THE NUCLEI OF NATURAL CLOUD FORMATION PART II: THE SUPERSATURATION IN NATURAL CLOUDS AND THE VARIATION OF CLOUD DROPLET CONCENTRATION

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Summary — Calculations are performed to determine the time variation of supersaturation during cloud formation. It is shown that a simple expression can be used to obtain the maximun supersaturation (and hence the number of nuclei activated) as a function of updraught velocity and nucleus spectra insert omitted portion of copy reported in Part 1 and it isshown that they lead to cloud droplet concentrations which agree with those actually observed.

1. *Introduction* — The importance of the supersaturation spectra of condensation nuclei, particularly of the part of the spectrum below 1% supersaturation, was discussed in Part I of this paper. In this part the relationship between nucleus spectrum, supersaturation and cloud droplet concentration will be discussed quantitatively, and computations made to determine the probable range of maximum supersaturation in natural clouds, and hence the number of droplets formed when condensation occurs on nuclei with supersaturation spectra similar to those described in Part I.

2. *Time Variation of Supersaturation* — The variation with time of the supersaturation in a volume of air ascending at uniform speed, or being cooled adiabatically at a uniform rate, is determined by two opposing factors: first the effect of cooling, which tends uniformly to increse the saturation, and secondly the effect of condensation, which abstracts water vapour and therefore tends to reduce the supersaturation. The rate of growth by condensation of a single droplet is proportional to the supersaturation and to the radius of the droplet. Providing the droplets are large enough for capillarity and solute effects on vapour pressure to be neglected, the equation obeyed by the supersaturation is of the form [1]:

$$\frac{dS}{dt} = a - bS \Sigma r . \tag{1}$$

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In this equation and throughout the theoretical derivation, the supersaturation S is measured in ^{0}C (elevation of the dew point); r is the droplet radius (cm); the summation is carried out over all the droplets in unit volume; for temperature 10^{0} C, pressure 800 mb, $\alpha = 7.93 \cdot 10^{-5}\, V$, $b = 3.38$, V being the rate of ascent (cm sec^{-1}). The radius of a droplet is determined by the equation (1):

$$r \frac{dr}{dt} \approx 6.6 \cdot 10^{-8}\, S\,. \tag{2}$$

If the number of nuclei in unit volume with critical supersaturation between σ and $\sigma + \Delta\sigma$ is $\nu(\sigma)\,\Delta\sigma$, then combination of equations (1) and (2) gives

$$\frac{dS}{dt} = a - \beta S \int_0^S \nu(\sigma) \left[\int_\tau^t S dt \right]^{1/2} d\sigma \tag{3}$$

where $\beta = 3.63 \cdot 10^{-4}\, b = 1.23 \cdot 10^{-3}$, and $S = \sigma$ at $t = \tau$. From a physical point of view, α represents the initial rate of rise of supersaturation, while the negative factor on the right hand side of equation (3) represents the increasing effect of condensation in removing water vapour (which slows down, and eventually revers the rise in S). In the absence of condensation supersaturation would increase linearly with time, following the line $S = \alpha t$ in Figure 1. When condensation occurs, the supersaturation curve deviates increasingly from the linear, following a curve similar to that shown in the figure. Equation (3), which determines the course of the curve, cannot be solved except by tedious stepwise numerical computation; however by making certain approximations solutions can be obtained which give upper and lower bounds for the supersaturation. In Figure 1 it is readily seen that for any point P on the supersaturation curve, the following inequality is satisfied:

$$\text{Area } OXY > \int_0^t S dt > \text{Area } PXZ\,.$$

From this it is readily deduced that:

$$\frac{1}{2} \alpha (t^2 - \tau^2) > \int_\tau^t S dt > \frac{1}{2\alpha} (S^2 - \sigma^2)\,. \tag{4}$$

Overestimation of $\int S dt$ in equation (3) is equivalent to overestimation of the effect of condensation and hence to *underestimation* of S in the final solution. It therefore $\frac{1}{2} \alpha (t^2 - \tau^2)$ is substituted for this integral in equation (3) to obtain an approximate solution S', then this solution will underestimate the supersaturation; similarly, a solution S'' obtained by substitution of $(S^2 - \sigma^2)/2\alpha$ will overestimate the supersaturation; thus $S'' > S > S'$.

