

## Numerical Solution for Condensation of Atmospheric Vapor on Soluble and Insoluble Nuclei

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### ABSTRACT

The growth of cloud droplets by condensation of vapor upon soluble and insoluble nuclei was computed for different nuclei distributions. The set of equations used takes into account the interdependence of vapor condensation and changes of temperature and pressure in the vertically moving air. The results show that the spectrum of cloud droplets is broader if the nuclei distribution includes hygroscopic and insoluble nuclei together, rather than different sizes of salt nuclei only. If different numbers of nuclei are assumed in neighboring air parcels, then the size spectrum in the cloud is even broader. The width of the spectrum obtained in this study seems to be broad enough to initiate the process of raindrop growth by coalescence. Results for different updraft velocities and a non-constant vertical velocity of the air parcel show that the final sizes of the drops are almost the same when measured at a certain altitude  $z$ , no matter what the vertical velocity was.

### 1. Introduction

Several investigators have dealt theoretically with the problem of cloud droplet growth by condensation of water vapor upon salt nuclei. Numerical results have been obtained by Howell (1949), Mordy (1959), and Neiburger and Chien (1960) for droplet spectra formed by condensation of water vapor upon different distributions of salt nuclei. Their spectra of drop sizes, however, are too narrow to permit initiation of raindrop growth by coalescence.

An attempt to explain the formation of large droplets has been made by Mason and Ghosh (1957) who show that large droplets may be formed by condensation on giant salt nuclei. This explanation, although acceptable for clouds over oceans, does not seem plausible for clouds inland, where giant salt nuclei are rarely found.

The results presented in this paper suggest a more realistic explanation for the formation of a broader spectrum of cloud droplets; namely, condensation on a heterogeneous population of condensation nuclei, including soluble and insoluble nuclei. Non-uniform distribution of the number density of nuclei in the atmosphere may also be an important factor.

McDonald (1964), in a theoretical study of cloud nucleation on insoluble particles, has considered the case of silicates in the form of thin circular discs. The model used in the present work also considers insoluble wettable disc-shaped condensation nuclei, but as part of a distribution which includes nuclei of other types.

In addition, the present study, unlike most of the previous studies, takes into account the interdependence of vapor condensation and changes of temperature and pressure in the vertically moving air. Somewhat different models for such interdependence have been studied by Mordy (1959) and by Warshaw (private communication).

### 2. The model

Let us consider a parcel of air with vertical motion which initially has a volume of  $1 \text{ cm}^3$ . The parcel of air expands while rising but its weight remains unchanged. We assume that the parcel includes dry air, water vapor, and condensation nuclei. Our equations are written for constant weight (not constant volume) of the air parcel and include phase change of the water constituent. Following a parcel of constant weight insures that during the whole process the number of condensation nuclei remains the same and the decrease in water vapor increases the liquid water content by the same amount. Diffusion of nuclei or vapor into or out of the parcel is not considered in the model.

### 3. Moist-adiabatic process

The heat released by condensation of the amount,  $-dG_w$ , of vapor is <sup>3</sup>

$$\delta Q_1 = -LdG_w. \quad (1)$$

If the temperature changes by  $dT$ , the liquid water present takes up sensible heat from the parcel in the

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<sup>3</sup> See Appendix for list of symbols.

amount

$$\delta Q_2 = -sG_w dT. \tag{2}$$

Except for these two sources of heat, the rising parcel expands in a manner that is adiabatic. Hence, the heat balance (see Byers, 1965, p. 8) is

$$-LdG_v - sG_w dT = (G_a + G_v) \left[ c_p dT - R_m T \left( \frac{dp_m}{p_m} \right) \right]. \tag{3}$$

Combining the equation of state for an ideal gas,

$$p = \rho \frac{R}{M} T, \tag{4}$$

with the hydrostatic equation,

$$dp = -\rho g dz, \tag{5}$$

we obtain for moist air

$$\frac{dp_m}{p_m} = -\frac{g dz}{R_m T}. \tag{6}$$

Substituting (6) in (3) and rearranging, we obtain the general temperature lapse rate

$$\frac{dT}{dz} = \frac{-(G_a + G_v)g - L(dG_v/dz)}{(G_a + G_v)c_p + sG_w}, \tag{7}$$

where

$$G_v = G_v^0 - G_w. \tag{8}$$

Before condensation starts we have

$$G_w = 0, \quad \frac{dG_w}{dz} = 0;$$

and for this case Eq. (7) reduces to

$$\frac{dT}{dz} = -\frac{g}{c_p}, \tag{9}$$

which is the dry-adiabatic lapse rate.

