Chapter Three | Cloud Microphysics

"...fleecy piles dissolved in dew drops..."[42]

As noted in the Introduction, cloud physics consists of two branches: cloud microphysics and cloud dynamics. While the topic of this book is the latter, it is impossible to divorce a discussion of the dynamics from a knowledge of the microphysics. Just as the discussions in Chapters 5–12 assume a certain level of background knowledge of atmospheric dynamics, so do they assume some background in cloud microphysics. To provide this background, the present chapter summarizes the aspects of cloud microphysics that are crucial to the discussions of later chapters. First, we describe some of the basic microphysical processes that are involved in the formation, growth, shrinkage, breakup, and fallout of cloud and precipitation particles.[43] In Sec. 3.1, we describe the microphysics of *warm clouds,* where the temperature is everywhere above 0°C. Section 3.2 extends the review of microphysical processes to *cold clouds,* in which the temperature drops below 0°C and both ice and liquid particles may exist. After this review of the individual microphysical processes that may occur in clouds, we consider in Secs. 3.3–3.6 how these microphysical processes occur simultaneously in a real cloud and how they may be linked to the cloud dynamics through a set of water-continuity equations.

3.1 Microphysics of Warm Clouds

3.1.1 Nucleation of Drops

The particles in a cloud form by a process referred to as *nucleation,* in which water molecules change from a less ordered to a more ordered state. For example, vapor molecules in the air may come together by chance collisions to form a liquid-phase drop. To see how this process takes place, consider the conditions required for the formation of a drop of pure water from vapor. This case is called *homogeneous nucleation* to distinguish it from the case of *heterogeneous nucleation,* which refers to the collection of molecules onto a foreign substance. If the

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[42] Goethe realizes that the clouds are composed of microscopic particles.

[43] These microphysical processes are described in more detail in basic texts on cloud microphysics, such as Fletcher (1966), Mason (1971), Pruppacher and Klett (1978), and Rogers and Yau (1989). The physics of ice is presented comprehensively by Hobbs (1974).

embryonic drop of pure water has radius R, then the net energy required to accomplish its nucleation is

$$\Delta E = 4\pi R^2 \sigma_{vl} - \frac{4}{3}\pi R^3 n_l(\mu_v - \mu_l) \qquad (3.1)$$

The first term on the right is the work required to create a surface of vapor–liquid interface around the drop. The factor σ_{vl} is the work required to create a unit area of the interface. It is called the *surface energy* or *surface tension*. The second term on the right of (3.1) is the energy change associated with the vapor molecules going into the liquid phase. It is expressed as the change in the Gibbs free energy of the system. The Gibbs free energy of a single vapor molecule is μ_v, while that of a liquid molecule is μ_l, and the factor n_l is the number of water molecules per unit volume in the drop. If the work required to create the surface exceeds the change in Gibbs free energy ($\Delta E > 0$), the embryonic drop formed by chance aggregation of molecules has no chance of surviving and immediately evaporates. If, on the other hand, the work required to create the surface is less than the change in Gibbs free energy ($\Delta E < 0$), then the drop survives and is said to have been nucleated.

It can be shown[44] that

$$\mu_v - \mu_l = k_B T \ln\frac{e}{e_s} \qquad (3.2)$$

where k_B is Boltzmann's constant, e is the vapor pressure, and e_s is the saturation vapor pressure over a plane surface of water. Substituting this expression into (3.1), seeking the condition for which the work required to change the drop's surface is exactly matched by the change in Gibbs free energy ($\Delta E = 0$), and rearranging terms, we obtain an expression for the critical radius R_c at which this equilibrium condition holds. This expression is

$$R_c = \frac{2\sigma_{vl}}{n_l k_B T \ln(e/e_s)} \qquad (3.3)$$

and is referred to as *Kelvin's formula*.[45] This radius is evidently crucially dependent on the *relative humidity* (defined as $e/e_s \times 100\%$). Air is said to be *saturated* whenever the relative humidity is 100% ($e/e_s = 1$). However, it is clear from (3.3) that it is impossible for a cloud droplet to form under saturated conditions since $R_c \rightarrow \infty$ as $e/e_s \rightarrow 1$. Rather, the air must be supersaturated ($e/e_s > 1$) for R_c to be positive. The greater the *supersaturation* [defined, in percent, as $[(e/e_s - 1) \times 100\%]$, the smaller the size of the drop that must be exceeded by the initial chance collection of molecules.