(i) *Derivation of* S': Many observed nucleus spectra were close to logarithmic distributions; the number of nuclei active at supersaturation σ will therefore be taken to be $N(\sigma) = c\sigma^k$; the function $\nu(\sigma)$ then becomes $kc\,\sigma^{k-1}$. Inserting this distribution function into equation (3) and putting $\int_\tau^t S dt = \frac{1}{2} \alpha (t^2 - \tau^2)$ and

$\sigma = \alpha\tau$, one obtains an equation for S':

$$\frac{dS'}{dt} = \alpha - 2^{-1/2}\, \beta\alpha^{k+1/2}\, ck\, S' \int_0^t \tau^{k-1} (t^2 - \tau^2)^{1/2}\, d\tau$$

$$= \alpha - 2^{-3/2}\beta\alpha^{k+1/2}\, ck\, S'\, t^{k+1} \int_0^1 \left(\frac{\tau^2}{t^2}\right)^{k/2-1} \cdot \left(1 - \frac{\tau^2}{t^2}\right)^{1/2} d\left(\frac{\tau^2}{t^2}\right)$$

thence

(5) $$\frac{dS'}{dt} = \alpha - AS'\, t^{k+1}$$

where $A = 2^{-3/2}\, \alpha^{k+1/2}\, \beta ck\, B\,(3/2, k/2)$ and B is the complete Beta-function. The

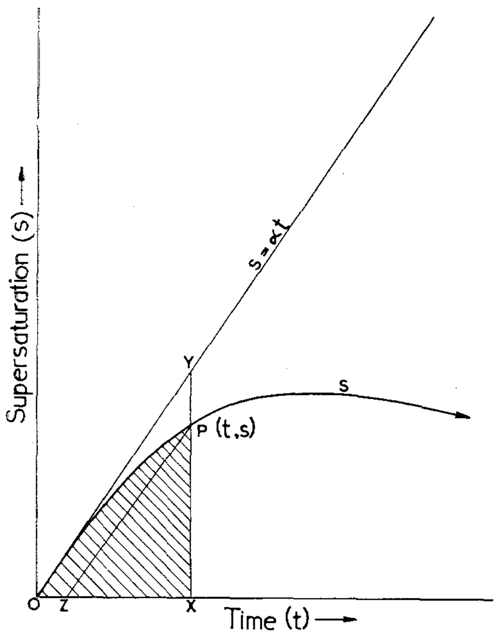

Fig. 1 - *Time variation of supersaturation.*

required solution of equation (5) is

(6) $$S' = \alpha \exp\left(-\frac{A}{k+2}\, t^{k+2}\right) \int_0^t \exp\left(\frac{A}{k+2}\, t^{k+2}\right) dt$$

which can be expressed also in series form:

(7) $$\begin{cases} S' = \alpha t \Big[1 - \dfrac{am}{m+1}\, t^m + \dfrac{a^2 m^2}{(m+1)(2m+1)}\, t^{2m} - \\ \quad - \dfrac{a^3 m^3}{(m+1)(2m+1)(3m+1)}\, t^{3m} + \ldots\ldots \Big] \end{cases}$$

where $m = k + 2$, $a = A/m$. In (7), therefore, α is proportional to the rate of ascent; m is determined by the distribution index k, and a is a function of the distribution constants c and k (which determine the supersaturation spectrum of the nuclei), and of the upward velocity V. The full expression for a is:

$$a = 4.35 \cdot 10^{-4} [7.93 \cdot 10^{-5} \, V]^{k+1/2} \frac{ck}{k+2} \; B\left(\frac{3}{2}, \frac{k}{2}\right).$$

For a given distribution, equation (7) is used to find S', which provides a lower bound for the supersaturation. Since the maximum supersaturation determines the number of droplets formed, computation of S' need not be continued beyond the maximum.

