

THE GROWTH OF CLOUD DROPS IN UNIFORMLY COOLED AIR

By *Wallace E. Howell*¹

Blue Hill Meteorological Observatory, Harvard University²

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ABSTRACT

Recent studies of precipitation, aircraft icing, and visibility through fog have focussed attention on the physical constitution of clouds, a subject to which knowledge of the drop-size spectrum and its origin would be an important contribution. The drop-size spectrum resulting when air containing condensation nuclei is uniformly cooled may be computed, leading to a differential equation for the growth of a cloud drop which cannot be integrated analytically. A numerical method of integration is therefore employed.

Three integrations of drop growth under simulated natural conditions are described and compared with data for natural clouds, leading to the following conclusions:

The computed drop-size spectra agree with observation regarding the relatively uniform clouds most frequently observed. Convergence and mixing of fine-grained turbulence are suggested as influences broadening the distribution in the less uniform clouds. Even for the latter, the computed spectra agree with observation on the shape of the spectrum curve. It is not clear what conditions of cooling most favor a broad spectrum. The computed mean drop sizes agree very well with observation, indicating that growth by collision is not ordinarily significant in uniform clouds.

The concentration of drops is determined primarily by the rate of cooling during the initial stage of condensation. It depends, as a rule, only slightly on the concentration of condensation nuclei. With continued uniform cooling, the drop concentration diminishes slightly toward a fixed constant value. It can be increased, once the cloud has formed, only through a great increase in the rate of cooling.

Supersaturation during cloud formation ordinarily reaches about 0.1 per cent and can surpass 1 per cent only under extreme circumstances. Subsaturations in descending air currents have the same order of magnitude. Observations of lower relative humidities in clouds, if real, must indicate clear spaces.

Operation of the equation of growth on nuclei of modal sizes proposed by Dessens cannot explain the modal drop sizes reported by Köhler.

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1. Introduction

Several questions in recent years have focussed attention on the physical processes which underlie the formation of clouds, changes in their constitution, and their conversion into precipitation. The origin of natural rain, the artificial dissipation of fog, the protection of aircraft from icing, seeing through cloudy air, and most recently the artificial release of precipitation from clouds, all have led to demands for funda-

mental knowledge about how clouds are constituted and how they came to be that way.

The constitution of a cloud may be described by specifying the drop-size "spectrum," the amount of water per unit volume of cloud in drops in each portion of the range of sizes. Infinitesimal subdivision of the size range yields a continuous spectrum which may be used to find other properties of the cloud, such as concentration of drops, the average drop size, a measure of the breadth of distribution, and other data that may be required for one purpose or another.

This paper addresses itself in general to the problem of the physical origin of the drop-size spectrum and in particular to considering the adequacy of condensation processes operating alone in uniformly cooled air as an explanation for observed drop-size spectra.

2. The equation of growth of a droplet

It has been established by Wilson [26], Wigand [27], Köhler [17], Aitken [1] and others that the water drops of natural clouds condense upon nuclei consisting of minute quantities of hygroscopic chemicals which are normally present in large concentrations. At the relative humidity where cloud begins to form, the nuclei are always liquid, the chemicals being

¹ Abbott Lawrence Rotch Research Fellow, Blue Hill Observatory, Harvard University.

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dissolved in water. The balance of hygroscopic forces against the force of surface tension combines with the relative humidity of the surrounding air to determine whether the droplet will grow or diminish in size. The rate of growth depends not only upon the resultant of these forces but also upon the processes of diffusion and thermal conduction which transport mass and heat to and from the droplet.

The derivation here presented of the equilibrium radius of a drop differs only in detail from those previously given by Köhler [17], Wright [29] and others, and is repeated only for the sake of investigating all pertinent assumptions. The equations are put in dimensionless form, and the gram molecular weight or *mole* is used as the unit of mass of the hygroscopic material in the nucleus.

A. The effect of surface tension on vapor pressure over a drop.—A spherical drop of water is under pressure exerted by the surface tension. If the drop grows, work is done. Lord Kelvin [16] showed that this work must be supplied by an excess of vapor pressure over the surface of the drop in obedience to the formula³

$$\ln \frac{e_w'}{e_w} = \frac{F_1}{r} \approx \frac{e_w' - e_w}{\bar{e}_w}, \quad (1)$$

where $F_1 \equiv 2\rho_a\gamma/\rho_w e_w r_0$ and \bar{e}_w is a mean of the saturation vapor pressures. Values of F_1 are shown in table 1 for several different temperatures.

TABLE 1. Values of F_1 .

T	273	268	263	258	253	248	243	238	233
F_1	1.200	1.234	1.269	1.308	1.348	1.392	1.439	1.492	1.552

B. The effect of dissolved substances.—Raoult's law may be stated in the form

$$\frac{e_w'' - e_w}{\bar{e}_w} = - \frac{M}{M + M'}.$$

This law is valid only for solutions so dilute as to be virtually completely dissociated. Since, in the integrations that follow, only conditions very near 100 per cent relative humidity are ever considered, the law may be used for even the smallest nucleus, 10^{-17} moles, likely to be involved. Furthermore, for the same reason, the M in the denominator may be neglected against the much larger M' , and

$$\frac{e_w'' - e_w}{\bar{e}_w} = - \frac{F_2}{r^3}, \quad (2)$$

where $F_2 \equiv 3m_w\nu/4\pi\rho_w r_0^3$. The quantity F_2/ν is shown in table 2 for various temperatures.

A corollary of equation (2) is that in terms of molecular concentration the nature of the solute, if it

be dissociated in the solution, has no influence whatsoever upon the vapor pressure over the drop. Nuclei composed of only slightly hygroscopic substances are at no disadvantage with respect to more hygroscopic ones. With supersaturation, condensation will occur most rapidly on the drops containing the most molecules of solute regardless of the substance in solution.

C. Equilibrium relative humidity over a drop of solution.—Addition of equations (1) and (2) yields

$$\frac{e_w''' - e_w}{\bar{e}_w} = \frac{F_1}{r} - \frac{F_2}{r^3}, \quad (3)$$

from which is constructed the graph, fig. 1, showing the equilibrium conditions over drops growing from nuclei of various sizes. Fig. 1 is identical in form to similar ones published by Köhler, Wright, and Houghton. To the left of the broken line the drops may be in stable equilibrium; to the right of it the equilibrium is unstable.

D. Diffusion of water vapor about a drop.—Mass transfer by diffusion may be stated dimensionally in its customary form as

$$\frac{dq}{dt'} = \frac{4\pi r'^2 D \rho_v}{\bar{e}} \frac{de}{dr'},$$

where \bar{e} is a mean vapor pressure in the vicinity of the drop. When this equation is integrated with respect to e and r' from the surface of the drop to an infinite distance, the well-known result is obtained that

$$r' \frac{dr'}{dt'} = \frac{D \rho_v}{\rho_w} \left(\frac{e - e_w'''}{\bar{e}} \right).$$

However, in arriving at this result, several assumptions have been made. One of them, that drops in a cloud do not affect one another, has been discussed at length by Langmuir [19] and shown to be permissible. Another is that the amount of vapor enclosed between two spherical shells concentric with the drop is constant. Calculation of the actual mass divergence in a sample case shows it to be of the order of 6×10^{-13} gm per drop, less than a thousandth the mass of an average drop and hence negligible.

TABLE 2. Values of F_2/ν .

T	273	268	263	258	253	248	243	238	233
F_2/ν	4.30	4.30	4.30	4.31	4.32	4.34	4.36	4.39	4.45

A third assumption not so easily dismissed is that the law of diffusion in its usual form holds for any system. This is not true, for when the size of the particle about which diffusion is occurring is of the same or smaller order of magnitude than the mean free path of the vapor, mass particles are exchanged directly over a major portion of the range of integration. For

³ A table of symbols is appended at the end of the paper.

