The Microstructure and Colloidal Stability of Warm Clouds

Part II — The Causes of the Variations in Microstructure

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Abstract

An attempt is made to explore the causes of the differences in microstructure between the Hawaiian orographic cloud, maritime cumuli and continental cumuli, observations on which were discussed in Part I of this series.

1. Introduction

In Part I of this series evidence has been presented to show that the Hawaiian orographic cloud, maritime cumuli, and continental cumuli form a series of cloud types, each of which is more colloidally stable that the one before; further, from the point of view of their microstructure, these cloud types form a series, each of which has typically a narrower droplet spectrum, smaller maximum and average droplet sizes, and a higher droplet concentration than the preceding one.

This parallel between colloidal stability and microstructure would be expected from the theory of raindrop growth by coalescence. It seems reasonable therefore to draw the conclusion that the variations in stability from one cloud type to another are the result of the variations in microstructure. This raises the question of the causes of these variations in microstructure.

It has been pointed out elsewhere (SQUIRES 1952b) that, although the final formation of raindrops in warm rain must occur by coalescence, the factors which determine whether a cloud shall be colloidally stable or not are most likely to operate during the condensation phase of the growth of the droplets.

In Part I of this series it was shown that the systematic differences in the type of droplet spectrum which distinguish these three kinds of cloud are associated with differences in their typical droplet concentrations. It is evident that, if only a few droplets are formed in a cloud, these must be larger than if many are formed, other things being equal. The calculations of DAS (1956) indicate that coalescence of droplets has little or no effect on the droplet spectrum as a whole. Thus it seems reasonable to suppose that the differences in microstructure are not only related to, but are very largely caused by, differences in the number of droplets which form per unit volume; this point of view forms the basis of the following discussion.

The number of droplets which form per unit volume is determined by two factors: the maximum value reached by the supersaturation, and the spectrum of critical supersaturations of the condensation nuclei present in the air. Only nuclei whose critical supersaturations are exceeded can grow by condensation to form cloud droplets. The maximum value of the supersaturation itself is determined partly by the spectrum of nuclei, and partly by the speed of the upcurrent. These two

factors are therefore likely to be among the chief determinants of the microstructure and colloidal stability of warm cloud.

Unfortunately neither of these can be measured in a manner which is effective for the present purpose. With regard to the nuclei, what is required is a spectrum of critical supersaturations of cloud nuclei; since the supersaturations in cloud are extremely small, this presents great difficulties. As for upcurrents, what is required is a knowledge of their speed in the critical region of the cloud where the supersaturation is passing through its maximum. This in itself presents difficulties over and above the instrumental ones, which are formidable.

It seems, therefore, that these crucial factors in the problem of the microstructure and colloidal stability of warm cloud are, for the present, unobservable. It will therefore often be necessary in the following exploratory discussion to rely on crude estimates of these factors.

Table 1 summarizes the data on droplet concentrations derived from the observations discussed in Part I of this series. It shows the contrast between the three types of clouds, and constitutes the quantitative basis of the following discussion.

Table 1. Data on the frequency distribution of values of the droplet concentration in three types of cloud. (Number of droplets per cm^3).

Cloud Type	Hawaiian Orographic Cloud	Maritime Cumuli	Continental Cumuli
1st quartile droplet concentrations...	4	22	119
Median droplet concentration.....	10	45	228
3rd quartile droplet concentration....	30	70	310
Maximum droplet concentration observed.....	370	470	2,800
Total number of observations.....	123	438	403

2. The effect of the upcurrent — the contrast between the Hawaiian orographic cloud and maritime cumuli

The Hawaiian orographic cloud forms in an unmodified maritime air mass. The difference in microstructure between this cloud and maritime cumuli must therefore reflect the influence of factors other than that of the spectrum of condensation nuclei. Chief among these would appear to be (a) the rate of rise of the air forming the cloud, and (b) the total time available for the subsequent evolution of the droplet spectrum, that is, the average lifetime of a parcel of cloud. This average lifetime may be considerably longer in the orographic cloud than in cumuli. Since the cloud mass usually fills the space between the ground and the trade-wind inversion, it is reasonable to take this lifetime to be (at a maximum) some 2 to 4 hours, for the width of the cloud along the wind is some 20 nautical miles, and typical horizontal wind speeds during the period of the observations were 5—10 kt.

