Models for reconstruction of long-term aerosol fallout fields

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Few-parameter models for estimation of the local and regional pollution of the environment by aerosol sources have been developed. Similarity of pollutant dissipation processes and statistical characteristics of wind and turbulent regimes in the surface and boundary atmospheric levels has been used. The models were tested with the field data on pollution of snow blanket by heavy metals near the Norilsk Copper-Smelting Plant.

Introduction

Under conditions of permanent growth of the anthropogenic impact on the environment, it is desirable to have various and detailed data on its actual state. When organizing network of observation, one should use the data on existing and planned sources of atmospheric pollution, pollutant characteristics, hydrometeorological and climatic conditions, results of observations in the past, and the information about the long-range transport of pollutants.

The theory and techniques are now developed most thoroughly for calculation of the fields of one-time (mostly 20–30 min) concentrations.[1,2] To determine the fields of seasonally and annually mean concentrations from the one-time concentrations, far more voluminous detailed information on the parameters of atmospheric diffusion, meteorological conditions, intra-year volume of emissions from a source, disperse composition, and transformation of pollutants is necessary. That is why the most papers dealing with calculation of annually mean concentration restrict the consideration to a rough estimation or take into account only individual meteorological factors. Most complete schemes require the use of sampled multidimensional distribution functions, which are determined with a low reliability from data of meteorological observations.[3]

1. Reconstruction of local pollution of a territory

The models for estimation of long-term aerosol pollution of a territory by point and distributed sources are based on the following equation[3]:

$$\bar{q}_{\bar{\tau}} = \int_0^\infty q \rho_{\tau,\bar{\tau}}(q) \mathrm{d}q, \qquad (1)$$

which expresses the relation between the mean concentration $\bar{q}_{\bar{\tau}}$ for a long period (season, year) corresponding to the time interval $\bar{\tau}$, and one-time concentrations q corresponding to a time interval $\tau \ll \bar{\tau}$; $\rho_{\tau,\bar{\tau}}$ is the probability density for one-time concentrations. The value of q is determined from solution of the turbulent diffusion equation.

Frequently occurring meteorological conditions are of key significance in calculation of the mean concentration in the surface atmospheric layer. These conditions include, in particular, so-called normal meteorological conditions, for which the following power approximation of the wind velocity and the coefficient of vertical turbulent exchange is valid[1,2]:

$$u(z) = u_1 \left(\frac{z}{z_1}\right)^n; \quad K_z = k_1 \frac{z}{z_1}, \qquad (2)$$

where u_1 and k_1 are the values of u and K_z at $z = z_1$.

In this case, for a point source we obtain the following representation for the mean near-surface concentration:

$$\bar{q}(r, \varphi) = \iint_\Omega q(r, \varphi, k_1, u_1) p_1(k_1, u_1) \mathrm{d}k_1 \mathrm{d}u_1, \qquad (3)$$

where r, φ are polar coordinates; $p_1(k_1, u_1)$ is the joint probability density of u_1 and k_1 for the period of averaging; Ω is the area of real change of u_1 and k_1;

$$q(r, \varphi, k_1, u_1) = \frac{P(\varphi + 180^\circ) q_\lambda(r, k_1, u_1)}{r}. \qquad (4)$$

Here $P(\varphi)$ is the surface wind rose; q_λ is the one-time concentration for a linear source.

The use of power approximation (2) for the wind velocity and the turbulent exchange coefficient allows $q_\lambda(r, k_1, u_1)$ to be represented in an analytical form for a light pollutant:

$$q_\lambda = \frac{Q}{r(1+n) k_1 \varphi_0 \sqrt{2\pi}} \exp\left[-\frac{u_1 H^{1+n}}{k_1 (1+n)^2 r} \right], \qquad (5)$$

where Q, H are the emission rate and the height of the source; φ_0 is the variance of the wind velocity.

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For a monodisperse pollutant, the concentration q_w is determined by the equation

$$q_w = q_\lambda \chi\left(\frac{k_1}{u_1}r, \frac{w}{k_1}, H\right); \quad \chi = \frac{\left[\frac{u_1}{(1+n)^2 k_1}\right]^\omega H^{(1+n)\omega}}{\Gamma(1+\omega)r^\omega}, \quad (6)$$

where $\omega = w/[(1+n)k_1]$; $\Gamma(1+\omega)$ is the gamma function.