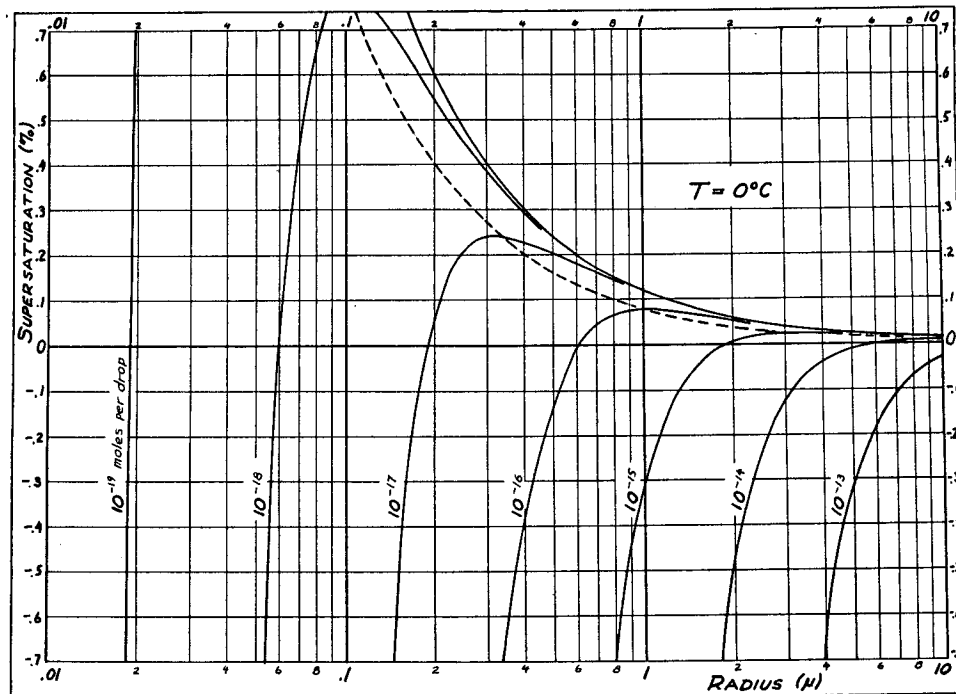


FIG. 1. Equilibrium supersaturation over drops of water containing given amounts of solute.

this case, Langmuir [19] gives the law of diffusion as

$$r' \frac{dr'}{dt'} = \frac{\Delta \rho_v}{\rho_w} \left(\frac{e - e_w'''}{\bar{e}} \right),$$

where

$$\Delta \equiv \left[\frac{r'}{D(r' - \alpha \lambda_w)} + \frac{1}{r'} \sqrt{\frac{2\pi \rho_v}{\bar{e}}} \right]^{-1}.$$

The quantity Δ may be regarded as a compensated diffusion coefficient. At a temperature of -20°C it is given within ± 3 per cent by

$$\begin{aligned} \Delta &= D, & \text{when } r > 5.9 \\ \Delta &= r' \sqrt{2\pi \bar{e} / \rho_v}, & \text{when } r < 0.018. \end{aligned}$$

In order to put the above equation in dimensionless form it is necessary to introduce a reference time interval, $t_0 = 1$ sec, dividing the time t' by t_0 to give t , the dimensionless number of seconds. The diffusion equation then becomes

$$\frac{e - e_w'''}{\bar{e}} = F_4 r' \frac{dr'}{dt}, \quad (4)$$

where $F_4 \equiv \rho_w r_0^2 / \rho_v t_0 \Delta$.

E. The heat balance and thermal conduction.—The heat balance requires that within a fixed interval of time the liberated heat of condensation and the heat contained in the added water together equal the heat stored within the drop and that escaping from it by conduction. The second and third terms cancel one another except for a term containing the rate of change of drop temperature; computation using typical values shows that this is negligible for cloud drops,

although it may be of concern for rain. The first and fourth terms remain:

$$\begin{aligned} \frac{dQ_1}{dt'} &= 4\pi r'^2 \rho_w L \frac{dr'}{dt'}, \\ \frac{dQ_4}{dt'} &= -4\pi r'^2 k \frac{dT}{dr'}. \end{aligned}$$

This last expression may be integrated in T and r' subject to the same assumptions as in the case of mass diffusion, and the mutual interference between drops and the divergence of heat may be similarly neglected. In the case of small drops, it is likewise necessary to use a compensated coefficient of thermal conductivity which may be deduced from the kinetic theory of gasses. It is assumed that the heat is transported by the individual molecules which, when they collide with the particle, undergo a temperature change proportional to the difference between their initial temperature and the temperature of the particle. The factor of proportionality is called the accommodation factor, f . Given the area of the particle, the number of collisions on it may be computed, and there is derived the equation

$$\frac{dQ_4}{dt'} = \pi r'^2 \rho_a c_p f (T_w - T) \sqrt{\frac{3\bar{e}}{\rho_v}}.$$

Between the realms of the two conductivity laws, an expression comparable to that for diffusion may be used, so that

$$dQ_4/dt' = 4\pi r' K (T_w - T),$$

where

$$K \equiv \left[\frac{r'}{k(r' + \alpha\lambda_a)} + \frac{4}{r'\rho_a c_p f} \sqrt{\frac{\rho_v}{3\bar{e}}} \right]^{-1}.$$

At -20°C , within ± 3 per cent, the compensated coefficient of thermal conductivity is

$$K = k, \quad \text{when } r > 7.9$$

$$K = \frac{1}{4} r' \rho_a c_p f \sqrt{3\bar{e}/\rho_v}, \quad \text{when } r < 2.4 \times 10^{-2}.$$

Equating the heat gain and loss and expressing the result in dimensionless form, we have

$$\frac{T_w - T}{T} = F_5 r \frac{dr}{dt}, \quad (5)$$

where $F_5 \equiv L\rho_w r_0^2 / K T t_0$.

F. Variation of vapor pressure with temperature.—Clapeyron's equation, if the specific volume of liquid water is neglected in comparison with that of vapor, may be written

$$\frac{de_w}{dT_w} = \frac{JL\rho_v}{T_w}.$$

Integrating this expression between the limits T_w , e_w at the drop and T , $e_{w(T)}$ in the surrounding air gives

$$\ln \frac{e_w}{e_{w(T)}} = - \frac{\rho_v \bar{L} J T}{\bar{e}_w} \left(\frac{1}{T_w} - \frac{1}{T} \right),$$

where $e_{w(T)}$ is the saturated vapor pressure of water at the temperature of the ambient air and \bar{L} is chosen as the mean value of L through the temperature interval. Because of the small proportional difference between the vapor pressures, an approximate expression may be substituted for the logarithm,

$$\frac{e_w - e_{w(T)}}{\bar{e}_w} = F_6 \frac{T_w - T}{T_w}, \quad (6)$$

where $F_6 \equiv \rho_v \bar{L} J / \bar{e}_w$.

G. The equation of growth in differential form.—The conditions of growth are now compassed, and the rate of growth of the drop in terms of its size and the prevailing conditions of temperature and supersaturation is now obtainable by combining equations (5) and (6) to give

$$\frac{e_w - e_{w(T)}}{\bar{e}_w} = F_5 F_6 r \frac{dr}{dt},$$

and adding this to equations (3) and (4) to yield

$$\frac{e - e_{w(T)}}{\bar{e}_w} = \frac{S}{100} = (F_4 + F_5 F_6) r \frac{dr}{dt} + \frac{F_1}{r} - \frac{F_2}{r^3}. \quad (7)$$

The assumption has been made here that the proportional difference between \bar{e}_w and \bar{e} is small, which we will see is justified. This is the equation of growth in

differential form. Unfortunately analytical integration of it is not possible without approximations which eliminate precisely those factors which it is the purpose of this paper to study. Stepwise integration was therefore undertaken, but the tedious nature of the process made it necessary to limit the number of independent variables as much as possible. Accordingly it was assumed that the ambient temperature and pressure do not change significantly during the cloud-forming process. Time, radius, supersaturation, and size of the condensation nucleus were the independent variables considered. The equation of growth was written in the form of finite increments,

$$\Delta t = r \Delta r (F_4 + F_5 F_6) \left[\frac{S}{100} - \left(\frac{F_1}{r} - \frac{F_2}{r^3} \right) \right]^{-1}. \quad (7.1)$$

The increment of radius, Δr , was taken as a tenth of a tenfold change in r ; that is, from $r = \text{antilog}(\log r_i - 0.05)$ to $r = \text{antilog}(\log r_i + 0.05)$. The value of Δt was then computed for a network of points in the r, S -plane for each of several nucleus sizes, with the temperature set at 0°C and the pressure at 800 mb. A selection of the results is shown in table 3. Fig. 2 comprises two maps of equation (7.1) for two different values of nucleus size. The equilibrium curves of fig. 1 reappear on these graphs as the isochrones $\Delta t = \infty$. With supersaturations well above equilibrium, the isochrones have a uniform slope where diffusion follows the ordinary law, increasing at small radii where the diffusion law changes. Where the distortion from uniform slope is small, the map for one nucleus size may be superimposed upon that for another size by a simple displacement along a line of slope $= -1$. In this way a single graph with a suitable overlay (such as fig. 3) suffices over a range of nucleus sizes. The actual integrations will be described in a later section.

