THEORETICAL MODEL OF TRANSPORT OF POLLUTANTS IN THE ATMOSPHERIC BOUNDARY LAYER

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Received November 15, 1993

A single–layer model of transport of pollutants in the atmospheric boundary layer is considered. The model is applied to calculate the long– (up to 1000 km) and short–range (about 50 km) transport of various pollutants (regional and local models). Results of model calculations are presented and discussed for both models.

1. INTRODUCTION

As a result of industrial and vital activities of human society large amounts of gaseous (e.g., carbon monoxide CO) and solid (e.g., soot) pollutants are emitted into the atmosphere.[8,9] Evaporation and erosion cause part of industrial wastes, initially collected on the surface or spilled over various water areas, enter the atmosphere. Products of volcanic eruptions also enter the atmosphere in large volumes.

All that results in changes in the composition of the atmosphere,[7–9] which from now on includes to a larger and larger extent such pollutants of both industrial and natural origin. While in the atmosphere, pollutants react with natural atmospheric constituents like oxygen O_2, water H_2O, and so on, as well as with each other forming new substances. For example, sulfur dioxide SO_2 reacts with oxygen and then with water to form sulfuric acid. Those reactions may be described as follows:

$$SO_2 + O_2 \to SO_3 + O, \quad SO_3 + H_2O \to H_2SO_4 .$$

Some atmospheric pollutants and particularly acids, being formed in chemical reactions, are exceptionally detrimental to all the fauna and flora (fishes perish, forests dry up), ferroconcrete structural members (metal is dissolved by acids), and harmful to the health of men.

Atmospheric diffusion (turbulence) results in fast spread of pollutants around their sources, and air currents transport them at hundreds and thousands of kilometers. To undertake counter measures that would prevent undesirable consequences of atmospheric pollution, one must calculate transport of such pollutants at different distances. This problem is treated in the present paper.

2. PROPAGATION OF POLLUTANTS: PROBLEM FORMULATION

Two basic approaches to the solution of the above–indicated problem have found wide use: statistical and hydrodynamic.[1–6,11,13–15] Further we consider only the hydrodynamic approach.

The hydrodynamic approach is based on the solution of the equation of transport (balance) of a pollutant, which we consider in the form

$$\frac{\partial s}{\partial t} + u\frac{\partial s}{\partial x} + v\frac{\partial s}{\partial y} + w\frac{\partial s}{\partial z} + \frac{\partial w_a s}{\partial z} - \frac{\partial}{\partial x}k_x\frac{\partial s}{\partial x} - \frac{\partial}{\partial y}k_y\frac{\partial s}{\partial y} - \frac{\partial}{\partial z}k\frac{\partial s}{\partial z} = \varepsilon_a, \quad (1)$$

where s is the concentration of a given pollutant a; $\varepsilon_a = \varepsilon_a(x, y, z, t)$ is the source (sink) of that pollutant, that is, the rate at which it enters (vanishes from) a unit volume; k_x, k_y, and k are the eddy diffusion coefficients in the directions of the x, y, and z axes; w_a is the vertical velocity of pollutant.

In addition to the variable s Eq. (1) includes the components of air velocity u, v, and w that may be determined independently of s either by direct measurements or by solving the equations of hydrodynamics. We assume in the present study that the components u, v, and w have been already known. We also assume the function ε_a, the quantity w_a, and the coefficients k_x, k_y, and k to be prescribed. Then Eq. (1) contains only one unknown variable s. To solve that equation, one has to set the initial and boundary conditions.

We take the initial condition in the form

$$s = s^0(x, y, z) \ \text{at} \ t = 0, \quad (2)$$

where s^0 is a prescribed function of the spatial coordinates.

Equation (1) includes second derivatives with respect to the three spatial coordinates x, y, and z. Therefore, to solve it, we have to specify two conditions for each coordinate. Consider first the conditions at the side boundaries of the computational region. For simplicity we assume that Eq. (1) is integrated over the region above the horizontal surface $z = 0$ and the region of integration has the shape of a rectangular parallelepiped bounded by the planes $x = x_1$, $x = x_2$, $y = y_1$, $y = y_2$, $z = z_1 = 0$, and $z = z_2$.

