SOME QUESTIONS IN THE THEORY OF ATMOSPHERIC CONDENSATION NUCLEI

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1966

SovietRxiv

Abstract

Full Text

UDC 551.574:551.510

GEOPHYSICS

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SOME QUESTIONS IN THE THEORY OF ATMOSPHERIC CONDENSATION NUCLEI

(Presented by Academician E. K. Fedorov on 24 XII 1965)

The formation of the droplet phase in clouds is determined to a substantial extent by the properties of condensation nuclei. In the present work, the relation is established between the distribution functions of nuclei with respect to sizes and to supersaturations; the change in the spectrum of nuclei and in the associated parameters of atmospheric aerosol with changing supersaturation is found. The question of the mechanism of droplet formation is considered, and relations are derived that determine the regular rate of droplet formation and the rate of formation caused by the fluctuation mechanism.

Analyzing numerous experiments, one may conclude that at relative humidity above 70% the condensation nuclei are already sufficiently hydrated, and the supersaturation δ over the surface of a drop of radius r may be represented in the form

$$\delta = \delta_0 - (1 - \delta_0)(Bx - Cx^3). \tag{1}$$

Here $x = 1/r$; δ_0 is the supersaturation over a plane surface; $B = 2\sigma/\rho R T \approx 1.2 \cdot 10^{-7}$ cm; C is a quantity that we call the activity of the nucleus and which, under a number of assumptions, can be expressed as

$$C = Am = br_0^{2(1+\alpha)}, \tag{2}$$

where m is the mass of the soluble part of the nucleus; r_0 is the radius of the dry nucleus; b and α are certain parameters. For nuclei with a constant ratio of soluble and insoluble parts, the parameter $\alpha = 1/2$. If this ratio is of the order of 20%, the coefficient $b = 0.25$. For nuclei with a soluble part proportional to the surface (the adsorption case), the parameter $\alpha = 0$. For completely insoluble nuclei $b = 0$, and, consequently, $C = 0$.

Fig. 1

Figure 1: Fig. 1

A visual representation of the behavior of the function $\delta(x)$ is given in Fig. 1. From the figure and from equation (1) it is easy to obtain that, for $\delta_0 > 0$, there exists a limiting value of the activity

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

The parabolas of equation (1) for $C < C_{\text{gr}}$ intersect the abscissa axis at two points x_1 and x_2, corresponding to unstable and stable equilibrium states of the nuclei. For $C > C_{\text{gr}}$, the nuclei have no equilibrium points and grow without bound. For $\delta_0 < 0$, there is only one point of stable equilibrium, existing for any C. Since nuclei with a given C cannot be smaller than a certain size, the family of parabolas is bounded in the region of small $r_0 = 1/x$ by a curve. This curve, for the model under consideration with equations (1) and (2), and under the assumption that the end value $x_3 = 1/r_0$, corresponds to the equation

$$\delta_3 = \delta_0 - (1-\delta_0)[Bx_3 - bx_3^{1-2\alpha}]. \tag{4}$$

It is not difficult to see that for $\delta_0 > 0$ a particle with the given activity C, after a sufficiently long interval of time, will either grow without bound, being to the left of the point x_1, or will have size x_2 (if $x_2 < x_3$) or x_3 (if $x_2 > x_3$). Accordingly, the spectrum of particles is divided into three parts. Particles with $C > C_{\text{gr}}$ grow without bound; particles with $C_{\text{gr}} < C < \bar{C}$ either grow without bound (if $x < x_1$) or have size x_2. For $C < \bar{C}$, the particles have size $x_3 = 1/r_0$. The boundary separating the zone of growing nuclei (drops) from nuclei in equilibrium ($x = x_2, x_3$) is the value x_{gr}

Fig. 1

$$x_{\text{gr}} = \frac{3\delta_0}{2(1-\delta_0)B} = 1/r_{\text{gr}}. \tag{5}$$

It is important to note that the limiting radius r_{gr}, separating "nuclei" and "drops," is a universal quantity, independent of the properties of the particles (C), and is determined only by the value of δ_0. The equilibrium radius $r_2 = 1/x_2$ is determined from equation (1) and can be specified in the form

$$r_2 = r_0 \quad \text{for } C < \bar{C}; \qquad r_2 = f(C/C_{\text{gr}})\sqrt{\bar{C}/B} \quad \text{for } C > \bar{C},$$

$$\frac{2}{\sqrt{3}}\, f(C/C_{\text{gr}}) = \begin{cases} \cos^{-1}\left(\pi/3 - \frac{1}{3}\arccos\sqrt{C/C_{\text{gr}}}\right), & \delta_0 > 0,\ C < C_{\text{gr}}, \\ \cos^{-1}\left(\frac{1}{3}\arccos\sqrt{C/C_{\text{gr}}}\right), & \delta_0 < 0,\ C < C_{\text{gr}}, \\ \operatorname{ch}^{-1}\left(\frac{1}{3}\operatorname{ar\,ch}\sqrt{C/C_{\text{gr}}}\right), & \delta_0 < 0,\ C > C_{\text{gr}}. \end{cases} \qquad (6)$$

