1. Formation and Growth of Ice Crystals

When a cloud extends to altitudes where the temperature is colder than $0^{\circ}C$, ice crystals may form. Cold clouds can consist of supercooled droplets or ice particles or a mixture of both. The probability of ice particles being present in a cold cloud increases as the temperature decreases below $0^{\circ}C$.

- Cloud tops between $0^{\circ}C$ and $-4^{\circ}C$ generally consist entirely of supercooled droplets
- At about $-10^{\circ}C$ there is a 50% chance of ice
- Below $-20^{\circ}C$ 95% chance of ice

Growth may occur due to:

- 1. Direct *deposition* of vapor to the solid phase
- 2. Drops colliding with and freezing on the ice particles *riming*
- 3. Ice crystals can grow by collision with other crystals *clumping*

Ice crystals in a cloud of liquid droplets are in a favorable environment to grow by diffusion because the environment is supersaturated with respect to ice. If supercooled droplets exist with ice particles, the latter grow at the expense of the liquid droplets. Similar to liquid droplets there is diffusion followed by coagulation, but for ice, diffusion is more significant than for drops.

Homogeneous nucleation depends on the surface free energy of a crystal/liquid interface. Droplets smaller than $4\mu m$ will freeze spontaneously at a temperature of about $-40^{\circ}C$. Water vapor molecules can form a stable ice embyro by chance collisions, this should only occur for extreme conditions of supersaturation.[Figure 6.29 Wallace and Hobbs]]

Ice crystals appear below $-15^{\circ}C$ indicating *heterogeneous nucleation*. The probability of nucleation depends strongly on the properties of the substrate material. The more tightly-bound the water molecules are to the substrate, the greater will be the probability of ice nucleation. Also, if the crystal structure of the substrate resembles that of an ice crystal plane, it will increase the chances of ice nucleation. Only a very small fraction of aerosol particles can serve as ice nuclei.

It is important to keep in mind that the onset of ice nucleation occurs at lower temperatures for water supersaturation than for water subsaturation. [Fig 6.30 Wallace and Hobbs]

1a. Growth habit of ice crystals

Ice crystals appear in a large variety of shapes or habits. The most important shapes are plate, dendrite, column. Laboratory experiments show that growth habit varies with temperature and supersaturation. See Figure 9.6 of Rogers and Yau. For large vapor density excess the ice crystal shape changes with decreasing temperature from plane to needle to column to secotr plate to dendrite to sector plate and finally back to column.

1b. Diffusional Growth of Ice Crystals

When the first ice crystals nucleate in a cloud, they find themselves in an environment in which the vapor pressure is equal to or slightly greater than the equilibrium vapor pressure e_s over liquid water. The saturation ratio relative to ice may be written:

$$S_i = \frac{e}{e_i} = \frac{e}{e_s} \frac{e_s}{e_i} = S \frac{e_s}{e_i}$$
(1)

Where S is the saturation ratio with respect to water. A water-saturated cloud has high supersaturation relative to ice and is in a favorable environment for rapid growth by diffusion and deposition. This will continue as long as there are liquid droplets available to evaporate and maintain vapor pressure a equilibrium relative to water.

The equations for ice-crystal growth were derived by analogy to electrostatics. (For a spherical conductor of radius a the rate of change of charge q on the conductor in terms of the difference in potential V is $\frac{dq}{dt} = 4\pi C(V_{\infty} - V_s)$. Where C = a is the capacitance, this is valid for any type of conductor if we change C

The generalized growth equation becomes:

$$\frac{dm}{dt} = 4\pi C D(\rho_v - \rho_v r) \tag{2}$$

C denotes the electrical capacitance, with length of units, a function of size and shape of the particle. For a sphere, C = r and this reduces to the growth equation for a droplet.