However, according to the observaions of SQUIRES and WARNER (communicated), there is only a slight change in the droplet concentration in the orographic cloud from near the coast to near its western or inland edge. This indicates that a difference in rate of rise may be the major cause of the contrast between maritime cumuli and the orographic cloud, at least in its lower reaches. Indeed, since the slope of the ground is nearly constant, about 5 %, the average vertical velocity of the air forming the orographic cloud must have been in the region 15—25 cm sec^{-1}. In contrast, the vertical currents in trade wind cumuli have been found by MALKUS (1953) to be normally some metres per second; BYERS (1956) states that the average growth rate of cumulus clouds and of the radar echoes found in some of them over the Caribbean Sea was "of the order of 300 to 400 ft per minute".

HOWELL (1949) has illustrated by numerical examples how, in a steadily rising air parcel, the supersaturation (S) of the air at first increases rapidly, but soon reaches a maximum (S_m) and begins to decrease. HOWELL found that, once a nucleus is activated, that is, grows past its critical size and forms a cloud droplet, there is very little tendency for it to re-evaporate to a nucleus, even though S decreases as the air parcel continues to rise. Since the value of S_m determines how many nuclei will be activated, it also determines essentially the concentration of droplets which will be found at later stages of the process. It is evident that S_m will be influenced by the speed of the up-

current. One of HOWELL's examples illustrates the dependence of this maximum supersaturation (S_m) on the upcurrent; with a fixed number of homogeneous nuclei, S_m increases by a factor of about 5 when the upcurrent is increased ten times. In the atmosphere, with its heterogeneous content of nuclei, the number of drops formed will of course increase as S_m increases, since more nuclei will be activated. These additional drops will restrain the rapid rise of S, so that one might expect that a ten-fold increase in upcurrent would result in an increase in S_m, but probably rather less than five-fold.

As applied to a cumulus, these ideas refer specifically to the region of active upcurrent; in surrounding regions, smaller or even negative upward velocities no doubt occur, but these regions play no part in the original formation of the cloud mass, and are not likely to exert much influence on the microstructure of the cloud as a whole. It is therefore reasonable to discuss this problem in terms of the typical upcurrents found in the cloud, as given above.

The supersaturation, S, is related at any moment to the difference between the liquid water which has become available since saturation was reached, and the total mass of the cloud droplets already formed. The characteristic maximum of S is partly due to the shape of the equilibrium vapour pressure curve over a nucleus. It is also partly due to the fact that the absorptive power of the forming cloud drops is small at first; consequently, water is not removed from the vapour phase rapidly enough to prevent the supersaturation increasing as the air ascends. Leaving aside for the moment the complications which arise from the unknown sizes of the nuclei which form droplets, let us suppose that the droplets are of uniform size, grow from zero radius at the condensation level, and that they have equilibrium vapour pressures equal to that over a plane water surface. Equations relating to the behaviour of S have been developed elsewhere (SQUIRES, 1952a). Using the same terminology and symbols, let r be the radius of the droplets, n their number per gram of air, v the upcurrent, z the height above the condensation level, and S the supersaturation expressed as the excess of the dew-point of the air over its temperature in deg. C.

