# Simulation of surface accumulation of anthropogenic polydisperse aerosol

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A mathematical model of pollutant sedimentation on the surface is considered with allowance made for climatic characteristics. Transition probabilities are constructed based on the Fokker–Planck–Kolmogorov equation, which is solved numerically. Specific calculated results for Irkutsk are presented, and the influence of emissions from Irkutsk industrial plants on the water area of Southern Baikal is estimated.

Pollution of the environment with anthropogenic emissions has a negative effect on living organisms, soil, buildings, architectural monuments, and constructions. It causes metal corrosion and decreases atmospheric transparency. Under the effect of gravity, pollutants deposit onto the surface (soil, water bodies) from the atmosphere. Surface run-off causes secondary pollution of water bodies (partial washout of pollutants from soil). This is why estimation of pollutant flow from the atmosphere to the surface is of great importance. One of the means of such estimation is mathematical simulation that permits one to find the optimal variant from the viewpoint of minimizing anthropogenic load by simulating possible situations on a computer.

A light pollutant in a homogeneous medium is described in Refs. 1 and 2. In this paper, we consider a passive pollutant characterized by a certain sedimentation rate in an anisotropic medium. The diffusion equation for this pollutant has the form

$$\frac{\partial s}{\partial t} + \frac{\partial u_i\, s}{\partial x_i} - \frac{\partial w_g\, s}{\partial x_3} = F + \frac{\partial}{\partial x_i} \nu_{ij} \frac{\partial s}{\partial x_j}, \qquad (1)$$

where $s$ is the pollutant concentration; $u_i$ is the component of medium velocity along the corresponding coordinate $x_i$ ($i = \overline{1, 3}$); $w_g$ is the gravitational sedimentation rate which is different for particles of different fractions; $F$ is the intensity of pollutant sources; $t$ is time; $\nu_{ij}$ are the coefficients of turbulent diffusion. Repeating indices mean summation.

Let us represent $s$, $u_i$, $\nu_{ij}$, and $F$ as a sum of mean values and deviations. Then Eq. (1) takes the form

$$\frac{\partial(\bar{s} + s')}{\partial t} + \frac{\partial(\bar{u}_i + u_i')(\bar{s} + s')}{\partial x_i} - \frac{\partial w_g(\bar{s} + s')}{\partial x_3} =$$

$$= \bar{F} + F' + \frac{\partial}{\partial x_i}(\bar{\nu}_{ij} + \nu_{ij}') \frac{\partial(\bar{s} + s')}{\partial x_j}.$$

Transforming the right-hand side of the last equation, we obtain

$$\frac{\partial(\bar{s} + s')}{\partial t} + \frac{\partial(\bar{u}_i + u_i')(\bar{s} + s')}{\partial x_i} - \frac{\partial w_g(\bar{s} + s')}{\partial x_3} =$$

$$= \bar{F} + F' + \frac{\partial \bar{\nu}_{ij}}{\partial x_i}\frac{\partial \bar{s}}{\partial x_j} + \bar{\nu}_{ij}\frac{\partial^2 \bar{s}}{\partial x_i\, \partial x_j} + \frac{\partial \bar{\nu}_{ij}}{\partial x_i}\frac{\partial s'}{\partial x_j} + \bar{\nu}_{ij}\frac{\partial^2 s'}{\partial x_i\, \partial x_j} +$$

$$+ \frac{\partial \nu_{ij}'}{\partial x_i}\frac{\partial \bar{s}}{\partial x_j} + \nu_{ij}'\frac{\partial^2 \bar{s}}{\partial x_i\, \partial x_j} + \frac{\partial \nu_{ij}'}{\partial x_i}\frac{\partial s'}{\partial x_j} + \nu_{ij}'\frac{\partial^2 s'}{\partial x_i\, \partial x_j}.$$

Let us now average the last equation using the properties of mean values:

$$\frac{\partial \bar{s}}{\partial t} + \frac{\partial \bar{u}_i\, \bar{s}}{\partial x_i} - \frac{\partial w_g\, \bar{s}}{\partial x_3} = \bar{F} - \frac{\partial\, \overline{u_i' s'}}{\partial x_i} +$$

$$+ \frac{\partial \bar{\nu}_{ij}}{\partial x_i}\frac{\partial \bar{s}}{\partial x_j} + \bar{\nu}_{ij}\frac{\partial^2 \bar{s}}{\partial x_i\, \partial x_j}. \qquad (2)$$