It should be noted that R_c is also a function of temperature. Not only does T appear in the denominator of (3.3) explicitly, but σ_{vl} and e_s are functions of T. However, at atmospheric temperatures, the dependence of R_c on temperature is comparatively weak. In view of the primary dependence of R_c on ambient humid-

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[44] See problem 2.19 of Wallace and Hobbs (1977).
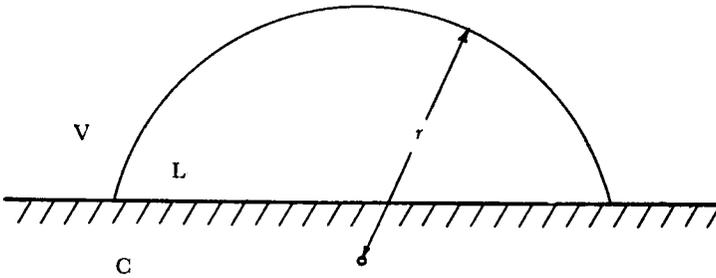[45] Named after Lord Kelvin, who first derived it.

Figure 3.1 A spherical-cap embryo of liquid (L) in contact with its vapor (V) and a nucleating surface (C). (From Fletcher, 1966. Reprinted with permission from Cambridge University Press.)

ity, it is not surprising that the rate of nucleation of drops exceeding the critical size R_c is a strong function of the degree of supersaturation. The rate at which the vapor molecules collide to form aggregates of various sizes can be computed using principles of statistical quantum mechanics applied to an ideal gas whose molecules are in a state of random motion.[46] This rate of formation of drops exceeding the critical size is the *nucleation rate*. It is found to increase from undetectably small values to extremely large values over a very narrow range of e/e_s. The value of e/e_s at which this rise occurs is in the range of 4–5. Thus, the air must be supersaturated by 300–400% for a drop of pure water to be nucleated homogeneously. Since supersaturation in the atmosphere seldom exceeds 1%, one concludes that homogeneous nucleation of water drops plays no role in natural clouds. However, the physics of the process are nonetheless relevant, as will become evident below.

Heterogeneous nucleation is the process whereby cloud drops actually form. The atmosphere is filled with small *aerosol particles,* and molecules of vapor may collect onto the surface of aerosol particles as illustrated ideally in Fig. 3.1. If the surface tension between the water and the nucleating surface is sufficiently low, the nucleus is said to be *wettable,* and the water may form a spherical cap on the surface of the particle. A particle onto which the molecules collect in this manner is referred to as a *cloud condensation nucleus (CCN).*

If a CCN is insoluble in water, the physics governing the survival of an embryonic cloud droplet are the same as in the case of homogeneous nucleation. It can be shown that Eq. (3.3) still applies, but R_c has the more general interpretation in that it refers to the critical radius *of curvature* of the embryonic drop. Since the radius of curvature of the droplet forming on a particle is greater than what it would be if the same number of molecules were to aggregate in the absence of the particle (Fig. 3.1), the aggregation of the vapor molecules has a greater chance of producing a drop exceeding the critical radius. If the aggregated water molecules form a film of liquid completely surrounding a particle, then a complete droplet is formed whose radius is larger than it would be in the absence of the nucleus. Clearly, the larger such a nucleus is, the more likely is the survival of a drop

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[46] See Chapter 2 of Fletcher (1966).

formed by a film around it. For this reason, the larger the aerosol particle, the more likely it is to be a site for drop formation in a natural cloud.

If the cloud condensation nucleus happens to be composed of a material that is soluble in water, the efficacy of the nucleation process is further enhanced. Since the saturation vapor pressure over the liquid solution is generally lower than that over a surface of pure water, e/e_s is increased. According to (3.3), the critical radius is then reduced, and nucleation is easier to achieve at the ambient vapor pressure.