Rearranging Eq. (6), we obtain the hydrostatic relationship

$$\frac{dp_m}{dz} = -\frac{gp_m}{R_m T}, \tag{10}$$

where

$$R_m = \left[ 1 + \left( \frac{1}{\beta} - 1 \right) \left( \frac{w}{1+w} \right) \right] R_a, \tag{11}$$

(see Fieagle and Businger, 1963).

#### 4. Growth equation of a drop

The growth equation of a drop by condensation is

$$\frac{dm_w}{dt} = 4\pi C(e - e_s^*) \left/ \left( \frac{R_v T}{D} + \frac{L^2 e_s^*}{R_v K T^2} \right) \right., \tag{12}$$

a form similar to that given by Koenig (1968). Since it is given here for an arbitrarily shaped drop, the electrostatic capacity  $C$  enters into the equation. For the mathematical analogy between the diffusion problem and the electrostatic problem, see, for example, Byers (1965).

By definition,

$$w = \frac{G_v}{G_a}. \tag{13}$$

Using Eq. (4), we thus have

$$w = \frac{e}{p_m - e}, \tag{14}$$

whence, the ambient vapor pressure,

$$e = \frac{w}{\beta + w} p_m. \tag{15}$$

Over a cloud drop, the saturation vapor pressure is given by

$$e_s^* = e_s k R_c, \tag{16}$$

where, as given in Murray (1967),

$$e_s(T) = 6107.8 \exp \left[ \frac{17.2694(T - 273.16)}{T - 35.86} \right], \tag{17}$$

and where the Kelvin corrector is

$$k = \exp \left[ \frac{2\sigma}{TR_v r \rho_e} \right]. \tag{18}$$

The electrostatic capacity is estimated by

$$C = \left( \frac{A}{4\pi} \right)^{\frac{1}{2}}. \tag{19}$$

For the special case of a sphere, (19) gives the known relation  $C = r$ .

#### 5. Raoult corrector

The vapor pressure over a solution is different from the vapor pressure over pure water, the difference being given by the Raoult corrector. If the solvent is an electrolyte, the Raoult corrector, as shown by McDonald (1953), is

$$R_c = \left[ 1 + i \frac{m_n M_w}{m_w M_n} \right]^{-1}, \tag{20}$$

where  $m_n$  is the mass of dissolved salt. In one gram of water we can dissolve  $\alpha$  grams of salt. Therefore, as long as there is not enough water on a drop to dissolve the whole amount of salt, the amount that will be dissolved is  $m_n = \alpha m_w$ . As soon as there is enough water, we shall

TABLE 1. Characteristics of the four distributions of condensation nuclei used in the computations.

Type of nuclei	I		Distribution II		III		IV	
	Size of nuclei	No. of nuclei	Size of nuclei	No. of nuclei	Size of nuclei	No. of nuclei	Size of nuclei	No. of nuclei
A (salt)	$r_0 = 1 \mu$	30	$r_0 = 1 \mu$	30			$r_0 = 1 \mu$ $r_0 = 0.5 \mu$ $r_0 = 0.25 \mu$	10 60 30
B (disc of insoluble material)	$a = 1 \mu$	50	$a = 1 \mu$ $a = 0.5 \mu$	35 15	$a = 5 \mu$	25		
C (type B with salt)	$a = 5 \mu,$ $\nu = 1.8125 \times 10^{-13}$ moles	20	$a = 5 \mu,$ $\nu = 1.8125 \times 10^{-13}$ moles	20	$a = 5 \mu$	25		
					$\nu = 2 \times 10^{-13}$ moles	25		
					$\nu = 2 \times 10^{-14}$ moles	25		
					$a = 5 \mu,$ $\nu = 2 \times 10^{-15}$ moles	25		

have  $m_n = m_0$ . Substituting those values of  $m_n$  into Eq. (20), the Raoult corrector can be written as

$$R_c = \begin{cases} \left[ 1 + i\alpha \frac{M_w}{M_n} \right]^{-1}, & m_w \leq \frac{m_0}{\alpha} \\ \left[ 1 + i \frac{m_0 M_w}{m_w M_n} \right]^{-1}, & m_w > \frac{m_0}{\alpha} \end{cases} \quad (21)$$

If we substitute  $m_0 = 0$  in the second part of (21), we obtain the expected value of  $R_c = 1$  for pure water.

6. Condensation nuclei

In this study we deal with three types of nuclei. Type A are spherical nuclei of the soluble material NaCl, with radius  $r_0$ . Type B nuclei are discs of an insoluble wettable material. The radius of a disc is  $a$  and its width is  $\theta a$ , with  $\theta = 0.1$  (see Fig. 1). Type C nuclei are discs as described above, but with some amount  $\nu$  of salt attached to both surfaces, half on each surface.

