in the accumulated number with the height of the cloud top is not so clear in the cloud with weak vertical air current as in the cloud with stronger one. This is due to the fact that the drop cannot reach high level in the former cloud. In the cloud with vertical current stronger than 2 m/s , the accumulated number of ice crystals colliding for 1 hour ($N_{1\text{hr.}}$) and the height of cloud top almost satisfy a relation

$$\log N_{1\text{hr.}} = 1.6 \times 10^{-3} H - 5.6 \quad (17)$$

where H is expressed in unit of m . The difference of the height of the top by 1000 m causes the accumulated number to increase 10 to 50 times.

(f) The effects of the initial radius of the supercooled water drop.

All results mentioned above are concerned with the drop of 20μ in initial radius. Let us consider the effects of the initial radius of the drop on the collision. It is natural that the drop larger at the cloud base grows more quickly, becoming large enough to fall to the ground. Therefore, as shown in Fig. 7, the effects of initial radius are exceedingly similar to that of the water content of cloud droplets. Fig. 7 shows only the cases of 2000 m in the height of cloud top. In the case of the updraft of 1 m/s the drop with initial radius of 20μ has more chances of collision with ice crystals for both 0.5 hour and 1 hour than that with larger initial radius. The maximum accumulated number in the case of 2 m/s can be found on the drop of about 50μ in initial radius. Most cases of 3 m/s have the maximum value on the drop of 100μ in initial radius.

Thus, although the drop larger at initial radius is in some cases favourable for the collision with ice crystals, it tends to fall out of clouds rapidly and it cannot collide with so many ice crystals unless the strong vertical

air currents carry up and suspend it at the high level for a long time.

These effects of the velocity of updraft (w), the height of the cloud top (H) and the water content of cloud droplets (L) on the collision of a supercooled water drop with ice crystals are summarized in Fig. 8. Each line represents the lower bound of the values of w and H which are required for the drop to collide with more than one or 0.01 ice crystal for 0.5 hour (dotted lines), 1 hour (solid lines) and 1.5 hour (chain lines), after the drop of 20μ in initial radius left the cloud base. Thin and thick lines indicate the values required in the clouds of 0.5 g/m^3 and 1 g/m^3 in L , respectively. The values in the clouds of L between 0.5 g/m^3 and 1 g/m^3 are generally found between thin line and thick one. The figure shows that it is nearly impossible for each drop to collide with one ice crystal for 0.5 hour in the clouds which have not very high cloud top and very strong updraft. If the drop can stay in these clouds (especially at high level) more than 1.0 hour,

Fig. 8. The lower bound of the values of w and H which are required for a supercooled water drop to collide with more than 1 or 0.01 ice crystal for 0.5 hour (dotted lines), 1.0 hour (solid) and 1.5 hour (chain) after the drop of 20μ in initial radius left cloud base. Thin and thick lines indicate the values required in clouds of 0.5 g/m^3 and 1 g/m^3 in L , respectively.

it is possible for each drop to collide with one ice crystal and to freeze. However, such a long stay of the drop in the convective cloud, is considered to be very rare, because the life of convective cloud itself does not generally exceed 1.0 hour. The conclusion is that, in clouds lower than 3000 m in H and a few m/s in w , at most, one drop of 10 to 100 supercooled drops can collide with one ice crystal and freeze for 1.0 to 1.5 hour. Although the life of stratus is longer than that of convective cloud, the updraft in it is weak and water drops will be unable to stay for a long time.

In all cases shown in figures, the number density of atmospheric ice nuclei corresponds to that expressed by equation (5) of 10^{-5} /litre in A and 0.6 in β (one active nucleus per litre at -21°C). Of course, the number density of atmospheric ice nuclei varies remarkably with time and place. However, as far as the temperature spectrum on the activation of atmospheric ice nuclei can be expressed approximately by equation (5), the results of computations can be easily interpreted to the case with other values of A than 10^{-5} /litre because the difference of one order in A changes the freezing probability by one order.

