THEORETICAL MODEL OF THE POLLUTANTS TRANSPORT FROM THE BOUNDARY LAYER TO THE UPPER ATMOSPHERE

P.N. Belov and V.S. Komarov

M.V. Lomonosov State University, Moscow
Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
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In this paper we propose a method for calculating the pollutants transfer from the atmospheric boundary layer to the upper atmosphere. The hypothesis about a vertical profile of the pollutants in the lower part (up to about 5 km *altitude) of the atmosphere is essential for this method. The results of computations of variations in the pollutants concentration under the effect of this transport are presented.*

To compute the pollutants distribution in the atmosphere using any multilayer numerical model, the data about pollutants concentration at the initial moment of time are needed. Nevertheless, such data are unavailable and there is no hope for their obtaining in the nearest future. In this connection, this distribution at a given time moment should be presented by some model. Such a model is also needed when describing the pollutants transport within a single layer, since in this case the boundary conditions at the layer top are essential, for example, at the top of the atmospheric planetary boundary layer (PBS). In the atmosphere there always exist mechanisms for the pollutant transport from the lowest atmospheric layers to the upper ones. Thus, for example, in the emergency at the Siberian Chemical Plant in Tomsk the radionuclides emitted near the earth's surface were then detected in the upper atmosphere over Sweden.[6]

Spatial distribution of pollutants is rather complex. It is governed by a number of factors, such as a location of pollution sources, which practically everywhere are near the surface, horizontal and vertical air motions, atmospheric stratification, etc. Complications come also from the unavailability of data on pollutants concentration in the atmosphere, since measurements are normally conducted near the earth's surface. There are only few measurements of pollutants concentrations from an airplane, as well as from high towers. Shown in Figs. 1*a* and *b* are the examples of the profiles of sulfurous gas up to a 5-km altitude.[5,8,9]

These examples allow us to conclude that the vertical profile of a pollutant concentration can be described using exponential functions of the following form: $s(z) = s_0 e^{-\gamma z}$, where s_0 is the pollutant concentration near the earth's surface, γ is the parameter.

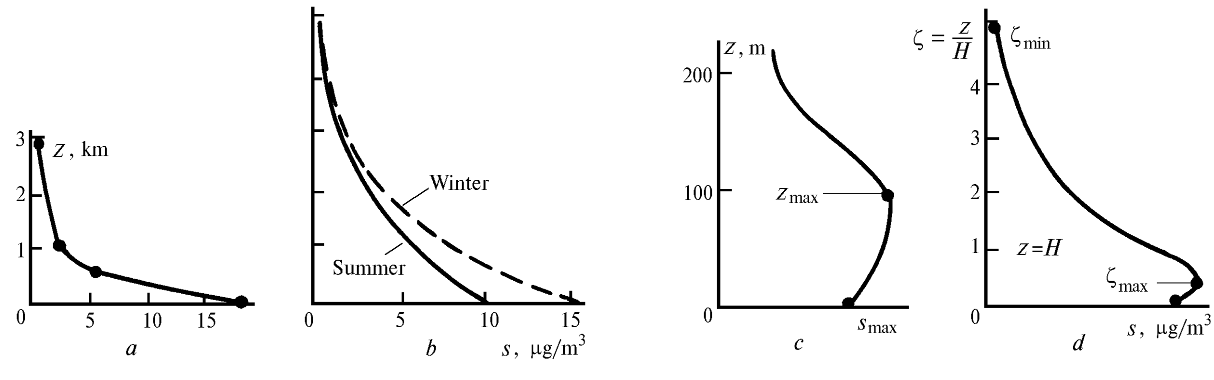

FIG. 1. Vertical profiles of the pollutants concentration: a) from the airborne measurements over the territory of Hungary,[8] *b) from the airborne measurements over the territory of Germany,*[8] *c) over Moscow,*[5] *d) calculated by the technique proposed in the form of a polynomial of the third order (*$\zeta = z/H$*, H is the height of the mixing layer,* ζ_{max} *is the relative height of the level with the maximum concentration in the PBL).*

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Such a form of the distribution can most likely be considered valid for some averaged conditions, under which all the pollution sources are exactly on the earth's surface. However, most strong emissions of pollutants occur from the stacks of industrial plants, whose mouths are at some altitude z_{max}. The layer with maximum concentration of pollutants should be at that altitude. Just in this case the turbulent transport of pollutants toward the earth's surface is possible (the term $k\,\partial s/\partial z$) in the boundary condition near the surface[3] and, consequently, the process of dry adsorption of a pollutant by the surface (the term βs, see Ref. 3).