As in Refs. 1 and 2, to construct the probability model, we turn to the second Kolmogorov equation written in the phase coordinate $s$:

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial s}[A(t, s)\, p] + \frac{\partial^2}{\partial s^2}[B(t, s)\, p], \qquad (3)$$

where $p = p(t, s)$ is the differential distribution law for the parameter $s$; $A = \frac{\partial \bar{s}}{\partial t}$ and $B = \frac{1}{2}\frac{\overline{\partial s'^2}}{\partial t}$ are respectively the mean rate of variation of the mean concentration and the intensity of random oscillations about this mean value in the interval $t \in [0, T]$.

The initial state is $p(0, s) = p_0(s)$. The boundary conditions are as follows:

$$\frac{\partial(Bp)}{\partial s} - Ap = 0 \text{ at } s \to \infty, \quad \int_0^\infty p(t, s)\, \mathrm{d}s = 1. \qquad (4)$$

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The problems of solvability of Eqs. (3) and (4) under certain restrictions placed on the coefficients $A$ and $B$ were considered by Kolmogorov in Ref. 3. In particular, proofs were performed for the so-called Bachelier case when $A(t) = 0$ and $B(t) = 1$, i.e., the Kolmogorov equation is reduced to the classical equation of heat transfer (at $s = x$), for the case when the coefficient $A$ varies linearly and $B$ is an arbitrary constant, and for the case $A(t, x) = 0$ and $B(t, x) = x$.

The unknown values in Eq. (3) are $p$, $A$, and $B$. To close the equation, let us find $A$ and $B$. To determine the coefficient $A$, we perform the following calculations. First, Eq. (2) is subtracted from Eq. (1):

$$\frac{\partial s'}{\partial t} + \frac{\partial}{\partial x_i}(u_i s - \bar{u}_i \bar{s}) - \frac{\partial w_g s'}{\partial x_3} =$$

$$= F' + \frac{\partial \overline{u'_i s'}}{\partial x_i} + \frac{\partial}{\partial x_i} \nu_{ij} \frac{\partial s}{\partial x_j} - \frac{\partial \bar{\nu}_{ij}}{\partial x_i}\frac{\partial \bar{s}}{\partial x_j} - \bar{\nu}_{ij}\frac{\partial^2 s}{\partial x_i \partial x_j}.$$

Transforming both sides of the last equation, we obtain

$$\frac{\partial s'}{\partial t} = -u_i \frac{\partial s'}{\partial x_i} - u'_i \frac{\partial \bar{s}}{\partial x_i} - \left(\bar{s}\frac{\partial u'_i}{\partial x_i} + s'\frac{\partial u_i}{\partial x_i}\right) + \frac{\partial w_g s'}{\partial x_3} +$$

$$+ F' + \frac{\partial \overline{u'_i s'}}{\partial x_i} + \frac{\partial}{\partial x_i}\bar{\nu}_{ij}\frac{\partial s'}{\partial x_j} + \frac{\partial}{\partial x_i}\nu'_{ij}\frac{\partial s}{\partial x_j}. \qquad (5)$$

For an incompressible liquid, every summand in parentheses vanishes because it is divergence of the velocity field and its fluctuations.

Following Refs. 4 and 5, we can neglect the influence of a passive pollutant on the velocity field of the medium in the linear approximation, i.e., the turbulent velocity field is assumed to be independent of pollutant concentration. Let us introduce a designation

$$q_k = \overline{u'_i s'}, \qquad (6)$$

where $s'$ is unknown.