For each computation, a distribution of nuclei from the three types described was selected (see Table 1). Each distribution included nuclei of different sizes. For

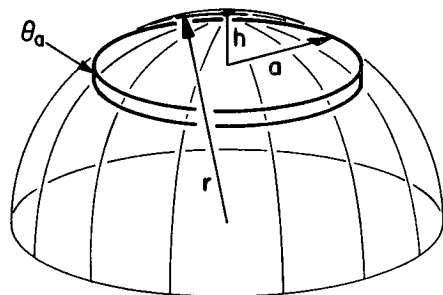


FIG. 1. A disc of radius  $a$  and width  $0.1a$ . On each disc surface there is a cup of water which may include salt, partially or completely dissolved. The cup is spherical, with height  $h$  and radius of curvature  $r$ .

Type C nuclei, the amount of salt was varied as well as the disc size.

7. Initial conditions

We start our computations at  $t=0$ , at a reference altitude denoted  $z=0$ . Initial conditions include the initial temperature  $T^0$ , pressure  $p_m^0$  and the relative humidity  $S^0$ . By choosing these parameters, we may obtain all other necessary initial values by known relations.

The initial ambient vapor pressure is

$$e^0 = \frac{e_s(T^0)S^0}{100} \quad (22)$$

Using  $G_a$  and  $G_v^0$  as the mass of dry air and the initial mass of vapor, respectively, of a parcel of air which initially has a volume of  $1 \text{ cm}^3$ , we can write

$$G_a = \frac{p_m^0 - e^0}{R_a T^0} \quad (23)$$

$$G_v^0 = \frac{e^0}{R_v T^0} \quad (24)$$

for the initial densities of dry air and vapor, respectively.

We assume for computational convenience that the condensation nuclei already have a certain amount of water upon them at the initial altitude,  $z=0$ . This amount of water is taken to be very small (e.g., see Figs. 2 and 4) and is not included in that term of (7) which is defined by (2). Experimental computations suggest that changing the initial amount of water does not affect the final results.

The initial amount of water on the wettable discs is assumed to form spherical cups on both surfaces of the disc. Initially, the cups have a height  $h$  small compared

to  $a$  ( $h/a=0.05$ ). For nuclei of Type C, these cups are assumed to include some amount of salt which is partially or completely dissolved.

**8. The set of equations and their numerical computation**

The set of differential equations to be solved includes Eq. (7) for temperature, Eq. (10) for pressure, and a growth equation for each size of nucleus. The growth equation (12) is transformed to be dependent on  $z$  rather than  $t$  which, for a nucleus of size  $i$ , has the form

$$\frac{dm_{wi}}{dz} = 4\pi C_i(e - e_{si}^*) / \left( \frac{R_v T}{D} + \frac{L^2 e_{si}^*}{R_v K T^2} \right) u. \quad (25)$$

Using Eq. (25), we can write for the second term in the numerator of Eq. (7)

$$\frac{dG_v}{dz} = -\sum N_i \left( \frac{dm_{wi}}{dz} \right). \quad (26)$$

For Type A nuclei, (25) is transformed to compute directly the radius of the drop, using the relation

$$m_w = (4/3)\pi(r^3 - r_0^3). \quad (27)$$

For Type B and C nuclei, we use (25) as given. In the beginning the drop does not have a spherical shape, but is rather a disc with cups of water on both surfaces. As these cups grow, their radius of curvature decreases. When the height of the cup reaches the value  $h = a$ , the drop attains its spherical shape (see McDonald, 1964).

The volume of both cups in terms of  $h$  is

$$v = (\pi/3)(h^3 + 3ha^2), \quad (28)$$

while in terms of mass, we have

$$v = m_w + \frac{m_0}{\rho_n}. \quad (29)$$

As in all our computations, the volume of the solution is assumed to be the sum of the volume of water and the volume of salt.

From Eqs. (28) and (29) we obtain a cubic equation for  $h$ ; solving it we obtain the radius of curvature, using the relation

$$r = (1/2) \left( h + \frac{a^2}{h} \right). \quad (30)$$

When the drop reaches its spherical shape, the radius of the drop is given by

$$r = [(3/4\pi)(v + \pi a^3 \theta)]^{1/3}, \quad (31)$$

where  $\pi a^3 \theta$  is the volume of the disc.

The set of equations is solved by the Runge-Kutta method, with control on the integration step  $dz$  to insure the desired accuracy. The equations also allow evapora-

tion. However, evaporation of the initial amount of water on the drops at  $z=0$  was not allowed in the computations.

**9. The van't Hoff factor**

For the results presented here, the value  $i=2$  for NaCl was used. For comparison, the table of experimental values given in McDonald (1953) was used and linear interpolation for intermediate values made. The results with  $i$  from that table do not differ substantially from those computed with  $i=2$ .

