REGIONAL MODEL OF TRANSPORT OF CONSERVATIVE IMPURITIES IN THE ATMOSPHERE

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A regional mathematical model of pollution transport in the atmosphere is described at length. The model is based on the equation of conservative impurity transport in a turbulent medium. The schemes of parametrization of humid washout, sedimentation, and self–induced vertical lift are considered. The model is constructed numerically using the method of separation for physical processes. At the stage of advection the scheme that does not increase the total variation is used. Advantages of the model allow its use in the problems of regional ecology.

Mathematical models which describe propagation of gaseous and aerosol impurities in the atmosphere have found a wide application to the solution of the problems of atmospheric physics, environmental protection, and forecasting the level of atmospheric air pollution. The semiempirical equation of turbulent diffusion, as a rule, provides the basis for these models. The overwhelming majority of the mathematical models of impurity transport are kinematic models since the components of the wind velocity vector and the other meteorological characteristics, which are the parameters of transport models, are determined from the measurement results or are calculated using an atmospheric prognostic model. In view of the fact that most anthropogenic impurities are concentrated in the lower two–kilometer layer of the atmosphere the mathematical models of transport of impurities by air are generally constructed for the boundary layer of the atmosphere. However, when aerosols that absorb short–wave solar radiation very intensively (smoke, dust, soot) are discharged into the atmosphere, the mathematical models of transport must describe physical processes occurring over the entire depths of the troposphere and lower stratosphere. This can be caused by the tendency of strongly absorbing aerosol to self–induced vertical lift.[1]

This paper describes a three–dimensional nonstationary regional model of impurity transport in the atmosphere. It represents one block of the ecological–economic model of the region. Impurities are assumed to be multicomponent and chemically inert.

Formulation of the problem. The processes of propagation of pollutants in the atmosphere are described by the transport equation with allowance for turbulent exchange, sedimentation, humid washout, and exchange processes between the atmosphere and the underlying surface

$$\frac{\partial \mathbf{c}}{\partial t} + \mathbf{U}\,\mathrm{grad}\,\mathbf{c} - \mathrm{div}_s(\mu\,\mathrm{grad}_s\,\mathbf{c}) - \frac{\partial}{\partial p} g^2 \rho^2 \nu \frac{\partial \mathbf{c}}{\partial p} + \frac{1}{\rho}(\mathbf{I}_c + \mathbf{I}_c^*) = \mathbf{f}, \quad (1)$$

where $\mathbf{c} = \{c_i\ (i = \overline{1, N})\}$ is the vector of normalized concentration of an impurity; c_i is the normalized concentration of one of its components; $\mathbf{U} = (u, v, w)$ is the vector of wind velocity in the coordinate system (x, y, p); μ and ν are the horizontal and vertical coefficients of turbulent diffusion; g is the acceleration of free fall; ρ is the air density; $\mathbf{I}_c$ and $\mathbf{I}_c^*$ are the vector functions, which take into account aerosol generation and sink due to transformation processes; and, $\mathbf{f}$ is the vector–function of the source. The subscript s denotes the operators of gradient and divergence in horizontal directions.

The components of the velocity vector, the coefficients of turbulent exchange, and the air density are the input parameters of the model. They are assigned based on the measurement results or are calculated using any prognostic model.

The tropopause, being a thick barrier layer, can be considered as the upper boundary for the transport model for majority of natural and anthropogenetic aerosols. Then the corresponding boundary condition is written as[2]

$$g\rho\nu \left.\frac{\partial c_i}{\partial p}\right|_{p = p_{\text{T}}} = k_{ci}\left(c_i\big|_{p=p_{\text{T}}} - c_i^*\right), \quad i = \overline{1, N}, \quad (2)$$

where k_{ci} $(i = \overline{1, N})$ are the exchange coefficients; c_i^* is the concentration of the ith impurity in the stratosphere; p_{T} is the isobaric level that is conditionally assumed to be a tropopause level.

The expression (2), due to weak convective air flow near the tropopause, is sufficiently good approximation for an impurity flux to the stratosphere.

