A CONVENIENT NUCLEUS PARAMETER FOR CONSIDERATIONS OF DROPLET GROWTH

by

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RÉSUMÉ

La quantité r_0 est définie comme le rayon d'une gouttelette quand elle est en équilibre avec son entourage à une humidité de 100 %. On montre ici que r_0 est un paramètre fondamental des noyaux et que r_0 permet une représentation de la croissance des gouttelettes servant d'exemple. Cette représentation est utile à des fins pédagogiques.

ABSTRACT

The quantity r_0 is defined as the radius of a droplet when it is in equilibrium with its environment at 100 % humidity. It is shown here that r_0 is a fundamental nucleus parameter and that r_0 allows an illustrative representation of droplet growth. This representation is useful for pedagogical purposes.

INTRODUCTION.

In dealing with the growth of cloud droplets by condensation it is illustrative to formulate the growth equation in terms of the equilibrium radius r_0 of the droplet at 100 % humidity. This can easily be done with the equations as used by Howell [1], Mordy [2], and several textbooks, where the assumption is made that a spherical water surface has already formed about the condensation nucleus. When the droplet radius is large enough the adjustment of the droplet's vapor pressure by surface tension and dissolved foreign molecules may be simply approximated. This approximation leads directly to the use of r_0, which results in a picturesque illustration of the growth of a distribution of droplets on a distribution of condensation nuclei.

Following in a general way the derivation of the droplet growth equation by Mordy [2], we define

$$\text{Surface tension term, } A = 2\sigma/\rho RT \doteqdot 10^{-3}\ (\mu), \tag{1}$$

$$\text{Raoult's law term, } B = 3iM/4\pi\rho \doteqdot 10^{11}M\ (\mu^3), \tag{2}$$

$$\text{and } U = A/r - B/r^3, \tag{3}$$

and write the applicable equations for droplet condensation,

$$\text{vapor diffusion: } r\dot{r} = (D/\rho RT_\infty)\ (e_\infty - e_r), \tag{4}$$

$$\text{droplet vapor pressure: } e_r = e_{\Gamma}\ (1 + U), \tag{5}$$

Clausius-Clapeyron : $e_T = e_s - (Le_s/T^2_\infty R)(T_\infty - T)$, (6)

heat diffusion : $(T_\infty - T) = (1/4\pi rk)\,q$, (7)

heat continuity : $(1/4\pi r)\,q = (\rho c/3)\,r^2T - L\rho rr$, (8)

where

σ	=	surface tension
ρ	=	water density
R	=	gas constant
i	=	van't Hoff factor
M	=	mass of nucleus in gm.
r	=	radius of drop in microns
D	=	vapor diffusion coefficient
T_∞	=	temperature of the environment
T	=	temperature of the drop
e_∞	=	vapor pressure of the environment
e_r	=	vapor pressure near droplet surface
e_T	=	saturation vapor pressure at temperature T
e_s	=	saturation vapor pressure at temperature T_∞
L	=	latent heat of condensation
k	=	heat diffusion coefficient
c	=	specific heat of water
q	=	heat content of droplet

The dot above a quantity indicates its time derivative. Using (4), (5) and (6) to eliminate e_r and e_T, and (7) and (8) to eliminate $\dot{q}$, we have

$$r\dot{r} = (De_s/\rho RT_\infty)\left\{e_\infty/e_s - [1 + U]\,[1 - (L/RT^2)(T_\infty - T)]\right\}, \quad (9)$$

$$r^2\dot{T} + (3k/\rho c)(T_\infty - T) + (3L/c)\,r\dot{r}. \quad (10)$$

From (10) we find that the time response τ of the temperature (T) of the droplet, to changes in the temperature (T_∞) of its environment is

$$\tau_T = \rho c r^2/3k \doteqdot 10^{-4}\,r^2 \text{ (sec)}, \quad (11)$$

assuming $\dot{r} = 0$.

Reasoning from this we conclude that the term $(L/RT^2)(T_\infty - T) \doteqdot 20\,(T_\infty - T)/T$ in (9) has an absolute value much less than 1 under all regular conditions in cloud and laboratory. Also the terms in U are taken to be much smaller than 1 since they result originally from an expansion requiring just this. Therefore (9) may be rewritten to give

$$r\dot{r} = D'\,[E - U + (L/RT^2)(T_\infty - T)], \quad (12)$$

where

$$E \equiv e_\infty/e_s - 1 = \text{supersaturation}, \quad (13)$$

$$D' \equiv De_s/\rho RT_\infty \doteqdot 10^2\ (\mu^2/\text{sec}) \quad (14)$$

The supersaturation of the environment is denoted by E. It is independent of the parameters of the droplet but is affected as nearby droplets grow. The term U is a function only of the droplet parameters and is independent of the environment.

