

## ON PARTICULATE FALLOUT OF POLLUTANTS FROM A LARGE-SIZED CLOUD IN A STABLY STRATIFIED ATMOSPHERE

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The paper deals with ecological problems. Failures at chemical factories and atomic power plants cause discharges of large amounts of substances that are harmful to every life form. Similarly, hazardous substances are ejected in ground tests of large solid-propellant rocket engines which are conducted in many countries. The typical feature of the above processes is that large amounts of pollutants ejected into the atmosphere in a short period of time form a cloud. The Archimedean force causes the cloud to ascend to an altitude usually called the cloud hanging altitude. The cloud contains, as a rule, liquid and solid particles (inclusions), whose sizes and masses exceed by far the sizes and masses of the gas molecules in air and of the molecules of the ejected substance. To calculate the amounts of pollutants fallen onto the ground, two theories have been proposed so far: the diffusion theory and the gravitational theory [1]. The former is developed similarly to the kinetic theory of gases, while the latter considers the motion of a particulate conglomerate as the motion of a liquid in the gravity field. The limits of applicability of either theory are usually not indicated in publications, but it is clear that there must be a parameter that would determine whether and when the above theories are applicable, depending on specific situations.

One can note that there is a parameter called the sluggishness index [2]. It is a product of the typical frequency of gas-flow turbulent pulsations and the typical time of the relative particle-velocity change in viscous (Stokes) gas flow. Although this index turns out to be an important parameter in considering, from the general standpoint, the particle motion in a turbulent medium by gravity, it does not, in fact, determine the limits of applicability of the diffusion and gravitational theories. This can be seen from the fact that free-falling acceleration does not enter the sluggishness index. The index in question determines the proximity of numerical values of the coefficients of turbulent diffusion of particles and of gaseous (liquid) medium [2].

Evidently, the parameter we need is the ratio  $v_*/u'$  ( $v_*$  is the particle-fall velocity in a laminar-flow medium and  $u'$  is the mean-square value of the gas-velocity pulsation). It characterizes the ratio of the gravity force  $F_g$  and the force  $F_t$  of viscous resistance at a randomly changing velocity of the air flow around the particle. Here  $F_g \sim d^3 \rho_p g$ ;  $F_t \sim \nu_g d \rho_g u'$ ;  $d$  is the typical particle size;  $\rho_p$  is the particle density;  $g$  is the acceleration of gravity;  $\nu_g$  is the kinematic gas viscosity; and  $\rho_g$  is the gas density.

If  $v_*/u' \gg 1$ , the particles settle on the ground by the gravity mechanism. The diffusion spread in the particle path is relatively small. If  $v_*/u' \ll 1$ , turbulent diffusion becomes the determining factor. One can easily see that the sluggishness index enters the above ratio of the velocities.

For turbulent flows, one can take the mean flow velocity  $u'$  (in this work, the mean wind velocity) instead of  $v_g$ . For sufficiently small wind velocities (for a specified  $v_*$ ), the turbulent transfer of particles is insignificant.

In this work, a somewhat peculiar form of the gravitational theory is employed: the particle motion from the cloud to the ground is regarded not as the liquid motion (for this to be the case, the particles must interact strongly) but as the motion with a specified velocity field. The physicomathematical content of the model reflects the essence of natural phenomena such as snow, rain, or hail falls. Its simplicity allows one quickly to obtain data — rough as they may be — on the pollution level of the ground surface in the case

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of still weather. This can be important when one faces a failure at a chemical factory or atomic power plant and has to make a decision.

Further considerations are based on the assumption that the ground surface under the discharged cloud of gas and particles is smooth and uniform. This allows one to assume that the atmospheric layer above the ground surface is stably stratified, provided that the wind velocities are small.

Solving the problem of the evolution of such a cloud would allow one to assess more or less exactly the actual scale of a catastrophe and take effective steps both to prevent failures and eliminate their consequences under the above conditions.

**1. Evolution of a Turbulent Large-Sized Cloud in a Stably Stratified Atmosphere.** The cloud discharged into the atmosphere in a short period of time is a mixed liquid. It spreads (collapses) under the action of propelling intrusion forces [3, 4]. If the typical time of particulate fallout is far greater than that of cloud formation and of ascent to the hanging altitude, one can assume the cloud to have ascended to the hanging altitude instantaneously. The physical mechanism of intrusion was studied experimentally in [3] and described in detail in [4]. Three pronounced stages of collapse are pointed out in [3]:

(1) the initial (significantly nonsteady) stage, in which the propelling intrusion force exceeds by far the inertial forces;

(2) the intermediate stage, in which the propelling intrusion force is counterbalanced by the shape and wave resistance; and

(3) the final stage, in which the propelling intrusion force is counterbalanced by viscous resistance.