If there are observational data concerned with the number density of frozen drops in the cloud (not the number density of ice crystals), it is possible to compare the freezing probability of a supercooled water drop obtained computationally with that in the actual cloud. It is regrettable that such satisfactory observations have not been carried out. However, if the growth rate of a frozen drop due to sublimation and the capture of cloud droplets is studied numerically and if there are observational data concerned with the fall-out rate of graupels on the ground, the number densities of giant nuclei and atmospheric ice nuclei, the indirect comparison of the both may be possible. These discussions will be made in the next paper of the author.

In computations mentioned above, it was assumed that the shapes of all ice crystals were approximated by a circular disk and their fall velocities were zero. However, the shapes of ice crystals are various in the actual clouds. Their growth rates, their fall

velocities and the efficiency of the collision of a water drop with them change with their shapes. The effects of the shapes of ice crystals on the freezing probability of a supercooled water drop will have to be studied by a further refined model.

4. Collection of ice nuclei by a supercooled water drop

If a supercooled water drop contains an ice nucleus in itself, it will freeze at the temperature at which the ice nucleus is activated. In this section let us consider the possibility that an ice nucleus is contained in a water drop before activated. Two processes may exist in the contact of nuclei with the drop; that is, in one process, the nucleus collides with the drop due to the difference in the fall velocities and in the other process, the nucleus is attached to the drop as the result of its Brownian motion. However, it is clear from Langmuir's formula that, since the size of the nucleus is negligibly small in comparison with the drop and its inertia is very weak, the former process is considered to hardly occur. Therefore, only the latter process will be studied here.

The problem to be solved is the number of ice nuclei attached to the water drop, which is assumed to be stationary, due to their Brownian motion in unit time. It is assumed that the shape of the nucleus is spherical and it adheres to the drop when the distance between its centre and that of the drop is equal to the sum of their radii. Under these assumptions, the probability of the coagulation of spherical aerosols due to their Brownian motions which is already solved in detail (see Fuchs, 1964) can be applied to the solution of the above mentioned problem if slight modification is given.

The number of nuclei (Q_0) which collide with the drop due to their Brownian motion in unit time is expressed by

$$Q_0 = 4\pi(R + r_c)D'N_c \left[1 + \frac{2(R + r_c)}{\sqrt{\pi D'}} \right] \quad (25)$$

Here,

R : the radius of the water drop

r_c : the radius of the nucleus

D' : the diffusion coefficient of nuclei

N_c : the number density of nuclei.

The change in Q_0 with the radii of the drop and ice-nuclei can be directly computed from the equation (25), the size dependency of the diffusion coefficient of nuclei being derived from the mobilities of aerosol particles with various sizes which were given by Laby (1942).

The results are shown in Fig. 9 in which the abscissa and ordinate represent the radius of the drop and the number of nuclei colliding with the drop in unit time due to their Brownian motion, respectively. As seen from equation (25) and Fig. 9, Q_0 is directly proportional to the number density of ice nuclei and nearly proportional to the square of drop radius, while it decreases as the radius of ice nucleus increases. In this figure, the number density of nuclei is taken to be one per litre. Then, the number of nuclei with radius of 0.5μ , which come into contact with the drop of 100μ in radius, is about 10^{-9} per second and it is 2×10^{-7} per second in the case of 0.1μ and 1 mm in radii of the nucleus and the drop.

Fig. 9. The number of ice nuclei coming into contact with a water drop due to their Brownian motion and the diffusion coefficient of nuclei.

In the previous computations concerned with the collision of ice crystals, the adopted number density of ice nuclei produces one activated nucleus per litre at -21°C . Therefore, the results obtained in this chapter can be directly compared with the results of the previous computations for clouds with top-height of 3000 m. In the clouds 3000 m high in which the vertical velocity of air-current is 1 m/s to 3 m/s and the water content of cloud droplets is 0.5 g/m^3 to 2 g/m^3 , the accumulated number of ice crystals colliding with the drop for 0.5 hour or 1 hour is 10^{-4} to 10^{-2} or 10^{-3} to 1. Meanwhile, the probability of collision of ice nuclei with radius of 0.1μ to 0.5μ which are to be activated at the temperature warmer than -21°C is 10^{-8} to 10^{-9} per second, if the radius of the drop is 100μ to 300μ . The accumulated number of these ice nuclei colliding with the drop before they are activated is 10^{-4} to 10^{-5} even for 10^4 second. It can be said from these comparisons that the number of ice nuclei contained in the drop is negligibly small compared with the number of ice crystals colliding with the drop and that the freezing process of the supercooled water drop due to the activation of ice nuclei, which were contained in the drop before activation, hardly takes place in comparison with that due to the collision of ice crystals.