Plate-type crystal $C = 2r/\pi$

Needles (approx. prolate spheroid) $C = \frac{A}{ln[(a+A)/b]}$ where $A^2 = a^2 + b^2$ Oblate spheroid $C = a\epsilon/arcsin\epsilon$ where $\epsilon^2 = 1 - b^2/a^2$

As the ice crystal grows, its surface is heated by the latent heat of sublimation and the value of ρ_{vr} is raised above the value that would apply without heating. Under stationary conditions, there is a balance between the rates of latent heating and heat transfer away from the surface:

$$\frac{\rho_v - \rho_{vr}}{T_r - T} = \frac{K}{l_s D} \tag{3}$$

Here, l_s is the latent heat of sublimation. In an analogous fashion to the liquid droplet case:

$$\frac{dm}{dt} = \frac{4\pi C(S_i - 1)}{\left(\frac{l_s}{R_v T} - 1\right)\frac{l_s}{KT} + \frac{R_v T}{e_i(T)D}} \tag{4}$$

Equation 4 is the basic model for the *Wegener-Bergeron-Fiendeisen mechanism* of precipitation formation. According to this mechanism, ice crystals grow by vapor diffusion at the expense of supercooled drops.

Vapor molecules cannot unite with an ice crystal in any haphazard way, but must join up, molecule-by-molecule, in such a manner that the crystal pattern is maintained. Consequently ρ_{vr} may not be the same over all points of the crystal surface. The rate of growth of an ice crystal will tend to be slower than given by this equation. Fukuta (1969) indicated that at temperatures between about 0°C and $-10^{\circ}C$ the growth of small crystals is about half that of Eq. 4. Larger particles do obey this equation more closely. The dependence of growth on temperature indicates that the temperature for maximum growth is about -15C over a wide range of pressures.

Ambient conditions determine not only the growth rate, but also the shape that a growing crystal takes. The main crystal types are column, plate and dendrite. A molecular-kinetic approach is required to explain the different shapes of ice or its transitions from one shape to another.

1c. Further Growth by Accretion

Accretion is reserved for the capture of supercooled droplets by an ice-phase precipitation particle.

- If droplets freeze immediately on contact they form a coating of rime leading to rimed crystals or graupel.
- If freezing is not immediate, denser structures are created, of which hail is an extreme example.

Aggregation is the clumping together of ice crystals to form snowflakes.

Fall speed of ice crystals is important for ice crystal growth by collision and capture. Graupel is the fastest falling crystals (frozen droplets). Rimed structures fall at about 1m/s, but all the pure crystal types fall slower than 1m/s. $u = 343D^{0.6}$ for graupel. u in cm/s and D, the diameter of the sphere which just circumscribes the particle, in cm. $u = kD^n$ for snowflake fall, where D is the melted diameter (cm) u in cm/s. $k \approx 160$ and $n \approx 0.3$ for dendrites. $k \approx 234$ and $n \approx 0.3$ for for columns and plates.

For the process of accretion, since ice crystals fall more slowly than water droplets, it seems that collision efficiencies are higher...but this is a complex problem. Because freezing is likely to occur on contact with supercooled droplets, coalescence efficiency is close to unity.

For the process of aggregation, collision efficiencies are less well understood and seem to be related to the shape and the temperatures (sticking more likely for temperatures higher than $-10^{\circ}C$).

Bearing these uncertainties in mind, the equations of accretional growth, leading to graupel is:

$$\frac{dm}{dt} = \overline{E}M\pi R^2 u(R) \tag{5}$$

m is the mass of particle, M cloud liquid water content, R radius, u(R) fall speed and \overline{E} mean collision efficiency.

The equations of aggregational growth for snowflakes:

$$\frac{dm}{dt} = \overline{E}M\pi R^2 \Delta u \tag{6}$$

m is the mass of particle, M cloud ice content, R radius, Δu is the difference in fall speed of the snowflake and the ice crystals, essentially a constant, \overline{E} mean collision efficiency. Sometimes the population of ice crystals is more conveniently characterized by the number density N than by the density of frozen water M = $Nv\rho$ where v is the average volume of the crystals and ρ is their density. If we assume that the snowflake has the same density then $m = \rho V$, where V denotes the volume:

$$\frac{dV}{dt} = B\overline{E}V^{2/3}Nv\Delta u \tag{7}$$

 $B^3 = 9\pi/16$. These equations have been found to give results in reasonable accord with observations on graupel and snowflakes.

Snowflakes must develop because a few of the crystals, which formed and grew by diffusion become larger than their neighbors. The larger crystals are in a favorable position to grow by the sweepout process.

- Aggregation/accretion in the ice phase or coalescence in the all-water process are necessary processes for precipitation particles of appreciable size to develop in the available time. Condensation-diffusion alone cannot explain the formation of large particles in realistic times.
- Diffusion is more effective for ice crystals than for water droplets (saturation with respect to water is supersaturation with respect to ice).