Before passing to the next subject, a word is in order about the simplifications of the equation of growth which may be affected by neglecting one term or another. If the effect of the solute is neglected, as may be done in the case of cloud drops which have considerably exceeded the critical size, then equation (7.1) becomes

$$t = \int (F_4 + F_5 F_6) \left(\frac{S}{100} - \frac{F_1}{r} \right)^{-1} r dr. \quad (7.2)$$

At such drop sizes, $K = k$ and $\Delta = D$, so that F_4 and F_6 are invariant with r . This is the case considered by Langmuir [19] and Tribus⁴ and has been integrated analytically by them. A still cruder approximation is reached by neglecting the effect of surface tension, justifiable only when F_1/ν is small with respect to $S/100$ and most likely to be the case in rapid ascent

⁴ M. Tribus, "Corrections to Langmuir's 'Time of Rise' theory for the growth of droplets in a fog," informal report, 8 pp., 1945.

of air carrying relatively large drops. The equation may then be very easily integrated to give

$$t = (F_4 + F_5 F_6)(r_2^2 - r_1^2)(50/S). \quad (7.3)$$

This is an elaboration of the parabolic law of drop growth presented by Houghton [10] and others, now in a form which takes account of the heat balance of the drop.

3. Condensation nuclei

In order to establish a reasonable starting point for the integrations of the equation of growth, it is necessary to know something about the size spectra and concentrations of condensation nuclei in the atmosphere. Despite many studies, no approach to precision has been made in measuring these ubiquitous particles,

TABLE 4. Nucleus sizes (in moles) according to various authors.

Author	Minimum	Mean	Maximum
Bennett [3]	1.2×10^{-19}	3.5×10^{-18}	10^{-16}
Coste & Wright [6]	—	1.5×10^{-17}	—
Junge [13]	2×10^{-22}	1.4×10^{-19}	10^{-16}
Köhler [17]	—	1.2×10^{-17}	—
Jacobs [12]	10^{-12}	3×10^{-12}	10^{-11}
Bradbury & Fryer [4]	10^{-17}	2.2×10^{-17}	5×10^{-17}
Nolan & Guerrini [21]	—	2×10^{-17}	—
Wright [29]	—	3×10^{-18}	—
Yunker [30]	—	3×10^{-18}	—
Dessens [7]	10^{-16}	3×10^{-14}	10^{-13}

and the best that can be said is that there is reasonable agreement as to orders of magnitude.

A. Size.—Table 4 compares the sizes stated by several authorities. For this tabulation other units of mass have been converted to moles per nucleus. It was assumed, as Dessens [7] has shown is often the case,

TABLE 3. Values of Δt (seconds) for selected values of the radius r' (cm), the supersaturation S , and the mass ν (moles) of the condensation nucleus. The computations were performed for a temperature 273K and pressure 800 mb. (The tabular entries are to be interpreted thus: $9.510^4 = 9.510 \times 10^4$, $1.494^{-3} = 1.494 \times 10^{-3}$, etc.)

$-\log r'$	$S = 0.0$	0.01	0.02	0.05	0.1	0.2	0.5	1.0	2.0
$\nu = 0$									
2.5	-9.51^4	3.81^4	2.23^4	7.82^3	3.76^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-3.05^3	-1.83^4	4.58^3	9.64^2	4.16^2	1.95^2	7.51^1	3.71^1	1.845^1
3.5	-1.000^2	-1.357^2	-2.11^2	3.15^2	6.10^1	2.34^1	8.20	3.94	1.935
4.0	-3.53	-3.85	-4.23	-6.05	-2.12^1	5.29	1.115	4.81^{-1}	2.25^{-1}
4.5	-1.494^{-1}	-1.535^{-1}	-1.578^{-1}	-1.721^{-1}	-2.03^{-1}	-3.16^{-1}	4.73^{-1}	9.15^{-2}	3.50^{-2}
5.0	-8.78^{-3}	-8.85^{-3}	-8.93^{-3}	-9.15^{-3}	-9.57^{-3}	-1.052^{-2}	-1.50^{-2}	-5.11^{-2}	1.335^{-2}
$\nu = 10^{-18}$									
2.5	-9.51^4	5.81^4	2.23^4	7.82^3	3.76^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-3.05^3	-1.83^4	4.58^3	9.64^2	4.16^2	1.95^2	7.51^1	3.71^1	1.845^1
3.5	-1.000^2	-1.357^2	-2.11^2	3.15^2	6.10^1	2.34^1	8.20	3.94	1.935
4.0	-3.54	-3.87	-4.25	-6.08	-2.16^1	5.27	1.115	4.18^{-1}	2.25^{-1}
4.5	-1.550^{-1}	-1.595^{-1}	-1.640^{-1}	-1.855^{-1}	-2.13^{-1}	-3.41^{-1}	4.24^{-1}	8.95^{-2}	3.47^{-2}
5.0	-1.356^{-2}	-1.372^{-2}	-1.392^{-2}	-1.450^{-2}	-1.580^{-2}	-1.826^{-2}	-3.80^{-2}	4.72^{-2}	8.54^{-3}
$\nu = 10^{-17}$									
2.5	-9.51^4	5.82^4	2.23^4	7.82^3	3.76^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-3.05^3	-1.83^4	4.58^3	9.64^2	4.16^2	1.95^2	7.51^1	3.71^1	1.845^1
3.5	-1.002^2	-1.364^2	-2.13^2	3.11^2	6.10^1	2.34^1	8.20	3.94	1.935
4.0	-3.67	-4.01	-4.43	-6.45	-2.70^1	5.03	1.103	4.80^{-1}	2.25^{-1}
4.5	-2.31^{-1}	-2.41^{-1}	-2.52^{-1}	-2.91^{-1}	-3.92^{-1}	-1.287	2.20^{-1}	7.45^{-2}	3.21^{-2}
5.0	3.34^{-3}	3.33^{-3}	3.32^{-3}	3.29^{-3}	3.24^{-3}	3.14^{-3}	2.88^{-3}	2.53^{-3}	2.03^{-3}
$\nu = 10^{-16}$									
2.5	-9.51^4	5.82^4	2.32^4	7.82^3	3.76^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-3.06^3	-1.87^4	4.55^3	9.64^2	4.16^2	1.95^2	7.51^1	3.71^1	1.845^1
3.5	-1.037^2	-1.425^2	-2.28^2	2.83^2	5.97^1	2.32^1	8.18	3.94	1.935
4.0	-5.49	-6.31	-7.42	-1.57^1	1.84^1	3.42	1.000	4.58^{-1}	2.20^{-1}
4.5	5.74^{-2}	5.68^{-2}	5.62^{-2}	5.46^{-2}	5.20^{-2}	4.76^{-2}	3.80^{-2}	2.84^{-2}	1.89^{-2}
5.0	1.92^{-4}	1.92^{-4}	1.92^{-4}	1.92^{-4}	1.91^{-4}	1.91^{-4}	1.90^{-4}	1.87^{-4}	1.83^{-4}
$\nu = 10^{-15}$									
2.5	-9.55^4	5.81^4	2.32^4	7.82^3	3.76^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-3.17^3	-2.33^4	4.34^3	9.52^2	4.14^2	1.95^2	7.51^1	3.71^1	1.845^1
3.5	-1.56^2	-2.64^2	-8.71^2	1.43^2	5.01^1	2.16^1	7.97	3.88	1.920
4.0	1.365	1.320	1.280	1.173	1.030	8.28^{-1}	5.21^{-1}	3.22^{-1}	1.83^{-1}
4.5	3.97^{-3}	3.97^{-3}	3.96^{-3}	3.96^{-3}	3.94^{-3}	3.91^{-3}	3.82^{-3}	3.69^{-3}	3.45^{-3}
4.7	3.63^{-4}	3.63^{-4}	3.63^{-4}	3.63^{-4}	3.63^{-4}	3.62^{-4}	3.60^{-4}	3.57^{-4}	3.50^{-4}
$\nu = 10^{-14}$									
2.5	-9.87^4	5.70^4	2.21^4	7.80^3	3.75^3	1.845^3	7.29^2	3.63^2	1.81^2
3.0	-4.75^3	1.595^4	2.98^3	8.66^2	3.97^2	1.905^2	7.44^1	3.69^1	1.84^1
3.5	3.86^1	3.50^1	3.21^1	2.56^1	1.91^1	1.272^1	6.33	3.43	1.81
4.0	9.90^{-2}	9.88^{-2}	9.85^{-2}	9.78^{-2}	9.66^{-2}	9.44^{-2}	8.84^{-2}	7.98^{-2}	6.71^{-2}
4.3	2.96^{-3}	2.96^{-3}	2.96^{-3}	2.96^{-3}	2.95^{-3}	2.94^{-3}	2.92^{-3}	2.87^{-3}	2.80^{-3}
$\nu = 10^{-13}$									
2.5	-1.485^5	4.77^4	2.06^4	7.60^3	3.70^3	1.83^3	7.25^2	3.62^2	1.81^2
3.0	1.182^3	8.93^2	7.18^2	4.52^2	2.80^2	1.59^2	6.90^1	3.55^1	1.805^1
3.5	2.70	2.68	2.66	2.60	2.51	2.34	1.960	1.535	1.073
4.0	7.61^{-3}	7.61^{-3}	7.61^{-3}	7.59^{-3}	7.59^{-3}	7.58^{-3}	7.53^{-3}	7.45^{-3}	7.28^{-3}