The atmosphere –– underlying surface interaction in the formalized representation can be described by the formula[3]

$$g\rho\nu \left.\frac{\partial c_i}{\partial p}\right|_{p = p_s} = k_{si}\left(c_{si} - c_i\big|_{p = p_s}\right) + Q_{si}, \quad i = \overline{1, N}, \quad (3)$$

where c_{si} is the ith impurity concentration above the underlying surface which provides a balance in an exchange process between the underlying surface and the atmosphere; $c_i\big|_{p=p_s}$ is the ith impurity concentration in air over the underlying surface; k_{si} are the turbulent exchange coefficients which are the functions of a soil type, vegetation cover, and temperature; and, Q_{si} is the source of the ith impurity located on the underlying surface.

The initial conditions for unambiguous solution of Eq. (1) are formalized as follows:

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$$\mathbf{c}(x, y, p, 0) = \mathbf{c}_0(x, y, p), \tag{4}$$

where $\mathbf{c}_0$ is the given vector –function.

Method of solution. The impurity transport equation (1) with the given boundary [Eqs. (2) and (3)] and initial [Eq. (4)] conditions can be solved using numerical methods, if the components of the wind velocity vector, the turbulent exchange coefficients, and the aerosol sinks and sources are known.

The problem of impurity transport in the atmosphere is classified among the problems centered around the solution of the hyperbolic–type equations. This solution becomes discontinuous at a certain moment of time (or it can be smooth but with large spatial gradient). The so–called monotonic numerical schemes[4] are used to solve the equations which describe evolution of positive functions. However, there are no monotonic schemes with the spatial approximation order higher than the first order. Therefore, such schemes possess strong diffusion, the unknown function profiles are too broad, and the accurate solutions can be obtained only using an extremely fine–structure grids.

We can improve the accuracy without loss of rigourous theoretical substantiation replacing the condition of monotony by the condition of nonincreasing total variation.[5] The total variation of numerical solution denoted as $TV(c^n)$ is defined as follows:

$$TV(c^n) = \sum_{i=-\infty}^{\infty} \left| c^n_{i+1} - c^n_i \right| , \tag{5}$$

where n is the serial number of a time step, and i is the serial number of a grid node.

Therefore, the numerical scheme is the TVD (total variation diminishing) scheme, when

$$TV(c^{n+1}) \le TV(c^n). \tag{6}$$

The TVD schemes do not result in formation of nonphysical oscillations and enable one to attain the second (and higher) order of accuracy in the regions of smooth variations of the solution.

For the one–dimensional equation

$$\partial c/\partial t + u\,(\partial c/\partial x) = 0$$

the TVD scheme can have the form

$$c^{n+1}_i = c^n_i - u\frac{\Delta t}{\Delta x}(c^n_i - c^n_{i-1}) - (f^n_{i+1/2} - f^n_{i-1/2}), \tag{7}$$

where $f^n_{i+1/2} = \varphi(r_i)\left(u\frac{\Delta t}{2\Delta x}\right)\left(1 - u\frac{\Delta t}{\Delta x}\right)(c^n_{i+1} - c^n_i)$, and $f^n_{i-1/2}$ is given by the analogous expression. The function $\varphi(r_i)$ is called a finitary function. Its parameter r_i is found from the gradient relation

$$r_i = (c_i - c_{i-1}) / (c_{i+1} - c_i).$$

The finitary function $\varphi(r_i)$ is chosen so that scheme (7) could satisfy conditions (6). In Ref. 6 the author offers the following type of finitary function:

$$\varphi(r) = \begin{cases} \min(2, r), & \text{for } r > 1, \\ \min(2r, 1), & \text{for } 0 < r \le 1, \\ 0, & \text{for } r \le 0. \end{cases} \tag{8}$$

The scheme (7) is single–step (in contrast with, for example, flux correction). In this case the finitary function has a sufficiently simple structure that makes the numerical scheme efficient.

Equation (1) is solved by the method of separation for physical processes. The equation of advection, the equation of turbulent diffusion, and the equation which describes transformation processes are solved at the first, second, and third stages, respectively.

The numerical TVD scheme is used at the stage of advection, and an implicit one is used at the stage of turbulent exchange.

