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## Abstract

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## GEOPHYSICS

**E. L. ALEKSANDROV, Yu. S. SEDUNOV**

# A METHOD FOR ESTIMATING THE MAGNITUDE OF SUPERSATURATION ARISING DURING THE FORMATION OF A DROPLET-LIQUID PHASE IN CLOUDS

*(Presented by Academician E. K. Fedorov, 31 III 1969)*

It is known that the process of formation of a droplet-liquid phase during the emergence of clouds is substantially connected with the rate of cooling of the air mass and with the presence in it of condensation nuclei. Since in the atmosphere there is always a sufficient number of condensation nuclei, their influence is manifested through the distribution function of condensation nuclei with respect to activity. A peculiarity of atmospheric processes is that droplet formation is caused by slow cooling of air in the presence of a sufficient number of condensation nuclei. The magnitude of the supersaturation that is thereby attained is very small and is unlikely to exceed tenths of a percent. Unfortunately, direct methods for measuring such small supersaturations still do not exist, and therefore we can judge the magnitude of supersaturation during the formation of the droplet-liquid phase in clouds only on the basis of theoretical estimates. Meanwhile, the magnitude of supersaturation is of great importance for understanding the processes occurring in clouds, since it is the principal factor determining the rate of condensational growth of droplets. It should be noted that in theoretical calculations of supersaturation ([1–4]) such quantities as the rate of cooling (ascent) of the air mass and the spectrum of nuclei with respect to supersaturation are used directly. In themselves these quantities are measured insufficiently reliably. Thus, for example, the rate of ascent of the air mass during the formation of layer clouds is determined from theoretical considerations and has not yet been measured. As may be judged from the literature ([5]), instruments for measuring the spectrum of nuclei with respect to supersaturation are still imperfect, and the methodology of such measurements requires refinement and analysis. It follows from this that, even having a good theoretical scheme for calculating the magnitude of supersaturation, we cannot be confident in the results obtained.

Below a method is proposed for estimating the magnitude of supersaturation that is free from the above shortcomings.

It is well known that at humidities above 70% nuclei become hydrated, while at humidities above 100% some of the hydrated nuclei acquire the possibility of unlimited growth, and cloud droplets are formed from them. If one starts from the model of condensation nuclei proposed in ([6], [7]), then the limiting radius of a hydrated nucleus can be determined from the solution of the system

$$\varphi = 1 - \frac{1}{1-\delta_0} \exp\left(-\frac{B}{r_{\text{rp}}}\right), \qquad (1-\varphi)\frac{B}{r_{\text{rp}}^2} = \frac{3r_{\text{rp}}^2\varphi^2}{aR_c^3}, \tag{1}$$

where $B = 2\sigma M/\rho RT$; $\varphi = aR_c^3/[r_{\text{rp}}^3 - R_0^3 + (1-b)R_c^3]$; $\sigma$ is the surface tension of the solution; $M$ is the molecular weight of water; $\rho$ is the density of the solution; $R$ is the universal gas constant; $T$ is the temperature; $R_0$ and $R_c$ are, respectively, the radius of the dry nucleus and the equivalent radius of its soluble part; $a$ and $b$ are certain constants, selected empirically and taking into account the ability of various substances to lower the elasticity of saturat–

ing vapor over the solution; $\delta_0$ is the magnitude of the supersaturation of the vapor over a flat water surface.

Table 1 gives (above the line) the results of calculations of the value $r_{\text{gr}}$ for nuclei whose soluble part consists of sodium chloride at different values of the parameter $G = m_c/m_0$ ($m_c$ is the mass of the soluble

**Table 1**

**Values of the limiting radius of hydrated nuclei $r_{\text{gr}}$ (in $\mu$) at supersaturation $\delta_M$ (in %) (above the line), and the size of these same nuclei at supersaturation $\delta_0 = 0$ (below the line)**