**10. Non-constant velocity**

Let us assume that the velocity of the updraft is not constant, but that

$$u = c_1 + f(t). \quad (32)$$

In this case the hydrostatic equation (5) must be used in the form

$$\frac{dp}{dz} = -\rho \left[ g + \frac{df(t)}{dt} \right]. \quad (33)$$

For the case of non-constant velocity, the equations are transformed to be functions of  $t$ , rather than  $z$ , by the transformation

$$\frac{dz}{dt} = u. \quad (34)$$

The growth equations then have the form of Eq. (12), and the equations for temperature and pressure are rewritten as

$$\frac{dT}{dt} = \frac{-(G_a + G_v)[g + df(t)/dt]u - L(dG_v/dt)}{(G_a + G_v)c_p + sG_w}, \quad (35)$$

$$\frac{dp_m}{dt} = \frac{-p_m[g + df(t)/dt]u}{R_m T}. \quad (36)$$

Computations were performed with velocity of the form

$$u = c_1 + c_2 \sin \omega t. \quad (37)$$

**11. Results**

Computations were made with the initial values  $T^0 = 283.16\text{K}$ ,  $p_m^0 = 900\text{mb}$ , and  $S^0 = 82.277\%$ , the latter being just below that at which condensation can begin. The constant updraft velocity was chosen to be  $u = 100\text{cm sec}^{-1}$ , except for one case where  $u = 400\text{cm sec}^{-1}$ . The non-constant velocity was chosen to have a mean velocity of  $100\text{cm sec}^{-1}$ .

Fig. 2 shows the growth of water mass on each size of nucleus for distribution I (see Table 1), while Fig. 3 represents results for the same computation, but shows radii of drops or radii of curvature of cups. The curves (a) and (c) decrease in the beginning, showing the

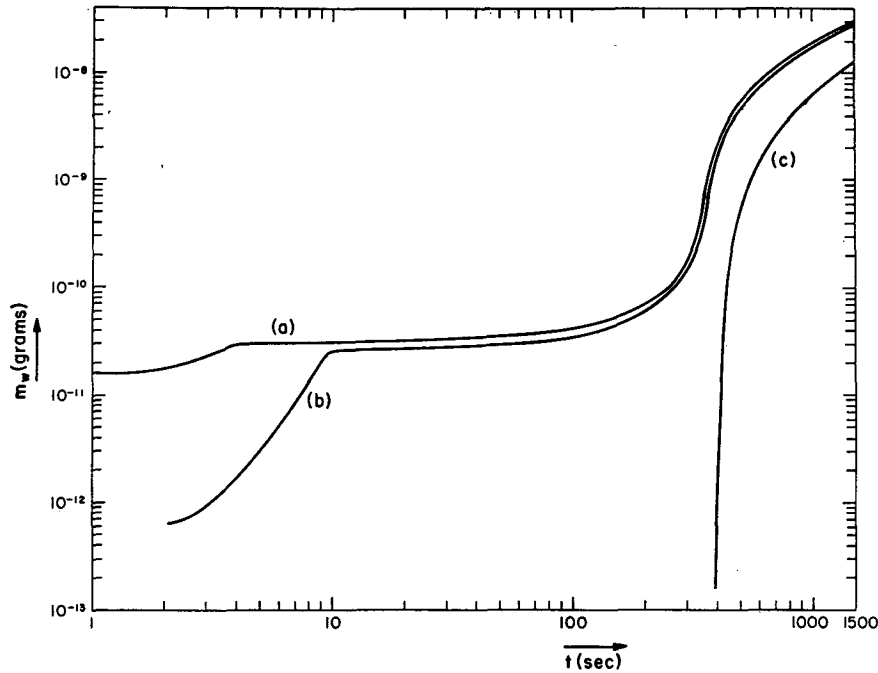


FIG. 2. Mass of water on one drop as a function of time for nuclei of distribution I. Curve (a) is for discs of radius  $a = 5 \mu$ , with  $\nu = 1.8125 \times 10^{-13}$  moles of salt attached to their surfaces; (b) for salt nuclei of radius  $r_0 = 1 \mu$ ; (c) for discs of radius  $a = 1 \mu$ .

behavior of radii of curvature of the cups before the drops attain a spherical shape.

Fig. 4 shows the results for distribution II, which includes the same sizes of nuclei as in distribution I, but adds small discs of radius  $a = 0.5 \mu$ . The curves (a), (b) and (c) behave as they do in Fig. 2, but curve (d) of Fig. 4 shows that a droplet with high vapor pressure over it starts at some point to evaporate while the others are still growing.

Fig. 5 shows the results for distribution III. All the nuclei in this case are discs of the same size, but with different amounts of salt attached to them in order to

show how the amount of salt in drops that initially have the same size affects their growth.

