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# INDIRECT DETERMINATION OF THE TURBULENT DIFFUSION COEFFICIENTS

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*By solving the inverse problem, the method of indirect determination of the turbulent diffusion coefficients (TDCs) has been developed. The application of this method to the case where the TDCs are constant and where the altitude dependence of the vertical TDC is present has been considered. The requirements imposed on the number of measurement points of the pollutant concentrations at which the TDCs can still be recovered have been established. The proposed method provides determination of the TDCs in a wide range of values under different meteorological conditions.*

**Introduction.** Turbulent diffusion coefficients (TDCs) of an atmospheric boundary layer are important parameters for mathematical models associated with a study of the processes of propagation of pollutants in the atmosphere. As a rule, different empirical dependences are used in applied calculations for determining the values of the TDCs. Direct experimental measurement of these coefficients is a rather complicated problem, since their values are influenced by specific local conditions. In the present work, the values of the TDCs are calculated by solving the inverse problem of advection and diffusion of air pollutants involving photochemical reactions and sources. It is assumed here that the main source of pollution is municipal motor transport [1]. The test problem with the simplifying assumption of the coordinate independence of the TDCs in space was solved earlier in [2]. This assumption is frequently used in describing the propagation of pollutants from high-altitude point sources [3, 4]. In the general case, this approximation is inapplicable. Recovery of the dependence of the vertical TDC on the altitude above the earth's surface was the subject of further investigations. In test calculations, the values of the pollution concentrations at certain points of the computational domain which have been found in solving the direct problem with the known TDCs serve as overdetermination conditions for the inverse problem. The recovery of the TDCs is a result of the minimization of the residual functional by the method of stochastic approximation. In solving the direct problems, use was made of the data on the motor transport emission and the wind field in Mexico City (Mexico).

**Direct Problem.** The procedure of finding the TDCs is based on solution of direct problems which describe the processes of emission of primary air pollutants, chemical transformations, advection, and turbulent diffusion (TD). In the present work, we used the model of photochemical smog formed as a result of motor-transport pollution; this model includes into consideration $n$ = 11 pollutants participating in the following $s$ = 12 chemical reactions [1, 5]:

$$\mathrm{NO_2} + h\nu \xrightarrow{K_1} \mathrm{NO} + \mathrm{O}\,, \quad K_1 = 0.02\exp(-0.507\sec\theta)\,,\ \mathrm{sec}^{-1}\,;$$

$$\mathrm{O} + \mathrm{O_2} + \mathrm{M} \xrightarrow{K_2} \mathrm{O_3} + \mathrm{M}\,, \quad K_2 = 0.036\cdot10^{-5}\,,\ \mathrm{sec}^{-1}\cdot\mathrm{ppm}^{-1}\,;$$

$$\mathrm{NO} + \mathrm{O_3} \xrightarrow{K_3} \mathrm{NO_2} + \mathrm{O_2}\,, \quad K_3 = 0.44\,,\ \mathrm{sec}^{-1}\cdot\mathrm{ppm}^{-1}\,;$$

$$\mathrm{RH} + \mathrm{OH} \xrightarrow{K_4} \mathrm{RO_2} + \mathrm{H_2O}\,, \quad K_4 = 62.92\,,\ \mathrm{sec}^{-1}\cdot\mathrm{ppm}^{-1}\,;$$

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$$RCHO + OH \xrightarrow{K_5} RC(O)O_2 + H_2O\ , \quad K_5 = 390.17\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$RCHO + h\nu \xrightarrow{K_6} RO_2 + HO_2 + CO\ , \quad K_6 = 5.55\cdot 10^{-5} \exp\,(-\,0.495 \sec\theta)\ , \quad sec^{-1}\ ;$$