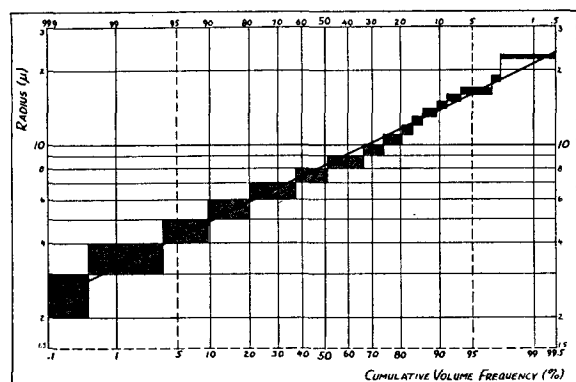


FIG. 4. Cumulative frequency of occurrence at Mount Washington of clouds according to the volume median radius of their drops. The shaded areas are interpreted according to the following example: the range between 6 and 7 microns radius corresponds to the range between 20 and 37 per cent cumulative liquid-water content. The sloping line is the best-fitted normal distribution.

defined spectrum which suggests a range of a hundred fold in size between the first and third quartiles of the distribution.

The shape of the distribution curve is virtually unknown. The Gaussian distribution would be a favored guess except that the unilateral limit at zero radius becomes serious when the distribution is so broad. The picture is more reasonable if a Gaussian distribution of the logarithm of the radius is assumed. Either assumption raises another objection, that giant nuclei larger than 10^{-13} moles are indicated in numbers larger than would be expected on the basis of observations. For the purpose of the present study it has been assumed that there is some factor highly effective in removing nuclei larger than about 10^{-13} moles.

C. Concentration.—More is known about the concentrations of nuclei, for they are easily counted with instruments, such as the Aitken counter, which are in widespread use. Reference has already been made to the study of their vertical distribution by Wigand [27]. Based on his data, a concentration of 2000 cm^{-3} has been taken as an average at the 800-mb level for the purposes of this study. He also indicates that the concentration varies less widely aloft than it does at the ground. Since even the average concentration of 2000 cm^{-3} is far larger than that of drops in most clouds, it makes little difference what the maximum is, but the minimum may be significant. In fresh, unstable maritime polar air at sea level, concentrations as low as 440 cm^{-3} were measured by Kahler and Zegula [15]; this gives the probable order of the minimum, considering the connection already pointed out between the richness of the vapor supply and the concentration of nuclei.

For the integrations of the equation of growth, initial conditions were assumed based on considerations here presented. These conditions are tabulated in a later section.

4. Drop sizes in natural clouds and fogs

The equation of growth deduced in section 2 is only one of the laws which operate on cloud particles. The present study can at best establish only the adequacy or inadequacy of the equation in explaining the observed results.

A. Size.—Observers are in substantial agreement on the size of fog and cloud drops, even though they have used a wide variety of measuring techniques. Notable contributors in this field include Köhler [17], Houghton and Radford [11], Diem [8], Arenberg,⁵ Schaefer [22], and Clark [6;8].⁶ The most suitable measure of the average drop size for our purpose is the volume median size; half of the water is contained in larger drops, half in smaller ones.

The volume median radius of cloud drops varies from about 3 to a maximum of about 15 microns; fog drops lie in the range from 15 to 30 microns median radius. In turbulent clouds the median radius seldom exceeds 10 microns. Fig. 4 shows the range of volume median radii observed in one year at Mount Washington as summarized by Conrad [9].

B. Spectrum.—Fig. 5 shows the volume spectrum measured by Houghton and Radford [11] (curve AA) in sea fog and by Vonnegut, Cunningham, and Katz [25] (curve BB) in a cloud. The close correspondence of both these curves with the Gaussian probability is remarkable, and it permits the use of the inverse of the "modulus of precision," here called the modulus

⁵ D. L. Arenberg, "Meteorological factors affecting the icing of aircraft," M.S. thesis, Massachusetts Institute of Technology, Cambridge, 64 pp., 1942.

⁶ V. F. Clark, "Analysis of Mount Washington Observatory cloud base data," *Mount Washington Observatory Icing Report*, July 1945, 8-18.

—, "The multicylinder method," *Mount Washington Observatory Icing Report*, June 1945, 32 pp.

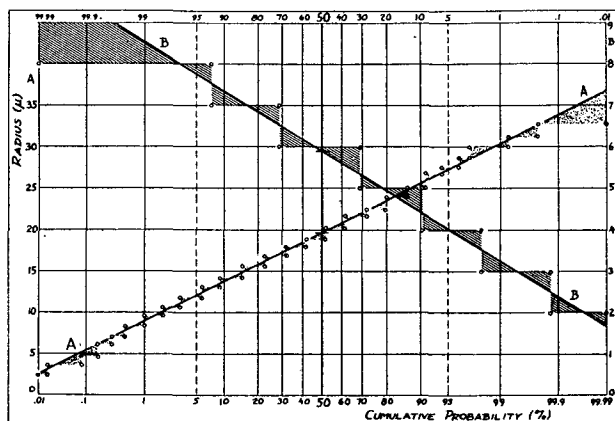


FIG. 5. Spectrum of drop size in sea fog (AA) according to Houghton and Radford, and in clouds (BB) according to Cunningham. The horizontal line segments are interpreted according to the following example (curve BB): drops measuring six microns radius, to the nearest micron, correspond to the range between 31 and 72 per cent cumulative liquid-water content. The sloping lines are the best-fitted normal distributions, and the shading is for emphasis only.

of distribution, to describe the breadth of the distribution. The agreement between the moduli of distribution for the two curves of fig. 5 is purely fortuitous, for the modulus is known to vary widely.

Fig. 6, prepared from data reported by Smith [24], shows the frequency with which various moduli of distribution have been observed at Mount Washington. The modulus is zero for the 'A' class, 0.5 for the 'B', and thence increases by increments of 0.25 to 0.75 for 'C', *etc.* More than half of all the clouds at Mount Washington are highly uniform in constitution, and more than 77 per cent fall in either the 'A' or 'B' group. Nevertheless, there remains a significant proportion of clouds with broader spectra, ranging up to the limit of the observational method at a modulus of 2.25.

C. Concentration.—The concentration of cloud drops, derived indirectly by Conrad [5] from the Mount Washington observations of volume median radius and liquid-water content, follows a logarithmic Gaussian probability law very closely (fig. 7); only the highest points fall an appreciable distance from the straight line. The measurements in fog by Houghton and Radford, corroborated by others, show that the range is further extended on the low end to concentrations of the order of 50 cm^{-3} when fog is considered.

5. Integrations of drop growth

To integrate the equation of growth, use was made of the concept of total supersaturation, defined as the supersaturation that would exist if all liquid water in the air were vapor at the same temperature. It is thus related to the concentration of drops and to their radii, imposing one of the conditions which must be met at

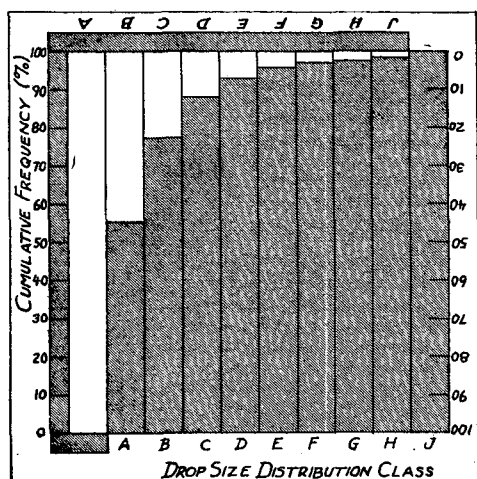


FIG. 6. Cumulative frequency of occurrence at Mount Washington of clouds according to the modulus of distribution of their drop sizes. The shaded columns show the percentage of clouds with a modulus at least as great as the one designated; when the figure is inverted, the clear columns show the percentage having a modulus at least as small as the one designated.