On this basis we propose a model for the vertical profile of a pollutant concentration with the maximum at the altitude z_{max} and further decrease upward approximately following the profile of an exponential function. The polynomial of the third order:

$$s(z) = s(\zeta) = s_0 + a_1\zeta + a_2\,\zeta^2 + a_3\,\zeta^3, \qquad (1)$$

where $\zeta = z/H$, H is a given height (from here on we will take it equal to the mixing layer[7] height or the height of PBS), satisfies the above said. The coefficients a_1–a_3 from Eq. (1) are found from the following conditions:

1) The ratio of the concentration at some height H_5 (hereinafter we take $H_5 = 5H$, about 5 km) to the concentration near the earth's surface s_0, i.e. the ratio $a_s = s_{5H}/s_0$ is known. Based on the data from Ref. 8 we can assume that $a_s = 0.1$. This means that below H_5 90% of the total mass of pollutants are contained.

2) The level with maximum concentration in PBS: $z_{max}(\zeta_{max})$ is known. This level is assumed to be determined by the heights of stack mouths through which the main bulk of pollutants is emitted. Obviously, at this level $\partial s/\partial z = \partial s/\partial\zeta = 0$.

3) At the height H_5 the pollutant concentration is minimum. Consequently, at this level $\partial s/\partial\zeta = 0$ (the distribution of pollutants above this layer is not considered in this case).

Using these three conditions we can compose a system of three equations. Solving this system, we find the coefficients of Eq. (1), i.e. a_1, a_2, and a_3. In so doing the value of s_0 is not fixed, it enters into the coefficients found as a parameter.

Having determined these coefficients, using Eq. (1) it is possible to estimate the value of concentration at any level $z(\zeta)$, expressed now through an arbitrary value s_0, in particular, to estimate s_H, the concentration at the altitude $z = H$. Differentiating Eq. (1) with respect to $z(\zeta)$, we obtain the derivatives

$$\frac{\partial s}{\partial z} = \frac{1}{H}\frac{\partial s}{\partial\zeta} = \frac{1}{H}\,(a_1 + 2a_2\,\zeta + 3a_3\,\zeta^2). \qquad (2)$$

needed for the further calculations.

Having integrated Eq. (1) over z from $z = 0$ to $z = H$, we obtain the mean concentration of a pollutant in the layer 0–H:

$$\bar{s} = \frac{1}{H}\int_0^H s(z)\mathrm{d}z = s_0 + \frac{a_1}{2}\,H + \frac{a_2}{3}\,H^2 + \frac{a_3}{4}\,H^3. \qquad (3)$$

Now we can find the values of the coefficients $\alpha_0 = s_0/\bar{s}$ and $\alpha_H = s_H/\bar{s}$.

In Fig. 1*d* we present schematically the vertical profile of the pollutant concentration obtained following the above procedure.

Let us now turn our attention to a single-layer trajectory model of transport of pollutants in the atmospheric boundary layer.[1–3] In this model we consider the vertical transport of pollutants through the PBL top being at the altitude H into the upper atmosphere.

Equation for the pollutant $\bar{s}$ averaged over the PBL is written for the x axis directed along the trajectory of particles which was predetermined[2] in the form[9]:

$$\frac{\partial\bar{s}}{\partial t} + \bar{u}\,\frac{\partial\bar{s}}{\partial x} - \frac{\partial}{\partial y}\,k_y\,\frac{\partial\bar{s}}{\partial y} + \sigma\bar{s} = \bar{\varphi}, \qquad (4)$$

where $\varphi = \varphi(x, t) = F - (1/H)f$; F is the pollutant inflow into the atmosphere from the elevated sources, f is the upward pollutant inflow from the surface sources (for example, motor transport); only the sources located within the zone of particle trajectory are taken into consideration. The term σs (the bar atop is omitted) in Eq. (4) can be presented in the form:

$$\sigma\,s = \sigma_1 s + \sigma_2 s + \sigma_3 s + \sigma_4 s + \sigma_5 s + \sigma_6 s. \qquad (5)$$

The terms $\sigma_1 s$ and $\sigma_2 s$ describe the process of dry sedimentation of pollutants onto the earth's surface and their washing out from the atmosphere with precipitation, the term $\sigma_3 s$ describes the processes of chemical transformations of pollutants, and the term $\sigma_6 s$ describes the falling out of heavy pollutants. All these processes are not discussed below.

Let us consider now the terms $\sigma_4 s$ and $\sigma_5 s$ which describe the vertical transport of a pollutant through the horizontal boundary $z = H$, i.e. pollutant transport between the PBL and upper layers by the turbulent exchange and vertical motions.