$$HO_2 + NO \xrightarrow{K_7} NO_2 + OH\ , \quad K_7 = 202.33\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$RO_2 + NO \xrightarrow{K_8} NO_2 + RCHO + HO_2\ , \quad K_8 = 187.83\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$RC(O)O_2 + NO \xrightarrow{K_9} NO_2 + RO_2 + CO_2\ , \quad K_9 = 187.83\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$OH + NO_2 \xrightarrow{K_{10}} HNO_3\ , \quad K_{10} = 268.83\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$RC(O)O_2 + NO_2 \xrightarrow{K_{11}} RC(O)O_2NO_2\ , \quad K_{11} = 114.88\ , \quad sec^{-1}\cdot ppm^{-1}\ ;$$

$$RC(O)O_2NO_2 \xrightarrow{K_{12}} RC(O)O_2 + NO_2\ , \quad K_{12} = 35.72\cdot 10^{-5}\ , \quad sec^{-1}\cdot ppm^{-1}\ .$$

A change in the concentration of the pollutant in the reacting mixture due to chemical transformations is described by a system of ordinary differential equations of the form

$$\frac{dC_i}{dt} = R_i\,(K_1, ..., K_s;\, C_1, ..., C_n)\ , \quad i = 1, ..., n\ .$$

For example, $\frac{d[\mathrm{RH}]}{dt} = -K_4\cdot[\mathrm{RH}]\cdot[\mathrm{OH}] \equiv R_4$, where [RH] = $C_4$; [OH] = $C_5$ (the chemical formula of the pollutant, enclosed in the square brackets, denotes its concentration).

The initial boundary-value problem in this simplified approximation, ignoring the deposition of impurities and some other processes, is written in the form of the following system of equations [1]:

$$\frac{\partial C_i}{\partial t} + \mathrm{div}\,(\mathbf{U}C_i) = \frac{\partial}{\partial x}\left(K_x \frac{\partial C_i}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial C_i}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z\,(z) \frac{\partial C_i}{\partial z}\right) + R_i\ , \quad i = 1, ..., n\ , \tag{1}$$

$$C_i\,(x, y, z, t = 0) = \varphi_i\,(x, y, z)\ , \tag{2}$$

$$-\,K_z \frac{\partial C_i}{\partial z}\bigg|_{z=0} = S_i\ , \quad \frac{\partial C_i}{\partial z}\bigg|_{z=L_z} = 0\ , \tag{3}$$

$$\frac{\partial C_i}{\partial x}\bigg|_{x=0} = \frac{\partial C_i}{\partial x}\bigg|_{x=L_x} = 0\ , \quad \frac{\partial C_i}{\partial y}\bigg|_{y=0} = \frac{\partial C_i}{\partial y}\bigg|_{x=L_y} = 0\ . \tag{4}$$

We determined the intensity of the surface sources by applying the values of the motor transport emissions. Problem (1)–(4) was solved numerically by the method of splitting by physical processes (advection and diffusion along three coordinates and a chemical cycle of 12 reactions) with the use of the IMSL library in the FORTRAN programming language.

**Inverse Problem.** The inverse problem lies in recovering the turbulent diffusion coefficients $K_x$, $K_y$, and $K_z(z)$ provided that the so-called overdetermination conditions, i.e., the pollutant concentrations $C_i$ at certain points of the region as functions of the time (values of the concentrations measured at meteorological stations), are known. The determination of the TDC is based on solution of a set of direct problems with iteration refinement of the coefficients $K_x$, $K_y$, and $K_z(z)$.

This method was first applied to the simplest case of the coefficients constant over the entire region: $K_x = K_y \equiv K_\mathrm{h}$ = const and $K_z(z) \equiv K_\mathrm{v}$ = const. In solving the inverse problem, these coefficients were considered to be unknown and were generally calculated by minimization of the residual functional at all points of the region:

$$\Phi(\mathbf{K}) = \sum_{i=1}^{n} \int_0^{\tau} \int_0^{L_z} \int_0^{L_y} \int_0^{L_x} (C_i(x, y, z, t; \mathbf{K}) - \overline{C}_i(x, y, z, t))^2 \, dxdydzdt\,.$$