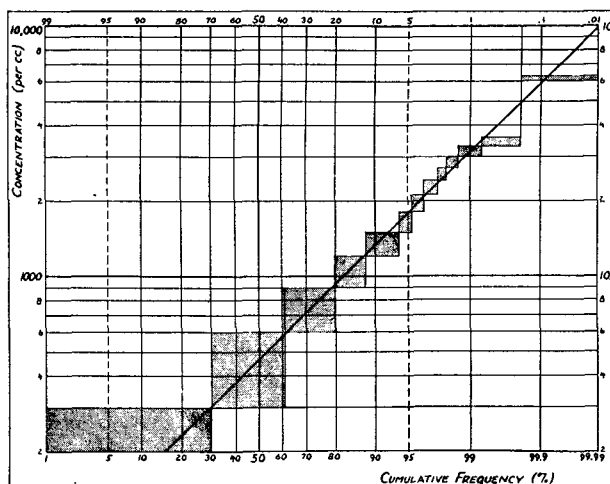


FIG. 7. Cumulative frequency of occurrence of clouds at Mount Washington according to the concentration of drops, cm^{-3} . Interpretation is similar to fig. 4.

each step of the integration. It was then assumed that the total specific humidity of an air parcel remains constant during cloud formation. The rate of increase of the total supersaturation under assumed conditions of temperature and pressure may then be easily related to the rate of cooling of the air or to the rate of pseudo-adiabatic ascent. At a temperature of 0°C and a pressure of 800 mb, if S_z is the total supersaturation, S_L the liquid supersaturation, and z the height,

$$\frac{dz}{dt} = 3.03 \times 10^3 \frac{dS_z}{dt},$$

$$S_L = \sum_{i=1}^n \frac{4\pi r_i^3}{3w\rho_a} \times 10^{-7}.$$

Each stepwise integration was begun at a low enough relative humidity so that the liquid supersaturation was negligible, and the steps were chosen small enough so that linear interpolation could be used on the graphs between the ends of the step. Each step was accomplished by a series of approximations in which values for two of the three variables Δt , r , and S were assumed and the third was computed which, when plotted on the graph yielded a second approximation for one of the other two. Over most regions of the graph a little experience enables the computer to select the proper independent variables so that the approximations converge very rapidly. This is done for each drop considered. The solution thus obtained for the end point of the step is then tested against the condition

$$S = S_z - S_L.$$

If it is not fulfilled, a new value or set of values is selected for the third of the variables listed above and the process is repeated to gain a second approximation,

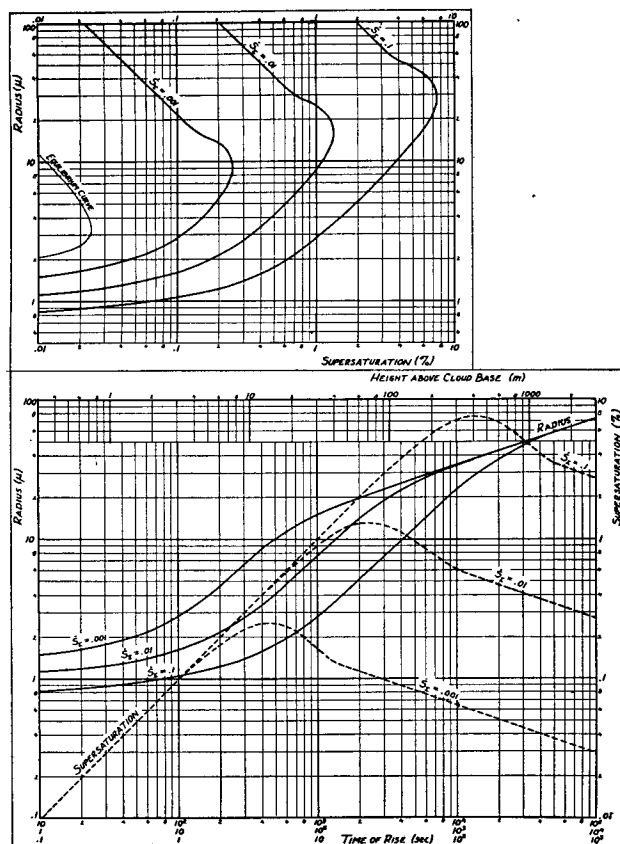


FIG. 8. Growth of drops from uniform nuclei of 10^{-17} moles, for three different rates of cooling corresponding to three rates of pseudo-adiabatic ascent. The upper graph shows curves of condition traced by the drops during their growth, while the lower one traces the history of radius and supersaturation. The time scales descend from the lowest to the highest rate of ascent.

and so on until the condition is met as accurately as may be represented on the graph.

Integrations were completed in this fashion for several cases of uniform drops in various concentrations, nucleus sizes, and rates of cooling; for a single case with two groups of nuclei of different sizes in different concentrations; and for three cases with five to seven classes of nuclei in sizes and concentrations selected to approximate conditions in the natural atmosphere. The result of these computations is shown in figs. 8-17.

Figs. 8 and 9 show that a group of uniform drops

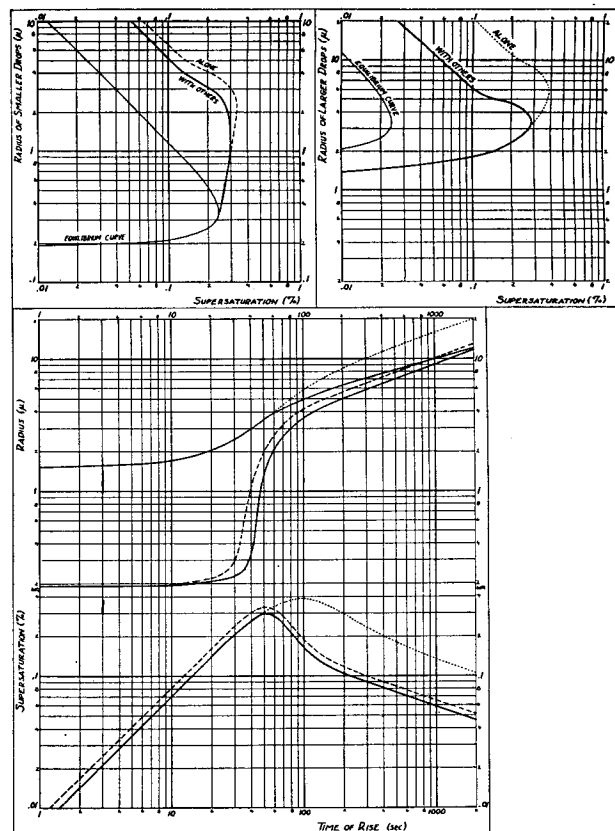


FIG. 9. Growth of drops from uniform nuclei of 10^{-15} moles.

undergoes four distinct phases of growth. In the first phase, before the supersaturation reaches the value critical for activation of the nucleus, the drops grow slowly and do not lag far behind their equilibrium size except in the case of large nuclei at high rates of lift. The second phase begins with rapid growth when the critical supersaturation is surpassed, and is terminated when increased condensation on the rapidly expanding liquid surface puts an end to further rise in the supersaturation. In the third phase, the rate of growth moves toward equilibrium with the rate of moisture supply at a level governed by the drop concentration and the rate of lift. The vapor supersaturation adjusts itself at a value just sufficient to cause the vapor to condense as fast as it is supplied above equilibrium pressure. The fourth and final phase is that of drop

TABLE 5. Initial conditions.