The climatological analysis of the data of gradient observations at a network of thermal-balance stations reveals more detailed structure and properties of the function $p_1(k_1, u_1)$. The following representation takes place[3]:

$$p_1(k_1, u_1) = p'(u_1)p''(\lambda), \quad (7)$$

where $\lambda = k_1/u_1$; p', p'' are one-dimensional probability densities of u_1, λ for the period of averaging.

In the first approximation, we can take that

$$p''(\lambda) = \delta(\lambda - \bar{\lambda}), \quad (8)$$

where $\delta(\lambda)$ is the delta function.

To take into account the stability parameter λ more thoroughly, $p''(\lambda)$ can be specified as a Weibull gamma distribution[3]:

$$p''(\lambda) = \frac{\alpha^{K-1}\lambda^{-K}}{\Gamma(K-1)}e^{-\alpha/K}. \quad (9)$$

For the case of a light pollutant with allowance for Eqs. (4), (5), (7), and (8), it follows from Eq. (3) that

$$\bar{q}(r,\varphi) = \frac{QP(\varphi+180°)}{\sqrt{2\pi}\varphi_0 r^2} \times$$

$$\times \iint_\Omega \frac{1}{n+1}\exp\left[-\frac{H^{n+1}}{\lambda(1+n)^2 r}\right]\lambda p'(u_1)p''(\lambda)\,d\lambda\,du_1 =$$

$$= \frac{QP(\varphi+180°)\bar{\lambda}}{\sqrt{2\pi}(1+n)\varphi_0 r^2}\exp\left[-\frac{H^{n+1}}{\bar{\lambda}(1+n)^2 r}\right]\int_0^u p'(u_1)\,du_1 =$$

$$= \theta_1 \frac{P(\varphi+180°)}{r^2}e^{-\theta_2/r}, \quad (10)$$

$$\theta_1 = \frac{Q\bar{\lambda}}{\sqrt{2\pi}(1+n)\varphi_0}\int_0^u p'(u_1)du_1, \quad \theta_2 = \frac{H^{1+n}}{\bar{\lambda}(1+n)^2}. \quad (11)$$

If observations of the pollutant concentration are available, θ_1 and θ_2 can be determined by the least-squares method.

Consider the case of a monodisperse pollutant. In the variables u_1 and λ, function χ takes the form

$$\chi = \frac{H^{(1+n)\omega_1}}{[(1+n)^2\lambda)]^{\omega_1}\Gamma(1+\omega_1)r^{\omega_1}}, \quad (12)$$

where $\omega_1 = w/[(1+n)\lambda u_1]$.

From Eq. (3) with allowance for Eqs. (4)–(8), it follows that

$$\bar{q}_w(r,\varphi) = \frac{QP(\varphi+180°)\bar{\lambda}}{\sqrt{2\pi}\varphi_0(1+n)r^2}\exp\left[-\frac{H^{n+1}}{\bar{\lambda}(1+n)^2 r}\right]\times$$

$$\times \int_0^u p'(u_1)\frac{H^{(1+n)\bar{\omega}_1} r^{-\bar{\omega}_1}}{[(1+n)^2\bar{\lambda}]^{\bar{\omega}_1}\Gamma(1+\bar{\omega}_1)}du_1, \quad (13)$$

where $\bar{\omega}_1 = w/[(1+n)\bar{\lambda}u_1]$.

Taking into account the similar property of Eq. (8) for $p'(u_1)$, we obtain from Eq. (13) the following regression equation:

$$q_w(r,\varphi) = \theta_{1w}P(\varphi+180°)r^{\theta_{3w}}e^{-\theta_2/r}, \quad (14)$$

where

$$\theta_{1w} = \frac{QH^{(1+n)\omega_2}}{\sqrt{2\pi}\varphi_0(1+n)^{2\omega_2+1}\bar{\lambda}^{\omega_2-1}\Gamma(1+\omega_2)},$$

$$\theta_{3w} = -2-\omega_2, \quad \omega_2 = w/[(1+n)\overline{\lambda_1}\,\overline{u_1}].$$

To estimate θ_{1w}, θ_2, and θ_{3w} in regression (14), observations at least in three sites are necessary. The number of sites can be smaller, if θ_2 is estimated previously for the considered source for the case of a poorly settling pollutant. Parameter θ_2 is, in some sense, external, and it can be estimated previously from the geometric height of the source and dynamic and thermal characteristics of the emitted air–gas mixture.

