DIFFUSION OF AEROSOLS IN THE SURFACE LAYER OF THE ATMOSPHERE

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Contamination of the atmosphere by harmful exhausts from industrial enterprises and transport vehicles, contamination by radioactive isotopes, the transport of pollen and seeds of plants, the controversy on bacteria, the use of poisonous chemicals in agriculture and forestry – these are some of a wide range of problems for which a knowledge of the law of turbulent diffusion and dispersal of the contaminant in the surface layer of the atmosphere is very important.

For studying these laws one uses either the semiempirical equation of turbulent diffusion (then the wind velocity and the coefficient of diffusion are given in the form of functions of spatial coordinates) or the statistical approach, in which the distribution of the impurity undergoing diffusion obeys a normal distribution law, while for determining the characteristics of the distribution some properties of the turbulent flow are used. The first approach is most fully developed in [1-4], and the second has been extensively used in [5-6]. A detailed discussion of these problems can be found in [7, 8].

Usually the final decision about using one or the other scheme of computation is taken only after comparing the computation with the experiment [9]. All theoretical estimates by both methods are obtained with an accuracy within some constant factors which are chosen with the aid of the experimental data. Both methods should be regarded essentially semiempirical, but in some cases the statistical method permits one to describe the phenomena in greater detail. Nonetheless, it is not always possible to make an unambiguous choice of the method of description.

The detailed analysis presented in [9] and a comparison of experimental results with the computations show that the currently available experimental data can be described by several existing models with an accuracy up to a factor of 2. The reliability of the values of the contaminant concentration and the density of deposition, measured in field conditions, is not better than ±50% [10].

Therefore it is sufficient to carry out the theoretical analysis of the law of diffusion of a contaminant in the surface layer of the atmosphere using the simplest model and then attempt to trace in it qualitative differences in the behavior of the diffused contaminant depending on the meteorological conditions, physical characteristics of the contaminant (primarily the rate of sedimentation), and the conditions of its injection into the atmosphere (height of the source above the ground and the duration of its operation, direction of motion relative to the wind, and so forth). According to the present state of knowledge, the coefficient of turbulent diffusion in the surface layer of the atmosphere is independent of time for time periods larger than the Lagrangian time scale [6, 8].

If the effect of molecular interaction is disregarded, then the equation describing the diffusion process coincides with the semiempirical equation of turbulent diffusion [8]

$$\frac{\partial c}{\partial t}+u\frac{\partial c}{\partial x}+v\frac{\partial c}{\partial y}-w\frac{\partial c}{\partial z}=k_x\frac{\partial^2 c}{\partial x^2}+k_y\frac{\partial^2 c}{\partial y^2}+k_z\frac{\partial^2 c}{\partial z^2}+f(x, y, z, t). \qquad (1)$$

Here c is the concentration of the diffusing substance at the point (x, y, z) at time t; u, v are the components of wind along x and y axes, respectively; w is the rate of sedimentation of the diffusing substance;

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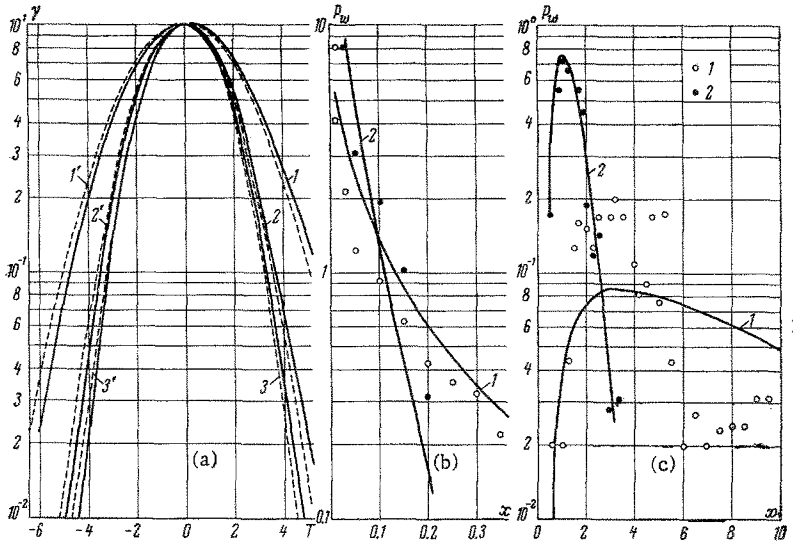

Fig. 1

k_x, k_y, k_z are the turbulent diffusion coefficients along the directions x, y, z; f (x, y, z, t) is the source function describing the operation of the generator.