The numerical scheme is realized using a grid with horizontal dimensions 40×40 with a 100 km step. A vertical step of the grid changes from 150 m in the boundary layer to 500 m in the free atmosphere.

A hydrometeorological regime can be specified either on the basis of real information from the available data banks or using a regional numerical prognostic model of the atmosphere.

Parametrization of self–induced vertical lift of aerosol. The aerosol strongly absorbing solar radiation (smoke, dust, soot) is known to be capable of self–induced vertical lift.[1] To estimate the velocity of vertical self–induced lift, i.e., to take into account this mechanism parametrically in the model, one must consider a stationary process of volumetric horizontally inhomogeneous heat release in a rotating stratified medium. Confining our attention to Boussinesq and hydrostatic approximations we write down a system of equations of hydrodynamics, continuity, and heat influx in the form

$$\frac{\partial\Phi}{\partial x} = \left(\nu\frac{\partial^2}{\partial z^2} + \Delta\right)u + l\,\upsilon, \quad \frac{\partial\Phi}{\partial y} = \left(\nu\frac{\partial^2}{\partial z^2} + \Delta\right)u - l\,u,$$

$$\frac{\partial\Phi}{\partial z} = g\alpha^*\theta, \quad \frac{\partial u}{\partial x} + \frac{\partial \mathrm{u}}{\partial y} + \frac{\partial w}{\partial z} = 0, \quad \Gamma w = \left(\nu\frac{\partial^2}{\partial z^2} + \Delta\right)\theta + Q, \tag{9}$$

where l is the Coriolis parameter, α^* is the coefficient of thermal expansion, θ is the potential temperature perturbation, $\Phi = p'/\bar{\rho}$ is the ratio of the pressure perturbation to background density, Γ is the variation of the potential temperature gradient from an equilibrium one, and Q is the given rate of heat release.

The system of equations (9) is considered within a horizontally infinite atmospheric layer $0 < z < H$ which is uniformly occupied by aerosol strongly absorbing solar radiation.

Let the optical density of turbid air and the solar zenith distance allow one to approximate the heat source by the expression

$$Q = Q_0 \sin\frac{\pi}{H} z, \tag{10}$$

where Q_0 is the maximum rate of heat release at $z = H/2$. It should be noted that the form of the approximate expression does not affect the final result.

Consider the limiting case in which horizontal variation of heat release is sufficiently slow so that the contribution of horizontal turbulent exchange is important only near lateral boundaries of the aerosol cloud. Then the initial system of equations takes the form

$$\frac{\partial\Phi}{\partial x} = \nu\frac{\partial^2 u}{\partial z^2} + l, \quad \frac{\partial\Phi}{\partial y} = \nu\frac{\partial^2 \nu}{\partial z^2} - l\,u, \quad \frac{\partial\Phi}{\partial z} = g\alpha^*\theta,$$

$$\frac{\partial u}{\partial x} + \frac{\partial \mathrm{u}}{\partial y} + \frac{\partial w}{\partial z} = 0, \quad \Gamma w = \nu\frac{\partial^2\theta}{\partial z^2} + Q. \tag{11}$$

Let us formulate the boundary conditions for solving the system of equations (11). At the lower boundary ($z = 0$) all the velocity components vanish and the temperature is assumed to be fixed $\theta\big|_{z=0} = 0$. At the upper boundary ($z = H$) the horizontal velocity components are finite and the potential temperature ratio is approximated by the expression $\frac{\partial\theta}{\partial z}\Big|_{z=H} = q\theta$ ($q \geq 0$ is the dimensional parameter).

As has already been noted, the problem is to determine the vertical velocity which can be found from the solution of the equation of continuity

$$w(z) = -\int_0^z \left(\frac{\partial u}{\partial x} + \frac{\partial u}{\partial y}\right) dz'. \quad (12)$$