We assume the concentration of the pollutant at side boundaries to be known, that is,

$$x = x_1,\ s = s_{x_1};\ x = x_2,\ s = s_{x_2};$$

$$y = y_1,\ s = s_{y_1};\ y = y_2,\ s = s_{y_2}.$$

In the particular case of integration over the infinite region we assume that the concentration of pollutant vanishes at infinity:

$$s \to 0, \ \text{for} \ x \to \pm\infty, \ y \to \pm\infty, \ z \to \pm\infty .$$

A value of pollutant concentration, its vertical eddy flux, and its vertical velocity are prescribed at the upper boundary of the region of integration by the relations

0235-6880/94/01 103-05 $02.00

$$s = s_H, \qquad k\frac{\partial s}{\partial z} = Q_H, \qquad w = w_H \text{ , for } z = z_2 = H. \tag{3}$$

The most difficult problem is prescribing the condition at the lower boundary. It is formulated depending on the properties of the underlying surface and other factors. The simplest form of the condition at the lower boundary is obtained for the water surface. It completely absorbs a pollutant reaching it. This property may be described as follows:

$$s = 0, \text{ for } z = z_1 = 0.$$

In the case of a solid underlying surface two opposite processes are simultaneously observed: absorption and reflection of a pollutant. Consider first the case $w_a = 0$. Then, provided the earth's surface totally reflects the pollutant, the condition will be written in the form

$$k_z \frac{\partial s}{\partial z} = 0 \text{ , for } z = 0.$$

When the earth's surface only partly reflects the pollutant reaching it, a part of that pollutant must be absorbed (in the process of "dry absorption"). That part of pollutant is equal to βs, where β is the coefficient accounting for this process ("accommodation" coefficient). In this case the boundary condition assumes the form

$$k\, z \frac{\partial s}{\partial z} - \beta\, s = 0 \text{ , for } z = 0.$$

Now we consider the general case in which a heavy pollutant sediments ($w_a < 0$), and there exists the surface source of pollutant $f_0\,(x, y)$. The boundary conditions for that case are written in the form

$$k_z \frac{\partial s}{\partial z} - w_a\, s_0 = \beta\, s_0 - f(x, y) \text{ , for } z = 0 \text{ ,} \tag{4}$$

where s_0 is the value of s for $z = 0$.

The equation of transport of pollutant (1) is nonlinear. Therefore, it can be solved by numerical techniques.[3] As a result of such numerical integration, we obtain a spatiotemporal distribution of concentration of the examined pollutant $s(x, y, z, t)$. In addition, the flux of pollutant from the atmosphere to the surface can be obtained

$$Q_0 = k \frac{\partial s}{\partial z} - w_a\, s = \beta\, s. \tag{5}$$

If several pollutants are simultaneously presented in the atmosphere, then the equation of transport is integrated separately for each pollutant.

3. EQUATION OF POLLUTANT TRANSPORT FOR THE ATMOSPHERIC BOUNDARY LAYER

The major part of various atmospheric pollutants (about 70–90% of them) is concentrated in the planetary boundary layer (PBL) whose height $z = H$ is about 1 km. At the same time, it is its lower part, the internal (surface) boundary layer (IBL), whose height $z = H_1$ is about 30–100 m, in which the vital activity of human society takes place. Food is produced in the upper layer of soil and in the air layer adjacent to it. That is why most studies dedicated to ecological problems consider the PBL.

Below we suggest a special model of transport and computational techniques for the calculation of the concentration of pollutants contained in that layer. First we derive the equation of transport of pollutants in the PBL.