Then, as shown in Appendix A, it results that the maximum value of the supersaturation, S_m, is given by:

$$S_m = 0.73\ v^{3/4}\ n^{-1/2}. \qquad (1)$$

Now it is obvious that when the upcurrent is increased, smaller nuclei will be utilized for cloud formation, on the average. Also, the more or less saline droplets already existing at the cloud base on the larger nuclei will have less time in which to exercise their restraining influence on the growth of S. This equation therefore tends to underestimate the relative effect of increasing upcurrent on S_m. In order to use it to compare the maritime cumuli with the Hawaiian orographic cloud, it is necessary to assume something about the spectrum of the nuclei, or rather, of the critical supersaturations of the nuclei. For this purpose we may take the law of distribution of aerosols given by JUNGE (1954) according to which the number of nuclei of radius r to $r+dr$ is proportional to r^{-4}. The number between r_1 and r_2 is then proportional to $(r_2^{-3} - r_1^{-3})$ The total number with radius $\geqslant r_2$ is therefore proportional to r_2^{-3}, provided r_1 is appreciably bigger than r_2. If we assume that the nuclei are all of similar material or mixture of materials, the effective molar mass (M) of a nucleus is proportional to r^3. The critical supersaturation of a nucleus, S_c, is proportional to $M^{-1/2}$. The number of nuclei with critical supersaturations less S_m is therefore proportional to S_m^2. Combining this with equation (1) gives the result that, in a given air mass,

$$\begin{aligned} S_m &\propto v^{3/8}, \\ n &\propto v^{3/4}. \end{aligned} \qquad (2)$$

If therefore the characteristic upcurrents in maritime cumuli exceed those in the orographic cloud by a factor of 5 to 10, the droplet concentrations should stand in a ratio of 3.3 to 5.5 or, in view of the underestimation of the effect of increasing v in this treatment, rather more.

Taking into account the rather wide fluctuations of droplet concentration observed in any one cloud, the approximate nature of JUNGE's law of distribution of aerosols, and the errors which can arise from using Raoult's law of vapour pressures, this result seems to be in reasonable agreement with the ratio of the

medians of the measured concentrations, as given in Table 1, namely 4.5. It may be concluded that the difference in microstructure between maritime cumuli and the Hawaiian orographic cloud is sufficiently well explained by the difference in upcurrent.

3. The contrast between maritime and continental cumuli

The contrast between continental and maritime cumuli is even more marked than that just discussed. BYERS (1955) attributes the notable difference in colloidal stability between cumuli over the Caribbean Sea and over the mid-west of the United States to the fact that cumuli grow rather more rapidly in the latter region, pointing out that the coalescence process needs time to operate. This difference in rate of growth no doubt reflects a systematic difference in upcurrent.

However, over the group of *warm* clouds sampled during the measurements of droplet spectra, no such systematic difference was in evidence. No measurements were made of upcurrents, but the observer's notes of turbulence, classified on a scale of 1 to 5, show that the maritime and continental cumuli sampled were about equally turbulent. On the whole, the maritime cumuli were slightly deeper than the continental ones; because of their higher bases, continental cumuli of any great depth are likely to reach far above freezing level, and form ice. The possibility therefore appears that, on the whole, the maritime cumuli might have had slightly stronger upcurrents than the continental ones. This difference, if it were real, would be in the wrong sense to explain the observed difference in microstructures. The only measurements which bear on this question are photographic records taken on the ground during some of the 1956 measurements of continental cumuli in inland Australia. These showed that the rates of growth of cumulus tops were mostly in the range 200 to 400 feet per minute; this is quite comparable with the values given by BYERS (1956) for cumuli over the Caribbean Sea, namely 300 to 400 ft per minute. Thus it seems unlikely that a systematic difference in upcurrent contributed in any notable way to the observed difference in microstructure between maritime and continental cumuli.

Cumuli vary considerably in their persistence as recognizable individual entities. It might be expected that, in a long-lasting cloud, the microstructure would undergo some evolution, presumably in the direction of a decrease in droplet concentration. The tendency for the droplet concentration to decrease upwards in cumuli (ZAITSEV, 1950) could be taken as suggesting this. However, it is quite possible that, even in such cloud, the lifetime of individual parcels of cloud is much the same as in less durable clouds. Certainly, no striking difference in microstructure has been found which related to the persistence of the cloud as a whole. There is indeed a tendency for the clouds of continental air masses to be more durable than those of maritime air masses; this is perhaps a reflection of some difference in the pattern of convection, and if it were relevant at all would apparently work in the wrong direction to explain the observed difference in microstructure.

