THE MODEL OF LONG-TERM TERRAIN CONTAMINATION BY EMISSIONS FROM AEROSOL SOURCES

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The model of reconstructing the terrain contamination by emissions from aerosol sources has been constructed based on the solution of inverse problem of atmospheric transport of the contaminants and the wind velocity data. The model proposed is tested based on the data of observations of heavy metal contamination of snow and soil in the vicinity of the Belovo Zinc Plant.

The problem of reconstruction of the environmental contamination based on the observation data is rather complicated since the characteristics of the environmental contamination sources, the disperse composition of aerosol emissions, and meteorological conditions are not always known. The approaches to solving this problem, taking into account the peculiar features of the contamination conditions, can be found in papers 1 – 6. In this paper we describe a model of the terrain long-term contamination by emissions from stationary aerosol sources based on the solution of the inverse problem of transport in the atmospheric boundary layer. For describing the process of aerosol impurity propagation we use analytical solutions of a semiempirical equation of turbulent diffusion for power approximations of wind velocity and coefficients of turbulent exchange. Such an approach, with the consideration for characteristics of the plume spread and occurrence of wind direction relative to the source, enables us to reduce the problem of reconstruction of the aerosol precipitation density field to a nonlinear regressive dependence with a relatively small number of required parameters.

The proposed model of reconstruction has been tested on the observation data of heavy metal sedimentation density in the vicinity of the Belovo Zinc Plant in Kemerovo Region. The calculations, made based on the snow sampling in the north and northwest directions from the emission source, show good agreement between the calculated and measured values of concentrations at reference points. Reconstruction of sedimentation density of zinc, lead, and cadmium in the soil was performed based on the dependence found for snow. The regressive dependences of sedimentation density have made it possible to estimate the emission of heavy metals during the winter season.

1. FORMULATION OF THE INVERSE PROBLEM

In this section the dependence has been constructed, describing the aerosol component density in the vicinity of stationary point sources over a long period of time and containing a moderate number of parameters. The following simplifying proposals should be made:

1) Emission of an aerosol component occurs from stationary sources, whose locations and heights are known.

2) The level of contamination of soil and snow at a definite local point over a period considered is proportional to the frequency of occurrence of wind directions relative to the source over this period.

The field of aerosol contamination density is determined by the following formula:

$$p(r, \varphi) = c\, \bar{q}(r, \varphi), \qquad (1)$$

where r and φ are the polar coordinates of the calculated point with the origin at the source, p is the aerosol contamination density, $\bar{q}$ is the mean surface concentration, and c is the parameter characterizing the contamination sedimentation to the underlying surface.

According to Refs. 1 and 6 the field $\bar{q}(r, \varphi)$ of averaged over a long period concentrations from the point source is determined by the following formula:

$$\bar{q}(r, \varphi) = q(r)\, g(\varphi) \int_{-\Delta}^{\Delta} e^{-r \sin^2\Psi/(4k_0)}\, d\Psi. \qquad (2)$$

Here $q(r)$ is the axial concentration from the point source, $g(\varphi)$ is the probability of the wind along the direction opposite to φ, Δ is a small angle characterizing the plume spread in the direction opposite to the wind direction, and k_0 is the parameter of the lateral broadening of the plume.

Because the angle Δ is small, assuming $\sin \Psi \approx \Psi$, Eq. (2) can be transformed as follows:

$$\bar{q}(r, \varphi) = [F(r)\, g(\varphi)\, q(r)] \,/\, r^{1/2}, \qquad (3)$$

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$$F(r) = 4\, k_0^{1/2} \int_0^{\xi(r)} e^{-v^2}\, dv, \qquad (4)$$

$$\xi(r) = \Delta \sqrt{r/(4k_0)}\ . \qquad (5)$$

For the characteristic values of $k_0 = 0.5 - 1$ m, $\Delta < 10 - 15°$ the function $F(r)$ is practically constant at $r > 1$ km.[1,6]