There are generally more than enough wettable aerosol particles in the air to accommodate the formation of all cloud droplets. However, the physics of the nucleation process just described indicate that the first droplets in a cloud will tend to form around the largest and most soluble CCN. The sizes and compositions of the aerosol particles in a sample of air thus have a profound effect on the size distribution of particles nucleated in a cloud.

3.1.2   Condensation and Evaporation

Once formed, water drops may continue to grow as vapor diffuses toward them. This process is called *condensation*. The reverse process, drops decreasing in size as vapor diffuses away from them, is called *evaporation*. Particle growth by condensation and evaporation may be represented quantitatively by assuming that the flux of water vapor molecules through air is proportional to the gradient of the concentration of vapor molecules.[47] In this case, the vapor density ρ_v (defined as the mass of vapor per unit volume of air) is governed by the diffusion equation

$$\frac{\partial \rho_v}{\partial t} = \nabla \cdot \left(D_v \nabla \rho_v \right) = D_v \nabla^2 \rho_v \tag{3.4}$$

where $D_v \Delta \rho_v$ is the flux of water vapor by molecular diffusion and D_v is the diffusion coefficient (assumed constant) for water vapor in air. The concentration of vapor around a spherical pure-water drop of radius R is assumed to be symmetric about a point located at the center of the drop, and the diffusion is assumed to be in a steady state. Under these assumptions, ρ_v depends only on radial distance r from the center of the drop, and (3.4) reduces to

$$\nabla^2 \rho_v(r) = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \rho_v}{\partial r} \right) = 0 \tag{3.5}$$

The vapor density at the surface is $\rho_v(R)$. As $r \to \infty$, the vapor density approaches the ambient or free-air value $\rho_v(\infty)$. The solution to (3.5) satisfying these boundary conditions is

$$\rho_v(r) = \rho_v(\infty) - \frac{R}{r} \left[\rho_v(\infty) - \rho_v(R) \right] \tag{3.6}$$

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[47] This assumption is called Fick's first law of diffusion.

If the drop has mass m, the flux of molecules causes its mass to increase or decrease at a rate given by

$$\dot{m}_{\text{dif}} = 4\pi R^2 D_v \frac{d\rho_v}{dr}\bigg|_R \tag{3.7}$$

where $D_v d\rho_v/dr|_R$ is the flux of vapor in the radial direction across a spherical surface of radius R. Substitution of (3.6) into (3.7) yields

$$\dot{m}_{\text{dif}} = 4\pi R D_v [\rho_v(\infty) - \rho_v(R)] \tag{3.8}$$

Since $m \propto R^3$, there are two unknowns in (3.8), $\rho_v(R)$ and either m or R. Conditions in the environment ($r = \infty$) are assumed to be known. To obtain a solution for m or R, other relationships are needed. First, a heat-balance equation is introduced. In the condensation of water vapor on a drop, latent heat is released at a rate $L\dot{m}_{\text{dif}}$, where L is the latent heat of vaporization. Assuming that heat is conducted away from the drop as rapidly as it is being released, we have by analogy to (3.8)

$$L\dot{m}_{\text{dif}} = 4\pi \kappa_a R[T(R) - T(\infty)] \tag{3.9}$$

where κ_a is the thermal conductivity of air and T is temperature.

The equation of state for an ideal gas applied to water vapor under saturated conditions over a plane surface of pure water is

$$e_s = \rho_{vs} R_v T \tag{3.10}$$

where R_v is the gas constant for a unit mass of water vapor, and e_s and ρ_{vs} are the saturation vapor pressure and density over a planar surface of water. Since e_s depends only on temperature,[48] it is evident from (3.10) that ρ_{vs} is a known function of T. If it is then assumed that the vapor density at the drop's surface is given by the saturation vapor density, we may write

$$\rho_v(R) = \rho_{vs}[T(R)] \tag{3.11}$$

and (3.8), (3.9), and (3.11) can be solved numerically for $\dot{m}_{\text{dif}}$, $T(R)$, and $\rho_v(R)$. These equations can, moreover, be combined analytically for the special case of a drop growing or evaporating in a saturated environment (i.e., for the case in which $e(\infty) = e_s[T(\infty)]$). In this special case, use is made of the Clausius–Clapeyron equation:[49]

$$\frac{1}{e_s}\frac{de_s}{dT} \cong \frac{L}{R_v T^2} \tag{3.12}$$

Combination of (3.10) and (3.12) yields

$$\frac{d\rho_{vs}}{\rho_{vs}} = \frac{L}{R_v}\frac{dT}{T^2} - \frac{dT}{T} \tag{3.13}$$

[48] See pp. 72–73 of Wallace and Hobbs (1977).
[49] See p. 95 of Wallace and Hobbs (1977).