Computation number	1		2		3	
Rate of lift, cm sec ⁻¹	60.4		1.51		30.2	
Total nuclei cm ⁻³	500		667		2000	
Nuclei by groups	log ν	cm ⁻³	log ν	cm ⁻³	log ν	cm ⁻³
	-15.0	9.0	-13.50	1.00	-13.80	28
	-16.0	65.0	-14.00	1.93	-14.25	32
	-17.0	175.0	-14.5	3.87	-14.55	46
	-18.0	175.0	-15.0	7.87	-14.90	94
	-19.0	65.0	-15.5	25.3	-15.30	110
			-16.0	38.6	-15.65	106
			< -16.0	573.7	-15.95	124
					< -15.95	1460

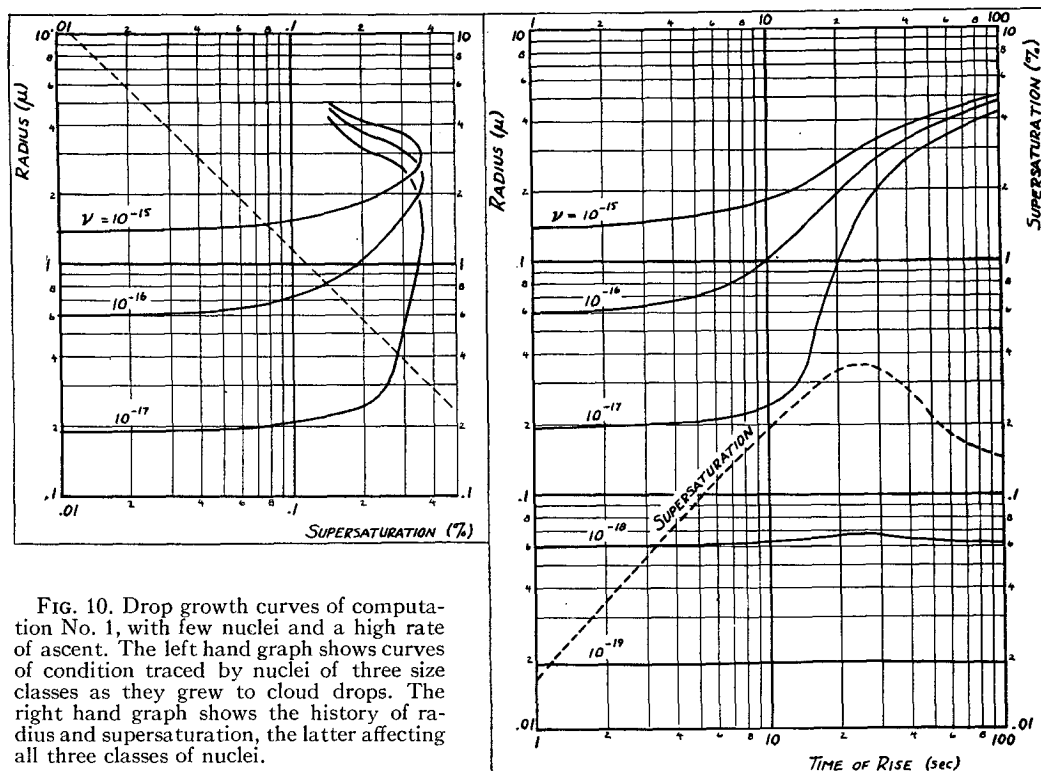


FIG. 10. Drop growth curves of computation No. 1, with few nuclei and a high rate of ascent. The left hand graph shows curves of condition traced by nuclei of three size classes as they grew to cloud drops. The right hand graph shows the history of radius and supersaturation, the latter affecting all three classes of nuclei.

growth after all transient effects of activation have become negligible.

When there are a few large nuclei and a considerable number of smaller ones present together, the same four phases of growth may be recognized, but for each group the second phase is shortened and the third made less distinct by the presence of the other group. The rate of growth in the fourth phase is established for each group on a lower level, and the radii approach each other continuously. Most of the liquid water is found in the large drops until shortly after the smaller ones are activated, but thereafter they share the water more and more according to their relative concentrations rather than their nucleus sizes.

For the integrations intended to approximate conditions in the atmosphere, the initial conditions shown in table 5 were selected. The first computation simulated conditions in rapid convection, with a rate of cooling equivalent to about 4 m sec^{-1} lift, and relatively few nuclei—conditions which were expected to lead to uniform drop growth. Fig. 10 shows that the supersaturation reaches its peak of 0.36 per cent within 25 seconds after saturation is reached, activating nuclei of 4.5×10^{-18} moles or larger. At the end of 100 seconds the drops are quite uniform in size. The size spectrum at three different moments is shown in fig. 11. Since the modulus of distribution is proportional to the slope of the spectrum curve divided by the median radius, the figure indicates increasing uniformity with time. This is more clearly shown in fig. 16,

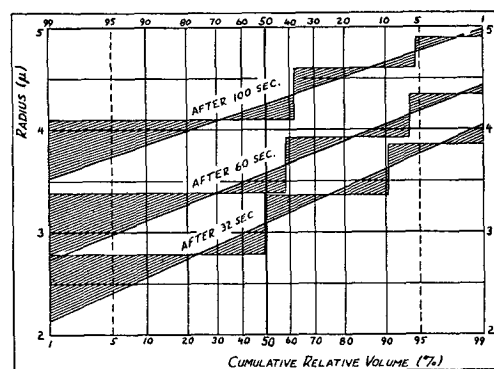


FIG. 11. Drop-size distribution at three stages during the growth of cloud drops according to computation No. 1. The horizontal line segments are interpreted as follows (lower curve): after 32 seconds, the nuclei grown to 3.4 microns radius corresponded to the range from 49 to 91 per cent of cumulative water content. The sloping lines are the best-fitted normal distributions, and the shading is for emphasis only.

which summarizes the spectrum widths for the three computations.

For the second computation, conditions were selected which were intended to simulate conditions favorable to the development of a wide drop-size spectrum, namely large nuclei and a slow rate of lift. Fig. 12 shows that the supersaturation reached a maximum of only 0.056 per cent. Nuclei in the last group to become active never grew much larger than their equilibrium size because of the growth-inhibiting effect of surface tension, but nevertheless they were not re-evaporated within the time computed nor does

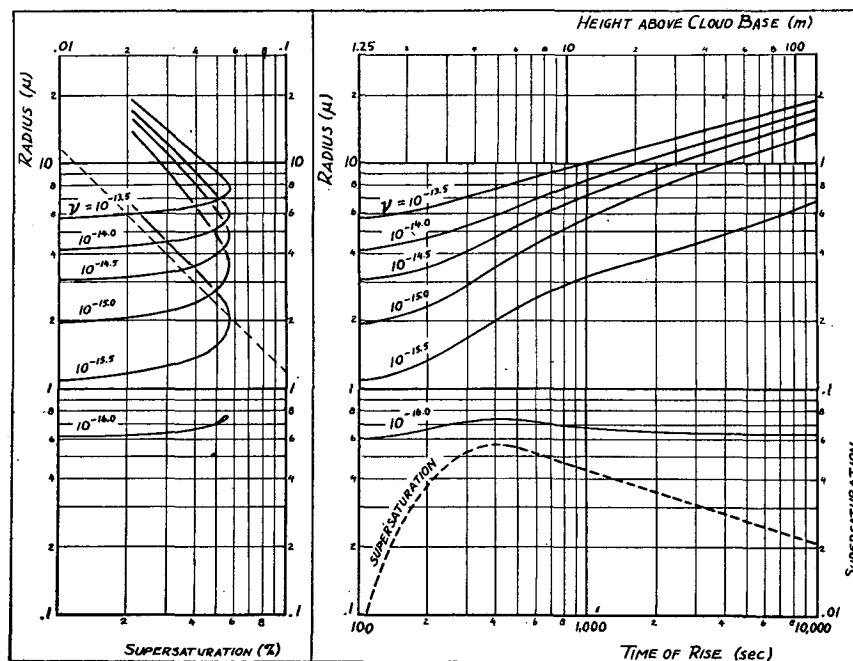


FIG. 12. Drop growth curves of computation No. 2, with large nuclei and a low rate of ascent.

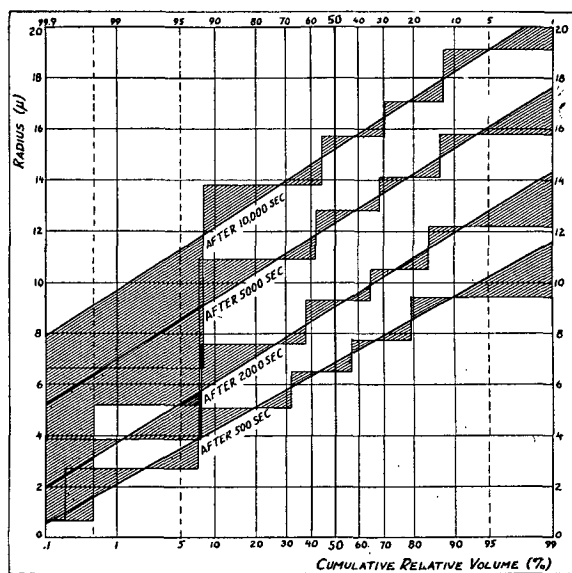


FIG. 13. Drop-size distribution, computation No. 2.

it seem likely that they would evaporate at a later time. A broader spectrum did result, as is shown by figs. 13 and 16, but it is still narrow enough to warrant classifying the cloud as a rather uniform one.