Figs. 6-8 show the results for distribution IV. All the nuclei in these cases are salt nuclei, varying only in size, Fig. 7 reflecting computations for non-constant velocity, where  $u = [100 + 200 \sin(0.1t)]$  cm sec $^{-1}$ . The behavior of the curves almost always shows that for  $u > 0$  the drops are growing and for  $u < 0$  they are evaporating. Nevertheless, the trend of growth in Fig. 7 is very similar to that shown in Fig. 6. In both cases, computations indicate that at a given altitude  $z$  we have essentially the same spectrum of drop sizes.

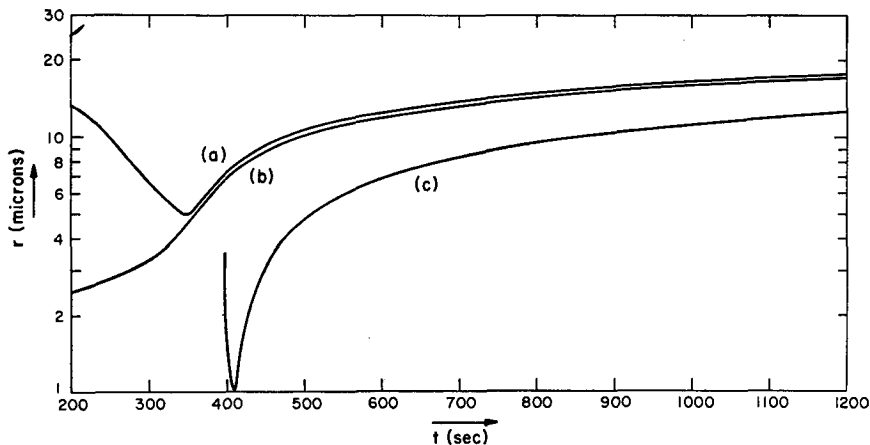


FIG. 3. Radii of drops as a function of time for nuclei of distribution I. Curves (a), (b) and (c) are the same as in Fig. 2.

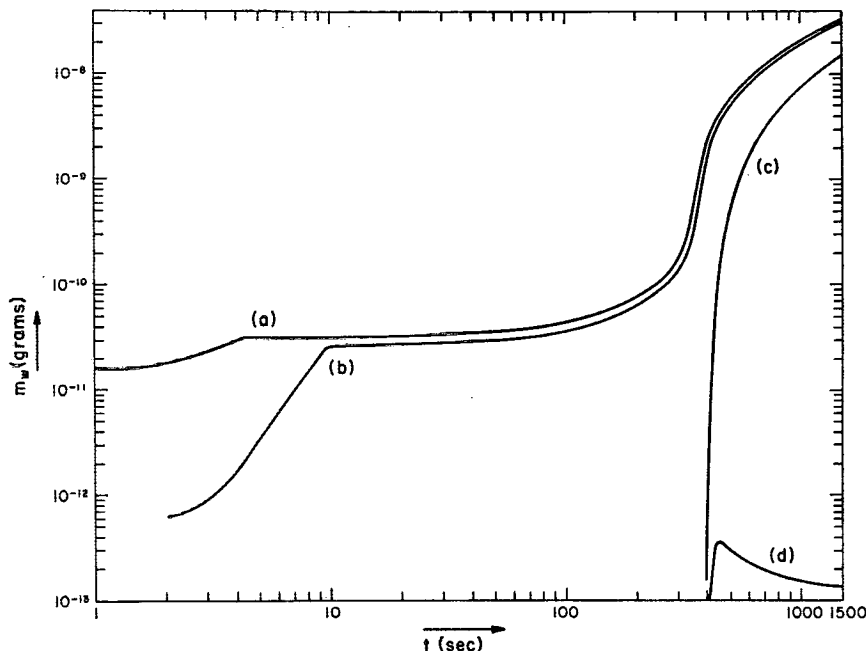


FIG. 4. Mass of water on one drop as a function of time for nuclei of distribution II. Curves (a), (b) and (c) are the same as in Fig. 2; curve (d) is for discs of radius  $a=0.5 \mu$ .

Fig. 8 shows results for an updraft velocity  $u=400$   $\text{cm sec}^{-1}$ . The curves are similar to those of Fig. 6 if we change the time scale in Fig. 8 to permit comparison of drop sizes at the same altitude  $z$ . For the same altitude the drops with the stronger updraft are somewhat smaller; condensation is not fast enough to consume all the excess vapor in the rising air parcel. Indeed, as shown by a comparison of Figs. 9 and 10, supersatura-

tion with a high updraft reaches a much higher maximum value than with a lower updraft.