It seems reasonable therefore to seek the cause in the spectra of the condensation nuclei; this spectrum, together with the upcurrent, must determine the course of events in the critical region near the cloud base where the supersaturation passes through its maximum, and the character of the microstructure of the cloud is determined. It is known that there are certain differences between the maritime and the continental aerosol. According to LANDSBERG (1938), the mean count of Aitken nuclei found over the continents, 9,500 cm^{-3}, is about ten times that found over the ocean, 940 cm^{-3}. WOODCOCK (1953) has measured the giant nuclei, that is, those of diameter about 1 micron and larger, in the maritime aerosol and found that they are predominantly sea salt. At a given height above the sea, the number of these giant nuclei is a function of the strength of the surface wind. TWOMEY (1954; 1955) has made similar measurements in a maritime air mass moving inland. He found that even far from the sea nearly all the giant particles consist of either sea salt or sodium chloride. The concentration of these particles at distances of up to 700 miles inland was found to be sometimes quite comparable with that measured at the coast; intensified mixing over the land had merely redistributed them in the vertical without seriously changing the total number present. However, convective showers

over the land were efficient in removing the salt particles from the atmosphere, and appeared to be capable of reducing the number of these particles by a factor of the order of 100. Thus continental air masses may often be deficient in giant nuclei compared with maritime air masses.

The Aitken count itself has of course no direct bearing on the microstructure of clouds, since the majority of the particles included in it never become cloud nuclei, that is, they are not utilized in the formation of cloud; if LANDSBERG's mean values were taken as typical of the sub-cloud layer, it would appear that on the average only about one Aitken particle in twenty succeeded in becoming the nucleus of a cloud droplet in maritime air and one in forty in continental air. LANDSBERG's figures do however indicate that the continents can be sources of Aitken nuclei; it is possible therefore that the continents also add to the atmosphere a number of particles which, while not as large as the giant sea-salt nuclei, are large enough to be utilized in cloud formation.

The giant sea-salt nuclei reach concentrations of 1 cm^{-3} only with very strong winds over the sea. They cannot account therefore in themselves for more than a small fraction of the cloud droplets. Nevertheless, they may be important in two ways. WOODCOCK (1952) has suggested that, by forming relatively large droplets, they may act to trigger off the formation of rain by the coalescence process. Since however, in the observations discussed in Part I of this series, a large proportion of the samples were taken in cumuli which were not raining, this process alone cannot account for the contrast in microstructure between maritime and continental cumuli. It seems possible however that sea-salt nuclei could influence the microstructure of cumuli in another way. KEITH and ARONS (1954) have shown that, at cloud base, these nuclei have already formed droplets with diameters of several microns, or even tens of microns. Thus the giant nuclei must have the effect of reducing the maximum value reached by the supersaturation a little above cloud base. In this way they could indirectly affect the microstructure by restricting the number of nuclei which become activated and grow to cloud droplets.

There would appear therefore to be two possible ways in which a difference between the maritime and the continental aerosol could account for the difference in microstructure between maritime and continental cumuli:

(a) The addition of cloud nuclei to the air from some continental surfaces.

(b) The reduction of the supersaturation in cloud by giant sea-salt nuclei, which are on the whole more numerous in maritime air.

The following discussion has as its aim to show that the second of these possibilities is improbable; in effect, to show that, with likely values of the upcurrent, the concentrations of giant sea-salt nuclei which occur at cloud base levels are not great enough to produce the effect proposed.

It will be assumed, for the moment, that the only significant difference between the maritime and the continental aerosols is that the former is rich in giant sea-salt nuclei and the latter devoid of them. The argument will be concerned with a comparison of the supersaturation of cloud air in maritime and continental cumuli. It will be shown that, in the region of active upcurrent, and above the level where S passes through its maximum, the supersaturation is typically greater in maritime than in continental cumuli. This is contrary to what one might expect if it were true that the more numerous giant nuclei of maritime air masses were effective in restricting the number of droplets forming in maritime cumuli by depressing the supersaturation.