As a rule, the concentrations are measured only at certain points $x_p$ and $y_p$ determined by the position of the meteorological stations. Then the problem is reduced to minimization of the following functional:

$$\Phi(\mathbf{K}) = \sum_{i=1}^{n} \sum_{p=1}^{N_p} \int_0^{\tau} (C_i(x_p, y_p, H_\mathrm{st}, t; \mathbf{K}) - \overline{C}_i(x_p, y_p, H_\mathrm{st}, t))^2 \, dt\,.$$

In this work, we used the method of stochastic approximation which is usually applied to search for the functional minimum which is determined by the quantities containing errors of random character. The iterations of the method of stochastic approximation can be written in the following form [6]:

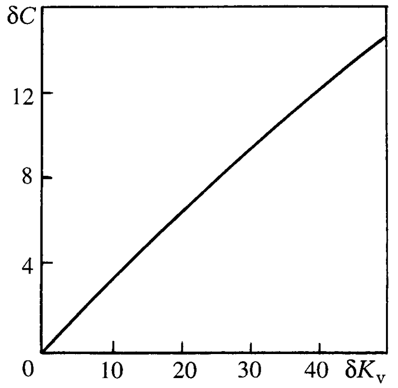
$$K_j^{k+1} = K_j^k - \beta_k \,\mathrm{sign}\left[\frac{\Phi(\mathbf{K}^k + f\,\mathbf{e}^j) - \Phi(\mathbf{K}^k - f\,\mathbf{e}^j)}{2f}\right], \quad j = \mathrm{h}, \mathrm{v}\,; \quad k = 1, 2, \ldots,$$

where the sign of $\beta_k$ coincides with that of the expression in square brackets, while the constant $f$ = 0.01 m$^2$/sec corresponds to the accuracy with which the TDC were recovered (the criterion of stopping of the iterative process, used in the calculations, is as follows: $|K_j^{k+1} - K_j^k| \le \delta$ = 0.01 m$^2$/sec).

**Results of Calculations of the Inverse Problem with Constant TDCs.** The simulated region was a parallelepiped with a rib length of $L_x$ = 65 km, $L_y$ = 75 km, and $L_z$ = 1 km along the coordinates $x$, $y$, and $z$ respectively. The region was subdivided into unit cells of dimensions $\Delta x = \Delta y$ = 5 km and $\Delta z$ = 50 m. The initial distributions of the concentrations of the considered pollutants and the other input parameters of the problem corresponded to the typical values obtained in Mexico City (Mexico).

The calculations were carried out for a different number of meteorological stations $N_p$ with a wind and without it. The diffusion coefficients for the first iteration were equal to $K_\mathrm{h} = K_\mathrm{v}$ = 1 m$^2$/sec. As can be seen from the calculations, the method of stochastic approximation leads to a stable determination of the TDC under various conditions. The numerical experiments have shown that the number of iterations depends on the difference between the true value of the TDC and the values chosen for the first iteration. To attain an acceptable accuracy in the variants of calculations carried out, it was required to perform no more than 20–30 iterations. The TDCs were recovered here with an accuracy no worse than 5%. It should be noted that the recovery of the vertical TDC was observed irrespective of the wind velocity, whereas the horizontal TDC can be found only in the case of a very weak wind or in its absence, which is a rarely realizable meteorological situation. However, in practical calculations, use is frequently made of the assumption of constancy of the horizontal turbulent diffusion coefficient (one takes it to be equal to $K_\mathrm{h} \approx 50$ m$^2$/sec [7]). This is associated with the fact that the horizontal TDC is in proportion to the first derivative of dispersion of the impurity distribution over the horizontal coordinate, whereas the dispersion itself in this direction is virtually a linear function (and, correspondingly, its derivative can be considered to be constant) [5, 8]. Therefore, the reconstruction was carried out only for the vertical TDC.

