3.2 Microphysics of Cold Clouds
3.2.l Homogeneous Nucleation of Ice Particles

Ice particles in clouds may be nucleated from either the liquid or vapor phase.

An embryonic ice particle can be considered a polyhedron of volume $\alpha_i 4\pi R^3/3$ and surface area $\beta_i 4\pi R^2$, where R is the radius of a sphere that can just be contained within the polyhedron, and α_i and β_i are both greater than unity but approach unity as the polyhedron tends toward a spherical shape. By reasoning analogous to that leading to (3.3), the
expression for the critical radius R_{ci} of the inscribed sphere is

$$R_{ci} = \frac{2\beta_i \sigma_{il}}{\alpha_i n_i k_B T \ln(e_s/e_{si})}$$

(3.37)

where σ_{il} is the free energy of an ice-liquid interface, n_i is the number of molecules per unit volume of ice, and e_{si} is the saturation vapor pressure with respect to a plane surface of ice. The saturation vapor pressures of liquid and
Theoretical and empirical results indicate that homogeneous nucleation of liquid water occurs at temperatures lower than about -35 to -40 , depending somewhat on the size of the drops being subjected to the low temperature. This threshold lies within the range of temperatures in natural clouds, which may have cloud-top temperatures below -80 . It is therefore possible, in a natural cloud, to have unfrozen liquid (i.e., supercooled) drops in the temperature range of 0 to about -40 . However, wherever the temperature in the cloud is below about -40 , any liquid drops that happen to be present freeze spontaneously by homogeneous nucleation. This conclusion is consistent with the fact that at temperatures below -40 atmospheric clouds are always composed entirely of ice.

In principle, an ice particle may be nucleated directly from the vapor phase. The critical size for homogeneous nucleation of an ice particle depends strongly on both temperature and ambient humidity. Theoretical estimates of the rate at which molecules in the vapor phase aggregate to form ice particles of critical size indicate, however, that nucleation occurs only at temperatures below -65 and at supersaturations ~ 1000 . Such high supersaturations do not occur in the atmosphere. Since liquid drops would nucleate from the vapor phase before these supersaturations were reached, and since the liquid drops thus formed would freeze homogeneously below -40 , it is concluded that homogeneous nucleation of ice directly from the vapor phase never occurs in natural clouds.

2 Heterogeneous Nucleation of Ice Particles

From observations of the particles in clouds it is readily determined that ice crystals form at temperatures between 0   and -40   . Since homogeneous nucleation does not occur in this temperature range, the crystals must form by a heterogeneous process. As in the case of heterogeneous nucleation of liquid drops, the foreign surface on which an ice particle nucleates reduces the critical size that must be attained by chance aggregation of molecules. However, in the case of drops nucleating from the vapor phase, the atmosphere has no shortage of wettable nuclei. In contrast, ice crystals do not form readily on many of the particles found in air. The principal difficulty with the heterogeneous nucleation of the ice is that the molecules of the solid phase are arranged in a highly ordered crystal lattice. To allow the formation of an interfacial surface between the ice embryo and the foreign substance, the latter should have a lattice structure similar to that of ice. Fig 1 illustrates schematically an ice embryo which has formed on a crystalline substrate with a crystal lattice different from that of the ice. There are two ways in which the embryo could form. Either the ice could retain its normal lattice dimensions right to the interface, with dislocations in the sheets of molecules, or the ice lattice could deform elastically to join the lattice of the substrate. The effect of dislocations is to increase the surface tension of the ice-substrate interface. The effect of elastic strain is to raise the free energy of the ice molecules. Both of these effects lower the ice-nucleating efficiency of a substance. These effects, moreover, are temperature dependent in the sense that the higher the temperature, the more the surface tension and elastic strain are increased.
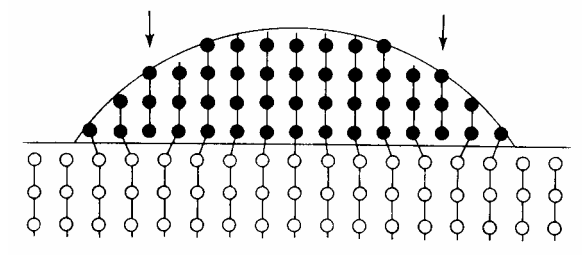

Fig1 Schematic illustration of an ice embryo growing upon a crystalline substrate with a slight misfit. Dislocations of the interface are indicated by arrows.