By approximating the wind velocity $u(z)$ and the vertical turbulent exchange $\nu(z)$ using power functions of the view

$$u(z) = u_1 (z/z_1)^n, \quad \nu(z) = k_1 (z/z_1), \qquad (6)$$

the concentration $q(r)$, taking into account the contaminant deposition, can be presented by the following analytical expression[1]:

$$q(r) = q_l(r)\, \chi(r, \omega), \qquad (7)$$

where

$$q_l(r) = \frac{M}{2(1+n)\, k_1 \sqrt{\pi k_0}\, r^{3/2}}\, e^{-2r_m/r},$$

$$\chi(r, \omega) = \frac{(2r_m/r)^{\omega}}{\Gamma(1+\omega)}\ , \quad r_m = \frac{u_1 H^{1+n}}{2k_1 (1+n)^2}\ ,$$

$$\omega = \frac{w}{k_1(n+1)}\ ,$$

M is the source power, r_m is the point of maximum surface concentration, H is the effective source height, w is the rate of gravitational sedimentation of aerosol particles, and $\Gamma(1+\omega)$ is the gamma-function.

If we limit our consideration by the mean annual or mean winter wind velocity and temperature and characteristic of these conditions coefficients of turbulent exchange, then according to Ref. 1, the quantity r_m can be estimated by the source geometric characteristics and the parameters of outgoing gas – air mixture.

Taking into account Eqs. (1), (3), (4), and (7), the mixture precipitation density can be represented by a simple regressive expression:

$$p(r, \varphi, \boldsymbol{\theta}) = g(\varphi)\, f(r, \boldsymbol{\theta}), \qquad (8)$$

$$f(r, \boldsymbol{\theta}) = \theta_1\, r^{\theta_2}\, e^{-2r_m/r}, \qquad (9)$$

$\boldsymbol{\theta} = (\theta_1, \theta_2)$ is the vector of unknown parameters;

$$\theta_1 = \frac{c\, M\, F\, (2r_m)^{\omega}}{2\,(1+n)\, k_1 \sqrt{\pi k_0}\, \Gamma\,(1+\omega)}\ , \quad \theta_2 = -\omega - 2.$$

As to these observations, with consideration for Eq. (8) we assume that

$$s_k = p(r_k, \varphi_k, \boldsymbol{\theta}) + \xi_k, \qquad (10)$$

where ξ_k are the measurement errors;

$$E[\xi_k] = 0, \quad E[\xi_k\, \xi_{k'}] = \delta_{kk'}\, \sigma_k^2, \quad k, k' = \overline{1,N}\ . \qquad (11)$$

Here E is the operation of mathematical expectation; δ_{ki} is the Kronecker symbol. By the solution of inverse problem (8)–(11) are meant the estimates of $\hat{\boldsymbol{\theta}}_N$ bythe method of the least squares and the function $\hat{p} = p(r, \varphi, \hat{\boldsymbol{\theta}}_N)$. The estimates of the method of the least squares present the minimum of the function:

$$J_N(\boldsymbol{\theta}) = \sum_{k=1}^{N} \sigma_k^{-2}\, [s_k - p(r_k, \varphi_k, \boldsymbol{\theta})]^2. \qquad (12)$$

The solution of the problem (8)–(12) is not very difficult if the linearity of the function (8) is considered with respect to θ_1. Then, by eliminating from (12) θ_1, due to the necessary condition of minimum we obtain the problem of minimizing of the function of one variable θ_2.

Comment 1. In the case of several sources the expression for total density of aerosol precipitation is of the form:

$$p(r, \varphi, \lambda) = \sum_{k=1}^{M} g(\varphi - \varphi_k)\, f_k(r - r_k, \Theta_k), \qquad (13)$$

where M is the number of sources; r_k, φ_k are the polar coordinates of these sources; λ is the vector of unknown parameters of $2M$ dimensionality consisting of the components of vectors Θ_k.

The number of unknown parameters may be reduced markedly, if the assumption is made that the mean size of aerosol particles is identical for all the sources. In this case, in view of Eq. (8) the number of unknown parameters in the regression (13) equals $M + 1$. Further decrease in the number of regression parameters (13) can be obtained based on *a priori* information on a relative emission of gaseous contamination for the sources considered.