In investigating problems related to the influence of the volume and accuracy of the input data on the solution of the inverse problem of recovery of the vertical TDC, it was primarily established how much the solution of direct



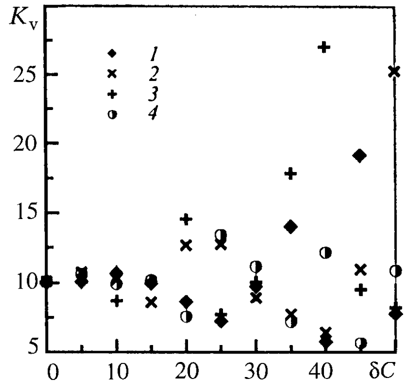


Fig. 1. Dependence of the error of computation of the ozone concentration on the error of determination of the vertical turbulent diffusion coefficient. $\delta C$, $\delta K_{\text{v}}$, %.

Fig. 2. Dependence of the values of the recovered vertical turbulent diffusion coefficient on the error of the concentrations: 1–4) correspond to four series of recovery of $K_{\text{v}}$ on introduction of random errors into the concentrations used under overdetermination conditions. $K_{\text{v}}$, m$^2$/sec; $\delta C$, %.

problem (1)–(4) depends on the accuracy of determination of the vertical TDC. From the calculations conducted it is evident that in order to calculate the concentrations with an accuracy of 5% (which is a standard error in measuring the concentrations of various substances at meteorological stations [9]), it is sufficient to determine the vertical TDC accurate to 10–15%. Figure 1 shows the error of computation of the ozone concentration as a function of the error of determination of the vertical TDC (the computational error was determined as the relative difference between the pollutant concentrations calculated with the use of the true diffusion coefficient and the vertical TDC changed by a certain percent). The influence of the accuracy in finding the pollutant concentrations used as an overdetermination condition is another question important for solving the inverse problem. As the calculations have shown, to recover the vertical TDC accurate to 10–15% it is necessary to assign overdetermination conditions with an accuracy of 15–20%. Figure 2 shows a few series of recovery of the vertical TDC for different errors in assigning overdetermination conditions (for each value of the maximum amplitude of the relative error in $\delta C$ we carried out four different series of recovery of $K_{\text{v}}$ with four different random sets of errors in the overdetermination conditions lying in the range $\overline{C} \pm \delta C$). Since the exact initial pollutant distributions are unknown in solving the inverse problem, as is evident from the calculations carried out, the assignment of zero initial concentrations in the limiting case leads to a systematic error in recovery of the vertical TDC of no more than 5–10%. If the initial concentration distributions in solving the inverse problem are taken with an error less than 30%, at least two points of overdetermination would suffice for successful recovery of the vertical TDC. At zero initial concentration distributions, at least ten points of overdetermination are required in order to recover the TDC with an accuracy of 5–10%. We also established that when the number of points $N_p$ exceeds the minimum required number, the values of the recovered TDCs remain unchanged, in practice. However, for $N_p = 1$ the vertical TDC cannot be recovered by the method proposed.

**Inverse Problem for a Variable Vertical TDC.** The value of the vertical TDC in the atmospheric surface layer can change rather strongly with altitude and can lead to large errors in the values of the concentrations (of the order of several tens of percent). In this case, the main differences are observed at night when the intensity of the motor-transport sources is low and photochemical reactions are virtually damped out, i.e., when the diffusion and transfer of impurities by the wind become the basic processes determining the concentration of a specific substance at any point of the computational domain.

At the present time, there are several empirical formulas approximately showing the form of the dependence of the vertical TDC on the altitude [5]. To describe the propagation of pollutants from motor-transport sources one most often uses the following relations [7, 10]:

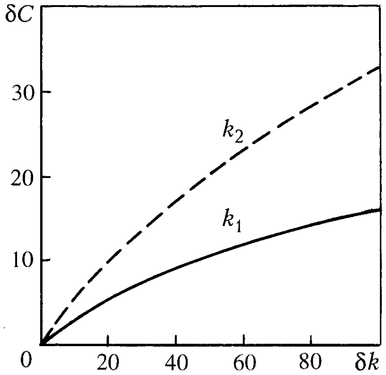


Fig. 3. Dependence of the error of the calculated concentrations of ozone on the error in determination of the coefficients $k_1$ and $k_2$. $\delta C$, $\delta k$, %.