For $\delta_0 < 0$, the function $f(C/C_{\text{gr}})$ can be well approximated by the following expressions:

$$f(C/C_{\text{gr}}) = 1 - 0.153\sqrt{C/C_{\text{gr}}} + 0.0192\, C/C_{\text{gr}} \quad \text{for } C/C_{\text{gr}} < 4,$$

$$f(C/C_{\text{gr}}) = \frac{3^{1/2}2^{1/3}(C/C_{\text{gr}})^{1/6}}{1 + 2^{2/3}(C/C_{\text{gr}})^{1/3}} \quad \text{for } C/C_{\text{gr}} > 4. \qquad (7)$$

To investigate the relationships between the spectrum and the humidity of the air, we take as a basis a power-law spectrum of condensation nuclei (for dry nuclei)

$$n_0(r_0)\, dr_0 = a r_0^{-\nu}\, dr_0. \qquad (8)$$

According to Junge' s data, $\nu = 4$, $a = 10^{-11}$ ([2]). Then it is easy to obtain that the spectrum of nuclei by activity is determined by the relation

$$n_C(C) = \frac{a}{2b(1+\alpha)} \left(\frac{C}{b}\right)^{-(\nu+1+2\alpha)/2(1+\alpha)} = PC^{-m}. \qquad (9)$$

Knowing the relation of r_2 to C, one can also construct the spectrum $n_r(r)$ by sizes for any value of δ_0. For example, for $\delta_0 = 0$,

$$n_r(r)\big|_{\delta=0} = \frac{a}{1+\alpha}\sqrt{\frac{B}{b}} \left[\sqrt{\frac{B}{b}}\, r\right]^{-(\nu+\alpha)/(1+\alpha)}. \qquad (10)$$

Of particular interest is the transition to spectra in terms of limiting supersaturations. Simple calculations lead to the formulas

$$n(\delta_0) = -\, n_C[C_{\text{cr}}(\delta_0)]\, \frac{dC_{\text{cr}}}{d\delta_0} = n_C\left[\frac{4B^3}{27}(\delta_0^{-1} - 1)^2\right] \frac{8B^3}{27}(\delta_0^{-1} - 1)\delta_0^{-2}. \qquad (11)$$

If relation (9) holds, we obtain (if $\delta_0 \ll 1$)

$$n(\delta_0) = 2P(4B^3/27)^{-m+1}\delta_0^{2m-3} = P_k\delta_0^k. \tag{12}$$

Here $k = (\nu - 2 - \alpha)/(1 + \alpha)$, and for $0 < \alpha < 1/2$ the exponent satisfies $2 > k > 1$ (for $\nu = 4$, i.e., for the Junge spectrum; the experiments of Twomey and Severins ([1]) give an exponent k lying within these limits; see Fig. 1 in ([1])).

The relations found between r and C and their spectra make it possible to find integral characteristics of the type $I_k = \int_0^\infty r^k n_r(r)\,dr$ (for example, for $\delta_0 < 0$)

$$I_k = 6\left(\frac{3}{4B}\right)^{k/2} C_{\text{cr}}^{k/2+1-m} P \int_\tau^\infty \frac{(4t^2-3)^{k-2m+1}(4t^2-1)}{t^{2m+1}}\,dt + \int_0^{1/x_k} r_0^k n(r_0)\,dr_0, \tag{13}$$

where

$$\tau = \cos\left(\tfrac{1}{3}\arccos\sqrt{\overline{C}}/C_{\text{cr}}\right), \quad C_{\text{cr}} > \overline{C};$$
$$\tau = \operatorname{ch}\left(\tfrac{1}{3}\operatorname{ar\,ch}\sqrt{\overline{C}}/C_{\text{cr}}\right), \quad C_{\text{cr}} < \overline{C}.$$

Using (13), one can calculate the dependence of I_k on the value of δ_0. However, the use of the approximation formulas (7) leads more quickly to quantitative results. Thus, for example, the second moment of the distribution (8)–(10), which determines visibility, changes in such a way that the meteorological visibility range

$$L = 3.9/2\pi I_2 \tag{14}$$

is equal to 0.8, 2.4, and 3.6 km at relative humidities of 99, 95, and 90%.

Such a sharp dependence of meteorological visibility range on relative humidity as it approaches 100% should lead to a significant increase in fluctuations of visibility when $\delta \to 0$. This means, in our view, that in denser hazes (humidities close to 100%) larger fluctuations of visibility should be observed than in optically less dense hazes.

On the basis of the concepts developed above, one can consider the question of droplet formation in the atmosphere. Since the appearance of new droplets is caused by the growth of δ_0, and the change in δ_0 may have a regular as well as a pulsating character, it is natural to assume the existence of two mechanisms of droplet formation. The first mechanism, caused by a regular growth of δ_0, has, generally speaking, been discussed in the literature. We shall attempt to establish analytical relations between dn_k/dt and $d\delta_0/dt$. Knowledge of the spectrum of nuclei with respect to supersaturations makes it possible to do this:

$$\Phi_1 = \frac{dn_k}{dt} = n_{\delta_0}(\delta_0)\frac{d\delta_0}{dt}\,\delta(r - r_{\text{cr}}) =$$

$$= 2P\left(\frac{4B^3}{27}\right)^{1-m}\delta_0^{2m-3}\frac{d\delta_0}{dt}\left(\delta_0 > 0;\ \frac{d\delta_0}{dt} > 0\right), \tag{15}$$

where P and m are determined from (10).