We introduce the parameters averaged over the altitude from $z = 0$ to $z = H$

$$\overline{s} = \frac{1}{H}\int\limits_0^H s(z)\,\mathrm{d}z, \quad \overline{u} = \frac{1}{H}\int\limits_0^H u(z)\,\mathrm{d}z, \text{ and so on .}$$

Let us now integrate Eq. (1) over the altitude with boundary conditions (3) and (4). As a result, we find for the average parameters

$$\frac{\partial \overline{s}}{\partial t} + \overline{u}\,\frac{\partial \overline{s}}{\partial x} + \overline{v}\,\frac{\partial \overline{s}}{\partial y}\,\frac{1}{H}\,w_H\,(s_H - \overline{s}) + \frac{1}{H}\,w_a\,s_H -$$

$$-\frac{1}{H^2}\,k_H\,(s_H - s_0) - \frac{\partial}{\partial x}\,k_x\,\frac{\partial \overline{s}}{\partial x} - \frac{\partial}{\partial y}\,k_y\,\frac{\partial \overline{s}}{\partial y} + \frac{\beta}{H}\,s_0 = \overline{\varepsilon}_a + \frac{1}{H}\,f_0.$$

We then introduce semiempirical coefficients

$$\alpha_0 = s_0 \,/\, \overline{s}, \quad \alpha_H = s_H \,/\, \overline{s}$$

and on their account rewrite the last equation in the form

$$\frac{\partial \overline{s}}{\partial t} + \overline{u}\,\frac{\partial \overline{s}}{\partial x} + \overline{v}\,\frac{\partial \overline{s}}{\partial y} - \frac{1}{H}\,w_H\,(1 - \alpha_H)\overline{s} + \frac{1}{H}\,\alpha_H\,w_a\,\overline{s} +$$

$$+\frac{\alpha_0 - \alpha_H}{H^2}\,k_H\,\overline{s} - \frac{\partial}{\partial x}\,k_x\,\frac{\partial \overline{s}}{\partial x} - \frac{\partial}{\partial y}\,k_y\,\frac{\partial \overline{s}}{\partial y} + \frac{\alpha_0\beta}{H}\,\overline{s} = \overline{\varepsilon}_a + \frac{1}{H}\,f_0. \tag{6}$$

The function ε_a entering the last equation is determined by the strength of atmospheric emissions from various sources (e.g., stacks of industrial complexes) F, washing out of pollutants by atmospheric precipitation W, and decomposition of the given pollutant in chemical reactions R. Therefore, we may write

$$\varepsilon_a = F - W - R\,. \tag{7}$$

In their turn, these functions are assumed to have the form

$$W = -\,\sigma_2\,s\,, \quad R = -\,\sigma_3\,s, \tag{8}$$

where σ_2 and σ_3 are the semiempirical coefficients.

The parameter σ_3 is related to the intensity of precipitation I (mm/h) via the expression $\sigma_3 = \alpha^* I$, where α^* is constant. It is assumed for sulfur and nitrogen oxides that $\sigma_2 = 0.01\ \mathrm{h}^{-1}$. When sulfur dioxide changes to the sulfate SO_4^{2-} ion, we have $\sigma_3 = 0.05\ \mathrm{h}^{-1}$.

Alternatively, Eq. (6) may be rewritten in the form

$$\frac{\partial \overline{s}}{\partial t} + \overline{u}\,\frac{\partial \overline{s}}{\partial x} + \overline{v}\,\frac{\partial \overline{s}}{\partial y} + \sigma\,\overline{s} - \frac{\partial}{\partial x}\,k_x\,\frac{\partial \overline{s}}{\partial x} - \frac{\partial}{\partial y}\,k_y\,\frac{\partial \overline{s}}{\partial y} = \varphi, \tag{9}$$

where

$$\varphi=\varphi(x,\ y,\ t)=F-\frac{1}{H}f_0,\ \sigma=\sigma_1+\sigma_2+\sigma_3+\sigma_4+\sigma_5+\sigma_6,$$

$$\sigma_1=\frac{\alpha_0\beta}{H},\ \sigma_4=-\frac{1-\alpha_H}{H}w_H,\ \sigma_5=-\frac{\alpha_H}{H}w_a,\ \sigma_6=\frac{\alpha_0-\alpha_H}{H^2}k_H\,. \quad (10)$$

4. REGIONAL SINGLE–LAYER TRAJECTORY MODEL

We now introduce the so–called natural coordinates, and first define the trajectory of a particle motion in the PBL down the average wind. The x axis follows the direction of motion at each point of trajectory, while the y axis is normal to it. We may denote the new coordinates by l and m, respectively. However, for clarity we shall use the old notation, assuming at each point of the trajectory that $x=l$, $y=m$, and $\overline{u}=\overline{u}_l$, $\overline{v}=0$.