Further, as in Ref. 1, Eq. (5) is integrated from $t$ to $t + \tau$, where $t \geq \tau$ ($\tau$ is the Eulerian time scale), in order to satisfy Eq. (6). Then both sides of the resulted equation are multiplied by $u'_k(t + \tau)$ and averaged within the interval $T - \tau$ ($T \gg \tau$):

$$q_k = \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\, s'(t+\tau)\, dt_1 =$$

$$= \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\, s'(t)\, dt_1 -$$

$$- \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} u_i \frac{\partial s'}{\partial x_i}\, dt_1\, dt -$$

$$- \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} u'_i \frac{\partial \bar{s}}{\partial x_i}\, dt_1\, dt +$$

$$+ \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} \frac{\partial w_g s'}{\partial x_3}\, dt_1\, dt +$$

$$+ \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} F'\, dt_1\, dt +$$

$$+ \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} \frac{\partial \overline{u'_i s'}}{\partial x_i}\, dt_1\, dt +$$

$$+ \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} \frac{\partial}{\partial x_i}\bar{\nu}_{ij}\frac{\partial s'}{\partial x_j}\, dt_1\, dt +$$

$$+ \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} \frac{\partial}{\partial x_i}\nu'_{ij}\frac{\partial(\bar{s}+s')}{\partial x_j}\, dt_1\, dt. \qquad (7)$$

The first summand in the right-hand side of the last equation vanishes because the integrands $u'_k(t + \tau)$ and $s'(t)$ are not correlated. Further, let us use the method of recursive inclusions.[4,5] As a result, we obtain the first approximation for Eq. (6):

$$\overline{u'_i s'}^{\,(1)} = -N_{ki}^{(1)}\frac{\partial \bar{s}}{\partial x_i} + Q^{(1)}, \qquad (8)$$

where

$$N_{ki}^{(1)} = \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} u'_i(t_1)\, dt_1\, dt;$$

$$Q^{(1)} = \frac{1}{T-\tau}\int_0^{T-\tau} u'_k(t+\tau)\int_t^{t+\tau} F'(t_1)\, dt_1\, dt.$$

Substituting Eq. (8) into Eq. (2), we obtain the closed equation for mean concentrations:

$$A = \frac{\partial \bar{s}}{\partial t} = -\frac{\partial \bar{u}_i \bar{s}}{\partial x_i} + \frac{\partial w_g \bar{s}}{\partial x_3} + \bar{F} +$$

$$+ \frac{\partial}{\partial x_i}\left(N_{ij}^{(1)}\frac{\partial \bar{s}}{\partial x_j} - Q^{(1)}\right) + \frac{\partial}{\partial x_i}\bar{\nu}_{ij}\frac{\partial \bar{s}}{\partial x_j}. \qquad (9)$$

Thus, the equation for the coefficient $A$ is derived.

Similarly, we can obtain the approximation of an arbitrary order. The error of the first approximation was estimated by comparison with analytical solutions (it does not exceed 20%).

To find the coefficient $B$, let us turn to Eq. (5). Multiply both its sides by $2s'$ and add to the equation of continuity of incompressible liquid (whose sides are pre-multiplied by $s'^2$). After averaging, in the first approximation we obtain

$$B = N_{ki}^{(1)} \left(\frac{\partial \bar{s}}{\partial x_i}\right)^2. \qquad (10)$$

Pulsations of the sources $F'$ are neglected in Eq. (10). This can be also done in Eq. (9).

The obtained coefficients $A$ and $B$ close Eq. (3). However, Eq. (9) for the coefficient $A$ is also of interest, as it permits one not only to calculate the field of mean pollutant concentrations in commonly accepted ways at typical or averaged situations but also to take into account fluctuation effects of the medium.

The obtained closed equations (the Kolmogorov equation and that for the coefficient $A$) with the corresponding initial and boundary conditions are solved numerically in the Cartesian coordinate system using the method of fictitious areas, which allow calculations with an arbitrary function describing the relief. For time quantization, the Crank–Nicolson scheme and two-cycle multicomponent splitting method can be used.[6]

Without changing the essence of the matter, for brevity we write Eq. (9) in the form

$$\frac{\partial C}{\partial t} + \mathrm{div}(\mathbf{V}\, C) = \Phi + \frac{\partial}{\partial x} K_x \frac{\partial C}{\partial x} + \frac{\partial}{\partial y} K_y \frac{\partial C}{\partial y} + \frac{\partial}{\partial z} K_z \frac{\partial C}{\partial z}, \qquad (11)$$