Then (3.8), (3.9), (3.11), and (3.13) may be combined[50] under saturated environmental conditions to obtain

$$\dot{m}_{\text{dif}} = \frac{4\pi R\tilde{S}}{F_\kappa + F_D} \tag{3.14}$$

where $\tilde{S}$ depends on the humidity of the environment, F_κ on the heat conductivity, and F_D on the vapor diffusivity. More specifically, $\tilde{S}$ is the ambient supersaturation (expressed as a fraction):

$$\tilde{S} \equiv \frac{e(\infty)}{e_s(\infty)} - 1 \tag{3.15}$$

The other factors are given by

$$F_\kappa \equiv \frac{L^2}{\kappa_a R_v T^2(\infty)} \tag{3.16}$$

and

$$F_D \equiv \frac{R_v T(\infty)}{D_v e_s(\infty)} \tag{3.17}$$

From (3.14)–(3.17), it is evident that the diffusional growth rate of a drop depends on the temperature and humidity of the environment and on the radius of the drop.

The relation (3.11) used in deriving (3.14) assumes that saturation at the drop's surface may be approximated as if it obtained over a plane surface of water (i.e., that the growing drop were large enough for the curvature of the drop's surface to have negligible influence upon the equilibrium vapor pressure). The drop has also been assumed to be sufficiently dilute with respect to dissolved nuclei or other impurities that the drop may be regarded as being composed of pure water. For very small drops, however, curvature and solution effects must be included. If a drop is growing on a water-soluble nucleus, $\rho_v(R)$ becomes

$$\rho_v(R) = \rho_{vs}[T(R)]\left(1 + \frac{\hat{a}}{R} - \frac{\hat{b}}{R^3}\right) \tag{3.18}$$

where the term $\hat{a}/R$ represents the effect of drop curvature on the equilibrium vapor pressure above the drop. The factor $\hat{a}$ is given by

$$\hat{a} = \frac{2\sigma_{vl}}{\rho_L R_v T} \tag{3.19}$$

where σ_{vl} is the surface tension of liquid–vapor interface and ρ_L is the density of liquid water. The term $\hat{b}/R^3$ represents the effect of salt dissolved in the drop on

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[50] See pp. 99–102 of Rogers and Yau (1989) for details of the derivation.

the equilibrium vapor pressure above the drop. The factor $\hat{b}$ is given by

$$\hat{b} = \frac{3i_{vH}m_s M_w}{4\pi \rho_L M_s} \tag{3.20}$$

where i_{vH} is the van't Hoff factor,[51] m_s and M_s are the mass and molecular weight of the dissolved salt, respectively, and M_w is the molecular weight of water.

Replacing (3.11) with (3.18) leads, following steps similar to those leading to (3.14), to the equation

$$\dot{m}_{dif} = \frac{4\pi R}{F_\kappa + F_D}\left(\tilde{S} - \frac{\hat{a}}{R} + \frac{\hat{b}}{R^3}\right) \tag{3.21}$$

which applies when the air is saturated. When the air is unsaturated, (3.8), (3.9), and (3.18) must be solved numerically to obtain $\dot{m}_{dif}$ for the evaporation rate of the drop.