It has been shown elsewhere (SQUIRES, 1952a) that, in the body of a cloud after S has passed through its maximum value and is slowly decreasing, a fairly good approximation to S is given by:

$$S = (0.024v + 1.7 \times 10^{-6}n - 64.4\Sigma(M/r^2))/\Sigma r \quad (3)$$

where M is the effective molar mass of the nucleus in a drop of radius r, and the summations are extended over all the droplets in 1 gram of air. The numerical values are for 10° C, 800 mb. The first term in the numerator is related to the rate at which water is becoming available, the second to the raising of the equilibrium vapour pressure over the droplets by capillarity, and the third to its reduction by the dissolved nuclei. The denominator is a measure of the absorptive power of the cloud for water vapour. The effect of ventilation of the droplets, which is small, is neglected. Consider a maritime cumulus, with $n \approx 4.5 \times 10^4$

per gram of air (see Table 1), and an upcurrent of 1 m sec^{-1}, or more. The first term dominates in the numerator. Its value is at least 2.4; that of the second is 0.076, but the third term is more difficult to evaluate. It is shown in Appendix B that, under the conditions occurring at the time when the maritime observations were made, it could not have exceeded 0.27.

Hence, from (3), in a typical maritime cumulus, with $n = 4.5 \times 10^4$ droplets per gram of air.

$$(0.024v + 0.08)/\Sigma r > S > (0.024v - 0.19)/\Sigma r \quad (4)$$

In a typical continental cloud, with $n = 2.3 \times 10^5$ droplets per gram of air (see Table 1) the second (capillarity) term in the numerator of (3) is more important, viz. 0.39. The third term, which is essentially negative, need not concern us. We have

$$S > (0.024v + 0.39)/\Sigma r \quad (5)$$

in a typical continental cumulus.

From the data of Table 1, it is obvious that, for a given liquid water content, Σr must be much larger in continental than in maritime cumuli. Thus in the maritime cumulus of Fig. 3, of Part I of this series, the average value of Σr was 71 cm per gram of air, and in the continental cumulus of Fig. 4 of Part I, the average value was 184 cm per gram of air, or 2.6 times greater. As mentioned in Part I, a large proportion of the observations in both maritime and continental cumuli were made using a magnesium oxide layer, so that the values of Σr cannot be deduced. In order to make use of the whole of the observations, one may estimate that, for a given liquid water content, Σr will be very roughly proportional to the two-thirds power of the droplet concentration, treating the drops for this purpose as homogeneous. Since the data of Table 1 indicate that a typical continental cumulus has a droplet concentration about five times as great as a typical maritime cumulus, this would indicate that Σr would be about three times greater in the continental cloud. As mentioned in Part I, the continental cumuli form a rather heterogeneous group. Thus, one may conservatively estimate that, over a substantial proportion, and probably a majority, of continental cumuli, the value of Σr will be at least 2.5 times greater than in a comparable maritime cumulus.

It may be remarked here that Σr does not show much change with height in a cumulus; the slow upward increase in liquid water content implies only a small change in mean droplet radius, and this is largely offset by a tendency for the droplet concentration to decrease slightly with increasing height.

On this basis then, comparing similar maritime and continental cumuli with the same upcurrent, we may write:

$$\frac{S_{mar}}{S_{cont}} > \frac{2.5(0.024v - 0.19)}{(0.024v + 0.39)} \quad (6)$$

from the inequalities (4) and (5). S_{mar} and S_{cont} represent the supersaturations at similar levels in the body of each cloud, that is, above the region where S passes through its maximum. As pointed out earlier, over the groups of clouds in which samples have been taken, it is justifiable to take the upcurrents as being equal, on the average.

The values of the ratio on the right-hand side of (6) are shown in Table 2, for various values of the upcurrent.

Table 2.

Upcurrent v cm sec^{-1}	$S_{mar}/S_{cont}>$
20	0.8
50	1.6
100	2.0
200	2.2
300	2.3

The measurements of upcurrent and cloud growth rates already mentioned indicate that a typical upcurrent is about 1 m sec^{-1} or slightly more; hence it would seem that, in the body of the cloud, in a region of active upcurrent, the supersaturation is typically about twice as great in a maritime as in a continental cumulus.