Note 1. The regression dependence for estimation of the concentration field for a monodisperse pollutant can be constructed without assuming *a priori* the form of $p'(u_1)$ by using the asymptotic expansion method. To do this, $r^{-\bar{\omega}_1}$ is represented as a power series in $\ln r$:

$$r^{-\bar{\omega}_1} = e^{-\bar{\omega}_1 \ln r} \approx 1 - \bar{\omega}_1 \ln r + \frac{\bar{\omega}_1^2 \ln^2 r}{2!}. \quad (15)$$

For low sedimentation rates of aerosol particles, the consideration can be restricted to the first terms of the series (15). For example, for two first terms of the series, we obtain the following regression dependence:

$$\bar{q}_w(r,\varphi) = \frac{\theta'_{1w} + \theta'_{3w}\ln r}{r^2}e^{-\theta_2/r}P(\varphi+180°), \quad (16)$$

where

$$\theta'_{1w} = \frac{Q\bar{\lambda}}{\sqrt{2\pi}\varphi_0(1+n)}\int_0^u p'(u_1)\frac{H^{(1+n)\bar{\omega}_1}}{[(1+n)^2\bar{\lambda}]^{\bar{\omega}_1}\Gamma(1+\bar{\omega}_1)}\mathrm{d}u_1,$$

$$\theta'_{3w} = \frac{-Q\bar{\lambda}}{\sqrt{2\pi}\varphi_0(1+n)}\int_0^u p'(u_1)\frac{H^{(1+n)\bar{\omega}_1}\bar{\omega}_1}{[(1+n)^2\bar{\lambda}]^{\bar{\omega}_1}\Gamma(1+\bar{\omega}_1)}\mathrm{d}u_1.$$

Note 2. For a polydisperse pollutant, estimation of mean concentration fields is not a much more complicated problem, if we know the distribution function of the sedimentation rates for the pollutant emitted by some aerosol source. Representing the distribution function $R(w)$ in the discrete form as N fractions with the mean sedimentation rates w_i and weights ρ_i, $i = \overline{1,N}$, we can write the problem of determination of the field of long-term aerosol fallouts as[4,5]

$$\bar{q}_w(r,\varphi) = \frac{\tilde{\theta}_1}{r^2}\mathrm{e}^{-\theta_2/r}P(\varphi+180^\circ)\sum_{i=1}^{N}\rho_i\frac{\theta_2^{\bar{w}_i\tilde{\theta}_3}}{\Gamma(1+w_i\tilde{\theta}_3)}r^{-\bar{w}_i\tilde{\theta}_3}, \tag{17}$$

where

$$\tilde{\theta}_1 = \frac{Q\bar{\lambda}}{\sqrt{2\pi}\varphi_0(1+n)}, \quad \tilde{\theta}_3 = \frac{1}{(1+n)\bar{\lambda}\bar{u}_1}.$$

If the distribution function $R(w)$ is unknown, the dimension of the problem of estimating $\overline{q_w}(r,\varphi)$ based on Eq. (17) increases significantly, if ρ_i, $i = \overline{1,N}$, are included in the list of the sought parameters. At small N, the solution of this problem does not require too voluminous experimental information about the concentration fields of aerosol pollutants. The method of estimation based on the *a priori* setting of $R(w)$ depending on a small number of parameters seems to be more realistic.

Separation of different zones of aerosol fallouts depending on the distance from some source is also a rather efficient method, indirectly taking into account the polydisperse property of a pollutant. In this case, the pollutant is assumed to be monodisperse within a zone, and parameters of model (14) are estimated locally with the use of observations from this zone.[6–8] The method proposed should be applied, if there is rather large number of observation sites.