| $G$ | $\delta_M =$ 0,01 | 0,02 | 0,05 | 0,1 | 0,2 | 0,5 | 1 |
|---|---|---|---|---|---|---|---|
| 0,01 | $\frac{7,18}{4,17}$ | $\frac{3,60}{2,10}$ | $\frac{1,45}{0,861}$ | $\frac{0,730}{0,445}$ | $\frac{0,371}{0,234}$ | $\frac{0,153}{0,104}$ | $\frac{0,0794}{0,0571}$ |
| 0,05 | $\frac{7,17}{4,15}$ | $\frac{3,59}{2,08}$ | $\frac{1,44}{0,835}$ | $\frac{0,719}{0,421}$ | $\frac{0,361}{0,214}$ | $\frac{0,146}{0,0894}$ | $\frac{0,0740}{0,0471}$ |
| 0,1 | $\frac{7,17}{4,14}$ | $\frac{3,58}{2,07}$ | $\frac{1,43}{0,832}$ | $\frac{0,718}{0,418}$ | $\frac{0,359}{0,211}$ | $\frac{0,145}{0,0866}$ | $\frac{0,0729}{0,0449}$ |
| 0,5 | $\frac{7,17}{4,14}$ | $\frac{3,58}{2,07}$ | $\frac{1,43}{0,829}$ | $\frac{0,717}{0,415}$ | $\frac{0,359}{0,208}$ | $\frac{0,144}{0,0841}$ | $\frac{0,0718}{0,0426}$ |
| 1,0 | $\frac{7,17}{4,14}$ | $\frac{3,58}{2,07}$ | $\frac{1,43}{0,829}$ | $\frac{0,717}{0,415}$ | $\frac{0,359}{0,208}$ | $\frac{0,143}{0,0837}$ | $\frac{0,0717}{0,0422}$ |

| $G$ | $\delta_M =$ 0,01 | 0,02 | 0,05 | 0,1 | 0,2 | 0,5 | 1 |
|---|---|---|---|---|---|---|---|
| Equation (2) | 7,17 | 3,58 | 1,43 | 0,717 | 0,359 | 0,143 | 0,0717 |
| Equation (7) | 4,14 | 2,07 | 0,828 | 0,414 | 0,207 | 0,0828 | 0,0414 |

substance in the nucleus, and $m_0$ is the total mass of the dry nucleus). The constants $a$ and $b$ are taken from work ([8]).

For comparison, Table 1 gives values of $r_{\text{gr}}$ calculated by the simplified formula proposed in ([6],[7] ):

$$r_{\text{gr}} = 4cM/3\rho RT\delta_0 \approx 0,72 \cdot 10^{-7}/\delta_0 \text{ cm.} \tag{2}$$

From the data of Table 1 it is evident that the maximum error when using the simplified formula (2) does not exceed 10%. Taking into account that supersaturations $< 1\%$ are more probable in the atmosphere, the error in determining $r_{\text{gr}}$ (or $\delta_0$) will be still smaller.

The comparatively weak dependence of the quantity $r_{\text{gr}}$ on the content and chemical nature of the soluble substance of the nucleus, which follows from formula (2) and is confirmed by the results of the calculations presented in Table 1, makes it possible to draw the fundamental conclusion that, for atmospheric condensation nuclei containing a broad set of different chemical compounds in various proportions, at positive supersaturations ($\delta_0 > 0$) the actual magnitude of the supersaturation $\delta_0$ can be determined solely from the measured value of $r_{\text{gr}}$.

If the process of formation of the droplet-liquid phase in a cloud is considered in time, then at the initial stage the supersaturation increases, reaches a certain maximum value, and then slowly decreases ([1]–[4]). Since the growth of the supersaturation is accompanied by the formation of new droplets owing to the successive realization of less and less active nuclei, the maximum supersaturation attained in the process determines the number of droplets formed. Subsequently, as the value of $\delta_0$ decreases, the particle spectrum separates into two parts. One part corresponds to droplets that continue to grow despite the decrease in $\delta_0$, and the other to hydrated nuclei, whose size begins to decrease. In this case a gap forms in the distribution function, which broadens with time. The possibility of the existence of this type of distribution has been pointed out more than once in the literature; however, because of the difficulties of measuring particles of small size, data for this part of the spectrum are—

very scant data are available. An exception is the work ([9]), in which the results are presented of measuring particles by means of a highly sensitive photoelectric

Fig. 1. Distribution of the number of particles by size in a stratocumulus cloud, 8.XII.1961. ([9]). $r_{\text{gr}} \approx 0.55\,\mu,\ \delta_0 \approx 0.13\%$

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counter down to tenths of a micron. Measurements carried out in stratocumulus clouds (see Fig. 1) give spectra with the above-noted gap in the size interval $0.43 \div 0.55\,\mu$, which, according to equation (2), corresponds to supersaturations of 0.13—0.17%. This result is quite reasonable and agrees with theoretical ideas.