In aerosol analysis the generator moves almost perpendicular to the mean wind direction (x axis). The z axis is directed vertically upward from the ground level. The origin of the coordinates lies at the point from which the generator moves. For a constant output of the generator the source function f (x, y, z, t) has the following form:

$$f(x, y, z, t) = \begin{cases} Q\delta(x)\,\delta(y - U_1 t)\,\delta(z - h) & (t \leqslant t_1) \\ 0 & (t < 0,\ t > t_1), \end{cases} \tag{2}$$

Here Q is the output of the generator (g/sec), U_1 is its speed (m/sec), δ (x) is the delta function, t_1 is the time of operation of the generator (sec), and h is the height of the source (m).

The solution of Eq. (1) for the initial and boundary conditions $c \to 0$ for x, $y \to \pm\infty$, $z \to \infty$ and

$$k_z \frac{\partial c}{\partial z} + wc = \beta c \quad \text{for} \quad z = 0, \qquad c(x, y, z, 0) = 0$$

can be obtained by the method discussed in [11]. We assume that the flux of the contaminant at the earth's surface due to turbulent diffusion is zero; then

$$(dc\,/\,dz)_{z=0} = 0 \tag{3}$$

The solution of the problem has the form

$$c(x, y, z, t) = \int_0^t d\tau \iint_{-\infty}^{\infty} d\xi\, d\eta \int_0^{\infty} f(x - \xi, y - \eta, \zeta, t - \tau)\, c_0(\xi, \eta, z; \zeta)\, d\zeta \tag{4}$$

$$c_0(\xi, \eta, z, \tau; \zeta) = {}^1/_8\,(\pi\tau)^{-3/2}(k_x k_y k_z)^{-1/2} \exp\left[-\frac{(\xi - u\tau)^2}{4k_x\tau} - \frac{(\eta - v\tau)^2}{4k_y\tau} - \frac{w(z - \zeta)}{2k_z} + \frac{w^2\tau}{4k_z}\right]\left\{\exp\frac{-(z-\zeta)^2}{4k_z\tau} + \exp\frac{-(z+\zeta)^2}{4k_z\tau}\right\} \tag{5}$$

$$-(4\pi\tau k_z)^{-1}(k_x k_y)^{-1/2} w \exp\left[\frac{(\xi - u\tau)^2}{4k_x\tau} - \frac{(\eta - v\tau)^2}{4k_y\tau} + \frac{w\xi}{k_z}\right]\Phi\left(-\frac{z + \xi + w\tau}{\sqrt{2k_z\tau}}\right)$$

Let us analyze the behavior of the concentration of the imponderable contaminant at the surface (z = 0) in greater detail. Let $\omega = z = v = 0$. The condition $v = 0$ can be fulfilled by an appropriate choice of the coordinate system. Substituting (2) and (5) in (4) and integrating with respect to ξ, η, and ζ, we get

$$c(x, y, 0, t) = q(x, y, 0, t) - q(x, y, 0, t - t_1) \tag{6}$$

$$q(x, y, 0, t) = {}^1/_4\, Q\pi^{-3/2}(k_x k_y k_z)^{-1/2} \int_0^t \tau^{-3/2} \exp\left\{-\frac{(x - u\tau)^2}{4k_x\tau} - \frac{[y - U_1(t - \tau)]^2}{4k_y\tau} - \frac{h^2}{4k_z\tau}\right\} d\tau \tag{7}$$

Since

$$\int_0^{\tau} t^{-3/2} \exp\left[-{}^1/_2 (A^2/t + C^2 t)\right] dt = \frac{\sqrt{2\pi}}{A}\left[e^{-AC}\Phi\left(-\frac{A - C\tau}{\sqrt{\tau}}\right) + e^{AC}\Phi\left(-\frac{A + C\tau}{\sqrt{\tau}}\right)\right]$$

the solution of (7) is of the form

$$q(x, y, 0, t) = \frac{Qe^B}{\pi A} 2^{-3/2}(k_x k_y k_z)^{-1/2}\left[e^{-AC}\Phi\left(-\frac{A - Ct}{\sqrt{t}}\right) + e^{AC}\Phi\left(-\frac{A + Ct}{\sqrt{t}}\right)\right] \tag{8}$$

where

$$A^2 = \frac{x^2}{2k_x} + \frac{(y - U_1 t)^2}{2k_y} + \frac{h^2}{2k_z} = A_0^2 + \frac{h^2}{2k_z}, \quad B = \frac{xu}{2k_x} - \frac{U_1(y - U_1 t)}{2k_y}, \quad C^2 = \frac{u^2}{2k_x} + \frac{U_1^2}{2k_y}, \quad \Phi(x) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{x} e^{-1/2\, t^2}\, dt.$$