where $x = x_1$, $y = x_2$, and $z = x_3$ are the axes of the Cartesian coordinate system; $x$ and $y$ are directed horizontally, $z$ is directed vertically upward; $C = \bar{s}$, $\mathbf{V} = \mathbf{v} - \mathbf{w}_{\mathrm{g}}$, $\mathbf{v} = \mathbf{v}\,\{u = u_1, v = u_2, w = u_3\}$; $K_{ij} = N_{ij}^{(1)} + \bar{\nu}_{ij}$; $\Phi = \bar{F} - \frac{\partial}{\partial x_i} Q^{(1)}$ with the boundary conditions guaranteeing uniqueness of the solution:

$$C = C_0 \text{ at } t = 0;$$

at the lower boundary

$$w_{\mathrm{g}}\, C + N_z \frac{\partial C}{\partial z} = \beta C - \Phi_0$$

for $z = \Delta$; at other boundaries, either (a) $C = C_{\mathrm{b}}$ or (b) $C = C_{\mathrm{b}}$ for $V_{\mathrm{n}} < 0$,

$$\frac{\partial C}{\partial x} = \frac{\partial C}{\partial y} = \frac{\partial C}{\partial z} = 0 \quad \text{for } V_{\mathrm{n}} \ge 0, \qquad (12)$$

where $C_{\mathrm{b}}$ is the background concentration; $V_{\mathrm{n}}$ is the projection of the velocity vector onto the external normal to the boundary surface; $\beta$ is the coefficient describing the interaction between the pollutant and the surface.

Let us consider the method for solving the problem presented by Eqs. (11) and (12). Since the antisymmetric form of the operator is preferable when constructing energetically balanced finite-difference approximations, Eq. (11) is transformed, using the equation of continuity for the incompressible atmosphere, to the following form:

$$\frac{\partial C}{\partial t} + \frac{1}{2}(\mathbf{V}\ \mathrm{grad}\ C + \mathrm{div}\ (C\ \mathbf{V})) = \frac{\partial}{\partial x} K_x \frac{\partial C}{\partial x} + \frac{\partial}{\partial y} K_y \frac{\partial C}{\partial y} + \frac{\partial}{\partial z} K_z \frac{\partial C}{\partial z} + \Phi.$$

Let us introduce a nonuniform grid with main nodes $x_i = i\,\Delta x$ $(i = 0, 1, \ldots, I + 1)$, $y_j = j\,\Delta y$ $(j = 0, 1, \ldots, J + 1)$, $z_k = \Delta z_k$ $(k = 0, 1, \ldots, K + 1)$, and $t_n = n\,\Delta t$ $(n = 0, 1, \ldots)$ and the steps $\Delta x$, $\Delta y$, $\Delta z_k$, and $\Delta t$. We also use auxiliary points $x_{i+1/2}$, $y_{j+1/2}$, $z_{k+1/2}$ situated at the center of the main intervals. Let us designate

$$C_{i,j,k}^n = C(x_i, y_j, z_k, t_n);\ \Delta_k = (\Delta z_{k+1} + \Delta z_k)/2;$$
$$u_{i+1/2,j,k} = (u_{i+1,j,k} + u_{i,j,k})/2;$$
$$v_{i,j+1/2,k} = (v_{i,j+1,k} + v_{i,j,k})/2;$$
$$w_{i,j,k+1/2} = (w_{i,j,k+1} + w_{i,j,k})/2.$$

The finite-difference analogs of the operators are the following:

$$(L_1^n\, C)_{i,j,k} = \frac{u_{i+1/2,j,k}^n\, C_{i+1,j,k} - u_{i-1/2,j,k}^n\, C_{i-1,j,k}}{2\Delta x} - \frac{1}{\Delta x^2}\,[K_{x_{i+1/2,j,k}}^n\,(C_{i+1,j,k} - C_{i,j,k}) - K_{x_{i-1/2,j,k}}^n\,(C_{i,j,k} - C_{i-1,j,k})],$$

$$(L_2^n\, C)_{i,j,k} = \frac{v_{i,j+1/2,k}^n\, C_{i,j+1,k} - v_{i,j-1/2,k}^n\, C_{i,j-1,k}}{2\Delta y} - \frac{1}{\Delta y^2}\,[K_{y_{i,j+1/2,k}}^n\,(C_{i,j+1,k} - C_{i,j,k}) - K_{y_{i,j-1/2,k}}^n\,(C_{i,j,k} - C_{i,j-1,k})],$$

$$(L_3^n C)_{i,j,k} = \frac{(w_{i,j,k+1/2}^n - w_{\mathrm{g}})C_{i,j,k+1} - (w_{i,j,k-1/2}^n - w_{\mathrm{g}})C_{i,j,k-1}}{2\Delta_k} - K_{z_{i,j,k+1/2}}^n \frac{C_{i,j,k+1} - C_{i,j,k}}{\Delta z_{k+1}\,\Delta_k} + K_{z_{i,j,k-1/2}}^n \frac{C_{i,j,k} - C_{i,j,k-1}}{\Delta z_k\,\Delta_k}.$$