$$K_{\mathrm{v}}(z) = \begin{cases} [2.5q(x,y,t) - 77.3]\dfrac{z}{L_z} + 30.9\,, & 0 \le \dfrac{z}{L_z} \le 0.4\,, \\ q(x,y,t)\,, & 0.4 < \dfrac{z}{L_z} \le 0.8\,, \\ 5\,[30.9 - q(x,y,t)]\dfrac{z}{L_z} + 5q(x,y,t) - 123.6\,, & 0.8 < \dfrac{z}{L_z} \le 1\,, \end{cases}$$

where $q(x, y, t) = 0.85\sqrt{[U_x^2(x,y,t) + U_y^2(x,y,t)]} + 232$, m$^2$/min; here $U_x(x, y, t)$ and $U_y(x, y, t)$ are the components of the wind-velocity vector along the $x$ and $y$ axes respectively. In the general case this dependence can be replaced by the piecewise-linear function

$$K_{\mathrm{v}}(z) = \begin{cases} \dfrac{(k_2 - k_1)}{0.4}\dfrac{z}{L_z} + k_1\,, & 0 \le \dfrac{z}{L_z} \le 0.4\,, \\ k_2\,, & 0.4 \le \dfrac{z}{L_z} \le 0.8\,, \\ \dfrac{(k_2 - k_1)}{0.2}\left(1 - \dfrac{z}{L_z}\right) + k_1\,, & 0.8 \le \dfrac{z}{L_z} \le 1\,, \end{cases}$$

where $k_1$ and $k_2$ are the minimum and maximum values of $K_{\mathrm{v}}(z)$ respectively. Figure 3 gives the dependence of the error of computation of the ozone concentration in the direct problem on the error of determination of the coefficients $k_1$ and $k_2$. All the attempts to recover these coefficients by minimizing the residual functional in accordance with the method stated in the previous sections turned out to be unsuccessful. The calculations performed have shown that in the case of simultaneous recovery of $k_1$ and $k_2$, the accuracy of determination of $k_2$ substantially depends on the accuracy of recovery of $k_1$. In this connection, into the above-described algorithm of solving the inverse problem we introduced the following changes: 1) the regularization method was used to minimize the residual functional; 2) first the coefficient $k_1$ was recovered and then the coefficient $k_2$. Thus, to recover the functional dependence of the vertical turbulent diffusion coefficient, it turned out to be essential to minimize the following functional [11]:

$$\Phi(k_1, k_2) = \sum_{i=1}^{n} \int_0^{\tau}\int_0^{L_z}\int_0^{L_y}\int_0^{L_x} \left(C_i(x,y,z,t; K_{\mathrm{h}}, K_{\mathrm{v}}(z)) - \overline{C}_i(x,y,z,t)\right)^2 dx\,dy\,dz\,dt + \alpha\Omega(k_1, k_2)\,.$$

As a stabilizer we used the functional [12]

$$\Omega(k_1, k_2) = \int_0^{L_z}\left[\frac{dK_{\mathrm{v}}(z)}{dz}\right]^2 dz\,.$$

For successful recovery of the coefficients $k_1$ and $k_2$ it was not sufficient to apply overdetermination conditions at the points on the surface of the computational domain ($z = 0$); therefore, the points above the surface must also be taken into account. It is sufficient to use overdetermination only at three of these points lying on one vertical line and in this way to minimize the functional

$$\Phi (k_1, k_2) = \sum_{i=1}^{n} \sum_{p=1}^{3} \int_0^{\tau} (C_i (x_g, y_g, z_p, t; K_h, K_v (z_p)) - \overline{C}_i (x_g, y_g, z_p, t))^2 \, dt + \alpha\Omega (k_1, k_2) .$$

To do this, in the present work we used the previously described method of stochastic approximation with descent in the parameter $\alpha$ [11].