If we neglect the region immediately adjoining the considered band, whose width is $l = U_1 t_1$, then the second term in formula (6) can be omitted. Then the surface concentration from the generator moving perpendicular to the wind is described by Eq. (8). For practical computations it is possible to derive and use simpler relations instead of (8), which approximate this dependence with an accuracy $\sim$10%. This is obtained in the following way. For $h = 0$ the maximum concentration is obtained for $t_m = x/u + y/U_1$, wherein at the point t_m the following relations hold:

$$B - A_0 C = 0, \quad A_0(t_m) = C\frac{x}{u}, \quad A_0'' C = \frac{u^3}{2k_x x}\left(1 + \frac{k_y u^2}{k_x U_1^2}\right)^{-1}.$$

Expanding the function $B - A_0 C$ in Taylor series around t_m and restricting the expansion to second-order terms, we get

$$B - A_0 C \approx -\frac{(t - t_m)^2}{2\sigma^2}, \quad \sigma^2 = \frac{2k_x x}{u^3}\left(1 + \frac{k_y u^2}{k_x U_1^2}\right).$$

The error introduced by replacing $A_0(t)$ by its value $A_0(t_m)$ in the neighborhood of the point $t_m \pm \sigma\sqrt{4.6}$ is close to

$$\frac{A_0'(t_m)\,\sigma\sqrt{4.6}}{A_0(t_m)} = \left[\frac{9.2\,k_x}{xu}\left(1 + \frac{k_y u^2}{k_x U_1^2}\right)^{-1}\right]^{1/2}$$

and decreases with the increase in the distance from the source (for $k_x = k_y = 5$ m^2/sec, $U_1 = 3$ m/sec, $u = 2$ m/sec at a distance of 1 km from the line of motion of the generator, the error due to this substitution is not more than 9%). It is not difficult to show that

$$\mu = \frac{A_0(t_m) - Ct_m}{\sqrt{t_m}} < 0$$

and starting from distances of a few hundred meters $|\mu| > 1$. As a consequence $\Phi(-\mu) \approx 1$. The variation of t in the range $t_m \pm \sigma\sqrt{4.6}$ does not affect the above estimates if $y > \sqrt{2k_y x u^{-1}}$. This last condition is clearly fulfilled in practice; therefore the replacement of the probability integral by unity is entirely admissible.

Since the following inequalities hold for the argument of the second probability integral in (8):

$$\frac{A_0(t_m) + Ct_m}{\sqrt{t_m}} > 0, \quad \frac{A_0(t_m) + Ct_m}{\sqrt{t_m}} \gg 1$$

the second term in formula (8) can be evaluated if we make use of the asymptotic expansion of the integral. For $(x \gg 1)$

$$\Phi(-x) \approx \frac{\exp(-{}^1/_2\, x^2)}{\sqrt{2\pi}\, x}$$

the second term is much smaller than the first for all parameters that are of interest in practice, and formulas (6) and (8) are simplified:

$$c(x, y, 0, t) \approx q(x, y, 0, t) \simeq \frac{2^{-3/2} Q}{\pi A_0(t_m)}(k_x k_y k_z)^{-1/2} \exp\frac{-(t - t_m)^2}{2\sigma^2} \tag{9}$$

Hence it follows that for given k_y/k_x and u/U_1 the curves for the variation of concentration with time are similar, if the time scale is chosen as $\sqrt{k_x x u^{-3}}$ and the concentration scale as $c(x, y, 0, t_m)$.