Using the Crank–Nicolson scheme at every fractional step $[t_n, t_{n+1}]$, we can write the splitting algorithm in the form

$$\left(E + \frac{\Delta t}{2} L_m^n\right) C^{n+m/4-1} = \left(E - \frac{\Delta t}{2} L_m^n\right) C^{n+(m-1)/4-1},$$
$$m = 1, 2, 3;$$
$$C^{n+1/4} = C^{n-1/4} + \Delta t\ \Phi^n;$$

$$\left(E + \frac{\Delta t}{2} L^n_{5-m}\right) C^{n+m/4} = \left(E - \frac{\Delta t}{2} L^n_{5-m}\right) C^{n+(m-1)/4},$$

$$m = 2,\ 3,\ 4,$$

where $E$ is the unit matrix.

The difference approximation of the problem (3) and (4) is also constructed based on the Crank–Nicolson scheme. Let us designate

$$s_\gamma = \gamma\, \Delta s_\gamma \quad (\gamma = 0,\ 1,\ 2,\ \ldots,\ \Gamma + 1),$$

$$\Delta s_{\gamma+1} = s_{\gamma+1} - s_\gamma,\quad ss_\gamma = (ds_\gamma + ds_{\gamma+1})/2,$$

$$(\Lambda^n\, p)_\gamma = \frac{A^n_{\gamma+1}\, p_{\gamma+1} - A^n_{\gamma-1}\, p_{\gamma-1}}{2ss_\gamma} - \frac{B^n_{\gamma+1}\, p_{\gamma+1} - B^n_\gamma\, p_\gamma}{ss_\gamma\, ds_{\gamma+1}} -$$

$$- \frac{B^n_\gamma\, p_\gamma - B^n_{\gamma-1}\, p_{\gamma-1}}{ss_\gamma\, ds_\gamma} \quad (\gamma = 0,\ 2,\ \ldots,\ \Gamma - 1).$$

The boundary condition $p_0$ is determined from the condition of fulfillment of the probability measure (4) by the trapezium rule.

At the right boundary of the integration domain

$$(\Lambda^n\, p)_\Gamma = \frac{A^n_{\Gamma-1}\, p_{\Gamma-1}}{2ss_\Gamma} + \frac{B^n_\Gamma\, p_\Gamma}{ss_\Gamma\, ds_{\Gamma+1}} - \frac{B^n_\Gamma\, p_\Gamma - B^n_{\Gamma-1}\, p_{\Gamma-1}}{ss_\Gamma\, ds_\Gamma}\ .$$

Thus, the finite-difference approximation has the form

$$\left(E + \Lambda^n \frac{\Delta t}{2}\right) p^{n+1} = \left(E - \Lambda^n \frac{\Delta t}{2}\right) p^n.$$

These finite-difference schemes are absolutely stable, and they are second-order approximations with respect to time and coordinates.

For numerical realization of the finite-difference equations, nonmonotonic simulation run[7] is used.

Thus, Eq. (9) describes the dynamics of mean pollutant concentrations with allowance for fluctuations of input meteorological information, and Eq. (3) estimates the probability of these concentrations (including the excess above the given norms) for the considered time interval and permits us to calculate the flow $\Pi$ of pollutants onto the surface:

$$\Pi = \sum_{i=1}^{k} s_i\, w_{gi}\,,$$

where $k$ is the degree of particle dispersion.

Numerical solutions were tested at the simplifications, which were used in the known analytical solutions. Besides, the calculated results were compared with the data obtained from processing of samples of stable snow cover.