2. Reconstruction of regional aerosol pollution

The concentration field from a point source averaged over some long period is determined as[6]:

$$\bar{q}(r,\varphi,z) = \frac{S(r,z)g(\varphi)}{2\sqrt{\pi k_0 r}}\int_{-\Delta}^{\Delta}\exp\left[-\frac{r\sin^2\psi}{4k_0}\right]\mathrm{d}\psi, \tag{18}$$

where r, φ are the polar coordinates originating at a source location; $g(\varphi)$ is the probability of the wind direction opposite to φ; Δ is some small angle characterizing the widening of a plume in the direction transversal to the wind; $S(r, z)$ is the solution of the semiempiric equation of turbulent diffusion in the boundary atmospheric layer.[1,2]

For typical $k_0 = 0.5$–1 m, $\Delta < 10^\circ$–15° at $r > 1$ km

$$\bar{q}(r,\varphi,z) = FS(r,z)g(\varphi)/r. \tag{19}$$

Here F is almost independent of r.

Equation (18) permits further simplifications for a poorly settling and inert pollutant. In this case, at a distance of 7–10 km from a source, $S(r, z)$ is almost independent of z. Then with allowance for Eq. (19), we obtain for the density of pollutant fallout onto the underlying surface

$$P(r,\varphi) = \theta g(\varphi)/r, \tag{20}$$

where $\theta = \beta Q/(2\pi uh)$, β is the coefficient of pollutant interaction with the underlying surface; u and h are the wind velocity and the thickness of the mixing layer, respectively.

Note 3. It should be noted that rather serious conditions for estimation of pollutant concentration fields are the optimal arrangement of sampling sites or selection of most informative observations.[9] The fulfillment of these conditions allows the influence of measurement errors on the accuracy of parameter estimation to be diminished. The methods and algorithms for construction of optimal plans, as well as the numerical analysis of sensitivity of estimation models for particular gas–aerosol sources and observation plans can be found in Refs. 4, 5, 10, 11.

3. Analysis of territory pollution by emissions from the Norilsk Copper-Smelting Plant

Atmospheric industrial emissions form the main source of environmental pollution in the Norilsk industrial region. Metallurgical industry is responsible for about 70% of all emissions.[12] The mass of dust emissions exceeds 30 000 ton a year. The dust pollution of the snow blanket changes its albedo and brightness characteristics, due to which pollution aureoles are rather clearly seen in space images. They may be up to 100 km long.[13] The chemical composition of the solid sediment of the snow water is characterized by the presence of heavy metals, such as iron, copper, nickel, manganese, chrome, zinc, lead, and cadmium.[14]

The data of field investigation of the snow blanket pollution near the Norilsk Copper-Smelting Plant[14] are used to test the proposed methods for estimation of the aerosol pollution of some territory. Local model (14) was used to reconstruct the fields of aerosol fallouts in the close zone. Fallouts in the far zone (> 10 km) were estimated by model (20). The results of numerical reconstruction of the content of heavy metals in snow along the sampling route near the plant are shown in Figs. 1–3.