When ν = const the first two equations of system (11) enable one to obtain the Ekman solution

$$u(z) = -\frac{1}{l}\frac{\partial\Phi}{\partial y}(1 - e^{-az}\cos az) - \frac{1}{l}\frac{\partial\Phi}{\partial x}e^{-az}\sin az,$$

$$v(z) = -\frac{1}{l}\frac{\partial\Phi}{\partial x}(1 - e^{-az}\cos az) - \frac{1}{l}\frac{\partial\Phi}{\partial y}e^{-az}\sin az, \quad (13)$$

where the parameter $a = (f/2\nu)^{1/2}$. With allowance for relations (13) expression (12) takes the form

$$w(z) = \frac{1}{l}\int_0^z \Delta\Phi e^{-az'}\sin az'dz' \approx \frac{1}{l}\sqrt{\frac{\nu}{2l}}\Delta\Phi_0. \quad (14)$$

Here Φ_0 is the value of the function Φ at $z = 0$. To obtain an analytical expression that in its explicit form takes into account the dependence of vertical velocity on the rate of heat release, we carry out the following operations. The equation of heat influx is integrated over altitude. With allowance for the boundary conditions we obtain

$$\theta(z) = \frac{\Gamma w}{2\nu}z^2 - \frac{Q_0 H}{\pi\nu}\left(z - \frac{H}{\pi}\sin\frac{\pi z}{H}\right) +$$
$$+\frac{(1 - qH/2)(2Q_0 H/\pi - \Gamma H w)}{\nu(1 - qH)}z. \quad (15)$$

Integrating the equation of statics over altitude and taking into account relation (15), we derive

$$\Phi(z) = \Phi_H - \frac{g\alpha^*\Gamma w}{6\nu}(H^3 - z^3) + \frac{g\alpha^*}{2\nu}\frac{Q_0 H}{\pi} -$$
$$-\frac{(1 - qH/2)(2Q_0 H/\pi - \Gamma H w)}{1 - qH}(H^2 - z^2) +$$
$$+ g\alpha^*\frac{Q_0 H^3}{\pi^3\nu}\left(1 - \cos\frac{\pi z}{H}\right), \quad (16)$$

where Φ_H is the value of the function Φ at $z = H$.

Let us introduce the following designations:

$$L_* = NH\left[\frac{DH}{2\nu}\left(\frac{1 - qH/2}{1 - qH} - \frac{1}{3}\right)\right]^{1/2},$$

$$R_* = NH\left[\frac{DH}{\pi\nu}\left(\frac{1 - qH/2}{1 - qH} - \frac{1}{2}\right)\right]^{1/2},$$

$$N = (\alpha^* g\Gamma)^{1/2}, \quad D = \frac{1}{l}(\nu/2l)^{1/2}, \quad A = (L_*/R_*)^2.$$

Acting by the Laplacian operator on Eq. (10) and substituting the resulting expression into Eq. (16) we obtain the Helmholtz equation for the vertical velocity

$$[\,\Delta(Q_0/A\Gamma) - w\,] - [\,(Q_0/A\Gamma - w)/L_*^2\,] = Q_0/\left(A\Gamma L_*^2\right). \quad (17)$$

This equation has been derived for $\Delta\Phi_H = 0$, in particular, when pressure at the upper level $z = H$ is constant. It should be noted that without pressure perturbation at $z = H$ in a hydrostatic atmosphere (it is just the model that is considered here) the pressure variation at the lower boundary is determined by heating of an atmospheric column between the levels $z = 0$ and $z = H$, i.e., by the heat source Q_0.

Equation (11) is solved via the Green's function which is here the cylindrical McDonald function K_0

$$w = \frac{Q_0}{A\Gamma} - \frac{1}{2\pi}\int\!\!\int_{-\infty}^{\infty}\frac{Q_0}{A\Gamma L_*^2}K_0\left[\frac{(x - x')^2 + (y - y')^2}{L_*}\right]dxdy. \quad (18)$$

The parameter L_* has a meaning of a characteristic horizontal scale, and for $q \approx 0$ (turbulent heat flux at the upper boundary of the layer under study is sufficiently small)

$$L_* = NH\left(\frac{DH}{3\nu}\right) \approx 150 \text{ km}.$$

In this case we assume $\alpha^* = 3\cdot10^{-3}$ deg^{-1}, $H = 10^3$ m, $\Gamma = 3\cdot10^{-3}$ deg/m, $l = 10^{-4}$ s^{-1}, and $\nu = 1$ m^2/s. Then the Brunt–Veisala frequency $N = 10^{-2}$ s^{-1}, and the parameter $A \approx 2.1$. If the problem is two–dimensional ($\partial/\partial x \approx 0$) and heat release is localized within the region $|y| \leq y_0$, then

$$w = \frac{Q_0}{A\Gamma}\begin{cases}\exp(-y_0/L_*)\cosh(y/L_*), & \text{for } |y| < y_0,\\ \exp(-|y_0|/L_*)\sinh(y/L_*), & \text{for } |y| > y_0.\end{cases} \quad (19)$$