This discussion applies only to the body of the cloud, that is, the region above the level where S, after having passed through a maximum, is now decreasing slowly. It does not follow from the above argument that the maximum value of the supersaturation (S_m) is twice as great in a maritime as in a continental cumulus. If however the supposition were true that the giant sea-salt nuclei, by lowering S_m, were the prime cause of the low droplet

concentrations in maritime cumuli, and that other differences between the continental and maritime aerosols had little influence on the microstructure of cumuli, the result found above would be surprising. For, as shown in the derivation of equation (2) above, JUNGE's law of distribution of aerosols (JUNGE, 1954) would in that case suggest that S_m should be about $5^{1/3}=2.2$ times *smaller* in the maritime cumuli.

If, as these arguments suggest, the giant sea-salt nuclei are not primarily responsible for the continental-maritime contrast in cumulus microstructure, it seems reasonable to suspect that the chief cause may be found in the first possibility mentioned above, namely, the addition of cloud nuclei to the air from some continental surfaces.

It seems possible, for example, that unmodified maritime air masses may be relatively poor in nuclei in the size range called "large particles" by JUNGE (1954) ($0.8\mu > d > 0.08\mu$) but that when such an air mass moves inland, it may pick up these particles. The conclusion reached by JUNGE (communicated) that the large particles are primarily of continental origin would suggest this. If the concentration of particles of this kind should rise to some hundreds per cm^3 over the continent, the high droplet concentrations found in continental cumuli with low supersaturations are readily explained.

4. Conclusions

An exploratory attempt has been made to evaluate the causes of the differences in microstructure between the Hawaiian orographic cloud, maritime cumuli and continental cumuli. As regards the contrast between the first two, which both form in an unmodified maritime air mass, it has been shown that the relatively slow upcurrent in the orographic cloud affords a sufficient explanation.

The striking difference between maritime and continental cumuli cannot be explained in terms of a difference in upcurrent, or in the total lifetime of parcels of cloud. It seems likely that it originates from a difference between the maritime and continental aerosol. On the basis of an approximate formula for the supersaturation of cloud air, it is concluded that the giant sea-salt nuclei are not primarily responsible for this difference; it seems more likely that the operative difference between the maritime and the continental aerosol occurs in the size region below that of the giant sea-salt particles, namely in that of the "large particles" continental air being richer in nuclei of this size than maritime air.

Unfortunately, a wide gap separates the, lower end of the spectrum of giant nuclei, which can be studied individually by impaction methods, from the size range where graduated expansions might be useful in yielding a spectrum of critical supersaturations. Observations of droplet spectra in natural clouds might possibly be a means of bridging a portion of this gap, provided upcurrents can be measured simultaneously.

If indeed it should be found that the contrast in microstructure between maritime and continental cumuli is largely caused by cloud nuclei originating over the continents, this could have important implications for those experiments where attempts are made to modify warm continental cumuli by seeding them with salt particles. The conclusions mentioned in the Introduction indicate that the marked colloidal stability of continental cumuli is at least partly due to the fact that the average and maximum droplet sizes found in them are small and rather uniform compared with those in maritime cumuli. Since natural sea-salt particle concentrations are inadequate to seriously affect the microstructure of cumuli, it does not seem likely that artificially produced hygroscopic particles would. On the other hand, it may perhaps be possible to modify to some extent the microstructure of cumuli forming in a maritime air mass in such a manner as to increase their colloidal stability. On the views outlined above, this could be attempted by adding to the cloud-forming air a sufficient number of cloud nuclei. The number of particles required, of the order of a hundred per cm^3, is much greater than is usually considered necessary in seeding clouds; on the other hand, the particle size required would certainly be much smaller.

Appendix A

An evaluation of the effect of upcurrent on the maximum value reached by the supersaturation.