There are several modes of action by which an ice nucleus can trigger the formation of an ice crystal. An ice nucleus contained within a supercooled drop may initiate heterogeneous freezing when the temperature of the drop is

lowered to the value at which the nucleus can be activated. There are two possibilities in this case. If the cloud condensation nucleus on which the drop forms is the ice nucleus, the process is called *condensation nucleation*. If the nucleation is caused by any other nucleus suspended in supercooled water, the process is referred to as *immersion freezing*. Drops may also be frozen if an ice nucleus in the air comes into contact with the drop; this process is called contact nucleation. Finally, the ice may be formed on a nucleus directly from the vapor phase, in which case the process is called deposition nucleation.

From the above considerations, it is evident that the probability of ice particle nucleation should increase with decreasing temperature and that substances possessing a crystal lattice structure similar to that of ice should be the most likely to serve as a nucleating surface. In this respect, ice itself provides the best nucleating surface; whenever a supercooled drop at any temperature 0°C comes into contact with a surface of ice it immediately freezes. Other than ice, the natural substances possessing a crystal lattice structure most similar to that of ice appear to be certain clay minerals found in many soil types and bacteria in decayed plant leaves. They may nucleate ice at temperatures as high as -4 but appear to occur in low concentrations in the atmosphere. Most ice particle nucleation in clouds occurs at temperatures lower than this. In general, particles in the air on which ice crystals are able to form are called *ice nuclei*. However, extensive measurements of this type indicate that the average number of ice nuclei N_I per liter of air generally increases exponentially with decreasing temperature according to the empirical formula

$$\ln N_I = a_I \left(253^{\circ}\text{K} - T \right)$$

where α_I varies with location but has values in the range of 0.3-0.8. Note that according to this relationship, there is only about one ice nucleus per liter at -20°C. For a value of $\alpha_I = 0.6$, the concentration increases by approximately a factor of ten for every 4 of temperature decrease.

3.2.3   Deposition and Sublimation

Growth of an ice particle by diffusion of ambient vapor toward the particle is called deposition. The loss of mass of an ice particle by diffusion of vapor from its surface into the environment is called sublimation. These processes are the ice-phase analogs of condensation and evaporation. However, since ice particles take on a variety of shapes, the spherical geometry assumed in evaluating the growth and evaporation of drops by vapor diffusion may not always be assumed in calculations of the change of mass of ice particles. Diffusion of vapor toward or away from nonspherical ice particles is accounted for by replacing R in (3.8), and thus in (3.14) and (3.22), by a shape factor $\widetilde{C}$, which is analogous to electrical capacitance. Thus, the analog to (3.8) is

$$\dot{m}_{\mathrm{dif}} = 4\pi\widetilde{C}D_v\left[\rho_v(\infty) - \rho_{vsfc}\right] \tag{3.39}$$

where ρ_{vsfc} is the vapor density at the particle's surface. It follows that the analogs to (3.14) and (3.22) are

$$\dot{m}_{\mathrm{dif}} = \frac{4\pi\widetilde{C}\widetilde{S}_i}{F_{\kappa i} + F_{Di}} \tag{3.40}$$

and

$$\dot{m}_{\mathrm{dif}} = \frac{4\pi\widetilde{C}V_F\widetilde{S}_i}{F_{\kappa i} + F_{Di}} \tag{3.41}$$

respectively. $\widetilde{S}_i$, F_{Ki} and F_{Di} are the same as $\widetilde{S}$, F_K, and F_D in (3.15)-(3.17)

except that L is replaced by the latent heat of sublimation L_s in (3.16), and $e_s(\ )$ is replaced by the saturation vapor pressure over a plane surface of ice $e_{si}(\ )$ in

(3.15) and (3.17). The relations (3.40) and (3.41) apply only when the air is saturated (in this case with respect to ice). As in the case of drops, $\dot{m}$ must be obtained numerically if the air is unsaturated.