(ii) *Derivation of* S'': The equation for S'' is obtained by substituting $1/2\alpha\,(S^2 - \sigma^2)$ for $\int_\tau^t S dt$ in equation (3). The resulting equation is:

$$\frac{dS''}{dt} = \alpha - (2\alpha)^{-1/2} \beta \, ck \, S'' \int_0^S \sigma^{k-1} (S^2 - \sigma^2)^{1/2} \, d\sigma$$

$$= \alpha - (8\alpha)^{-1/2} \beta \, ck \, (S'')^{k+2} \int_0^1 (\sigma^2/S^2)^{k/2-1} (1 - \sigma^2/S^2)^{1/2} \, d\,(\sigma^2/S^2) \, .$$

Hence

$$\frac{dS''}{dt} = \alpha - C\,(S'')^{k+2}. \tag{8}$$

Here $C = (8\alpha)^{-1/2} \beta \, ck \, B\,(3/2, k/2)$, B again being the complete Beta function. An integral or series solution for equation (8) can readily be obtained, as the variables are separable. However, the maximum attained by S'' can be derived from (8) without solving the equation. The maximum is evidently

$$S''_{max} = (\alpha/C)^{1/k+2} \tag{9}$$

or, substituting for α and C, and putting $\beta = 1.23 \, . \, 10^{-3}$ as before

$$S''_{max} = \left[\frac{1.63 \times 10^{-3} \, V^{3/2}}{ckB\,(3/2, k/2)} \right]^{\frac{1}{k+2}} . \tag{10}$$

It can now be predicted that the maximum supersaturation attained when a given distribution of nuclei act as condensation centres in air at 800 mb and 10° C, ascending at V cm sec^{-1}, will lie between S'_{max} and S''_{max}; the number of nuclei activated and the number of cloud droplets formed (N) will be between N' and N'', where

$$N' = c\,(S'_{max})^k \text{ and } N'' = c\,(S''_{max})^k \quad \text{i.e.} \quad c\,(S''_{max})^k > N > c\,(S'_{max})^k \, .$$

When various pairs of values for c and k were inserted in equations (7) and (10) for rates of ascent 10 cm sec^{-1}, 100 cm sec^{-1} and 1000 cm sec^{-1}, it was found that the upper and lower bounds of the supersaturation never differed by more than 30%. It follows that equation (10) can be used to obtain the maximum supersaturation to better than 30% accuracy; the number of droplets, being proportional to the maximum supersaturation to power k, where for observed spectra k usually lay in the range 0.2 - 0.5, are given to better than 15% accuracy by the relation

$$N = c\,(S''_{max})^{k} = c^{\frac{2}{k+2}} \left[\frac{1.63 \times 10^{-3}\, V^{3/2}}{kB\,(3/2,\, k/2)}\right]^{\frac{k}{k+2}} . \tag{11}$$

3. *Application to observed spectra* — As described in Part I, observed spectra showed a very wide range in number and shape, but it was found that in the absence of rainy conditions, spectra generally fell into three types, one of which (type C) was found only in continental air masses, and two other types, A and B, which were found both in maritime (southerly or easterly) air and in modified maritime (southwesterly) air. These spectrum types gave the following median concentrations at the supersaturations indicated:

TABLE 1 - *Numbers of nuclei active at various supersaturations.*

Spectrum	$S =$ 0.01° C	0.05° C	0.1° C	0.2° C	0.5° C	1° C
Type A .... (maritime or mod. mar.)	25	35	40	50	80	125
Type B .... (maritime or mod. mar.)	48	65	90	180	600	1200
Type C .... (continental)	330	560	720	900	1400	2000

The distributions of types A or C are not very different to the logarithmic, and reasonably close approximations to the median spectra are obtained by taking the distribution constants $c = 2000$, $k = 0.4$ for type C and $c = 125$, $k = 1/3$ for type A. Type B curves deviate considerably from the logarithmic, owing to the inflexion of these curves. Up to about 0.15° C supersaturation, an approximation to the median type B distribution can be obtained by taking $c = 160$, $k = 1/3$, but if the maximum supersaturation exceeds 0.15° C, the increased numbers of nuclei at higher supersaturations should be allowed for.

Inserting these values of c and k in equations (7) and (10), the upper bounds S'' and the lower bound S' for the maximum supersaturation were calculated, and from these the limits of cloud droplet concentration N' and N''. The values obtained for updraught velocities 10 cm sec^{-1}, 100 cm sec^{-1} and 1000 cm sec^{-1} are set out in Table 2. These updraught velocities are probably typical of stratiform clouds, small cumuli and cumulonimbus clouds, respectively.

(The values marked with an asterisk are those for which the supersaturation was too high to justify the logarithmic approximation for type B spectra. These supersaturations were therefore overestimated and the droplet numbers underestimated).