When drops are falling relative to the surrounding air, the diffusion of vapor and heat is altered. To account for this process, the right-hand sides of (3.8) and (3.9) may be multiplied by a ventilation factor V_F. In this case, (3.14) and (3.21), the growth/evaporation rate under saturated conditions become

$$\dot{m}_{dif} = \frac{4\pi R V_F \tilde{S}}{F_\kappa + F_D} \tag{3.22}$$

and

$$\dot{m}_{dif} = \frac{4\pi R V_F}{F_\kappa + F_D}\left(\tilde{S} - \frac{\hat{a}}{R} + \frac{\hat{b}}{R^3}\right) \tag{3.23}$$

respectively.[52]

3.1.3 Fall Speeds of Drops

Growing cloud droplets are subject to downward gravitational force. This force can lead to their fallout as precipitation particles. The gravitational force on a drop is, however, largely offset by the frictional resistance of the air. As a particle is accelerated downward by gravity, its motion is increasingly retarded by the growing frictional force. Its final speed is called the *terminal fall speed V*. For drops of water in air, *V* is a function of the drop radius *R*. Generally *V* is negligible until the drops reach a radius of about 0.1 mm. This is usually considered to be the threshold size separating *cloud droplets*, which are suspended in the air indefinitely, from falling *precipitation* drops. The smallest precipitation drops (taken by con-

[51] This factor is equal to the number of ions into which each molecule of salt dissociates. See p. 162 of Wallace and Hobbs (1977).
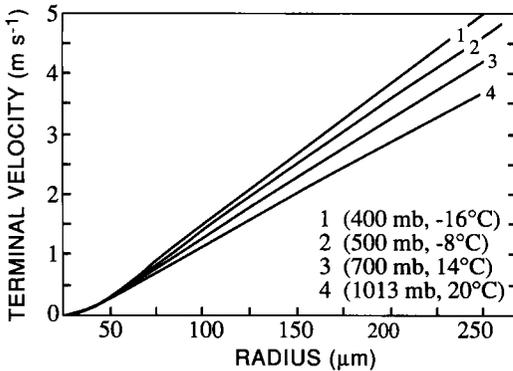[52] See pp. 440–463 of Pruppacher and Klett (1978).

Figure 3.2 Fall velocity of water drops <500 μm in radius for various atmospheric conditions. (From Beard and Pruppacher, 1969. Reprinted with permission from the American Meteorological Society.)
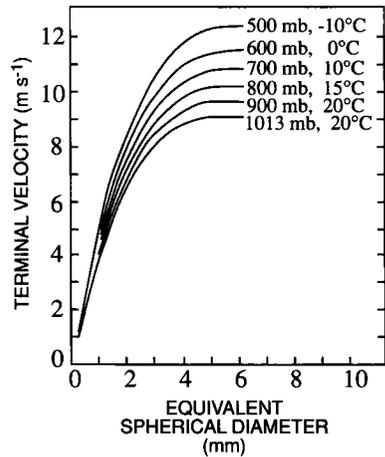
Figure 3.3 Fall velocity of water drops >500 μm in radius. (From Beard, 1976. Reprinted with permission from the American Meteorological Society.)

vention[53] to be those 0.1–0.25 mm in radius) are called *drizzle*. Drops >0.25 mm in radius are called *rain*. Drizzle and raindrops have terminal fall speeds that increase with increasing drop radius. We will represent this function as $V(R)$. For drops <500 μm in radius, V increases approximately linearly with increasing drop radius (Fig. 3.2). For larger drops, $V(R)$ increases at a lower rate (Fig. 3.3), becoming a constant at a radius of about 3 mm. This asymptotic behavior is associated with the fact that a drop becomes increasingly flattened, into the shape of a horizontally oriented disc, at larger sizes (see Fig. 4.2).

3.1.4 Coalescence

3.1.4.1 Continuous Collection

Cloud drop growth by coalescence with other drops can be envisioned in terms of a drop of mass m falling through a cloud of particles of mass m'. The water contained in the particles of mass m' is assumed to be distributed uniformly through the cloud with liquid water content $\rho q_{\mathrm{m}'}$ (g m^{-3}), where $q_{\mathrm{m}'}$ is the cloud water mixing ratio (mass of cloud water per mass of air). As it falls, the particle of mass m is assumed to increase in mass continually at a rate given by the *continuous collection equation*,

$$\dot{\mathrm{m}}_{\mathrm{col}} = A_{\mathrm{m}}|V(\mathrm{m}) - V(\mathrm{m}')|\rho q_{\mathrm{m}'}\Sigma_c(\mathrm{m},\mathrm{m}') \tag{3.24}$$

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[53] See the *Glossary of Meteorology* (Huschke, 1959).