**Results of Calculations of the Inverse Problem with a Variable Vertical TDC.** The test calculations were carried out for the same computational domain and the same meteorological conditions as for the case of constant TDC. The horizontal TDC was assumed to be constant (50 $m^2$/sec). In a stable atmosphere, the values typical of the coefficients $k_1$ and $k_2$ lie within the ranges (0, 1] and [1, 10] $m^2$/sec respectively. The calculations performed have shown that $\alpha = 0.1$ is the optimum starting value of the regularization parameter $\alpha$ and that after the realization of the first step of descent in the parameter $\alpha$, it is better to perform its further subdivision depending on the initial approximate value of the coefficient $k_1$ obtained at this step. When the parameter $\alpha$ reaches values of $10^{-9}$–$10^{-8}$, the coefficients $k_1$ and $k_2$ are recovered with an accuracy no worse than 5%. This error in the recovery of the coefficients leads to an error of more than 1% in the calculation of the concentrations.

## CONCLUSIONS

The method of indirect determination of a TDC by solution of the inverse problem has been developed. The recovery of this coefficient is based on the minimization of the residual between the concentration profiles measured and the values calculated at each iteration. It is shown that the constant diffusion coefficients can successfully be recovered using only the values of the pollutant concentrations on the lower surface of the computational domain, whereas the recovery of the altitude dependence of the vertical turbulent diffusion coefficient also requires information on the pollutant concentration in the near-surface layer.

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## NOTATION

$h$, Planck constant; $\nu$, frequency, Hz; $C_i(x, y, z, t)$, $C_i(x, y, z, t; \mathbf{K})$, and $\overline{C}_i(x, y, z, t)$, concentrations of the $i$th pollutant in the direct and inverse problems and the measured value, respectively, ppm; $f$, constant, $m^2$/sec; $H_{st}$, standard altitude of measurements, m; $K_x$, $K_y$, and $K_z(z)$, TDC along the coordinate axes, $m^2$/sec; $K_h$ and $K_v$, horizontal and vertical TDC, $m^2$/sec; $\mathbf{K} = (K_h, K_v)$; $K_1$, $K_2$, ..., $K_{12}$, constants of the chemical reactions; $k_1$ and $k_2$, minimum and maximum values of $K_v(z)$, $m^2$/sec; $L_x$, $L_y$, and $L_z$, dimensions of the investigated region along the $x$, $y$, and $z$ axes, respectively, m; $N_p$, general number of observation stations; $q(x, y, t)$, auxiliary function, $m^2$/sec; $R_i$, right-hand side of the ordinary differential equation describing the change in the concentration of the $i$th pollutant due to the chemical reactions; $S_i$, intensity of the surface sources of the $i$th pollutant, ppm·m/sec; $\mathbf{U} = (U_x, U_y, 0)$, wind velocity; m/sec; $\mathbf{e}^h = (1, 0)$ and $\mathbf{e}^v = (0, 1)$, unit vectors; $n$, number of pollutants; $t$, time, sec; $x_p$ and $y_p$, coordinates of the observation stations, m; $\alpha$, parameter of regularization; $\beta_k = 1/k$, iterative term of the sequence; $\Delta x$, $\Delta y$, and $\Delta z$, dimensions of the cells along the coordinates, m; $\delta$, constant determining the criterion of stopping of the iterative process, $m^2$/sec; $\varphi_i(x, y, z)$, initial distribution of the concentration of the $i$th pollutant, ppm; $\theta$, angle of incidence of the sunbeam, deg; $\tau$, time interval, sec; $\Omega(k_1, k_2)$, stabilizing functional; $\Phi(\mathbf{K})$, residual functional. Subscripts: g, index of the horizontal coordinates of the observation stations for which $z > 0$; h, horizontal; $i$, number of pollutants; $k$, number of the iteration; $p$, number of the observation station; v, vertical; $s$, number of reactions; st, standard.

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