2. RECONSTRUCTION OF THE HEAVY METAL CONTAMINATION PATTERN NEAR BELOVO ZINC PLANT

The proposed model of reconstruction of aerosol contamination density in snow and soil is being tested in the vicinity of Belovo Zinc Plant in Kemerovo Region. The Zinc Plant is located to the north from the down town area of Belovo. The major part of heavy metal aerosol plume from the plant industrial area is emitted from a stack of 45 height. In this case for the weightless component and the mean annual wind velocity the distance r_m from the stack is about 700 m.

Estimation of the snow cover contamination

In the spring of 1987 in the vicinity of the plant the snow was sampled for determining the concentrations of zinc, lead, and cadmium. The sampling points were located at 8 bearings, far removed from the main emission source, up to 10 km.

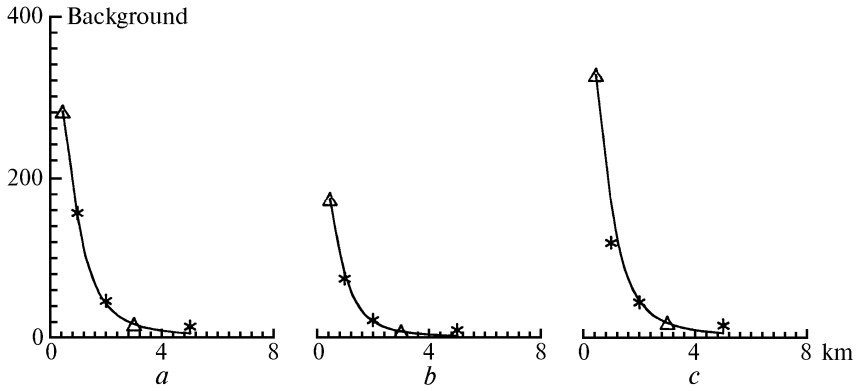

FIG. 1. Concentration of zinc (a), lead (b), and cadmium (c) (the ratio to the background concentration) in snow at a point to the north of the plant. Δ and ∗ present the data of observations.

The regression parameters (8) were estimated at two observation points, located at distances 0.5 and 3 km from the emission source to the north of the plant and denoted by triangles in Fig. 1.

As a result of solution of the set of equations (8)–(12) based on the observation data of zinc, lead, and cadmium concentrations in snow we have obtained the following estimates of the parameters:

$$\Theta_1^z = 619, \quad \Theta_1^l = 330, \quad \Theta_1^c = 694,$$
$$\Theta_2^z = -2.9, \quad \Theta_2^l = -3.1, \quad \Theta_2^c = -2.95, \qquad (14)$$

which enable one to reconstruct on the basis of Eqs. (8), (9) the level of heavy metal contamination of snow cover in the vicinity of the plant. Figure 1 shows the concentrations of zinc, lead, and cadmium aerosols to the north of the plant for the values of parameters (14). The data of observations at the reference points located at distances of 1, 2 and 5 km are denoted in Fig. 1 by asterisks. The analysis of estimations has shown that the agreement between the calculated and measured concentrations at reference points at a distance up to 3 km is rather good. The difference is of the order of 5%. At the point spaced at 5 km far from the source, this difference is larger, that is explained by an inadequate consideration of the influence of polydisperse nature of emitted aerosol at large distances.

Reconstruction of the level of soil contamination

Sampling of soil in the vicinity of the plant was performed by the research workers from the Institute of Soil Science and Agricultural Chemistry SB RAS in the second half of 1980ths, mainly in 1987. For the numerical analysis 13 sampling points were chosen at a 3.5 km from the stack. Such a sampling is due to a sufficiently good agreement of calculated and measured concentrations of heavy metals in snow for these distances. These points are mainly located at the center of Belovo. In this case, the parameter Θ_2, representing the degree of line curvature, given by Eq. (8), was not calculated. The parameter value was taken equal to the concentration value obtained for snow. The stretch coefficient Θ_1 was determined based on the observation data using the method of the least squares.

Figure 2 shows the data of observations and calculation of zinc concentration in soil normalized to the annual mean frequency of occurrence of wind directions. The analysis of the figure shows a good agreement between the calculations and observations.