The results show that the final spectrum of drop sizes is broader if the distribution of condensation nuclei includes different types of nuclei rather than nuclei of the same type, even if those are of different sizes. Comparing the results of Fig. 3 with those of Fig. 6, we find that after 800 sec of real time the heterogeneous

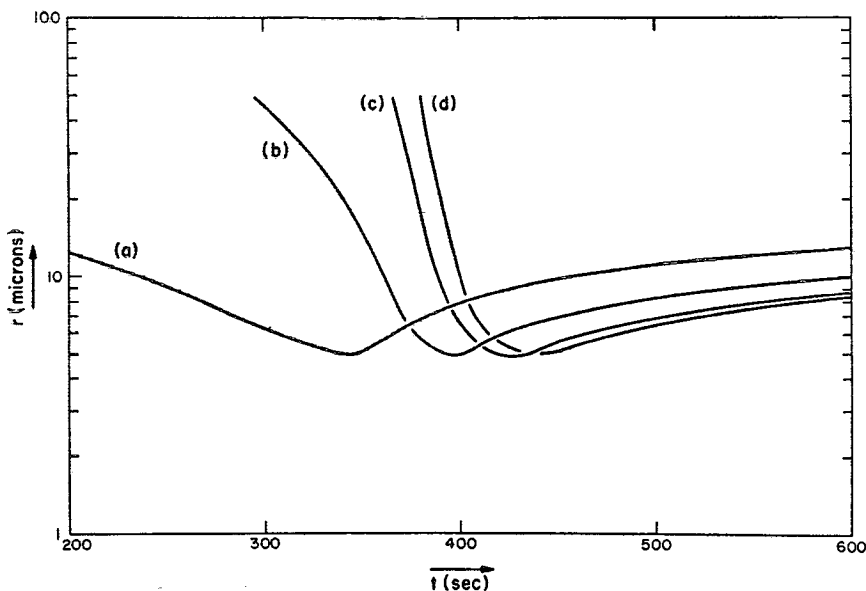


FIG. 5. Radii of drops as a function of time for discs of radius  $a=5 \mu$  with different amounts of salt attached to their surfaces for nuclei of distribution III. Curve (a) is for  $\nu=2 \times 10^{-13}$  moles of salt; (b) for  $\nu=2 \times 10^{-14}$ ; (c) for  $\nu=2 \times 10^{-15}$ ; (d) for no salt.

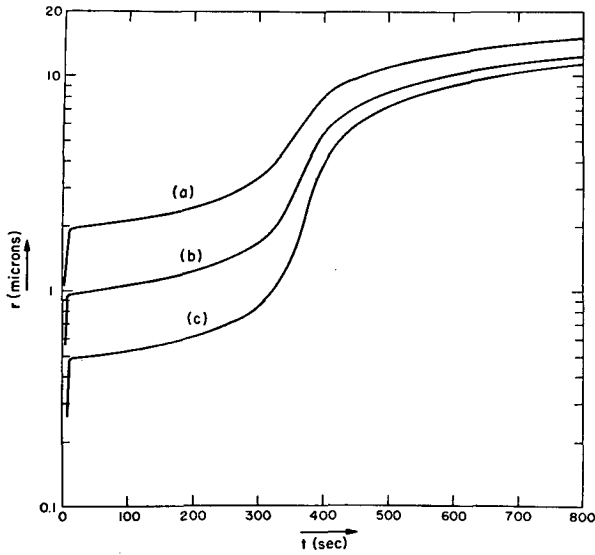


FIG. 6. Radii of drops as a function of time for salt nuclei of different sizes for nuclei of distribution IV. Curve (a) is for  $r_0=1 \mu$ ; (b) for  $r_0=0.5 \mu$ ; (c) for  $r_0=0.25 \mu$ .

distribution results in a difference of  $5.22 \mu$  between the biggest and smallest drop. The same value for the homogeneous distribution is  $3.60 \mu$ . All the computations represented in Figs. 2-10 are for 100 nuclei per  $1 \text{ cm}^3$  of initial volume of air. We can assume that different air parcels in the vicinity of each other may have different numbers of nuclei. It is very possible that somewhere, not too far from the parcel with 100 nuclei, there exists a parcel with the same sizes of nuclei, but with only 65 nuclei per  $1 \text{ cm}^3$  of initial volume.