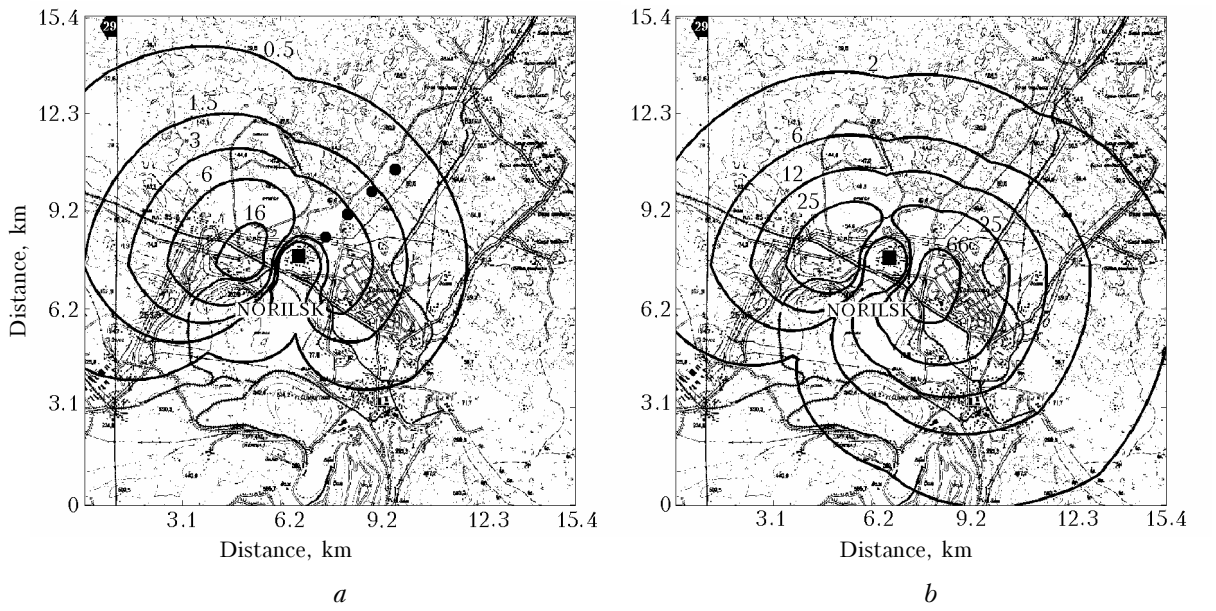

Fig. 1. Reconstructed fields of nickel aerosol fallout from the Norilsk Copper-Smelting Plant: (*a*) for winter period (mg/l); (*b*) for summer period (in % of the maximum value); emission source is shown by square ■, snow sampling points are shown by circles ●.