Using the physical model described in the main text in Section 2, and the symbols de-

fined there, it has been shown elsewhere (SQUIRES, 1952a) that:

$$S = az - bnr^3 \qquad \text{(A1)}$$

where $a = \frac{g}{J}\left(\frac{1}{c_p} - \frac{T}{L\varepsilon}\right)$

$$b = \frac{4\pi}{3}\left(\frac{L}{c_p} + \frac{\varrho R T^2}{JL\varepsilon^2 e}\right)$$

and, neglecting the very small effect of ventilation of the small droplets,

$$\frac{r dr}{dt} = ES \qquad \text{(A2)}$$

where $E = \varepsilon^2 LDJe/RT^3\left(1 + \frac{\varepsilon^2 L^2 DJe}{kR^2T^3}\right)$

where L is the latent heat of water vapour,
D the coefficient of diffusion of water vapour in air,
k the thermal conductivity of air,
ε the specific gravity of water vapour with respect to air,
ϱ the density of air,
R the gas constant of air per gram, and the other symbols have their usual meanings. At 10° C, 800 mb,

$$a = 7.9 \times 10^{-5}$$
$$b = 1.68 \times 10^{4}$$
$$E = 6.6 \times 10^{-8}$$

From (A1) and (A2) $r\frac{dr}{dt} = Eavt - Ebnr^3 \qquad \text{(A3)}$

if t is measured from the moment of passing the condensation level where $S = 0$, $r = 0$. Putting

$$r = y(Eav)^{1/4}\,(nEb)^{-1/2}$$
$$t = x(Eav)^{-1/4}\,(nEb)^{-1/2}$$

the equation (A3) becomes:

$$\frac{dy}{dx} = \frac{x}{y} - y^2. \qquad \text{(A4)}$$

A series solution of (A4) through the origin has been obtained. The value of S in terms of x and y is given by:

$$ES = (Eav)^{3/4}(nEb)^{-1/2}(x - y^3)$$

which is a maximum when

$$x = y^3 + \frac{1}{3y}$$

This curve intersects the solution curve of (A4) near $x = 0.86$, $y = 0.74$, so that

$$S_m = 0.46\ E^{-3/4}\ a^{3/4}\ b^{-1/2}\ v^{3/4}\ n^{-1/2}$$

or, substituting the numerical values already quoted for E, a and b

$$S_m = 0.73\ v^{3/4}\ n^{-1/2}.$$

Appendix B

An evaluation of the effect of condensation nuclei in depressing the supersaturation in the body of a maritime cumulus.

This effect is taken into account by the last term in the numerator of equation (3) of the main text. Consider first the contribution to $\Sigma(M/r^2)$ due to droplets containing giant sea-salt nuclei. WOODCOCK (1953) gives cumulative distribution curves for the spectra of sea salt over the sea near cloud base for various wind strengths and at a number of widely separated places. TWOMEY (1955) has given similar data which are in good agreement with WOODCOCK's. It seems that the spectrum of giant nuclei at cloud base level in a maritime air mass is fairly well defined by surface wind force alone, irrespective of geographical position. The wind strengths estimated over the sea during the measurements on maritime cumuli both off Australia and near Hawaii did not reach force 7. If therefore WOODCOCK's data for force 7 wind off Hawaii are used, an upper estimate of the effect of the giant nuclei can be derived. It is convenient to replace the observed spectrum by a distribution in groups as shown in Table B1, where the observed spectrum given by WOODCOCK is shown on the left, and the substituted group distribution on the right. It is obvious that the substituted spectrum dominates the observed spectrum for an air density of 0.98 g cm^{-3}, corresponding to 800 mb, 10° C.

Table B1

Observed spectrum of sea-salt nuclei for a force 7 wind near cloud base (after Woodcock)		Substituted group spectrum per gram of air	
Mass of nuclei $\mu\mu$g	Number of nuclei per m³ of mass greater than that shown in the first column	Mass of nuclei $\mu\mu$g	Number of nuclei of this mass per gram of air
1	800,000		
10	200,000	10	555
100	40,000	100	165
1,000	5,000	1,000	35
10,000	150	10,000	5
100,000	<1	100,000	0.15
		1,000,000	0.001

KEITH and ARONS (1954) have given curves for the growth of sea-salt particles in rising air, up to the cloud base. We may safely take their values as giving lower limits for the droplet radii inside the cloud. The values are quoted for upcurrents of 0.5, 1.0 and 2.0 m sec^{-1}; the effect of speed in this range is small. Taking the smallest values, those for $v=2$ m sec^{-1}, we get the results shown in Table B2.