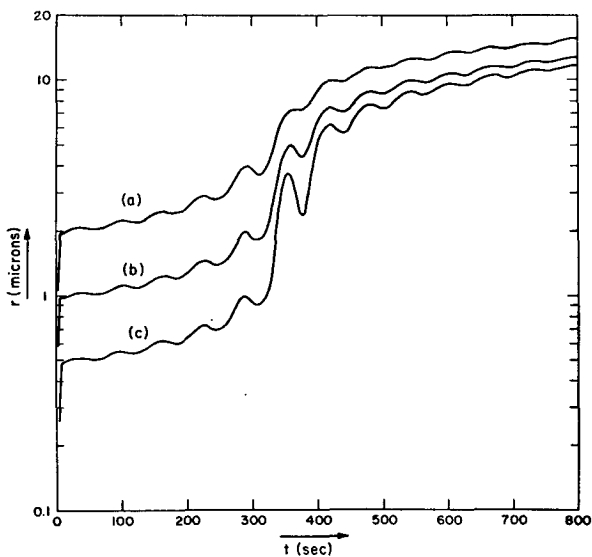


FIG. 7. Same as Fig. 6 except for the case where the updraft velocity  $u=[100+200 \sin(0.1t)] \text{ cm sec}^{-1}$ .

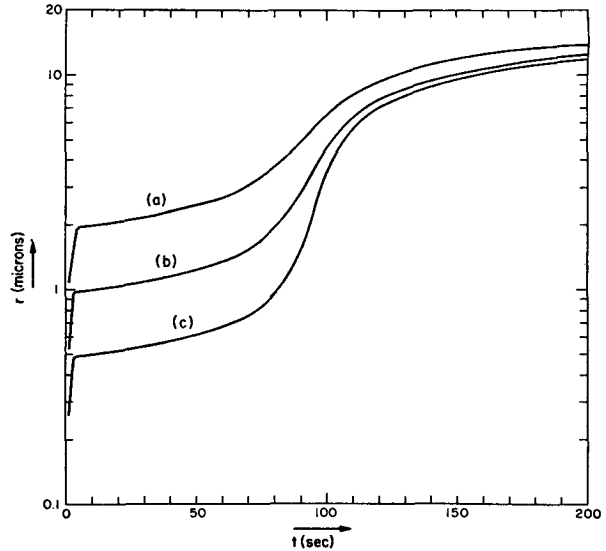


FIG. 8. Same as Fig. 6 except for the case where the updraft velocity  $u=400 \text{ cm sec}^{-1}$ .

Computations for 65 nuclei of the same kind as in distribution I show that after 20 min of real time, the biggest drop has a radius of  $r=19.9 \mu$ . The smallest drop in the parcel with 100 nuclei has a radius of  $r=12.8 \mu$ . This difference of  $7.1 \mu$  can be great enough to permit initiation of cloud drop growth by coalescence provided one adopts the Davis-Sartor (1967) collision efficiencies.

The computations were performed on the NCAR Control Data Corporation 6600 computer. The computation for one case, representing up to 800 sec real time, took 2-4 min of computer time. Computation in the region where drops are smaller was more time-

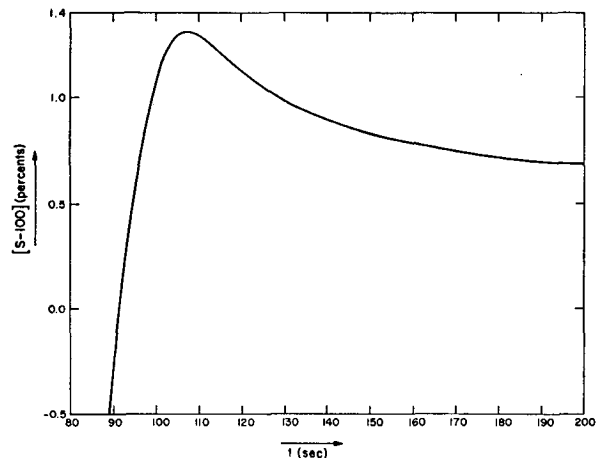


FIG. 9. Supersaturation as a function of time during the process of vapor condensation upon nuclei of distribution IV, for an updraft velocity  $u=400 \text{ cm sec}^{-1}$ .

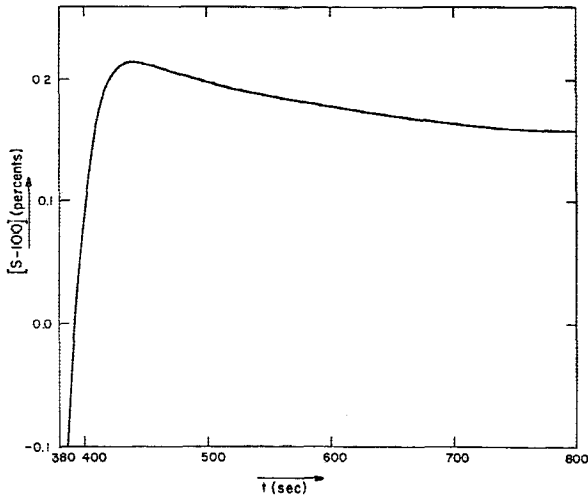


FIG. 10. Supersaturation as a function of time during the process of vapor condensation upon nuclei of distribution I.

consuming than computation in the region of larger drops.