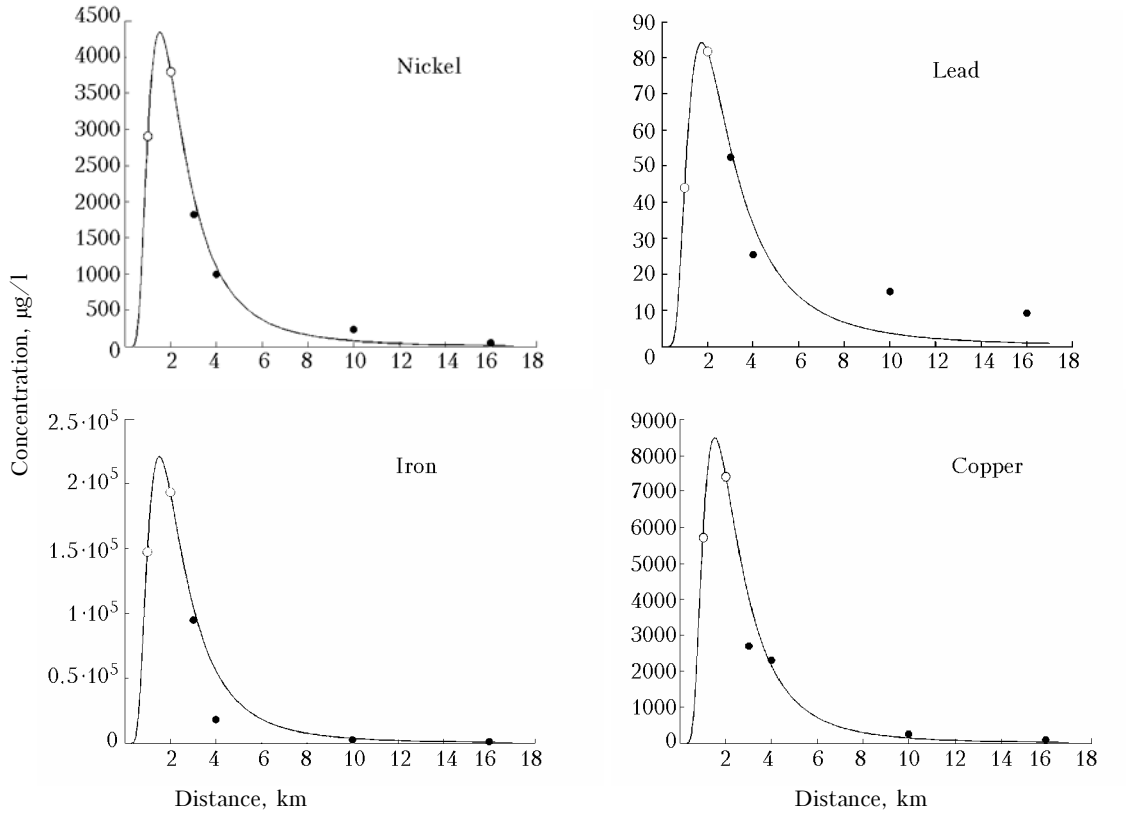

Fig. 2. Content of heavy metals in the solid sediments of snow water in the north-eastern direction from the Norilsk Copper-Smelting Plant. Nickel, lead, iron, and copper concentrations were reconstructed by the local model: reference points (○), control observation points (●), calculated concentration (——).

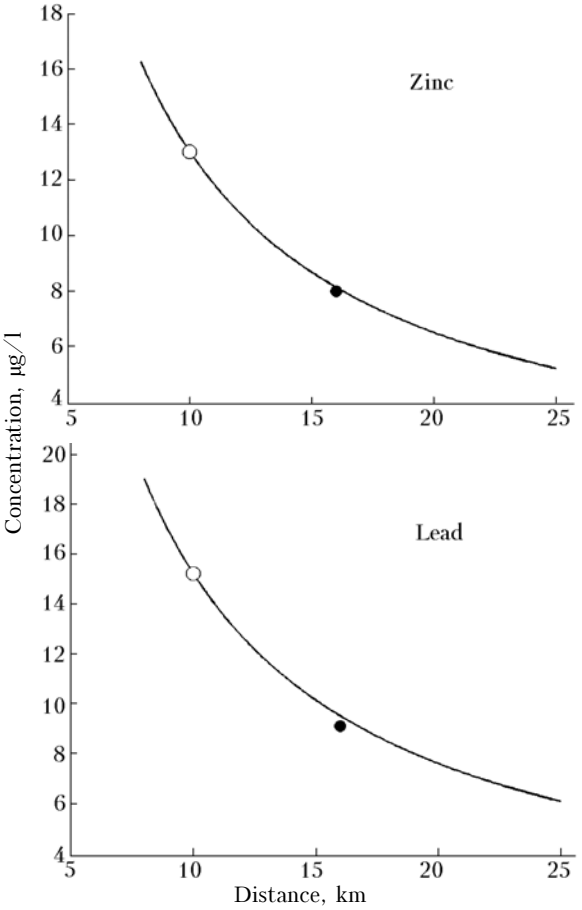

Fig. 3. Zinc and lead content reconstructed by the model of regional transport (20).

It should be noted that the experimental data (see Fig. 3) are insufficient to estimate the efficiency of the model of regional transport (20). In the case considered, the closeness of the observations to the calculated curve demonstrates the quantitative tendency of the concentration dynamics at long distances from the plant. To stronger confirm this regularity, additional experimental investigations are necessary. The more thorough experimental confirmation of model (20) for point and area sources is presented in Refs. 8 and 15.

The results of numerical simulation confirm rather high level of agreement between the measured and calculated concentrations. The fallout of heavy metals at short distances is presented mostly by coarse aerosol fractions, while in the zone farther than 10 km heavy metals are transported by poorly settling aerosol particles. The analysis of Fig. 1 shows that the Norilsk territory is subjected to the intense pollution by aerosol emissions from the Norilsk Copper-Smelting Plant in all seasons. To be noted is also the very bad mutual arrangement of the plant and the city.

Conclusions

The theoretical investigation and analysis of the data of field observations demonstrate that the fields of long-term pollution of some territory can be described quite adequately within the framework of rather simple regression dependences. The procedure of aggregation of unknown parameters significantly increases the efficiency of solution of the inverse problems of pollutant transport. With very limited input information, the quantitative models describing the formation of fields of long-term aerosol pollutant fallout near a particular anthropogenic source have been developed.

The obtained quantitative regularities of aerosol pollution near some anthropogenic source can be used to establish relations with brightness characteristics of snow blanket when analyzing the data of remote sensing of the Earth's surface in the visible region.

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