Let us consider particular estimates of sedimentation of solid particles onto the surface. It is well-known that aerosols emitted by anthropogenic sources are considerably polydisperse, and the polydisperse structure determines their physical properties. A particle size distribution is usually described as a part (per cent) $\mathrm{d}f$ of particles whose radii lay in the interval $(r,\ r + \mathrm{d}r)$, i.e., $\mathrm{d}f = f(r)\mathrm{d}r$, under the condition that the particle size distribution function has the property

$$P(0 < r < \infty) = \int_0^\infty f(r)\mathrm{d}r = 1.$$

Usually, the part of aerosols particles, whose radii lay within finite intervals, is experimentally determined, i.e., histograms are obtained instead of continuous probability curves. However, emissions are nonstationary in practice, and thus it is impossible to determine the particle size distribution exactly. So, the distribution is approximated by some analytical law. Theoretically, we can derive an equation describing all aerodisperse systems. However, this equation would contain a great number of coefficients, and the fit of these coefficients for every system would not be expedient. This is why the proposed equations contain as small number of coefficients as possible. As a rule, two coefficients are used: particle size and the degree of aerosol polydispersity. The Roller and Rozin–Rammler equations etc.[8] are examples.

Kolmogorov[8] showed that, proceeding from simple hypotheses about the character of breaking of solid particles, it can be proved that the particle size distribution asymptotically tends to the lognormal distribution as particles become finer. That is why we used the lognormal law to calculate the distribution of solid particles emitted by Irkutsk power plants into the atmosphere. Dusting of ash dumps, open pits, and other plants was ignored in our calculations. There are more than 300 boiler houses in Irkutsk, and 97% of them have smoke stacks from 8 to 35 m high. The diameter of emitted particles is $\le 4 \cdot 10^{-5}$ m, and the average density is 2800 kg/m$^3$. The gravitational sedimentation rate was calculated for every fraction by the Stokes equation

$$w_g = (2\rho_n\, g\, r_n^2)/(9\mu),$$

where $g$ is the free fall acceleration; $\mu$ is dynamic viscosity of the medium; $\rho_n$ and $r_n$ are density and radius of the particles. The gravitational sedimentation rate (calculated by the Stokes equation) ranges from 0.001 to 0.2 m/s depending on the particle size.

More than 80 000 kg/km$^2$ of pollutants deposit annually onto the underlying surface in the north-western part of Irkutsk (Fig. 1). This is the most polluted district of the city. In the south-western district, Novo-Irkutskaya power station provides the most part of thermal power. The smoke stacks of this station are 180 and 250 m in height. Therefore, its emissions are in the atmospheric boundary layer. Only large particles deposit near the station; other particles are transported very far by air flows. Figure 2 shows the estimate of annual accumulation of heavy particles (emitted by Irkutsk power plants) on the surface of Southern Baikal.