*Acknowledgments.* I am very grateful to M. H. Davis for suggesting the problem of this paper. I am also indebted to him for many helpful discussions during all phases of this work.

APPENDIX

List of Symbols

- $A$  surface area of drop ( $\text{cm}^2$ )
- $a$  radius of disc (cm)
- $C$  electrostatic capacity (cm)
- $c_p$  specific heat of dry air at constant pressure [ $= 1.00464 \times 10^7 \text{ ergs gm}^{-1}(\text{°K})^{-1}$ ]
- $c_1, c_2$  constants ( $\text{cm sec}^{-1}$ )
- $D$  diffusion coefficient of water vapor in air ( $= 0.226 \text{ cm}^2 \text{ sec}^{-1}$ )
- $e$  ambient vapor pressure ( $\text{dyn cm}^{-2}$ )
- $e_s$  saturation vapor pressure over flat surface of pure water ( $\text{dyn cm}^{-2}$ )
- $e_s^*$  saturation vapor pressure over cloud drop ( $\text{dyn cm}^{-2}$ )
- $G_a$  mass of dry air in air parcel (gm)
- $G_v$  mass of vapor in air parcel (gm)
- $G_w$  total amount of water condensed upon nuclei from vapor in air parcel (gm)
- $g$  acceleration of gravity ( $= 980.616 \text{ cm sec}^{-2}$ )
- $h$  height of cup (cm)
- $i$  van't Hoff factor
- $K$  heat conduction coefficient of air [ $= 2.42788 \times 10^3 \text{ ergs cm}^{-1}(\text{°K})^{-1} \text{ sec}^{-1}$ ]

- $k$  Kelvin corrector
- $L$  heat released by condensation of one gram of vapor ( $= 2.49234 \times 10^{10} \text{ ergs gm}^{-1}$ )
- $M$  molecular weight ( $\text{gm mole}^{-1}$ )
- $M_a$  molecular weight of dry air ( $= 28.966 \text{ gm mole}^{-1}$ )
- $M_n$  molecular weight of salt ( $= 58.548 \text{ gm mole}^{-1}$ )
- $M_w$  molecular weight of water ( $= 18.016 \text{ gm mole}^{-1}$ )
- $m_n$  mass of dissolved salt (gm)
- $m_w$  mass of pure water on drop (gm)
- $m_0$  mass of salt (gm)
- $N$  number of nuclei
- $p$  pressure ( $\text{dyn cm}^{-2}$ )
- $p_m$  pressure of moist air ( $\text{dyn cm}^{-2}$ )
- $R$  universal gas constant [ $= 8.3144 \times 10^7 \text{ ergs mole}^{-1}(\text{°K})^{-1}$ ]
- $R_a$  specific gas constant for dry air [ $= 2.8704 \times 10^6 \text{ ergs gm}^{-1}(\text{°K})^{-1}$ ]
- $R_c$  Raoult corrector
- $R_m$  specific gas constant for moist air [ $\text{ergs gm}^{-1}(\text{°K})^{-1}$ ]
- $R_v$  specific gas constant for water vapor [ $= 4.6156 \times 10^6 \text{ ergs gm}^{-1}(\text{°K})^{-1}$ ]
- $r$  radius of drop or radius of curvature (cm)
- $r_0$  radius of salt nuclei (cm)
- $S$  relative humidity (%)
- $s$  specific heat of water [ $= 4.186 \times 10^7 \text{ ergs gm}^{-1}(\text{°K})^{-1}$ ]
- $T$  temperature ( $\text{°K}$ )
- $t$  time (sec)
- $u$  vertical velocity of air parcel ( $\text{cm sec}^{-1}$ )
- $v$  volume of both cups ( $\text{cm}^3$ )
- $w$  mixing ratio
- $z$  altitude (cm)
- $\alpha$  solubility of salt in water ( $= 0.35$ )
- $\beta$   $M_w/M_a (= 0.622)$
- $\nu$  number of moles of salt attached to both surfaces of disc
- $\rho$  density ( $\text{gm cm}^{-3}$ )
- $\rho_n$  density of salt ( $= 2.16 \text{ gm cm}^{-3}$ )
- $\rho_s$  density of drop ( $= 1 \text{ gm cm}^{-3}$ )
- $\sigma$  surface tension ( $= 75 \text{ dyn cm}^{-1}$ )
- $\omega$  frequency ( $\text{rad sec}^{-1}$ )

The superscript zero indicates the value of the variable at  $t=0$ .

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