The shape, or habit, adopted by an ice crystal growing by vapor diffusion is a sensitive function of the temperature T and supersaturation $\widetilde{S^i}$ of the air These growth modes are known from observations in the laboratory and in clouds themselves. The basic crystal habits exhibit a hexagonal face. Let a crystal be imagined to have an axis normal to its hexagonal face. If this axis is long compared to the width of the hexagonal face, it is said to be prismlike. If this axis is short compared to the width of the hexagonal face, the crystal is said to be platelike. The habits change back and forth between prismlike and platelike as the ambient temperature changes (Table 3.1). The effect of increasing the ambient supersaturation is to increase the surface-to-volume ratio of the crystal. The additional surface area gives the increased ambient vapor more space on which to deposit. They may be thought of as hexagonal plates with sections deleted to increase the surface-to-volume ratio of the crystal. They occur in the temperature range where the difference between the saturation vapor pressure over water and the saturation vapor pressure over ice is greatest.

Temperature (^{o}C)	Basic habit	Types of crystal at slight water Supersaturation
0 to – 4	Platelike	Thin hexagonal plates
– 4 to – 10	Prismlike	Needles (– 4 to – 6 ^{o}C)
		Hollow columns (– 5 to – 10 ^{o}C)
– 10 to – 22	Platelike	Sector plates (– 10 to – 12 ^{o}C)
		Dendrites (– 12 to – 16 ^{o}C)
		Sector plates (– 16 to – 22 ^{o}C)
– 22 to – 50	Prismlike	Hollow columns

3.2.4　Aggregation and Riming

If ice particles collect other ice particles, the process is called aggregation. If ice particles collect liquid drops, which freeze on contact, the process is called *riming*. The continuous collection equation (3.24) may be used to describe the growth of ice particles by aggregation or riming.

Aggregation depends strongly on temperature. The probability of adhesion of colliding ice particles becomes much greater when the temperature increases to above -5 , at which the surfaces of ice crystals become sticky. Another factor affecting aggregation is crystal type. Intricate crystals, such as dendrites, become aggregated when their branches become entwined. The sizes increase sharply at temperatures above -5 , while aggregation does not appear to exist below -20 .

The collection efficiency for riming is not well known theoretically or empirically, but it is generally thought to be quite high and often assumed to be unity in calculations using (3.24) or (3.26). If the ice particle is viewed as the collector and the liquid drops as the collected particles in (3.24), the degree of riming that is achievable is determined primarily by the mixing ratio of the liquid water ($q_{m'}$).

3.2.5　Hail

Extreme riming produces *hailstones*. These particles are commonly 1 cm in diameter but have been observed to be as large as 10-15 cm. They are produced as graupel or frozen raindrops collect supercooled cloud droplets. So much liquid water is accreted in this fashion that the latent heat of fusion released when the collected water freezes significantly affects the temperature of the hailstone. The hailstone may be several degrees warmer than its environment. This temperature difference has to be taken into account in calculating the growth of hail particles, which is determined by considering the heat balance of the hailstone.

The rate at which heat is gained as a result of the riming of a hailstone of mass m is

$$\dot{Q}_f = \dot{m}_{col} \left\{ L_f - c_w [T(R) - T_w] \right\} \qquad (3.42)$$

The factor $\dot{m}_{col}$ is the rate of increase of the mass of the hailstone as a result of collecting liquid water. It is given by (3.26). The hailstone is assumed to be spherical with radius R. L_f is the latent heat of fusion released as the droplets freeze on contact with the hailstone. The second term in the curly brackets is the heat per unit mass gained as the collected water drops of temperature T_w come into temperature equilibrium with the hailstone. The factor c_w is the specific heat of water. If the air surrounding the particle is subsaturated, the temperature T_w is approximated by the wet-bulb temperature of the air, which is the equilibrium temperature above a surface of water undergoing evaporation at a given air pressure.[59] This temperature may be several degrees less than the actual air temperature when the humidity of the air is very low. If the air surrounding the particle is saturated $T_w = T(\quad)$.