Comparing the orders of magnitude of the second and fifth terms in the left–hand side of Eq. (9), we may conclude that the latter is small. Keeping that in mind and recalling that $\overline{v}=0$, Eq. (9) may be rewritten in a shorter form (below we omit the bar atop the symbols)

$$\frac{\partial s}{\partial t}+u\frac{\partial s}{\partial x}+\sigma s-\frac{\partial}{\partial y}k_y\frac{\partial s}{\partial y}=\varphi \quad (11)$$

or

$$\frac{\partial_r s}{\partial t}+\sigma s-\frac{\partial}{\partial y}k_y\frac{\partial s}{\partial y}=\varphi\ , \quad (12)$$

where $\frac{\partial_r s}{\partial t}=\frac{\partial s}{\partial t}+u\frac{\partial s}{\partial x}$ is the path derivative for particle moving along the trajectory.

Following Berlyand,[6] we assume for the eddy diffusion coefficient describing transverse pulsations

$$k_y=k_0\,u, \quad (13)$$

where k_0 is a coefficient depending on the atmospheric stratification. Then we obtain, instead of Eq. (12)

$$\partial_r s/dt+\sigma s-k_0\partial^2 s/\partial y^2=\varphi. \quad (14)$$

Further we rewrite expression (14) in the form

$$s(x,\ y,\ t)=s'(x,\ t)\ P(x,\ y)\ . \quad (15)$$

Separating the variables for the functions P and s', we obtain

$$\frac{\partial P}{\partial x}-k_0\frac{\partial^2 P}{\partial y^2}=0, \quad (16)$$

$$\frac{\partial_r s}{\partial t}+\sigma s'=\varphi/P. \quad (17)$$

We shall solve these two equations for isolated sources of pollutants that may be described by the Dirac δ–function.

Eq. (16) is solved with the following boundary conditions:

$$P=\delta(y)\quad \text{for}\quad x=0,\quad \text{and}\quad P\to 0\quad \text{for}\quad y\to\pm\infty\ . \quad (18)$$

We then have with these conditions

$$P(x,\ y)=1/\sqrt{2\pi k_0\,x}\ \exp(-\,y^2/4\,k_0\,x)\ . \quad (19)$$

The integration of Eq. (17) we make along the known trajectory, with the time step Δt and the step Δx along the x axis. Applying the explicit scheme in the step with number n, we have

$$s'^{(n)}=(1-\sigma\Delta t)s'^{(n-1)}+\Delta t(\varphi/P)^{(n-1)}, \quad (20)$$

where the superscript $(n-1)$ refers to the previous step of integration.

The height of the PBL H may be determined based on the theory of the boundary layer.[3,12] In the simplest case it is assumed that

$$H=\sqrt{k/l},$$

where $l=2\omega\sin\varphi$ is the Coriolis parameter after Panovskii.[12] Then we have

$$H=0.2\ u_*/l,$$

where $u_*=\kappa V/\ln z/z_0$, $\kappa=0.4$, V is the wind speed, and z_0 is the parameter of surface roughness.

Sometimes the height of the mixed layer[10] (HML) $k_{1\mathrm{M}}/l$, where $k_{1\mathrm{M}}$ is the eddy diffusion coefficient at an altitude of 1 m, is taken instead of the PBL height.

According to various estimates, the values of H and HML are close and vary from 1500 m or higher in the summer daytime to 400 m or lower in the winter nighttime.