Figure 3 presents the isolines of zinc concentration in soil in units of maximum permissible concentration (MPC) taking into account the obtained estimates of the average annual wind rose. The MPC value in soil for zinc concentration is 300 mg/kg. Analysis of Fig. 3 shows that the range of values of zinc concentration, exceeding MPC, covers practically all central part of Belovo. The zinc concentration grows rapidly in the vicinity of the source and can reach some tens of MPC. The area of very high concentration is located to the north of the plant depending on directions of winds prevailing over a period of one year.

Comment 2. Taking into account good agreement between the calculations and observations of heavy metal concentration in snow Eqs. (8), (9), and (14) enables us to estimate the total emission of zinc over a winter period by the formula

$$S = \rho \int_0^{2\pi} \int_{r_1}^{r_2} p(r, \varphi, \Theta)\, r\,dr\,d\varphi\,. \qquad (15)$$

Here S is the total emission; ρ is the surface density of a heavy metal in snow; r_1, r_2 are the lower and upper applicability limits of Eq. (8), respectively.

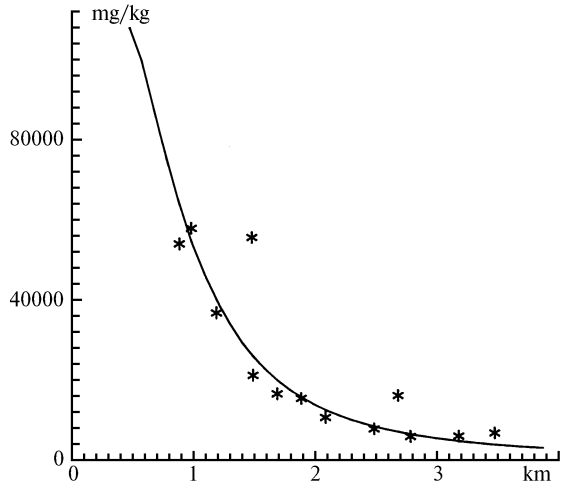

FIG. 2. Calculated and measured concentrations (mg/kg) *of zinc in soil,* * *denote data of observations.*

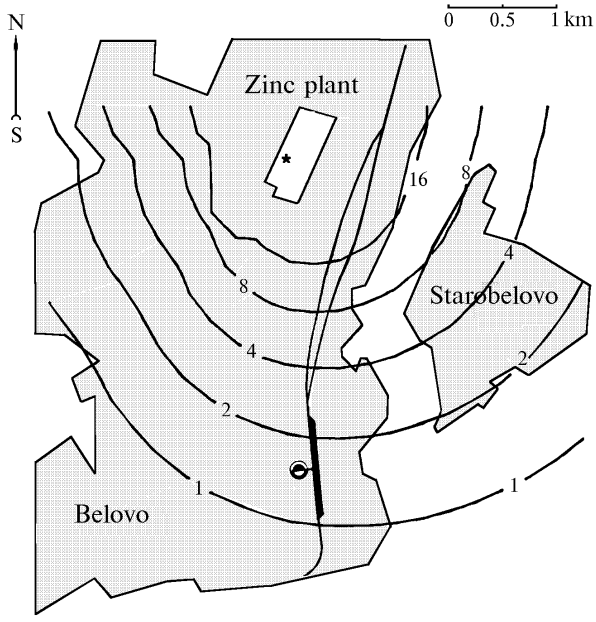

FIG. 3. Isolines of zinc concentration in soil in units of maximum permissible concentration. * *denotes the emission source.*

The value S obtained with the use of Eq. (15) should be considered as the lower estimate of the total emission since at large distances the contribution of emitted aerosol was not taken properly into account, and in the near area the contribution of low sources is not considered.

3. CONCLUSION

The numerical modeling performed on the basis of the data of field observations makes it possible to draw the following conclusions:

– the heavy metal contamination of snow and soil in the vicinity of the zinc plant mainly comes from the high aerosol source,

– the size spectra of the emitted aerosol is highly variable and for further studies of the environmental contamination the spectra should be determined,

– comparison of the observational data of heavy metal contamination of snow and soil essentially increases the reliability of the estimates. The reconstructed pattern of snow and soil contamination confirms the validity of the assumptions 1) and 2). Taking into account the location of the zinc plant, the relief, the climate conditions, the efficiency of the use of the snow cover monitoring should be noted for the control over heavy metal emissions and the contamination level in the vicinity of the plant.

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