The $(T_\infty - T)$ term in (12) is eliminated by (10). We neglect the $\dot{T}$ term for our present purposes and re-solve for $r\dot{r}$ to get

$$r\dot{r} = H\,(E - U), \tag{15}$$

where H is a combination of parameters that change slowly and is of the order of 10^2 $(\mu^2/\text{sec.})$. The $r\dot{r}$ term which appeared on the right before re-solving to get (15) accounts for the negative feedback upon droplet growth due to the raising of the droplet's vapor pressure by the heat of condensation. This feedback term is now included in H.

Rooth [3] reasoned that $\dot{r}$ cannot increase without limit as r is decreased and suggested a correction to the diffusion coefficient. When this is accounted for the r on the left of (15) is replaced by $(r + l')$, where l' is approximately 2μ. Thus we have

$$(r + l')\,\dot{r} = H\,(E - U). \tag{16}$$

The use of r_0.

If we re-write (3) in the form

$$U \equiv (A/r)\,(1 - r_0^2/r^2) \tag{17}$$

$$\text{where } r_0 \equiv \sqrt{B/A}, \tag{18}$$

we notice that the r_0 so defined is just the equilibrium radius for $E = 0$ and includes all of the parameters of the condensation nucleus used in the preceding, common account of condensation theory.

For the viewpoint of condensation theory, the value of r_0 is more convenient and representative than the various properties of the nucleus itself. While r_0 has here evolved from a form of condensation theory designed to treat salt nuclei, its usefulness is not so restricted. The fundamental consideration in B (equation 2, above), is how the nucleus lowers the droplet's vapor pressure by replacing some of the water molecules at the droplet's surface.

The term U is not really valid down to $r = 0$ since it is made up of the leading terms of the expansions of the Raoult's law and the surface curvature effects. Each of the terms of U must therefore be small compared to 1. If we set the maximum value of each of the terms in equation (3) equal to 1/3, then the minimum radius at which U is valid is given by the larger of

$$r_{min} = (3B)^{\frac{1}{3}} \doteq 0.05\ r_0^{\frac{2}{3}}, \tag{19}$$

$$r_{min} = 3A \doteq 3 \times 10^{-3}. \tag{20}$$

For all but the smallest salt nuclei (19) prevails, and in all but very dry atmospheric conditions the above criteria are satisfied so that the term U in the form of equation (3) is valid.

An Illustrative Representation of Droplet Growth.

Since the value of r_0 (along with r_{min}) contains all necessary information about a nucleus once condensation has begun, it seems natural to plot the function r_0 (r) and watch its behavior during condensation. Solving (17) for r_0 we have

$$r_0 = r \sqrt{1 - U' r}, \tag{21}$$

$$U' = U/A \doteq 10^3 U \tag{22}$$

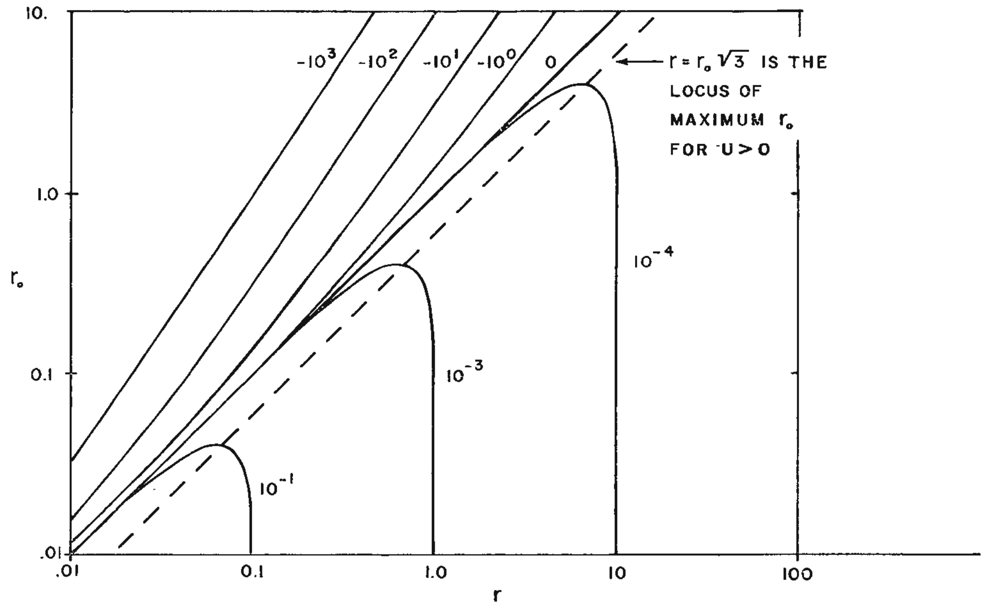

FIG. 1. — The iso-U lines ; which are also iso-E lines of the same value for $E \geqslant -1$.