Particle trajectories are calculated step by step from the expressions

$$\frac{\partial x}{\partial t}=\overline{u}\ (x,\ y,\ t),\quad \frac{\partial y}{\partial t}=\overline{v}(x,\ y,\ t)$$

using the values of wind velocity at the grid nodes and following a numerical technique.

Wind velocities at grid nodes, averaged over H, are calculated from the values of pressure at sea level and at a pressure altitude of 850 hPa, based on the theory of boundary layer.[4]

The model was used to calculate the spread of pollutant from various point sources for up to 24 hours and its average daily concentration at various trajectory points.

TABLE I. Pollutant concentration (μg/m^3) *vs. the length of the trajectory of motion l from a single source and the normal distance m from the trajectory at the instant t.*

t, h	l, km	m, meters			
		0	100	500	1000
0	0	100	–	–	–
2	70	0.142	0.137	0.058	0.004
6	201	0.074	0.073	0.055	0.021
12	434	0.042	0.042	0.037	0.024
18	690	0.028	0.028	0.025	0.019
24	997	0.020	0.019	0.018	0.015

Note: $s^0=100$ μg/m^3, $H=1000$ m, $\beta=1$ cm/s, and $\sigma=0.88\cdot 10^{-5}$ s^{-1}.

By way of example, Table I lists the results of calculations for individual source producing a value of mass concentration of 100 μg/m^3 at the initial trajectory point. Such data may be referred to any pollutant, e.g., to sulfur dioxide emitted by the Noril'sk metallurgical plant or to radioactive products of a nuclear explosion. Those two situations will only differ in strengths of their sources and trajectories of pollutant transport as well as in the parameters of the models.

One can see from that table, how the pollutant mass concentration changes while moving along the trajectory on the "jet axis" and across it.

For example, 24 hours later the mass concentration at a distance of 997 km from the source dropped to 0.02 μg/m^3, being equal to 0.02% of its initial value. It is also interesting to note that, due to cross–diffusion, mass concentration of the substance 24 hours later at a distance of 1 km from the jet axis is no longer different from its axial value.

5. LOCAL MODEL OF THE POLLUTANT TRANSPORT IN THE INTERNAL (SURFACE) LAYER

There exists in the lower part of the planetary boundary layer the so–called internal surface layer (ISL) whose depth is about 30–100 m. The theory of surface boundary layer holds that wind direction in it remains unchanged, and the wind speed is expressed by relation

$$u(z) = (u_*/\kappa) \ln (z + z_0)/z_0,$$

where $\kappa = 0.4$, z_0 is the parameter of roughness, $u_* = l\mathrm{d}u/\mathrm{d}z$ is the dynamic wind speed, $l = \kappa\,(z + z_0)$ is the mixing length (for stratifications close to neutral), and $k = l^2\,\mathrm{d}u/\mathrm{d}z$ is the eddy diffusion coefficient.

When the wind speed is known at some level z_2 (e.g., $z_2 = z_{10} = 10$ m is the level of wind vane), we have

$$u(z) = u_{10} \ln [(z + z_0)/z_0] \;/\; \ln [(z_{10} + z_0)/z_0]\ ,$$

$$k(z) = k_0 + az,\ k_0 = az_0,\ a = \kappa^2 u_{10} \ln [(z + z_0)/z_0]. \qquad (21)$$

According to Ref. 10, the depth of the IBL is

$$H_1 = H_{\mathrm{IBL}} = \frac{k_1}{10\,l}\,,$$

where k_1 is the eddy diffusion coefficient at an altitude of 1 m, which depends on the class of stability of stratification. Note that $k_1 \sim 0.1$ m^2/s for the daytime (neutral) stratification, while $k_1 \sim 0.05$ m^2/s for the nighttime (stable) stratification. To give an example, at $\varphi = 70°$, $l = 0.139{\cdot}10^{-3}$ s^{-1}, and $k_1 = 0.1$ m^2/s we find $H_1 = 72$ m. More accurate characteristics of the IBL may be found on the basis of the Monin–Obukhov theory.