When deriving Eq. (4) from the basic (unaveraged) equation of pollutant transport under boundary conditions $z = 0$ and $z = H$ (Ref. 3), the following expressions were obtained for the parameters σ_4 and σ_5:

$$\sigma_4 = \frac{1}{H}\left.\frac{\partial s}{\partial z}\right|_H k_H, \quad \sigma_5 = \frac{1}{H}\left.\frac{\partial s}{\partial z}\right|_H w_H, \qquad (6)$$

where k_H and w_H are the coefficients of turbulence and the vertical speed at the level $z = H$. By the vertical speed we mean here the ordered vertical motions determined from the general system of equations of hydrothermodynamics and the orographic motions calculated by the specialized procedure described, for example, in Ref. 1. Convective vertical motions are neglected here.

The derivative $\left.\frac{\partial s}{\partial z}\right|_H$ entering into Eq. (6) can be found using Eq. (2). In so doing, the value s_0 related to the value s by Eq. (3) enters into the derivative as a parameter.

Since at $z = H$ $\partial s/\partial z > 0$ according to Fig. 1d, then at $k_H > 0$ the concentration of a pollutant in the PBL will decrease under the influence of this factor, whereas in the upper layers it will increase, i.e., here we observe the pollutant transport aloft.

Taking into consideration the term $\sigma_5 s$ will result in a decrease in the pollutant concentration, i.e. the pollutant transport from the PBL to upper layers only with vertical motions ($w_H > 0$).

Let us now direct our attention to the solution of Eq. (4) in order to derive the formulas for calculating the changes in the pollutant concentration due to these two factors. We will consider the stationary process ($\partial s/\partial t = 0$) with the known coefficient k_y.

This equation will be solved under conditions that $u = \text{const}$, all pollution sources are along the particle trajectory (which was predetermined independently), and the conditions that

$$s \to 0 \text{ at } x \to \infty \text{ and } y \to \pm\infty, \quad s = s_0 \text{ at } x = y = 0. \quad (7)$$

The solution obtained has the form:

$$s(x, y) = P(x, y)\exp\left(-\frac{\sigma}{u}x\right)\left(s_0' + \int_0^x \exp\left(\frac{\sigma}{u}x'\right)\frac{\varphi(x')}{uP}\mathrm{d}x'\right), \quad (8)$$

where $s_0' = s_0 l_1$ (l_1 is a unit length);

$$P(x, y) = \frac{1}{\sqrt{2\pi}\,\sigma_y}\exp\left(-\frac{y^2}{2\sigma_y^2}\right), \quad (9)$$

$\sigma_y = ax$ (a is the coefficient of the order of 10^{-2}). When deriving the solution (8) it was taken that $k_y = a^2ux$.

To estimate quantitatively the contribution of these two factors, i.e. terms $\sigma_4 s$ and $\sigma_5 s$, into the pollutant transport through the boundary $z = H$, we assume in Eq. (8) that

$$\sigma = \sigma_4 + \sigma_5 = \frac{1}{H}\left.\frac{\partial s}{\partial z}\right|_H k_H + \frac{1}{H}\left.\frac{\partial s}{\partial z}\right|_H w_H$$

and that $\varphi(x') = 0$. Then, under the condition that $s = s_0$ at $x = y = 0$ at the "plume" axis ($y = 0$) the solution (8) takes the form

$$s(x) = s_0 \frac{l_1}{\sqrt{2\pi}\,\sigma_y}\exp\left(-\frac{\sigma_4}{u}x\right)\exp\left(-\frac{\sigma_5}{u}x\right). \quad (10)$$

Relative values of the decrease in the pollutant concentration in the P" L under the influence of turbulent exchange and vertical motions, calculated by Eq. (10), are presented in Table I. As follows from the table, the contribution from these two factors into the variation in the pollutant concentration at the distances from a source up to 200 km is significant. And the contribution from vertical motions exceeds that from the turbulent exchange. The decrease in the pollutant concentration in the P" L is naturally accompanied by its increase in the upper atmosphere.

TABLE I. Relative increase of the pollutant concentration in the PBL ($s(x)/s_0$, %) at different distances from a single source under the influence of the turbulent exchange (the term $\sigma_4 s$) and vertical upward motions (the term $\sigma_5 s$) (u = 10 m/s, k_H = 10 m^2/s, w_H = 5 cm/s).

x, km	36	72	196	392	784
t, h	1	2	6	12	24
$\sigma_4 s$	70	49	14	2	0.1
$\sigma_5 s$	84	70	38	16	2

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