In Figure 1 are sketched the iso-U lines, with r_0 and r plotted on logarithmic coordinates since several orders of magnitude need to be encompassed. This plot has several conveniences. $U = 0$ is just the line $r = r_0$. As U increases above zero its curve keeps the same shape while it slides down the line $U = 0$. The maximum r_0 for $U > 0$ lies on the line $r = r_0 \sqrt{3}$. The value of U/A for a given curve is just the recipricol of the r approached by the right leg of U for small r_0. When $U < 0$ its curve lies above $U = 0$ and its change of position occurs only slowly with change in U.

The curves of U are also the curves of E, so that for a given E, $U = E$ is an equilibrium curve. Equilibrium is stable when $r < r_0\sqrt{3}$ and unstable when $r \geqslant r_0\sqrt{3}$. Supersaturation is undefined for values less than -1, corresponding to perfectly dry air, and therefore we must have $E \geqslant -1$. It is apparent then that U may be much less than E even in dry air, which suggests a strong « potential » for droplet growth up to $U = -1$. It must be remembered, however, that the formulation used here ignores the important aspect of the initial water condensation on the nucleus and therefore tells nothing about the initiation of condensation. The formulation applies only after a complete water surface has formed and sufficient water has condensed to make the solution at the droplets surface reasonably dilute.

The changes in the van't Hoff factor [4] as the solution goes from concentrated to dilute could be accounted for by lines on this graph showing how r_0 changes with r. Here we have assumed that r_0 is a constant for a given nucleus.

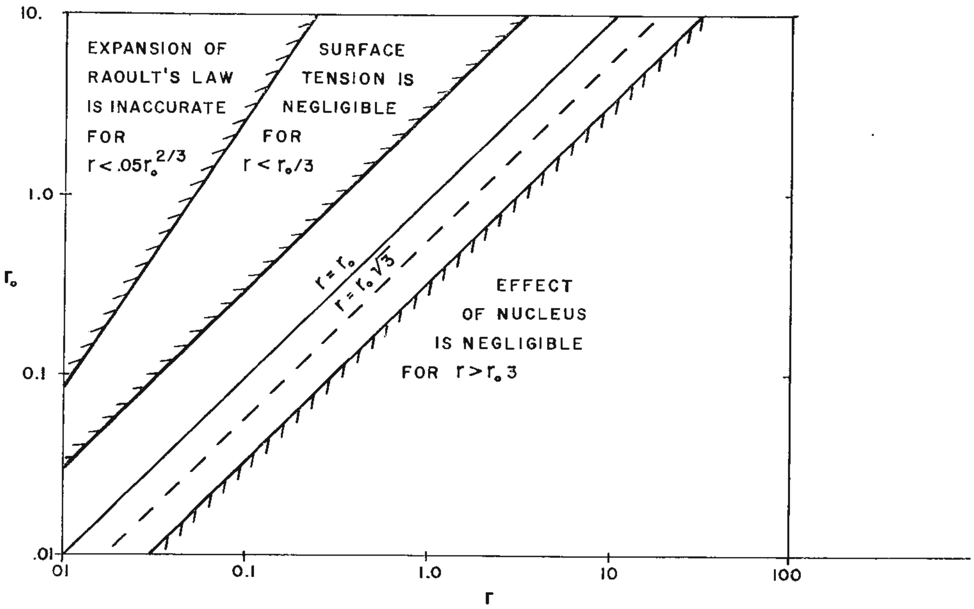

FIG. 2. — The regions where surface tension or nucleus effect become insignificant are shown along with the area where the dilute solution approximation becomes incorrect.

Figure 2 outlines the significant regions of the r, r_0-plane. The region excluded by (19) is shown in the upper left corner while the region excluded by (20) is far to the left of the range of r shown here.