The question of the reverse transition of droplets into nuclei is more complicated. Apparently, for $\delta_0 > 0$, $d\delta_0/dt < 0$, near the boundary between the droplet and nucleus regions there is no appreciable number of droplets, since the point x_1 (see Fig. 1) is a point of unstable equilibrium. Therefore, to a first approximation one may take

$$\Phi_1 = dn_k/dt = 0 \qquad (\delta_0 > 0,\ d\delta_0/dt < 0). \tag{16}$$

Nevertheless, more detailed calculations that take into account the reverse transition into nuclei present no particular difficulty.

The second mechanism of droplet formation that we propose is due to pulsations of δ_0. The character of these pulsations, under the assumption of quasi-equilibrium between the droplets and the vapor medium, was considered in [3]. Suppose that the mean supersaturation does not increase and is maintained at some mean value $\overline{\delta}_0 = \text{const}$.

In this case, from the standpoint of the generally accepted ideas about droplet formation, the appearance of new droplets is impossible. However, the presence of turbulence leads to pulsations of supersaturation, and the possibility arises of a pulsational transfer of nuclei across the forbidden size zone. This process is in a certain sense similar to the passage of particles through a potential barrier. The reverse process of transfer into the region of nuclei is less significant (because of the unstable character of the equilibrium point x_1).

In velocity space the distribution function with respect to the velocities W, in the Lagrangian sense, satisfies the Fokker–Planck equation [4],

$$dW/dt = D\Delta_u W, \qquad \text{where } D = d\varepsilon, \tag{17}$$

d is a certain constant; ε is the rate of dissipation of turbulent energy. Then the value of the flux $\vec{j}$ of probability density in the direction of increasing u'_z, under the assumption of a normal distribution of u'_z, can be represented in the form

$$j(u'_z) = \frac{Du'_z}{\sqrt{2\pi}\,[\overline{u'^2_z}]^{3/2}}\exp\left[-u'^2_z/2\overline{u'^2_z}\right], \tag{18}$$

where $\overline{u_z'^2}$ is the root-mean-square value of the velocity of vertical pulsations. Knowledge of the flux with respect to pulsations u_z' makes it possible to calculate easily the flux with respect to supersaturation δ and, correspondingly, the rate of droplet formation Φ_2*

$$\Phi_2 = \frac{D}{\sqrt{2\pi}\,(\overline{u_z'^2})^{3/2} S} \int_{\bar{\delta}0}^{\infty} (\delta_0 - \bar{\delta}_0)\, n_{\delta_0}(\delta_0) \exp\left[-\frac{(\delta_0 - \bar{\delta}_0)^2}{4S\overline{u_z'^2}}\right] d\delta_0, \qquad (19)$$

where

$$S = c_p(\gamma_a - \gamma_a')\rho/4\pi D_p \varphi_1 L \rho_p;$$

γ_a and γ_a' are the adiabatic and moist-adiabatic gradients; ρ, ρ_p are the densities of air and vapor; D_p is the coefficient of vapor diffusion; L is the heat of condensation; c_p is the heat capacity; φ_1 is the first moment of the droplet size distribution function.

As is seen from (19), the rate of droplet formation is determined by the turbulent characteristics of the medium ε and $\overline{u_z'^2}$, the mean supersaturation $\bar{\delta}_0$, the distribution function with respect to supersaturations $n_{\delta_0}(\delta_0)$, and the quantity S. In the case of a power-law spectrum $n_{\delta_0}(\delta_0)$, the integral reduces to incomplete Γ-functions.

An estimate shows that, for the case of a Junge spectrum with $u_z = 2$ cm/sec, $\sqrt{\overline{u_z'^2}} \sim 30$ cm/sec, expression (19) gives a formation rate of the order of 4 droplets/sec-cm^3. It should be noted that in more detailed calculations it is necessary to take into account the depletion of the nucleus spectrum, which, of course, will decrease the flux Φ_2 with time.

In conclusion, we note that the introduced concept of the activity of a nucleus and its quantitative characteristic C make it possible to connect functionally a number of parameters that determine the process of condensation on atmospheric nuclei. As a result of a rational comparison of this kind of relation with experimental data, it will be possible to refine the reality of the theoretical models of condensation nuclei introduced above. The analytical determination of the rate of droplet formation is of great importance in the study of the kinetics of cloud processes.

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Received
8 XII 1965

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* Expression (19) was obtained under the assumption that a change in the sizes of nuclei entails a change in supersaturation. Estimates show that, in the interval of scales considered, the characteristic times of the pulsations amount to tens of seconds.

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