TABLE 2(a) - *Bounds of supersaturation* S″ *(upper)*, S′ *(lower) for different spectrum types and rates of ascent* V.

Spectrum type Value taken for c Value taken for k		Maritime (A) 125 1/3	Maritime (B) 160 1/4	Continental (C) 2000 2/5
Supersaturation bounds (°C) at 10 cm/sec	S'_{max}	.022 (.14%)	.019 (.125%)	.007 (.045%)
	S''_{max}	.026 (.17%)	.02 (.13%)	.009 (.06%)
Supersaturation bounds (°C) at 100 cm/sec	S'_{max}	.095 (.62%)	.088 (.57%)	.031 (.2%)
	S''_{max}	.116 (.75%)	.093 (.61%)	.04 (.26%)
Supersaturation bounds (°C) at 1000 cm/sec	S_{max}	.42 (2.7%)	.41 (2.65%)	.13 (.85%)
	S''_{max}	.51 (3.3%)	.43 (2.8%)	.17 (1.1%)

Condensation upon nucleus spectra of the various types considered is therefore found to lead to droplet concentrations similar to those actually observed. The median spectra for maritime or modified maritime air were calculated to produce some 50-90 droplets cm^{-3} in an updraught of 1 metre sec^{-1}; the corresponding

TABLE 2(b) - *Droplet concentrations cm*$^{-3}$ *(nuclei activated) for conditions of Table* 2(a)

Spectrum type		Maritime (A)	Maritime (B)	Continental (C)
Droplet concentrations cm^{-3} at 10 cm/sec	N'	35	59	281
	N''	37	60	310
Droplet concentrations cm^{-3} at 100 cm/sec	N'	57	87	500
	N''	61	89	554
Droplet concentrations cm^{-3} at 1000 cm/sec	N'	93	128*	888
	N''	100	131*	985

figure for the median continental spectrum is about 500 cm^{-3}. SQUIRES ([2]) reported the following droplet concentrations:

Maritime clouds: Median 45 cm^{-3}, Maximum 470 cm^{-3}.

Continental clouds: Median 228 cm^{-3}, Maximum 2800 cm^{-3}.

There is obviously a good degree of agreement between the computed and observed concentrations, the computed values being somewhat higher. The spectra obtained during drought conditions and hot weather gave considerably higher nucleus concentrations than the median curve, for all supersaturations; computations based on these spectra yielded (for 1 m sec^{-1}) concentrations between 1000 and 4000 cm^{-3}, with maximum supersaturations between 0.015° C (0.1%) and 0.03° C (0.2%). The maximum of 2800 cm^{-3} (on a hot, dry midsummer day) reported by SQUIRES is consistent with these results. It is also of interest to note that clouds formed in southwesterly (modified southern maritime)air streams over south-eastern Australia have been found to contain consistently low numbers of cloud droplets, the concentrations being very similar to those found in maritime clouds (SQUIRES, private communication). This observation accords with the observation [Fig. 6(1), Part I] that modified southern maritime air contained somewhat fewer cloud nuclei than (easterly) maritime air.

It is well known that continental clouds, containing some hundreds, or even a thousand or more, droplets per cm^3, are markedly more stable than the maritime clouds of similar dimensions, and correspondingly more reluctant to precipitate. If this stability is found to be partly or entirely due to the presence of cloud nuclei in high concentrations, some basis may exist for the popular belief that a drought must be « broken » and that prolonged wet or dry spells tend to be self-propagating.

4. *Conclusion* — The observed variations in concentration of these cloud nuclei appear to be sufficient to account for observed variations in cloud droplet concentration, particularly the observed differences between maritime and continental clouds.

The results again emphasise that condensation nucleus counts at high supersaturations cannot be a reliable guide to cloud nucleus and cloud droplet concentrations. At 1 m sec^{-1} updraught, the maximum supersaturation attained in clouds was computed to be in the range 0.2%-0.8%, the higher supersaturations being attained with clean maritime air.

It is apparent that nuclei with critical supersaturation greater than 1% will seldom be activated in natural cloud formation.

To obtain a fuller understanding of the role of cloud nuclei in the determination of colloidal stability of clouds (particularly that of warm clouds), it is apparent that further observations of the distribution, properties and origin of cloud nuclei are desirable; simultaneous observations of cloud droplets and cloud nuclei are obviously essential and the initiation of such observations in the immediate future is intended.

REFERENCES

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