The vertical velocity associated with the source Q_0 can be estimated using formulas (19). In particular, if $y_0 \leqslant L$ (a smoke cloud transforms into a plume), then

$$w \approx \frac{Q_0}{AG}\begin{cases}1, & \text{for } |y| < y_0,\\ -\frac{y_0}{L_*}\exp\left(-\frac{y_0}{L_*}\right), & \text{for } |y| > y_0.\end{cases} \quad (20)$$

Let the solar radiation flux I_0, incident on an aerosol cloud, be 340 W/m^2 and be fully absorbed by aerosol. Then $Q_0 \approx I_0/(\rho c_p H) \approx 0.26\cdot10^{-3}$ deg/s. For $|y| < y_0$ we obtain the estimate $w \approx 4$ cm/s.

Since the optical thickness of the smoke cloud decreases with time, the quantity of absorbed solar radiation decreases as well, and so the vertical velocity of lifting of finely dispersed fraction of smoke particles also decreases.

Account of aerosol polydispersity. To take into account aerosol polydispersity, its particle size distribution is divided into individual fractions in which the particles are assumed to be monodisperse, and the aerosol transfer equation is integrated numerically for each fraction.

The mass $m(r_i, r_{i+1})$ of the particle whose radii are within the interval $r_a \in [r_i, r_{i+1}]$ is determined as follows. The formula[7]

$$M_t = \frac{4}{3}\pi\rho_a N \int_0^\infty r_a^3 f(r_a)\,dr_a = \frac{4}{3}\pi\rho_a N r_0^3 \exp(9\sigma_a^2/2) \quad (21)$$

is valid for the total mass M_t. Here ρ_a is the smoke aerosol particle density, N is the number of aerosol particles, $f(r_a)$ is the aerosol particle size distribution function (this function is described by the lognormal law[7]), r_0 is the mean geometric radius of particles, and σ_a^2 is the variance of the logarithm of r_a.

In its turn, the following expression can be written for mass $m(r_i, r_{i+1})$:

$$m(r_i, r_{i+1}) = \frac{4}{3}\pi\rho_a n(r_i, r_{i+1}) \int_{r_i}^{r_{i+1}} r_a^3 f(r_a)\,dr_a, \quad (22)$$

where $n(r_i, r_{i+1})$ is the number of smoke particles whose radii are within the interval $r_a \in [r_i, r_{i+1}]$.

It follows from comparison of Eqs. (21) and (22) that

$$m(r_i, r_{i+1}) = \frac{M_t}{r_0^3 \exp(9\sigma_a^2/2) \int_{r_i}^{r_{i+1}} f(r)\,dr} \int_{r_i}^{r_{i+1}} r^3 f(r)\,dr. \quad (23)$$

Thus to determine the mass of aerosol particles whose radii are within the interval $r_i - r_{i+1}$, it is necessary to assign the total mass M_t and the parameters of aerosol microstructure, namely, the modal radius r_m and variance s_a^2, since $r_0 = r_m \exp(\sigma_a^2)$ (see Ref. 7).

Parametrization of washout. Aerosol concentration in the atmosphere is significantly influenced by natural clouds, hazes, and precipitation. In this case microphysical characteristics, cloud amount, precipitation intensity, composition and morphology of aerosol particles, as well as dynamic factors play an important role. The washout rate also depends indirectly on the altitudes at which aerosol is concentrated in the atmosphere (the upper troposphere is much drier than the lower one). It should be noted that washout is minimum in the so–called Greenfield discontinuity region ($0.1 \le r_a \le 1.0$ μm). This minimum is caused by the fact that for these particles the effect produced by Brownian diffusion on particle sedimentation already becomes sufficiently weak, while the effect of entrainment is not yet sufficiently pronounced. Therefore, the aerosol particles whose sizes fall into the Greenfield discontinuity region have the longest lifetime in the atmosphere.