where V represents the fall speed of the drops of masses m and m' (Figs. 3.2 and 3.3), ρ is the density of the air, $\Sigma_c\,(m, m')$ is the *collection efficiency*, and A_m is the effective cross-sectional area swept out by a particle of mass m. The absolute value notation is used in (3.24) since it is only the relative motion of the particles that matters for collectional growth. For the case of a large drop collecting smaller drops, the absolute value symbol is redundant since the fall velocity of the larger drop always exceeds that of the smaller drops. However, (3.24) may also be used to calculate the increase of mass of a smaller drop coalescing with larger drops. If the absolute value were not used in that case, negative growth would be calculated. Moreover, as will be seen below, (3.24) is applied also to cold clouds where in some special cases (e.g., an ice particle collecting water drops) the fall velocity of the larger particle may not be the greater of the two.

For the purpose of calculating collectional growth, water drops are usually assumed to be spherical. In that case, the factor A_m in (3.24) is given by

$$A_m = \pi (R + R')^2 \tag{3.25}$$

where R and R' are the radii of drops of mass m and m', respectively. This area is based on the sum of the drop radii since any drop centered within a distance $R + R'$ of the center of the drop of radius R can be intercepted by that drop.

The collection efficiency $\Sigma_c(m, m')$ is the efficiency with which a drop intercepts and unites with the drops it overtakes. It is the product of a collision efficiency and a coalescence efficiency. The collision efficiency (Fig. 3.4) is determined primarily by the relative airflow around the falling drop. Smaller particles may be carried out of the path of a larger particle (efficiency <1), or small particles not in the direct path of a large particle may collide with the large particle if they are pulled into its wake (efficiency >1). The coalescence efficiency expresses the
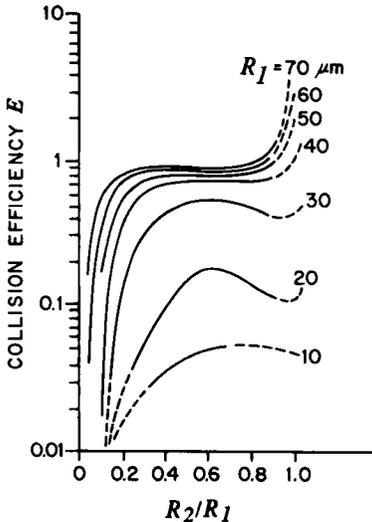

Figure 3.4 Collision efficiency for collector drops of radius R_1 with droplets of radius R_2. The dashed portions of the curve represent regions of doubtful accuracy. (From Wallace and Hobbs, 1977.)

fact that a collision between two drops does not guarantee coalescence; the drops may bounce off each other or remain united only temporarily. Under most conditions, coalescence efficiency is high, especially if the droplets are electrically charged or if an electric field is present. The electrical conditions are often met in clouds, and little else is known about the coalescence efficiency. Hence, the most common practice in theoretical or modeling studies is to assume a coalescence efficiency of unity. The collection efficiency then reduces to the collision efficiency.

A more general version of (3.24) may be written for the case in which a particle of mass m is falling relative to a population of particles of varying size. For that case, the generalized continuous collection equation is

$$\dot{m}_{col} = \int_0^\infty A_m |V(m) - V(m')| \, m' N(m') \Sigma_c(m, m') \, dm' \qquad (3.26)$$

where $N(m') \, dm'$ is the number of particles per unit volume of air in the size range m' to $m' + dm'$.

3.1.4.2 Stochastic Collection

Cloud drop growth by coalescence is actually not a continuous process, as assumed in (3.24), but rather proceeds in a discrete, stepwise, probabilistic manner. In a time interval Δt, drops of a given initial size do not grow uniformly. Some may undergo more than the average number of collisions and thus grow faster than others. Consequently, a drop size distribution develops.