Notice that the term r_0^2/r^2 in (17) gives the ratio of the nucleus effect to the surface tension effect. By putting this term either greater than or less than 9 we can define the regions where one effect or the other becomes negligible. These areas are also sketched in Figure 2.

Another requirement on r is that it be no smaller than the average radius of the nucleus itself. For salt, r_0 is large, and thus the r_{min} of (19) will always be much larger than the nucleus. However, once a water surface has formed around an insoluble nucleus, $B = 0$ and therefore $r_0 = 0$. Then (20) will have no effect and r_{min} will be determined by the size of the nucleus. The quantity r_0, even though less than r_{min}, will still govern the subsequent growth of the droplet by condensation.

A large insoluble nucleus will be represented by a point at low r_0 and large r_{min}. We see from Figure 2 that the nucleus effect would be negligible in condensation considerations, but also we see from Figure 1 that the r_{min} is very important. For if E were raised enough for its curve to go below the point (r_{min}, r_0) for this nucleus, the droplet on it may begin to grow. Thus the fact that a large r_{min} may initiate droplet growth is shown clearly on this diagram.

A population of nuclei is represented by some curve r_0 (r). Suppose that the $t = 0$ line in Figure 3 gives the initial size of the droplets about each nucleus. The line r_0 (r) is really a locus of points, each point representing a droplet and its nucleus. The projection of these points on either the r_0 or r axes gives a density of points which is proportional to the density functions f_0 (r_0) and f (r) of the nuclei and the droplets respectively.

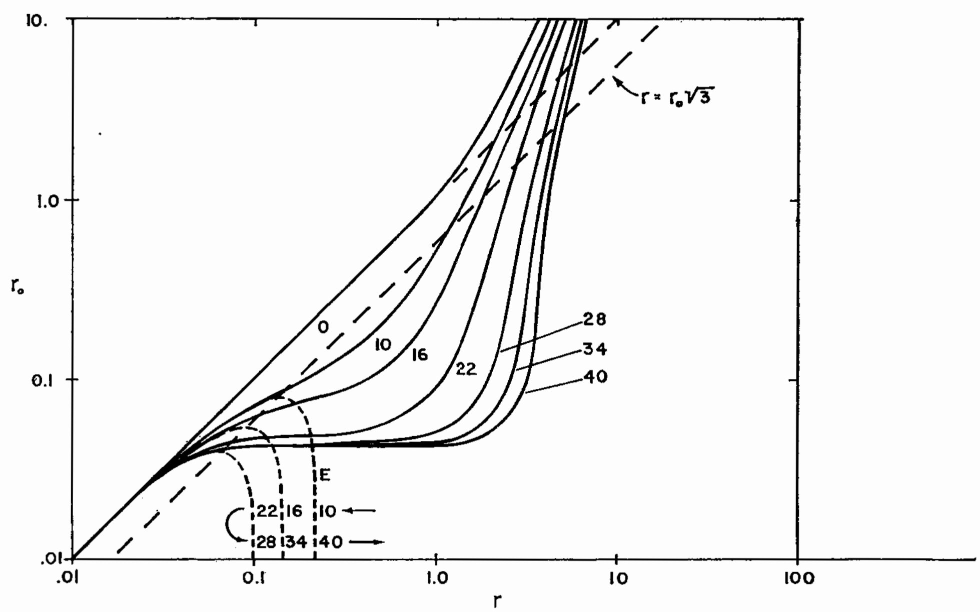

FIG. 3. — A plot of r_0 (r) for the droplet growth by condensation as computed by Mordy [2] for his distribution I at 1 m/sec vertical velocity.

Figure 3 illustrates the growth of droplets at a cloud base as computed by Mordy [2]. The results shown in his Figure 12 for 1 m/sec vertical velocity are here plotted on the r, r_0-plane. Curves of r_0 (r) and E are labeled with the elapsed time in seconds. The fact that as E is increased the droplets with lower r_0 are freed to grow is clearly shown. Those droplets with $r > r_0 \sqrt{3}$ have grown beyond the crest of their U and continued growth brings them into a region of lower U. They will continue to grow until E is decreased enough for its curve to lie above the point (r, r_0).

SUMMARY.

The equilibrium radius r_0 is a convenient parameter for the classification of condensation nuclei. This information must be supplemented by the nucleus radius itself when this has a greater value than r_0. The plot of r_0 (r) shows clearly the characteristics of condensation growth, including the regions where surface tension or nuclei effects predominate. The use of f_0 (r_0) is useful to the cloud physicist in the study and teaching of condensation and evaporation as it occurs in both the real cloud and the laboratory.

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