6. Summary

Under the assumption that a supercooled water drop freezes when the drop collides with an ice crystal, the freezing-probability of the supercooled water drop was numerically studied by simulating the growth of the water drop by the capture of cloud droplets and the collision of ice crystals with the drop in various clouds. All ice crystals were assumed to be formed on ice nuclei. Variable parameters were the vertical velocity of air-current, the water content of cloud droplets, the height of cloud top and the initial radius of the water drop.

Results of computations are summarized as follows;

- (1) In any case, the probability of the collision of ice crystals with the drop increases about 100 times during the stay of the drop at the cloud top and the probability de-

creases rapidly after the drop left the cloud top. Most of collisions take place during the stay of the drop near the cloud top.

- (2) The accumulated number of ice crystals colliding with the drop increases with the vertical velocity of the air-current, particularly remarkably in the clouds with high top.
- (3) The accumulated number of colliding ice crystals tends to increase with the water content of cloud droplets in early stage of the growth of the drop. However, after sufficiently long time, the accumulated number of ice crystals colliding with the drop in clouds of small water content is larger than that in the clouds of large water content, because the drop falls out of the latter cloud sooner on account of its more rapid growth. Most favourable water content for the collision with ice crystals is dependent upon the velocity of updraft and the time length.
- (4) The increase in the accumulated number of colliding ice crystals with the height of cloud top is very remarkable and nearly exponential.
- (5) Though the drop with large size at cloud base is favourable for the collision of ice crystals in some cases, it has a tendency to fall out of clouds rapidly and it cannot collide with so many ice crystals with the exception of clouds with strong vertical air current.
- (6) In clouds which are lower than 3000 m in the height of cloud top and which are a few m/s in the velocity of updraft, at most one drop of 10 to 100 supercooled water drops can collide with one ice crystal and freeze for 1.0 to 1.5 hour.
- (7) Although a supercooled water drop becomes frozen, if the drop contains an ice nucleus in itself, at the activation-temperature of the ice nucleus, the probability of this freezing is negligibly small in comparison with that due to the collision of ice crystals.

Thus, the freezing-probability of a supercooled water drop due to the collision of ice crystals are considerably variable depending upon the conditions of clouds and it can become unexpectedly high. The influences of

the collision of ice crystals with the supercooled water drop on the precipitation in supercooled cloud are quantitatively studied in another paper of the author (1968) concerned with the growth of solid particles.

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過冷却雲内の降水粒子 第1部

—氷晶との衝突による過冷却水滴の凍結—

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雲粒の捕捉により成長している過冷却水滴が、氷晶との衝突により凍結する確率を、種々な条件をもつ雲について数値計算により調べた。但し、氷晶は全て氷晶核の上に生成されたと仮定した。いずれの例でも、凍結確率は水滴が雲頂に滞在している間に100倍位増し、凍結の大部分が水滴が雲頂附近に滞在している間に起る。1個の水滴に衝突した氷晶の積算数は、上昇気流の速度及び雲頂の高さが増すにつれ増加する。一方、雲水量及び水滴の初期半径は大きいほど良いわけではなく、最大の積算数を与える値が存在する。その値は上昇気流の速度及び時間の長さにより変る。又、雲頂の高さ(雲底上の)が3000 m以下そして上昇気流の速度が数 m/sの雲では、雲底から上昇し始めて1~1.5時間に氷晶と衝突出来る過冷却水滴は、せいぜい10~100個のうちの1個である。