The probabilistic nature of collection may be accounted for by considering the size distribution $N(m,t)$, where $N(m,t) \, dm$ is the number of particles per unit volume of air in mass range m to $m + dm$ at time t. The change in $N(m,t)$ with time is computed as follows. The rate at which the space within which a particle of mass m' is located is swept out by a particle of mass m is given by the *collection kernel*, defined as

$$\hat{K}(m, m') \equiv A_m |V(m) - V(m')| \Sigma_c(m, m') \qquad (3.27)$$

The probability that a particular drop of mass m will collect a drop of mass m' in time interval Δt is

$$\hat{P} \equiv N(m',t) \, dm' \, \hat{K} \Delta t \qquad (3.28)$$

where it is assumed that Δt is small enough that the probability of more than one collection in this time is negligible. Making use of (3.27) and (3.28), we note that the mean number of drops of mass m that will collect drops of mass m' at time Δt is

$$\hat{P} N(m,t) \, dm = \hat{K}(m, m') N(m',t) N(m,t) \, dm \, dm' \, \Delta t \qquad (3.29)$$

Rearranging this expression we obtain

$$\frac{\hat{P} N(m,t)}{\Delta t} = \hat{K}(m, m') N(m',t) N(m,t) \, dm' \qquad (3.30)$$

which expresses the rate at which the number of drops of mass m is reduced as a result of coalescence with drops of mass m' per unit volume of air. It follows that the rate of decrease of the number concentration of drops of mass m as a result of their coalescence with drops of all other sizes is given by the integral

$$I_1(m) = \int_0^\infty \hat{K}(m,m')N(m',t)N(m,t)dm' \tag{3.31}$$

By reasoning similar to that given above we may express the rate of generation of drops of mass m by coalescence of smaller drops as

$$I_2(m) = \frac{1}{2}\int_0^m \hat{K}(m - m',m')N(m - m',t)N(m',t)dm' \tag{3.32}$$

where the factor of 1/2 is included to avoid counting each collision twice. The net rate of change in the number density of drops of mass m is obtained by subtracting (3.32) from (3.31) and may be written as

$$\left(\frac{\partial N(m,t)}{\partial t}\right)_{col} = I_2(m) - I_1(m) \tag{3.33}$$

This result is referred to as the *stochastic collection equation*.

   Computations may be made with (3.33) starting with some arbitrary initial drop size distribution $N(m,0)$. The result obtained by integrating (3.33) over time yields the drop size distribution altered by the stochastic collection process. In addition to the initial distribution, one must also assume reasonable values of the collection efficiencies and fall velocities appearing in (3.31) and (3.32). For realistic conditions, it is generally found that a large portion of the liquid water accumulates in the tail of the distribution. An example of such a calculation is shown in Fig. 3.5.
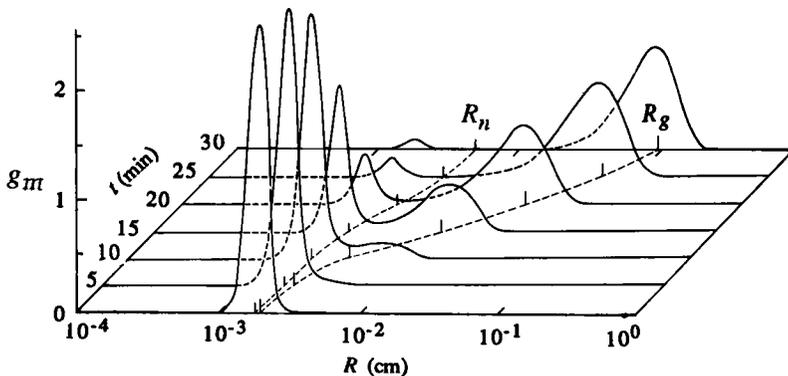

Figure 3.5 Example of the evolution of a drop size distribution as a result of stochastic collection. g_m is the mass distribution function; R is the drop radius. The two dashed lines show the radii (R_n and R_g) corresponding to the means of the number and mass distributions, respectively. (From Berry and Reinhardt, 1973. Reprinted with permission from the American Meteorological Society.)
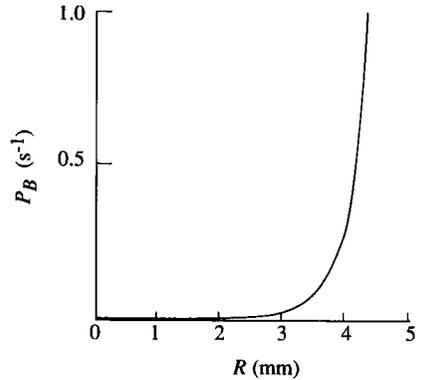
Figure 3.6 The probability $P_B(m)$ that a drop of radius R breaks up per unit time. Based on empirical formula of Srivastava (1971).