For small-sized clouds of mixed liquid, the third stage is the most prolonged one, as was shown by experiments in [3, 5]. Here small clouds correspond to Reynolds numbers  $Re$  which are far smaller than the critical  $Re_{cr}$ . In the initial stage of collapse, the Reynolds number is determined by the equality  $Re = NV^{2/3}/\nu$ , where  $N$  is the Brunt-Väisälä frequency (for the Earth's atmosphere,  $N = 0.01 \text{ sec}^{-1}$ )

$$N = \left[ \frac{1}{\rho} \left| \frac{d\rho}{dz} \right| g \right]^{1/2};$$

$\rho$  is the air density;  $z$  is the coordinate directed vertically to the ground surface;  $V$  is the cloud volume; and  $\nu$  is the kinematic gas viscosity in the cloud.

The problem of collapse for the viscous stage under the assumption that  $Re \ll Re_{cr} = 2300$  is solved in [6]. This solution is also presented in [4], and the results of [6] are confirmed experimentally in [5]. Our prime interest is in the case where  $Re \gg Re_{cr}$ , i.e., we assume failures at chemical factories or atomic power plants to be large-scale. We also assume that for both large and small  $Re$  values, the third stage is the longest.

Under the above assumptions, the statement of the problem has a form similar to [4]:

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} r v h = 0; \quad (1.1)$$

$$-\frac{\partial}{\partial r} p h = \tau_*; \quad (1.2)$$

$$p = \rho_1 \frac{N^2}{12} h^2. \quad (1.3)$$

Here  $h$  is the half-thickness of the cloud;  $r$  is the radial coordinate;  $v$  is the gas velocity in the cloud;  $p$  is the pressure;  $\tau_*$  is the turbulent stress; and  $\rho_1$  is the cloud-gas density which is equal to the air density at the cloud hanging altitude.

Contrary to [4], we use in (1.2) the turbulent stress instead of the viscous stress. The former can be easily found from analysis of dimensions, similarly as in [4]:

$$\tau_* = \rho_1 \alpha^2 v^2 \quad (1.4)$$

( $\alpha$  is an empirical constant).

From (1.2)–(1.4) we have

$$v = \frac{N}{2\sqrt{3}\alpha} \left( -\frac{\partial h^3}{\partial r} \right)^{1/2}. \quad (1.5)$$

Substituting relation (1.5) in (1.1), we readily find

$$\left( -\frac{\partial h}{\partial r} \right)^{1/2} h^2 \frac{\partial h}{\partial t} = \frac{1}{t_*} \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial h^5}{\partial r} \right) \quad (t_* = 20\alpha/N). \quad (1.6)$$

Equation (1.5) is supplemented by the condition of conservation of volume

$$2\pi \int_0^\infty h(r, t) r dr = V = \text{const} \quad (1.7)$$

( $V$  is half the volume of the cloud) and by the condition that the solution vanishes at infinity:

$$h(\infty, t) = 0. \quad (1.8)$$

Condition (1.7) is easily justified. When the cloud is large-sized, entrainment of the surrounding gas is a weak surface effect of the order of  $S/2V \sim 1/L$  ( $S$  is the cloud-surface area and  $L$  is its typical size).

This reasoning is valid for the initial stage of collapse in which the cloud shape is nearly spherical and the velocity  $v$  is a relatively large quantity. Later  $v$  decreases. The fact that the velocity tends to diminish is due to the parabolic character of Eq. (1.5) and to the absence of source-related terms in it. Below, we demonstrate that  $v$  decreases faster than  $r_0^2(t)$  [ $r_0(t)$  is the typical cloud radius]. Thus we are justified in ignoring entrainment of the surrounding air by the cloud at the later moments of time.