The rate at which the hailstone gains heat by deposition (or loses heat

by sublimation) is obtained from a modified form of (3.8)

$$\dot{Q}_c = 4\pi R D_v [\rho_v(\infty) - \rho_v(R)] V_{Fc} L_s \qquad (3.43)$$

where V_{Fs} is a ventilation factor for sublimation, and L_s is the latent heat of sublimation.

The rate at which heat is lost to the air by conduction is obtained from a modified version of (3.9), which may be written as

$$\dot{Q}_c = 4\pi R \kappa_a [T(R) - T(\infty)] V_{Fc} \qquad (3.44)$$

where V_{Fs} is a ventilation factor for conduction.

In equilibrium we have

$$\dot{Q}_f + \dot{Q}_s = \dot{Q}_c \qquad (3.45)$$

which upon substitution from (3.42)-(3.44) may be solved for the hailstone equilibrium temperature as a function of size. As long as this temperature remains below 0 , the surface of the hailstone remains dry, and its development is called *dry growth*. The diffusion of heat away from the hailstone, however, is generally too slow to keep up with the release of heat associated with the riming (depositional growth is much less than the riming). Therefore, if a hailstone remains in a supercooled cloud long enough, its equilibrium temperature can rise to 0 . At this temperature, the collected supercooled droplets no longer freeze spontaneously upon contact with the hailstone. Some of the collected water may then be lost to the warm hailstone by shedding. However, a considerable portion of the collected water becomes incorporated into a water-ice mesh forming what is called *spongy hail*. This process is called *wet growth*. During its lifetime, a hailstone may grow alternately by the dry and wet processes as it passes through air of varying temperature. When hailstones are sliced open, they often exhibit a layered structure, which is evidence of these alternating growth modes.

3.2.6 Ice Enhancement

When the concentrations of ice particles are measured in natural clouds, it is often found that there are far more ice particles present than can be accounted for by the typical concentrations of ice nuclei activated by lowering the temperature of air in expansion chambers. The latter concentration is calculated from (3.38). The cloud-top temperature is the lowest temperature anywhere in the cloud and hence provides an estimate of the maximum possible ice nucleus concentration in the cloud according to (3.38).

These high concentrations are found in many cold clouds. They do not, however, occur in uniform spatial and temporal patterns within a cloud. A common characteristic is that they occur in older rather than newly formed portions of clouds, and they are found in association with supercooled cloud droplets. They are most likely to be found when the size distribution of the droplets is broad, with largest drops exceeding about 20 μm in diameter. The high concentrations develop initially near the tops of clouds, and the high concentrations may develop suddenly (e.g., the concentration may rise from 1 to 1000 l^{-1} in less than 10 min).

The microphysical process, or processes, by which the concentrations of ice particles become so highly enhanced relative to the number of nuclei which would appear to be active according to (3.38) are not certain. Some hypotheses that have been suggested are

(i)   *Fragmentation of ice crystals*. Delicate crystals may break into pieces as a result of collisions and/or thermal shock.

(ii) *Ice splinter production in riming*. It has been found in laboratory experiments that when supercooled droplets >23 μm in diameter collide with an ice surface at a speed of   1.4 m s^{-1} at temperatures of -3 to –8   , small ice splinters are produced.[62]

(iii) *Contact nucleation*. It is thought that when certain aerosol particles come into contact with supercooled droplets they can cause nucleation at higher temperatures than they would through other forms of nucleation.

(iv) *Condensation or deposition nucleation*. There is evidence that the icenucleating activity of atmospheric aerosol particles by either condensation or deposition nucleation is greatly increased when the ambient supersaturation rises above 1% with respect to water. Ice nucleus counters whose data lead to the expression in (3.38) are usually operated near water saturation. A pocket of high supersaturation in a cloud might be favorable for the sudden appearance of a large number of ice particles.

The latter two mechanisms, (iii) and (iv), do not require the pre-existence of ice particles and may thus help to account for the sudden appearance of high concentrations in cloudy air at relatively high temperatures.