**Fig. 1.** Distribution of the number of particles by size in a stratocumulus cloud, 8.XII.1961. ([9]). $r_{\text{gr}} \approx 0.55\,\mu,\ \delta_0 \approx 0.13\%$

As already stated, the supersaturation, varying with time, reaches a maximum value $\delta_M$, and then slowly decreases. The size of the wetted nuclei that have not had the opportunity to grow into cloud droplets also decreases in this process and is determined by the supersaturation existing at the given moment. In accordance with ([6,7]), the equilibrium radius of a wetted nucleus is determined by the formula

$$r_p = \sqrt{\frac{C}{B}}\, f\left(\frac{C}{C_{\text{gr}}}\right), \tag{3}$$

where $f$ is a certain function defined in ([7]); $C$ is a characteristic of the nucleus, called the activity; $B$ is the characteristic quantity indicated above, and $C_{\text{gr}}$ characterizes the activity of nuclei which, at the value $\delta_0$, become droplets:

$$C_{\text{gr}} = \frac{4B^3}{27}\left(\frac{1-\delta_0}{\delta_0}\right)^2. \tag{4}$$

If we consider the deformation of the particle spectrum after the maximum supersaturation has been reached, then $r_{\text{gr}}$, separating nuclei from droplets, will correspond precisely to those nuclei whose activity was equal to $C_{\text{gr}}$ at the moment when the maximum supersaturation was reached. Then, taking into account that $\delta_0 \ll 1$, from (3) and (4), taking the function $f$ in explicit form, we obtain for $\delta_0 \geq 0$:

$$r_{\text{gr}}(\delta_0\delta_M) = \frac{2B}{3\sqrt{3\delta_M}}\, f\left(\frac{\delta_0^2}{\delta_M^2}\right) = \frac{B}{3\delta_M \cos\left(\pi/3 - \frac{1}{3}\arccos \delta_0/\delta_M\right)} = \frac{r_{\text{gr}}(\delta_M)}{2\cos\left(\pi/3 - \frac{1}{3}\arccos \delta_0/\delta_M\right)}. \tag{5}$$

The equation determining the equilibrium value $r_{\text{gr}}(\delta_0, \delta_M)$ can also be solved with respect to $\delta_M$:

$$\delta_M = \frac{2B\sqrt{B}}{3r_{\text{gr}}(\delta_0, \delta_M)\sqrt{3[B - \delta_0 r_{\text{gr}}(\delta_0, \delta_M)]}}. \tag{6}$$

In formulas (5) and (6) the unknown quantity is $\delta_0$—the supersaturation at the moment of measuring $r_{\text{gr}}$. However, it is easy to see that $r_{\text{gr}}(\delta_0, \delta_M)/r_{\text{gr}}(\delta_M)$ and $\delta_M$ depend rather weakly on the value of $\delta_0$, since the factor depending on $\delta_0$ varies within the limits from 1 to $1/\sqrt{3} \approx 0.58$ when $\delta_0$ changes from $\delta_M$ to zero. This means that, if $r_{\text{gr}}$ is used from direct measurements at a somewhat smaller supersaturation, then the value of the maximum supersaturation estimated by formula (2) will differ from the true value by no more than a factor of 1.73 in the direction of overestimation. Such accuracy is satisfactory, since for the time being the question can only be one of determining the order of magnitude of $\delta_M$. In principle, the accuracy of determining $\delta_M$ can be increased if the value of the limiting radius is measured at a certain

under controlled conditions when the value $\delta_0$ is known. The possibility of practical use of relations (5) and (6) is connected with the realism of the model of nuclei used. To verify the applicability of relation (5), we calculated the change in $r_{\text{gr}}(\delta_0, \delta_{\text{M}})$ as the supersaturation decreases, with a more accurate allowance for the effect of the dissolved substance on the pressure of saturated vapor over the solution at various weight contents of the soluble substance in the nucleus $G$. The results of the calculation, given in Table 1 (the underlined figures), show good agreement with the value $r_{\text{gr}}(\delta_0, \delta_{\text{M}})$ calculated from formula (5).

It should also be noted that the use of the simplified equation (2) leads to an underestimation of the values of $\delta_{\text{M}}$ by approximately 30%, while the use of formula (5) leads to an overestimation of the maximum supersaturation, so that the total error should not exceed 50%.

All this gives grounds to believe that:

1. The content of soluble substance in the nucleus and its chemical composition have practically no effect on the value of the limiting radius that determines the possibility of conversion of watered nuclei into droplets.
2. The value of the maximum supersaturation $\delta_{\text{M}}$ in the process of cloud formation can be determined from the upper boundary of the fine-dispersed part of the spectrum.
3. For estimating $\delta_{\text{M}}$, formula (2) can be used with sufficient accuracy.
4. The magnitude of the error in determining $\delta_{\text{M}}$ can be found with the aid of relations (5) and (6).

The proposed method for estimating the maximum supersaturation from the measured particle spectrum is of great importance for investigating processes occurring in clouds, since it makes it possible to dispense with data on the spectrum of nuclei, on supersaturation, and on the cooling rate of the air mass.

**Institute of Experimental Meteorology**
Obninsk, Kaluga Region

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*Note: Figure translations are in progress. See original paper for figures.*