Computations from formula (8) for $k_x = k_y = 2k_z$ equal to 0.5, 1.5, and 5 m^2/sec, U = 1, 2, and 4 m/sec, and $U_1 = 3$ m/sec showed that independently of the distances x and y (computation carried out for x = 1, 3,

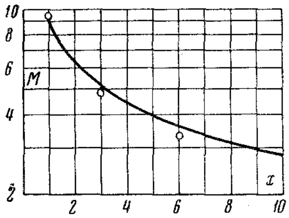

Fig. 2

5, 7, 10 km and y = 1 km) for a given wind velocity all the laws of variation of concentration in the coordinates

$$\gamma = \frac{q(x, y, 0, t)}{q(x, y, 0, t_m)}, \qquad T = \frac{t - t_m}{\sqrt{2k_x x u^{-3}}}$$

are stated by a single curve. The continuous curves 1, 2, 3 in Fig. 1a correspond to the values u = 1, 2, 4 m/sec.

The results of the computation from formula (9) for the same values of the parameters are also shown in the same figure by the dashed lines (curve 1' corresponds to curve 1 and so on). The maximum divergence between the corresponding curves for concentration equal to 0.1 of its maximum value does not exceed 15-25%. This inaccuracy is certainly compensated for by the simplicity of the computation. Adding $A'_0(t_m)(t-t_m)$ to the value $A_0(t_m)$ in formula (9), the divergence is reduced to a few percent.

The concentration of the contaminant from a high source usually differs from zero noticeably for distances equal to a few times the height. For $x \gg h$ formula (9) should be rewritten in the form

$$c(x, y, 0, t) \approx \frac{2^{-3/2} Q}{\pi A_0(t_m)} (k_x k_y k_z)^{-1/2} \exp\left(-\frac{h^2 u}{4k_z x} - \frac{(t - t_m)^2}{2\sigma^2}\right)$$

In estimating the efficiency of aerosol and aerochemical analysis perhaps the important quantity is not the concentration itself but the dose determined by the following formula:

$$D_w = \int_0^\infty c_w dt \tag{10}$$

The index w indicates that the dose D and the concentration c depend on the rate of sedimentation of the particles, i.e., on their size. The density of the sediment, which is most simply determined in experiments on scattering and turbidity of the contaminant, is linearly related to the dose by virtue of (3):

$$p_w = wD_w \tag{11}$$

For polydispersive contaminants Eqs. (10) and (11) are integrated over all sizes:

$$D = \int_0^\infty D_w f(w)\, dw, \quad p = \int_0^\infty p_w f(w)\, dw$$

Here $f(w)$ is the distribution function of the rate of sedimentation (over sizes), normalized to unity. Thus all the quantities of interest can be calculated if the expression for the dose of monodispersive aerosol is known. After substituting c_w from (4) and (5) into (10) the expression for the dose D_w is written in the following form:

$$D_w = \frac{QB_1}{4\pi^{3/2}\sqrt{k_x k_y k_z}} - \frac{QwB_2}{4\pi k_z \sqrt{k_x k_y}} \tag{12}$$

Changing the order of integration with respect to t and τ and introducing the new variable

$$\frac{y - U_1(t - \tau) - v\tau}{\sqrt{2k_y \tau}}$$

in place of t, for B_1 and B_2 we obtain the following expressions:

$$B_1 = \frac{2\sqrt{\pi k_y}}{U_1} \exp\frac{wh}{2k_z} \int_0^\infty \frac{e^{-\varphi(\tau)}}{\tau} \left[\Phi\left(\frac{y - v\tau}{\sqrt{2k_y\tau}}\right) - \Phi\left(\frac{y - l - v\tau}{\sqrt{2k_y\tau}}\right)\right] d\tau$$

$$B_2 = \frac{2\sqrt{\pi k_y}}{U_1} \exp\frac{wh}{k_z} \int_0^\infty \frac{1}{\sqrt{\tau}} \exp\frac{-(x - u\tau)^2}{4k_x\tau}\, \Phi\left(-\frac{h + w\tau}{\sqrt{2k_z\tau}}\right)\left[\Phi\left(\frac{y - v\tau}{\sqrt{2k_y\tau}}\right) - \Phi\left(\frac{y - l - v\tau}{\sqrt{2k_y\tau}}\right)\right] d\tau \tag{13}$$

$$\varphi(\tau) = \frac{(x - u\tau)^2}{4k_x\tau} + \frac{w^2\tau}{4k_z} + \frac{h^2}{4k_z\tau}$$