3.2.7   Fall Speeds of Ice Particles

The fall speeds of ice particles encompass a wide range. Observations show that these speeds depend on particle type, size, and degree of riming and that the more heavily rimed a particle, the more its fall speed depends on its size. Individual snow crystals (lower curves of Fig. 3.12) and unrimed to moderately rimed aggregates of crystals (Fig. 3.13) drift downward at speeds of 0.3-1.5 m s^{-1}, with the aggregates showing a tendency to increase slightly in fall speed as they approach 12 mm in dimension. Graupel fall speeds increase sharply from 1 to3m s^{-1} over a narrow size range of 1-3 mm (upper curve of Fig. 3.12 and Fig. 3.14). T
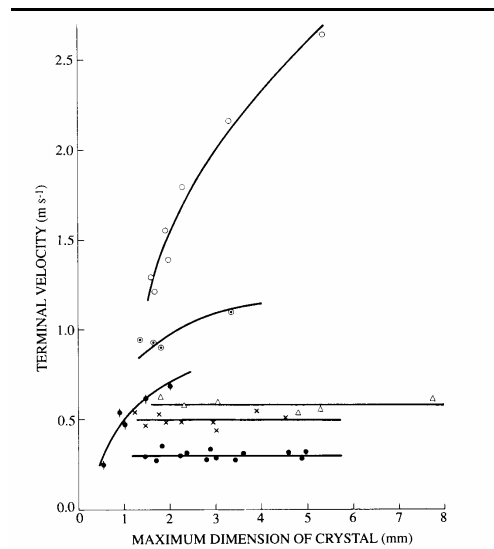

Figure 3.12   Terminal fail speeds of snow crystals as a function of their maximum dimensions. Open circle (uppermost curve) indicates graupel fall speeds. Other curves are for rimed crystals (dot in circle), needles (filled circle with slash), spatial dendrites (triangle), powder snow (×), and dendrites (filled circle).

Hailstone fall velocities are an order of magnitude larger than those for snow and graupel. At a pressure of 800 mb and a temperature of 0   , they obey the empirical formula

$$V\left(ms^{-1}\right) \approx 9D_h^{0.8} \qquad (3.46)$$

where D_h is the diameter of the hailstone in cm. This formula was obtained for hailstones in the size range 0.1-0.8 cm. Over this range, the fall speeds indicated by (3.46) are roughly 10-50 m s^{-1}. These large values imply that updrafts of comparable magnitude must exist in the cloud to support the hailstones long enough for them to grow.
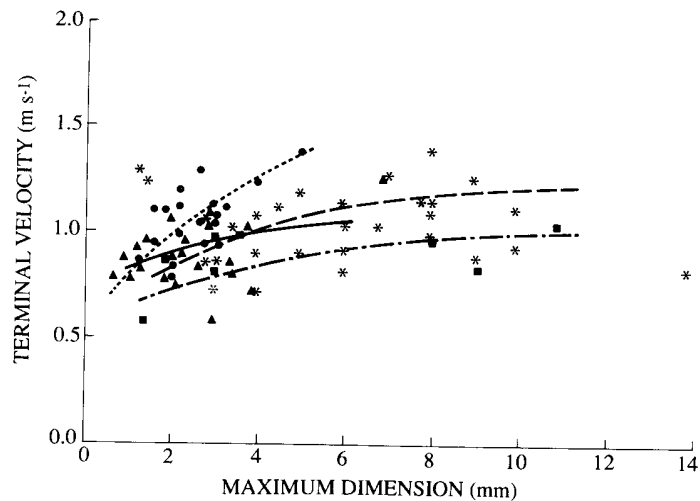
Figure 3.13 Terminal velocity and maximum dimension measurements for unrimed to moderately rimed aggregates. Combinations of: sideplanes (a type of branched crystal), bullets and columns (circles, dotted curve); sideplanes (triangles, solid curve); radiating assemblages of dendrites (asterisks, dashed curve); dendrites (squares, dash-dot curve).
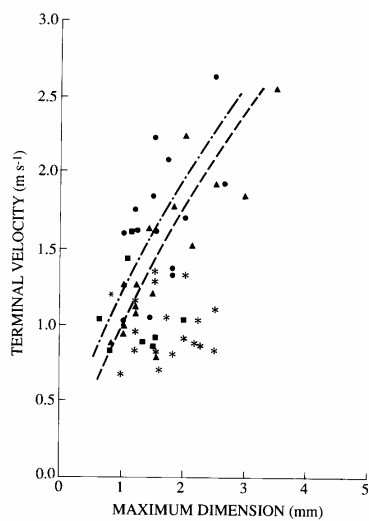

Figure 3.14 Terminal velocity and maximum dimension measurements for graupel and graupel-like snow. Cone-shaped graupel (circles, dash-dot curve); hexagonal graupel (triangles, dashed curve); graupel-like snow of hexagonal type (asterisks).