A solution of the problem (1.6)–(1.8) with arbitrary initial conditions can be found only numerically. One can expect that after a little while the collapse will proceed in a self-similar regime, as in [3–5]. To find a self-similar solution, we change the variables:  $h = h_0(t)Z(\xi)$ ,  $\xi = r/r_0(t)$ ,  $h_0(t) = (9/4)^{4/9} [\sqrt[3]{V}/(2\pi)^{5/3}] (t_*/(t+t_1))^{4/9}$ , and  $r_0(t) = \sqrt[3]{2\pi V(4/9)^{2/9}} ((t+t_1)/t_*)^{2/9}$  ( $t_1$  is the conditional onset of the self-similar regime).

Substitution of the above relations in (1.6)–(1.8), brings about the following problem:

$$Z^2 \left( -\frac{dZ}{d\xi} \right)^{1/2} \left( Z + \frac{\xi}{2} \frac{dZ}{d\xi} \right) + \frac{1}{\xi^2} \frac{d}{d\xi} \left( \xi^2 \frac{dZ^5}{d\xi} \right) = 0; \quad (1.9)$$

$$\int_0^\infty Z(\xi) \xi d\xi = 1, \quad Z(\infty) = 0. \quad (1.10)$$

Analysis of Eq. (1.9) shows that for  $\xi \rightarrow 0$ ,

$$Z(\xi) = C_1 - C_2 \xi^3. \quad (1.11)$$

If we take into account the second condition in (1.10), it follows that for  $\xi \rightarrow \xi_*$ ,

$$Z(\xi) = C_3(\xi_* - \xi)^{1/3} \quad (1.12)$$

( $C_1, C_2, C_3$ , and  $\xi_*$  are the numbers).

Since (1.9) contains the square root of the first derivative of the function  $Z(\xi)$ , we assume that  $Z(\xi)$  is a monotonically decreasing function in the interval  $[0, \xi_*]$ . Bearing the above in mind and taking into account the asymptotic behavior of (1.11) and (1.12), we seek a solution of (1.9) in the form

$$Z = a(\xi_*^3 - \xi^3)^{1/3}, \quad a = \text{const}. \quad (1.13)$$

Substituting relation (1.13) into (1.9), we easily derive the algebraic equation  $(1 - 20a^{3/2})(\xi_*^3 - (3/2)\xi^3) = 0$  from which, in view of the arbitrariness of  $\xi$ , we find  $a = 20^{-2/3} \approx 0.14$ .

The first of conditions (1.10) leads to the relation

$$\frac{1}{3} a \xi_*^3 \int_0^1 (1-\eta)^{1/3} \eta^{-1/3} d\eta = \frac{a \xi_*^3}{3} B\left(\frac{2}{3}; \frac{4}{3}\right) = 1,$$

where  $\eta = \xi/\xi_*$  and  $B$  is the Euler beta-function. Expressing  $B$  through  $\Gamma$  (the Euler gamma-function), we obtain

$$\xi_*^3 = \frac{3 \cdot 20^{2/3} \Gamma(2)}{\Gamma\left(\frac{2}{3}\right) \Gamma\left(\frac{4}{3}\right)} \approx 18.3 \quad \text{or} \quad \xi_* \approx 2.63.$$

It was noted above that the gas velocity  $v$  inside the cloud decreases more rapidly than  $r_0^2(t)$ . According to (1.5),  $v \sim r/t$ . The velocity reaches its maximum at  $r = \xi_* r_0(t)$ . The entrainment of air is of the order (the pulsation component of the velocity is proportional to  $v$ )  $r_0^2 v \sim r_0^3(t)/t \sim t^{-1/3} \rightarrow 0$  for  $t \rightarrow \infty$ . Thus, a decrease in entrainment of the surrounding gas occurs faster than the change in the typical size of the cloud, and the volume-constancy condition (1.7) is justified for large moments of time, too.

The two-dimensional problem is solved similarly. In this case, the formulation of the problem in dimensional variables has the form

$$h^2 \left( -\frac{\partial h}{\partial x} \right)^{1/2} \frac{\partial h}{\partial t} = \frac{1}{t_*} \frac{\partial^2 h^5}{\partial x^2}, \quad t_* = \frac{10a}{N}, \quad \int_0^\infty h(x, t) dx = S'/2 = \text{const}, \quad h(\infty, t) = 0.$$

Here  $S'$  is half the cross-sectional area of the cloud and  $x$  is the coordinate along the ground surface which is orthogonal to the cloud's axis.