Here $l = U_1 t_1$ is the path traversed by the generator. In the probability integral occurring outside the braces in formula (13) the argument is negative and takes the minimum value $\sqrt{2hwk_z^{-1}}$. Its absolute value increases with the particle size and the source height. For small particles the second term in formula (12) is small, since the factor w occurs before B_2. Therefore the probability integral may be replaced by its asymptotic expansion without introducing significant errors. Taking the first term of this expansion, we obtain

$$B_2 \approx \frac{2\sqrt{k_y k_x}}{U_1} \exp \frac{wh}{2k_z} \int_0^\infty \frac{\tau e^{-\varphi(\tau)}}{h + w\tau} \frac{d\tau}{\tau} \left[\Phi\left(\frac{y - v\tau}{\sqrt{2k_y\tau}}\right) - \Phi\left(\frac{y - l - v\tau}{\sqrt{2k_y\tau}}\right) \right]$$

The integrals occurring in the expressions for B_1 and B_2 are evaluated by Laplace's method. It is easy to see that B_2 differs from B_1 by the factor

$$\frac{\tau_m k_z^{1/2}}{\pi^{1/2}(h + w\tau_m)}, \quad \tau_m = \frac{(x^2 k_z + h^2 k_x)^{1/2}}{(u^2 k_z + w^2 k_x)^{1/2}}$$

Here τ_m is the root of the equation $\varphi'(\tau) = 0$. In practice the conditions

$$(h / x)^2 \ll 1, \quad (w / u)^2 \ll 1$$

are usually satisfied fairly well.

In this case $\tau_m = x/u$, and after substituting B_1 and B_2 in formula (12), the expressions for the dose become

$$D_w \approx \frac{Q}{U_1}\left(1 + \frac{xw}{ku}\right)^{-1} (\pi x u k_z)^{-1/2} \exp\left[-\frac{h^2 u}{4k_z x}\left(1 - \frac{xw}{hu}\right)^2\right]\left[\Phi\left(\frac{y - xv/u}{\sqrt{2k_y x/u}}\right) - \Phi\left(\frac{y - l - vx/u}{\sqrt{2k_y x/u}}\right)\right] \quad (14)$$

Let us compare the results of computations by these formulas with the available experimental results. Investigations of the dispersion of aerosol wave from a strong aerosol generator have shown that more than 90% of the aerosol mass is contained in drops of less than 10 μm in diameter. Since it follows from (14) that particles of less than 10 μm diameter for a single compactness of the matter forming them are practically weightless, the cloud produced by the generator can also be regarded as weightless.

The axis of the reactive nozzle of the generator is located at a height of about two meters from the ground, but the cloud may emerge at a height of a few tens of meters due to its initial temperature being higher than that of the surrounding air. When the height at which the cloud appears does not exceed 10 m, at distances more than 1 km from the line of motion of the generator this ascent can be neglected and the source can be taken as located on the ground. In this case the dose of the imponderable contaminant must be maximum, other conditions being equal.

Figure 2 shows the dependence of the dose on the distance for an imponderable contaminant from a ground source corresponding to average conditions of the use of the generator. Plotted along the ordinate is the quantity

$$M = D_w U_1 Q^{-1} 10^3 \; ([M] - \mathrm{sec/m^2})$$

and along the abscissa is the distance in km from the line of motion of the generator and along the wind direction. The maximum doses at distances of 1, 3, 6 km measured during the experiments are shown by the dots. The agreement between the experimental results and the computation is satisfactory.

The results of computation by formula (11) for the sediment density (p_w mg/m²), reduced to a single discharge ($Q/U_1 = 1$ g/m), are presented in Figs. 1b and 1c by the continuous curves as a function of the distance (km) to the line of motion of the generator and are compared with experimental points obtained in field conditions [12, 13].

In Fig. 1b the open circles and curve 1 refer to particles with 50 μm diameter; the black circles and curve 2 refer to particles with 117 μm diameter. The value of k_z was taken equal to $0.4u_*h$ (u_* is the friction speed, roughly equal to 0.2 wind speed measured at a height of 2 m from the ground). For the experiments with a high source (h = 100 m), for which the results are presented in Fig. 1c, the notation is the same. The value $k_z = 5$ m²/sec was estimated from the position of the maximum sediment density. Considering the appreciable scatter of the results of the field measurements of the sediment density, which was computed from the number of drops deposited on glass plates placed at different distances [14], the agreement between the experimental and computational results should be considered satisfactory.

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