The third computation was intended to simulate conditions of moderate convection with nuclei of average size and concentration. Six of the seven nuclei groups postulated became active, the last of these failing again to grow much beyond equilibrium size, as is seen in fig. 14. That the distribution was actually broader in this case (see figs. 15 and 16) than in the case selected to give a broad spectrum, was not anticipated.

6. Discussion

The results presented in the preceding section show that uniform cooling of air, when apparently reasonable assumptions regarding nuclei sizes and concentration are made, result in drop-size spectra in accord with those observed in the majority of clouds on Mount Washington. However, it is clear that the nucleus spectrum cannot alone explain most of the 45 per cent of clouds which had moduli of distribution greater than 0.25. The explanation for these spectra must be sought elsewhere.

The drop size follows with very little lag the rate of growth necessary to accommodate the water as it becomes available above saturation; supersaturation is merely the means of enforcing this conformity upon the drops. The concentration determined during the first few moments of cloud formation is thus a major determinant of drop size. Past speculations as to the largest size attainable by a drop due to condensation alone, based on the operation of a constant supersaturation, do not recognize the true limiting factor of drop growth, which is simply the amount of water available to be partitioned among the drops on hand and the discrimination in this partition against the larger drops.

To explain less uniform clouds, Arenberg [2] observes that inhomogeneity will result if cloud drops which have had different histories of growth are brought adjacent to one another. Prior to the final intimate mixing, turbulent or convergent flow on a large scale must have operated to bring dissimilar air parcels within mixing distance of each other. The last

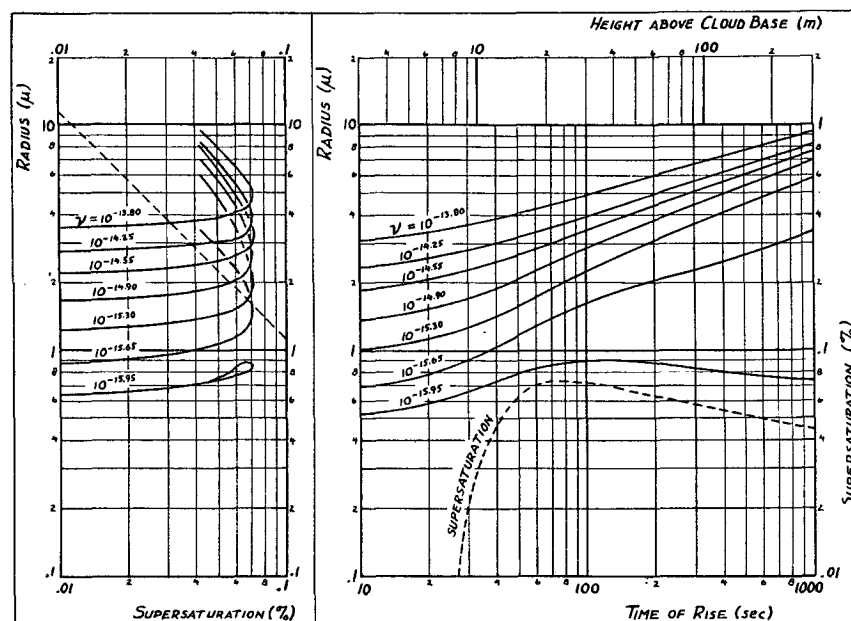


FIG. 14. Drop growth curves of computation No. 3, with average nuclei and moderate rate of ascent.

step is the intimate mixing of different bodies of air through fine-grained turbulence. The older the parcel of cloud since its general origin, the more its characteristics will be affected.

It is not surprising then that newly formed convective clouds are markedly homogeneous. Despite the pronounced convergence and turbulence that may be present; the main body of such a cloud is generally of so recent origin that these processes will not have had time to become effective. The equation of growth will then find its clearest expression. As the cloud ages, its uniformity is progressively destroyed. The tendency toward uniformity arising from the condensation processes is overcome by other influences at a rate depending in part at least upon the amount of convergent flow and fine-grained turbulence present.

A frontal zone thus affords favorable conditions for a broad drop spectrum. The relative translatory motion between the air masses brings parcels of widely different characters together, while the shear stresses in the front produce intimate turbulent mixing. The instance related by Houghton and Bemis [10] of a broadening of the spectrum just before the onset of rain does not necessarily indicate a causal relation; each phenomenon may have resulted independently from quite different aspects of the same weather situation.

Conditions of condensation favoring a broader drop spectrum have not been made clear. It had been supposed that slow cooling, with very small supersaturations, would permit the differential pressure effects of surface tension and solute to find greater expression. While true (though to a lesser extent than was anticipated) when cloud parcels are compared age for age,

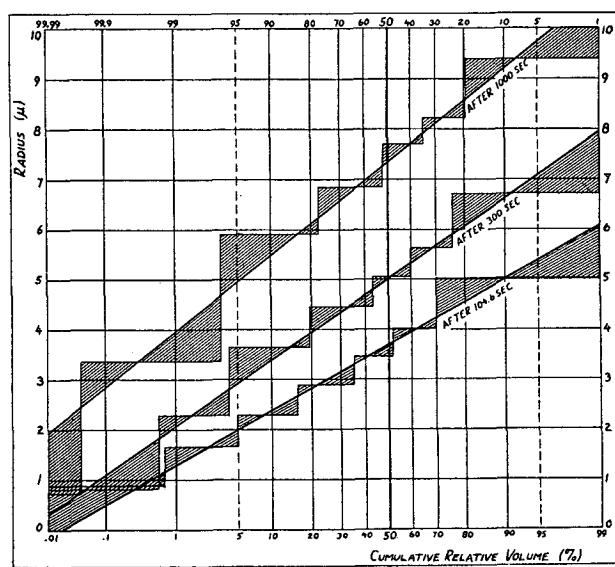


FIG. 15. Drop-size distribution, computation No. 3.

it is by no means true, and the situation may indeed be reversed, when cloud parcels are compared level for level between two clouds with different rates of lift. The almost equal distribution of water among the fewer drops in the slower rising cloud offsets and may overcome the other effect, as is seen in fig. 16. The slow cooling associated with fog is apparently not peculiarly favorable to a broad spectrum through condensation; even the spectrum breadth reported by Houghton and Radford [11] cannot be adequately explained on this basis.

The drop-growth integrations clarify the role played by the evaporation of smaller cloud drops in maintaining the drop-size distribution and in contributing to

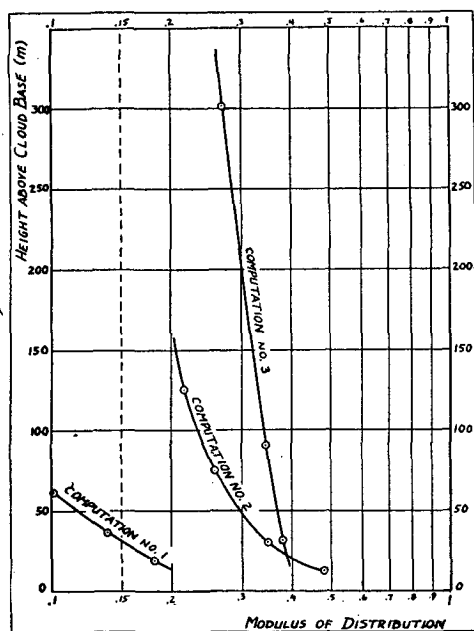


FIG. 16. Moduli of distribution at different heights above the base of uniformly lifted clouds for three different sets of conditions according to the three experimental computations.

the growth of the larger drops. On an S, r -diagram such as fig. 12 some curve may be drawn which the growth curve of a drop must cross if the drop is not eventually to re-evaporate. The equilibrium curve for pure water drops (the dashed line of slope -1 on fig. 12) may play the part of this critical curve. At any rate, the number of nuclei which are activated but fail to cross the critical curve is apparently a very small proportion of the total number activated, and the volume of water which they represent is even less significant. The drop concentration does reach a maximum when the supersaturation is highest and decreases thereafter, but only slightly, approaching a fixed value slightly less than the maximum. It is only when the rate of cooling decreases significantly after the first formation of the cloud that more drops can evaporate and the concentration be materially affected.