Table B2

Mass of nucleus (m_s) $\mu\mu$ g	Radius at cloud base (r) μ	m_s/r^2 10^{-4} ×	Number assumed per gram	Product
10	6	0.28	555	0.016
10^2	10	1.0	165	0.017
10^3	17	3.5	35	0.012
10^4	33	9.2	5	0.005
10^5	57	31	0.15	0.000
10^6	87*	132	0.001	0.000
				Total $= \Sigma\left(\frac{m_s}{r^2}\right) =$ 0.050

* A rough lower bound found by extrapolating by a tangent the curve given by KEITH and ARONS, which is concave towards the axis of r.

The total of $\Sigma m_s/r^2$ is therefore certainly less than 0.050. Treating sea salt as ionically dissociated sodium chloride $M=m_s/29$; hence $\Sigma M/r^2<0.0017$; the contribution to the last term in the numerator in equation (3) from droplets formed on giant sea-salt nuclei is therefore less than 0.11.

As mentioned earlier, only with very strong winds does the concentration of giant sea-salt nuclei reach 1 per cm³. This is small compared with the concentration of cloud droplets. It remains to make an estimate of the contribution to $\Sigma(M/r^2)$ due to the cloud drops formed on nuclei too small to be observed by the impaction method, that is, below about 1 $\mu\mu$g. JUNGE (communicated) has studied the maritime aerosol in the Hawaiian Islands in two size groups—giant particles ($r=8\mu$ to $0.8\,\mu$) and large particles ($r=0.8\ \mu$ to $0.08\ \mu$). He analysed the two components for NH_4, NO_3, Cl and SO_4. The average total mass of all these constituents in the form of giant particles was 5.8 μg per m³; in the form of large particles, 0.18 μg per m³. If, as the analyses of fog and cloud water made by HOUGHTON (1955) indicate, these components are an important fraction of the total aerosol in the "large particle" range, it is evident that the total mass of the large particles can hardly exceed ten times the concentration found by JUNGE—that is, 1.8 μg per m³. On the basis of measurements at various places, JUNGE (communicated) considers that the large particles are primarily of continental origin; their total mass concentration over the sea would not therefore be affected by wind strength, unlike that of the giant sea-salt particles. If therefore all the cloud drops not formed on giant nuclei are formed on large particles, the total mass of nuclear material cannot exceed 1.8 μ g per m³ or about 1.8×10^{-9} g per g of air.

It is easily shown that nuclei below JUNGE's lower limit for larger particles ($r=0.08\ \mu$) make no significant contribution. Their masses must be less than 4×10^{-15} g, if a density of 2 g cm^{-3} is assumed. Hence the total mass in the cloud drops (assumed not to exceed 4.5×10^4 per gram in number) in a gram of air could not exceed 1.8×10^{-10} g. Thus the total mass of the nuclei in the cloud droplets, apart from the giant particles, cannot exceed 2×10^{-9} g per gram of air. The effective molar mass must be less than about 10^{-10} per gram of air, since no likely material, even when completely dissociated, has an effective molecular weight of less than 20.

The observations of droplet spectra in maritime cumuli show that only a very small minority (about 1 %) of the droplets are smaller than $d = 4\ \mu$. Hence the total contribution to $\Sigma(M/r^2)$ from droplets formed on particles smaller than giant sea-salt nuclei must be less than $10^{-10}/4 \times 10^{-8} = 25 \times 10^{-4}$. Their contribution to the third term of the numerator in equation (3) is therefore less than 0.16; and the total value of the third term, including nuclei of all sizes, must be less than 0.27.

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