To pass to self-similar variables, we use the following relations:

$$h = h_0(t) Z(\xi), \quad \xi = \frac{x}{x_0(t)}, \quad h_0(t) = \frac{\sqrt{S'}}{\sqrt[3]{3}} \left( \frac{t_*}{t+t_1} \right)^{1/3}, \quad x_*(t) = \sqrt{S'} \sqrt[3]{3} \left( \frac{t+t_1}{t_*} \right)^{1/3}.$$

This leads to the problem

$$Z^2 \left( -\frac{dZ}{d\xi} \right)^{1/2} \left( Z + \xi \frac{dZ}{d\xi} \right) + \frac{d^2 Z^5}{d\xi^2} = 0, \quad \int_0^\infty Z(\xi) d\xi = 1, \quad Z(\infty) = 0.$$

Its solution can be written as

$$Z(\xi) = a(\xi_*^3 - \xi^3)^{1/3}, \quad a = 10^{-2/3}, \quad \xi_*^2 = \frac{3 \cdot 10^{2/3} \Gamma\left(\frac{5}{3}\right)}{\Gamma\left(\frac{4}{3}\right) \Gamma\left(\frac{1}{3}\right)}.$$

One can assess the atmospheric pollution by gaseous pollutants by employing the solution of the problem considered in this section. Recall that the theory presented is valid for small wind velocities, when the mean-square value of the pulsational velocity is far less than the velocity  $v$  of the cloud boundary.

**2. Model of Particulate Fallout of Pollutants from a Cloud to the Ground Surface.** As a rule, a cloud discharged to the atmosphere contains solid or liquid particulate pollutants, with the particle sizes changing from a few micrometers to tens of micrometers. An important quantity which can be used in estimating the level of ground-surface contamination is the amount  $\sigma(x, y)$  of pollutants fallen per unit area.

We shall first consider formula (1.13). The curve given by the formula changes little virtually in the entire interval  $[0 \leq \xi \leq \xi_*]$ . Only near  $\xi = \xi_*$  does this curve cut off abruptly. This allows one to replace (1.13) by an approximately rectangular profile with height  $h_0(t)$  and width  $\xi_* r_0(t)$ .

The change of  $\sigma$  at the point  $(x, y)$  at time  $t$  is then specified by the equation

$$d\sigma/dt = j, \tag{2.1}$$

where  $j$  is the particulate flow from the cloud  $j = \rho v_* \theta [\xi_* r_0(t) - \sqrt{(x - v_1 t)^2 + y^2}]$ ;  $\rho$  is the particle density

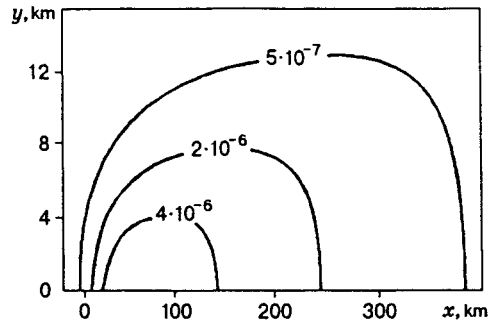


Fig. 1

in the cloud;  $v_*$  is the typical velocity of particulate fallout from the cloud which remains constant;  $\theta[z]$  is the Heaviside function

$$\theta[z] = \begin{cases} 1, & z \geq 0, \\ 0, & z < 0; \end{cases}$$

and  $v_1$  is the wind velocity at the cloud hanging altitude.

The Heaviside function fixes the projection of the cloud boundary onto the ground surface. Further,  $\rho = M/2V$  ( $M$  is the total mass of particles in the cloud).

There is an indirect assumption in (2.1) that the particles in the cloud are distributed nonuniformly. This assumption is rough but simplifies the model significantly, since it is no longer necessary to consider the particle dynamics in the cloud. For  $M$ , the equation

$$\frac{dM}{dt} = -\rho v_* S(t) = -\frac{M}{2V} v_* S(t) \quad [S(t) = \pi \xi_*^2 r_0^2(t)]$$

is valid. At  $t = 0$ , the latter equation with the initial condition  $M = M_0$  yields

$$M = M_0 \exp \left\{ -\lambda \left[ \left( \frac{t+t_1}{t_*} \right)^{13/9} - \left( \frac{t_1}{t_*} \right)^{13/9} \right] \right\}, \quad \lambda = \frac{v_* t_*}{3\sqrt{V}} (2\pi)^{5/3} \frac{9}{52} \left( \frac{4}{9} \right)^{4/9} \xi_*^2 = 17.9 \frac{v_* t_*}{3\sqrt{V}}. \quad (2.2)$$

To find  $\sigma(t, x, y)$ , we have to solve an ordinary differential equation of the first order

$$\frac{d\sigma}{dt} = \frac{v_* M}{2V} \theta \left[ \xi_* r_0(t) - \sqrt{(x - v_1 t)^2 + y^2} \right],$$

subject to the initial condition  $\sigma(t = 0) = 0$ . Here  $M$  is determined by expression (2.2).