The concentration is thus determined mostly in the first moments of cloud formation; it cannot increase further unless the supersaturation is forced, by a great increase in the rate of cooling, to exceed its initial peak. Cessation of cooling brings about the condition discussed by Langmuir [19] where the supersaturation establishes itself in equilibrium with the whole population of drops with the result that the largest ones grow while the smallest ones evaporate. The correlation described by Langmuir between drop size and drop age, according to these findings, should be interpreted as one between drop concentration and rate of cooling at initial condensation; the result is the same.

The concentration of nuclei of course sets an upper limit to the concentration of cloud drops. As a practical matter, the concentration of nuclei is generally

so great that only the largest ten or twenty per cent become drops. Usually each small increase in supersaturation results in an ever more rapid increase in the number of nuclei activated, which in turn tends to force a decrease in the supersaturation. The more rapid the cooling, the higher the supersaturation goes before it is forced to retreat, and the greater the drop concentration.

Activation of most of the nuclei, with consequent limiting effect on the drop concentration, would lead to abnormally large supersaturation and to unusually uniform drop growth. Such a situation is most likely with altocumulus lenticularis or cirrocumulus clouds, where very considerable rates of lift may be induced in relatively pure air by standing waves. The coronas and iridescence often observed in these clouds bespeak the uniformity of their constitution. The larger drops thus formed have significance for the problem of aircraft icing, since even a thin cloud of large drops may result in dangerous icing.

Warming of the air, or any other condition such as entrainment of dry air, reducing the relative humidity below 100 per cent, results in evaporation which affects the smallest drops most severely. The drop concentration decreases at first very rapidly, then more slowly as the cloud becomes depleted and only the larger drops remain. An incompleteness in the present study must be acknowledged in that no example of evaporation has been fully studied; it may well be that evaporation in the descending branches of turbulent air currents has much to do with the determination of the drop-size spectrum as the cloud ages. However, the rapidity of its evaporation does bring into question the possibility of the relative humidity falling much below 100 per cent within a true cloud, as appears frequently to be recorded by radiosondes. The similarity between the evaporative and condensative processes leads to the expectation that subsaturation as long as any significant concentration of drops remains must be of roughly the same order of magnitude as the supersaturation during condensation, say a fraction of a per cent up to two or three per cent in most cases. If the subsaturations of the order of 5 to 10 per cent or more observed within cloudy regions are real, they must represent spaces where downward motion or entrained dry air have evaporated substantially all the drops from the cloud. Nor is the presence of such cleared spaces to be thought unlikely on the basis of airplane flights through massive cloud systems, particularly where the general ascent of air is slow and turbulence considerable. It may be significant to note that Clark⁷ reports no depression of the wet-bulb

⁷ V. F. Clark, "Factors involved in considering relative humidity during icing," *Mount Washington Observatory Icing Report*, March 1945, 37-39.

thermometer ever being observed on Mount Washington when the summit was in unbroken clouds.

Dessens [7] has observed that when nuclei are exposed to low relative humidity they become supersaturated solutions of their constituent chemicals in water, and that such particles may crystallize with explosive suddenness. He proposes that regular subdivision of nuclei may result in modal class sizes of nuclei, and that the modal drop sizes observed by Köhler [17] may be explained as the result of condensation on these modal classes of nuclei. This proposition receives no support from our study, for the nucleus modes would need to have volume ratios of anywhere from 10:1 to 100:1 to result in drops with a mass ratio of 2:1, and even then that mass ratio would hold only at a certain unique stage in the life of the cloud.

The drop concentrations and sizes computed from the assumed nuclei sizes and spectra are in excellent agreement with those observed in natural clouds. At least as much growth must occur as is produced by condensation alone; other processes such as collisions may increase but not decrease the mean size so arrived at. The conclusion is strongly suggested that other growth processes are not significant in uniform clouds.

The operation of the equation of growth is remarkably successful in reaching agreement with the observed shape of the drop-size spectrum curve, even when the absolute breadth of the spectrum cannot be so explained. There is no *a priori* reason to expect the spectrum to follow a Gaussian probability law; in fact, since it is unilaterally limited by the impossibility of a negative drop radius, some other distribution would seem offhand more likely. While under certain assumptions Schumann [23] has predicted a distribution closely similar to the Gaussian one on the basis of growth by collisions, this is probably inapplicable. However, the drops with varying histories which may be carried by turbulence into a small parcel of cloud from time to time have considerable opportunity for mutual size adjustment with the population already there. This is most true of the smaller drops. It therefore seems likely that the condensative process is effective in controlling the shape of the spectrum curve even when its breadth is considerably extended by other processes.

7. Summary

1. The narrow drop spectra characteristic of young clouds may be adequately explained by the equation of growth operating in uniformly cooled air. As the cloud ages, this process diminishes in importance as a determinant of the spectrum, and other influences become paramount. In particular, it cannot explain in-

homogeneous clouds. Turbulence and convergence acting in concert may explain them. It is suggested that the combined operation of advection and fine-grained turbulence produced by shear may be important in broadening the spectrum within frontal zones of transition without any causal connection with the concomitant precipitation.

2. The observed shape of the drop-spectrum curve is in accord with the operation of the equation of growth on uniformly cooled air. It is probable that even when other influences are paramount in determining the breadth of the spectrum, the equation of growth still controls the shape.

3. The conditions under which operation of the equation of growth most favors a broad spectrum are not adequately clarified. With slow cooling, the almost equal distribution of water among fewer drops offsets and may overcome the broadening effect of the smaller vapor-pressure differential.

4. The drop concentration is determined primarily by the cooling rate during the initial stage of condensation. It is as a rule only slightly dependent on the concentration of nuclei, though this may be important in altocumulus lenticularis and cirrocumulus. With continued steady cooling, the concentration decreases only slightly toward a steady value. It cannot be increased except by a very great increase in the cooling rate, but it will be quickly and greatly decreased by warming of the air.

5. Supersaturation in natural clouds ordinarily is of the order of 0.1 per cent and can surpass 1 per cent only under extreme circumstances. Subsaturations larger than 2 or 3 per cent probably cannot occur in cloud except under extremely rapid warming. Radiosonde observations of lower humidity within cloud systems must, if real, indicate cleared spaces.

6. Modal sizes of nuclei are incapable of explaining the modal drop sizes reported by Köhler.

7. Observed mean drop sizes agree well with the sizes predicted by operation of the equation of growth in uniformly cooled air. The conclusion appears well founded that other modes of growth are not important in uniform clouds.

Acknowledgments.—The writer gratefully acknowledges the help of Prof. H. G. Houghton of the Massachusetts Institute of Technology, whose guidance, counsel, and encouragement were invaluable. He is also indebted to Dr. Irving Langmuir, Prof. C. F. Brooks, and Dr. Vincent Schaefer for useful suggestions and enlightening discussions. Thanks are extended to Prof. Isadore Amdur for information on the mean free path of water molecules in air and the accommodation factor for air-water molecular collisions.

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TABLE OF SYMBOLS AND CONSTANTS

c_p	= specific heat at constant pressure
D	= diffusion coefficient of water vapor through air
e	= prevailing vapor pressure of water
e_w	= equilibrium vapor pressure over flat water surface
e_w'	= equilibrium vapor pressure over curved water surface
e_w''	= equilibrium vapor pressure over a flat surface of aqueous solution
e_w'''	= equilibrium vapor pressure over a curved surface of aqueous solution
f	= accommodation factor of air molecules in collision with liquid water = 0.7
F_m	= a function determined by equation (m)
J	= mechanical equivalent of heat = 4.185×10^7 ergs cal ⁻¹
k	= diffusion coefficient of heat through air
K	= compensated coefficient of thermal conductivity
L	= latent heat of condensation of water vapor
m_w	= molecular weight of water = 18
q	= quantity of water
Q	= quantity of heat
r	= dimensionless radius in number of microns
r_0	= unit radius, one micron
r'	= radius of curvature; radial distance
S	= prevailing supersaturation with respect to a flat surface of pure water
t	= dimensionless time in number of seconds
t_0	= unit time, one second
t'	= time
T	= air temperature in degrees Kelvin
T_w	= water temperature in degrees Kelvin
w	= saturation mixing ratio in g kg ⁻¹
z	= height
α	= the Cunningham constant = 0.7
γ	= surface tension of water
Δ	= compensated diffusion coefficient
λ_w	= mean free path of water molecules in air
λ_a	= mean free path of air molecules
ν	= mass of condensation nucleus (gram molecular weights)
ρ_a	= density of air
ρ_v	= density of water vapor
ρ_w	= density of water.

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