We consider below a mesoscale region with the horizontal dimension $L \sim 50$ km. Wind direction in such a region may be assumed constant.

We direct the x axis along the wind and integrate the equation of transport from z_0 to H_1. We introduce the parameters

$$\overline{s} = \frac{1}{H_1}\int\limits_{z_0}^{H_1} s(z)\,\mathrm{d}z,\quad \overline{u} = \frac{1}{H_1}\int\limits_{z_0}^{H_1} u(z)\,\mathrm{d}z$$

averaged over the IBL.

Integrating the expression for $u(z)$, we find for the average speed

$$\overline{u} = u_{10} \ln \frac{H_1 + z_0}{z_0} \Big/ \ln \frac{z_{10} + z_0}{z_0}, \qquad (22)$$

where u_{10} is the wind speed at the vane level. Then in analogy with the regional model we obtain the equation for the average parameters of the IBL similar to Eq. (14), which is transformed, via substitution of Eq. (15), into Eqs. (16) and (17) for the functions P and s'.

For individual sources described by the δ–function, the solution of equation for the function P is expressed in the form of Eq. (19).

Now, provided the direction of wind remains constant, the solution of the equation for variable s' is found in analytic form.

We consider the stationary case ($\partial s/\partial t = 0$) first. Then the equation for s' assumes the form

$$\frac{\partial s'}{\partial x} + \frac{\sigma}{\overline{u}} = \frac{\varphi(x)}{\overline{u}(P)}.$$

For $x = 0$ and $s' = s^{0\prime} = s^0/P$ its solution has the form

$$s'(x) = \exp(-\sigma x/\overline{u})\left[s^{0\prime} + \int\limits_0^x \exp(\sigma x'/\overline{u})\,\frac{\varphi(x')}{\overline{u}P}\,dx'\right]. \qquad (23)$$

The solution for the nonstationary case is reduced to that for the stationary case by way of introducing the new variables

$$x_1 = x - u\,t, \qquad t_1 = t.$$

The corresponding solution for s' will be written in the form

$$s'(t_1) = \exp(-\sigma t_1)\left[s^{0\prime} + \int\limits_0^{t_1} \exp(\sigma t_1)\,\varphi(t_1')\,dt_1'\right].$$

Solution (23) on account of Eqs. (19) and (15) was used to calculate the mass concentration of reference pollutant (e.g., sulfur dioxide) when several ground–based sources were present on the trajectory. We assumed $s_0 = 0$ for $x = 0$, that is, at the origin of coordinates.

An example of such computational results is given in Table II.

TABLE II. Pollutant concentration (μg/m^3) *vs. the distance x from the origin of coordinates and the distance y from the jet axis for three ground–based point sources.*

x, km	f_0, μg/m^2·s	y, m				
		0	50	100	300	500
0	0	–	–	–	–	–
6	500	15.10	13.54	9.91	0.35	0
12	0	8.88	8.43	7.21	1.36	0.49
16	400	13.13	12.63	11.23	3.22	0.26
18	0	11.66	11.26	10.15	5.34	0.36
20	500	12.47	16.93	15.42	5.67	0.77
32	0	9.63	9.45	8.91	4.77	1.37
50	0	4.49	4.47	4.27	2.86	1.27

Note: $s^0 = 0$, $H_1 = 100$ m, $\beta = 1$ cm/s, $\overline{u} = 10$ m/s, and $z_0 = 10$ cm.

It follows from the table that when air moves over the ground–based sources, the mass concentration of pollutant sharply increases. For example, the mass concentration of pollutant on the jet axis reached 15 μg/m^3 above the first ground–based source whose strength was 500 μg/m^3 (a small heat–and–power station), and 17.5 μg/m^3 above the third ground–based source. The concentration quickly decreases with distance from the sources. At a distance of 30 km from the last source mass concentration dropped to 4.5 μg/m^3. It is seen from Table II that although mass concentration off the jet axis becomes much lower, it remains noticeable.

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