3.2.8   Melting

Ice particles can change into liquid water when they come into contact with air or water that is above 0 . A quantitative expression for the rate of melting of an ice particle of mass m can be obtained by assuming heat balance for the particle during the melting. According to (3.45), this balance may then be written as

$$-L_f \dot{m}_{mel} = 4\pi R \kappa_a [T(\infty) - 273\,\text{K}] V_{Fc} + \dot{m}_{col} c_w (T_w - 273\,\text{K}) + \dot{Q}_s \qquad (3.47)$$

where $\dot{m}'_{mel}$ is the rate of change of the mass of the ice particle as a result of melting. The first term on the right is the diffusion of heat toward the particle from the air in the environment. The second term is the rate at which heat is transferred to the ice particle from water drops of temperature T_w that are collected by the melting particle. The third term, $\dot{Q}_s$, is the gain or loss of heat by vapor diffusion [given by (3.43) in the case of a spherical hailstone]. If both the air and drop temperatures exceed 273 K, then both the first and second term in (3.47) contribute to the melting.

3.3   Types of Microphysical Processes and Categories of Water Substance in Clouds

From the foregoing review of cloud microphysics, it is evident that water substance can take on a wide variety of forms in a cloud and that these forms develop under the influence of *seven basic types of* microphysical processes:

1. Nucleation of particles
2. Vapor diffusion
3. Collection
4. Breakup of drops
5. Fallout
6. Ice enhancement
7. Melting

These individual processes may sometimes be isolated for study in numerical models or in the laboratory. However, in a natural cloud several or all of these processes occur simultaneously, as the entire ensemble of particles comprising the cloud forms, grows, and dies out. Thus, the various forms of water and ice particles coexist and interact within the overall cloud ensemble. It is the behavior of the overall ensemble that is of primary interest in cloud dynamics, and it is generally unnecessary to keep track of every particle in the cloud in order to describe the cloud's gross behavior. At the same time, it is also impossible to ignore the microphysical processes and accurately represent the cloud's overall behavior. To retain the essentials of the microphysical behavior, it is convenient to group the various forms of water substance in a cloud into several broad categories of water substance:

- *Water vapor* is in the gaseous phase.

- *Cloud liquid water* is in the form of small suspended liquid-phase droplets (i.e., drops that are too small to have any appreciable terminal fall speed constitute cloud liquid water and therefore are generally carried along by the air in which they are suspended).

- *Precipitation liquid water* is in the form of liquid-phase drops that are large enough to have an appreciable fall speed toward the earth. This water may be subdivided into drizzle (drops 0.1-0.25 mm in radius) and rain (drops >0.25 mm in radius), as defined in Sec. 3.1.3.

- *Cloud ice* is composed of particles that have little or no appreciable fall speed. These particles may be in the form of pristine crystals nucleated directly from the vapor or water phase, or they may be tiny particles of ice produced in some form of ice enhancement process.

- *Precipitation ice* refers to ice particles that have become large and heavy enough to have a terminal fall speed ~0.3 m s^{-1} or more. These particles may be pristine crystals, larger fragments of particles, rimed particles, aggregates, graupel, or hail. To simplify the description of the gross behavior of a cloud these particle types are sometimes grouped into categories according to their density or fall speed. Such groupings are arbitrary; however, a commonly used scheme (employed in discussions below) is to divide these particles into *snow*, which has lower density and falls at speeds of ~0.3 - 1.5 m s^{-1} (see Figs. 3.12-3.13); *graupel*, which falls at speeds of ~1 - 3 m s^{-1} (see Figs. 3.12 and 3.14); and *hail*, which falls at speeds of ~10 - 50 m s^{-1} [see Eq. (3.46)].