The above problem was solved numerically by the Runge-Kutta-Merson method. The Heaviside function was approximated by the relation  $\theta[z] = (1 + e^{-\chi z})^{-1}$ , where  $1/\chi$  is the typical length, which must be sufficiently small. In calculations, the typical length was set equal to 0.1 m. According to the Stokes formula [7], the typical sedimentation velocity in the laminary-flow medium is  $v_* = (2/9)\rho_p a_p^2 g / \mu$  ( $a_p$  is the particle radius and  $\mu$  is the dynamic gas viscosity in the cloud).

In the case of a turbulent medium, the relation has to be multiplied by a coefficient which would allow for the time during which the particles are delayed in the cloud. According to Monin and Yaglom [8], this can be the turbulence intensity  $\varepsilon$  in the cloud. In our opinion, this is an oversimplified approach. The problem of choosing the right coefficient remains unsolved. In our calculations we had to use  $\varepsilon$ , yet we were fully aware of the fact that what we were doing was imprecise (or, possibly, even incorrect). The value of  $\varepsilon$  for  $Re = 10^6 - 10^7$  is 0.03–0.05 [7, 8]. As for the coefficient  $\varepsilon$ , the reasoning presented in [8] shows that the coefficient is immediately related to the turbulent-diffusion processes in the cloud. This means that the model presented herein can be extended to the cases where intrusion is not the only mechanism by which the particles scatter in space. The cloud can actively interact with the surrounding air. We have not yet studied this aspect in detail.

As an example, Fig. 1 shows the result of calculation of the surface concentration  $\sigma$  in  $\text{kg}/\text{m}^2$ . The numerical values of the physical parameters are as follows:  $\rho_p = 3010 \text{ kg}/\text{m}^3$ ,  $a_p = 7 \cdot 10^{-6} \text{ m}$ ,  $g = 9.81 \text{ m}/\text{sec}^2$ ,  $v_1 = 10 \text{ m}/\text{sec}$ ,  $\mu = 1.8 \cdot 10^{-5} \text{ kg}/(\text{m} \cdot \text{sec})$ ,  $V = 4.2 \cdot 10^9 \text{ m}^3$ ,  $\alpha = 0.3$ ,  $\varepsilon = 0.03$ , and  $M_0 = 1.8 \cdot 10^4 \text{ kg}$ .

The calculations also show that the dimensional parameters that affect  $\sigma(x, y)$  most significantly are  $v_*$  and  $v_1$  (or the dimensionless complex  $v_*/v_1$ , to be precise).

Let us make several remarks. In practical application of the model considered, one has to bear in mind that upon leaving the cloud the particles pass a distance from the cloud to the ground surface. For this reason, if one takes the origin of coordinates as a discharge point, the calculated values of  $\sigma$  must be shifted at a certain distance  $x_0$  (for  $\sigma$ , this can be accomplished by making the substitute  $x \rightarrow x + x_0$  in the equation). The distance  $x_0$  should be determined with regard for the fact that the wind velocity changes with height.

Compared with the known models, the model presented above has the advantage of being simple and having only one parameter that needs adjusting, namely the coefficient of  $v_*$ .

In this work, we assumed the ground surface to be smooth and uniform. This allowed us to ignore large-scale turbulent processes which otherwise could be significant even under the assumption that the wind velocity is small. As a result, the particle distribution would be different from  $r_0 \sim t^{2/9}$  obtained above.

Mankind has wide experience in "encountering" radioactive clouds. Unfortunately, there are no classified experimental data on their evolution from which one could deduce the law of scattering, determine the role of large-scale processes, and check the validity of the above model.

Let us note in conclusion that Berlyand [9] uses the equation of turbulent diffusion for the case of small wind velocities. As has been mentioned above, the parameter  $v_*/u'$  draws a dividing line between the diffusion and gravitational theories. The approach developed in [9] is valid for sufficiently fine particles at nonzero wind velocities.

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