When solving the problems of mathematical modeling of aerosol formations it is expedient to use the approach which is based on studying two–component stochastic systems consisting of cloud (rain) particles and aerosols. In this case it is possible to solve the problem of coagulation[8]

$$dn_i/dt = -K(r_i, R_k)\, n_i N_k, \quad (24)$$

where n_i and N_k are the numbers of impurity particles and droplets (rain, cloud, or haze) of radii r_i and R_k per unit volume, respectively; $K(r_i, R_k)$ is the coefficient of proportionality which is expressed in terms of the coefficient of entrainment.[7]

Integration of Eq. (24), provided that N_k is constant, enables one to find a general law of aerosol particle concentration decrease in time

$$n_i(t) = n_i(0) \exp[-K(r_i, R_k) N_k t]. \quad (25)$$

Hence the constant of aerosol particle removal due to washout is determined from the expression

$$\Lambda_i = K(r_i, R_k) N_k.$$

Thus the problem of parametrization is reduced to the determination of the washout constant Λ_i. We constructed the plots of the washout constant vs. the cloud–cover index for different isobaric levels based on the statistical data processing (Fig. 1). These plots are used in numerical modeling of evolution of aerosol formations in the atmosphere.

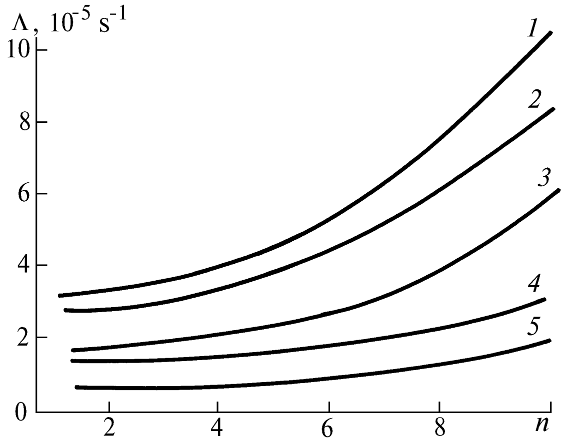

FIG. 1. Washout coefficient vs. the cloud–cover index at different isobaric levels. Not in the Greenfield discontinuity region: 1, 2, and 4) P = 850, 700, *and* 500 hPa, *respectively. In the discontinuity region: 3) P* = 850 *and* 700 hPa, *5)* 500 hPa.

Parametrization of sedimentation. The force of gravity affecting aerosol particles causes their sedimentation at a rate that primarily depends on particle size and density. We used analytic relations for calculating the rate of spherical particle sedimentation. When the particle diameter is larger than 150 μm, their sedimentation is described by the Newton formula

$$w_s = 174\left(g\,\frac{\rho_a - \rho}{\rho}\,d_a\right)^{1/2}, \quad (26)$$

where w_s is the rate of sedimentation of a particle of density ρ_a.

When the aerosol particle size varies from 3 to 150 μm, the rate w_s is estimated by the known Stokes formula

$$w_s = \frac{g(\rho_a - \rho)}{18\,\eta}\, d_a^2 , \qquad (27)$$

where η is the coefficient of molecular viscosity.

For small particles ($d_a < 3$ μm) the Stokes formula is refined by application of the dimensionless Kunninghem coefficient

$$w_s = w'_s\, [1 - a\,(\lambda_m/d_a)\,] \quad , \qquad (28)$$

where w'_s is the Stokes fall rate, λ_m is the mean free path of air molecules and $a \in [1.3, 2.3]$ is the constant.

The relations (26)–(28) allow one to calculate the sink of aerosol mass from aerosol clouds under gravity.

Conclusion. The present paper describes a regional model of pollution transport in the atmosphere which is algorithmically written as an independent block. The model was tested by solving, by way of example, the problem of spread of smoke clouds in the atmosphere.[9] The capabilities and advantages of the model allow its usage in the problems of regional ecology.

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