The drop size distribution at successive times is plotted as mass distribution $g_m \equiv m N(m)$, rather than number distribution $N(m)$, so that the area under each curve is proportional to the total liquid water content in the distribution. The mass distribution is plotted versus the radius of a drop of mass m on a logarithmic scale. This plotting convention emphasizes the result that a large portion of the liquid water becomes concentrated in the large drops as time progresses. The two peaks in the mass distribution after 30 min correspond to the amount of water contained in cloud droplets (radii $\sim 10^{-3}$ cm) and raindrops (radii $\sim 10^{-1}$ cm). The two dashed lines following the centers of the two peaks correspond to the means of the number and mass concentrations. The mean of the number distribution follows the cloud droplet peak. This result illustrates that the cloud droplets are far more numerous than the raindrops but that the latter nonetheless contain a large part of the liquid water after half an hour of stochastic collection. Stochastic collection can thus quickly convert cloud water to rainwater.

3.1.5 Breakup of Drops

When raindrops achieve a certain size, they become unstable and break up into smaller drops. Breakup has been studied in the laboratory, and empirical functions based on the experimental data are used to describe breakup quantitatively.[54] One empirical function is the probability $P_B(m)$ that a drop of mass m breaks up per unit time. It is nearly zero for drops less than about 3.5 mm and increases exponentially with size for radii greater than this value (Fig. 3.6). The function shown in the plot is

$$P_B(m) = 2.94 \times 10^{-7} \exp(3.4R) \tag{3.34}$$

where R is the radius in millimeters of a drop of mass m and $P_B(m)$ is in s^{-1}. A second empirical function is $Q_B(m',m)$, which is defined such that $Q_B(m',m)\,dm$ is the number of drops of mass m to $m + dm$ formed by the breakup of one drop of

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[54] The formulation of breakup presented in this subsection was developed by Srivastava (1971).

mass m'. $Q_B(m',m)$ is approximately exponential. It is given by

$$Q_B(m', m) = 0.1R'^3 \exp(-15.6R) \tag{3.35}$$

where the radii are in cm. The empirical functions $P_B(m)$ and $Q_B(m',m)$ can be used to determine the net effect of breakup on the drop size distribution $N(m,t)$. The net rate of production of drops of mass m by breakup implied by these functions is

$$\left(\frac{\partial N(m,t)}{\partial t}\right)_{\text{bre}} = -N(m,t)P_B(m) + \int_m^\infty N(m',t)Q_B(m',m)P_B(m')dm' \tag{3.36}$$

3.2 Microphysics of Cold Clouds

3.2.1 Homogeneous Nucleation of Ice Particles

Ice particles in clouds may be nucleated from either the liquid or vapor phase. Homogeneous nucleation of ice from the liquid phase is analogous to nucleation of drops from the 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

$$R_{ci} = \frac{2\beta_i \sigma_{il}}{\alpha_i n_i k_B T \ln\left(e_s/e_{si}\right)} \tag{3.37}$$

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 ice in the denominator and the free energy in the numerator are all functions of temperature. The critical radius is thus a function of temperature.

   Theoretical and empirical results indicate that homogeneous nucleation of liquid water occurs at temperatures lower than about -35 to $-40°C$, depending somewhat on the size of the drops being subjected to the low temperature.[55] This threshold lies within the range of temperatures in natural clouds, which may have cloud-top temperatures below $-80°C$. It is therefore possible, in a natural cloud, to have unfrozen liquid (i.e., *supercooled*) drops in the temperature range of $0°C$ to about $-40°C$. However, wherever the temperature in the cloud is below about $-40°C$, any liquid drops that happen to be present freeze spontaneously by homogeneous nucleation. This conclusion is consistent with the fact that at tempera-

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[55] Larger drops freeze homogeneously at slightly higher temperatures than smaller ones (Rogers and Yau, 1989, p. 151).