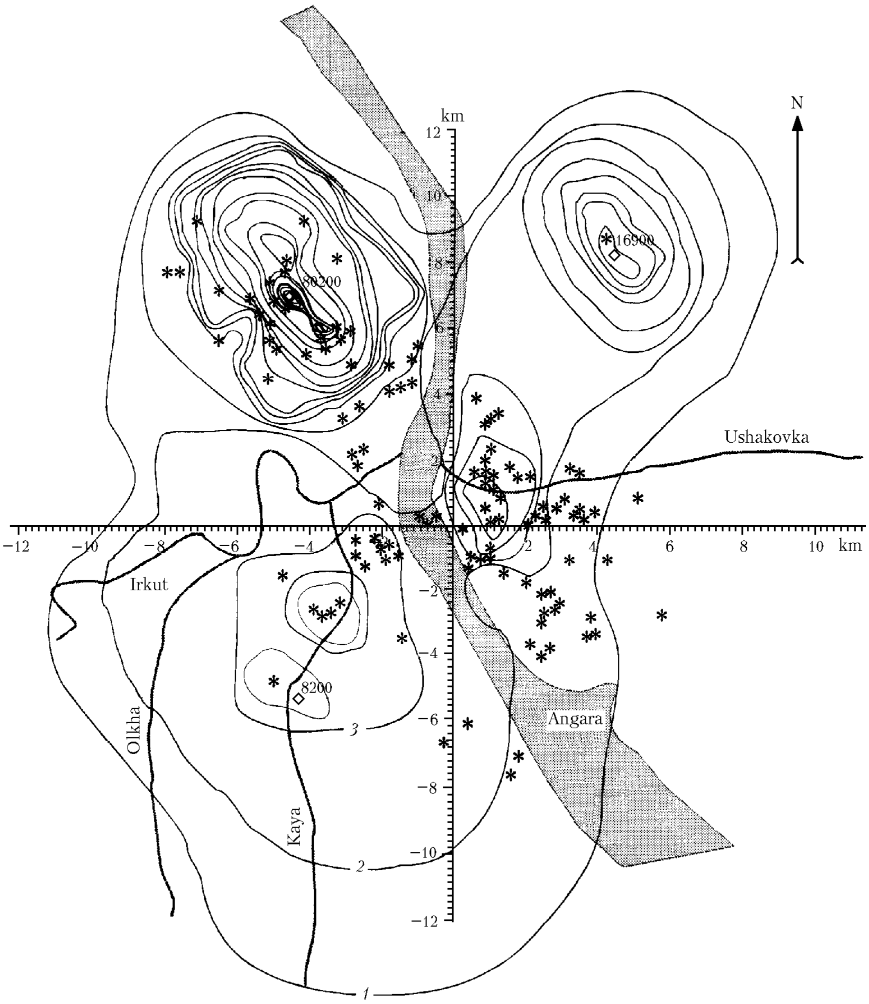


**Fig. 1.** Accumulation of an anthropogenic polydispersive pollutant on the underlying surface in Irkutsk during a year: pollutant sources (∗); local maximums ( ). The isoline *1* corresponds to 2000 kg/km$^2$. Isoline step is 2000 kg/km$^2$.

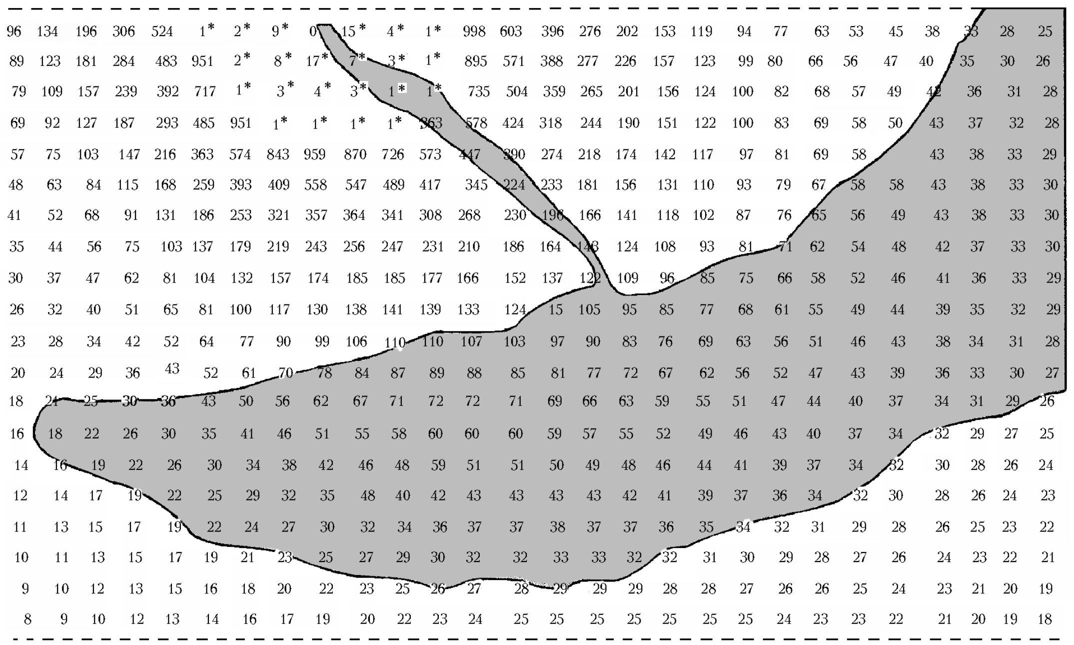


**Fig. 2.** Accumulation of an anthropogenic pollutant (kg/km$^2$) on the underlying surface of Southern Baikal during a year; ∗ is the order of the number $10^3$.

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