Analysis of aerosol fallouts in the vicinity of Novosibirsk heat and power stations

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The data of experimental study of snow cover contamination in the vicinity of some basic heat and power production plants of Novosibirsk are discussed. The fallout fields of dust, heavy metals, macro components, and polyaromatic hydrocarbons are reconstructed based on models of long-term pollution and experimental observations. A good agreement is demonstrated between the calculated and measured concentrations in areas located close to and far from the sources.

Introduction

The air in many Siberian cities is highly polluted in winter. In the first turn, this is true for the cities, which are heated by burning coal. When large amounts of toxic combustion materials are emitted to the atmosphere and the conditions are unfavorable for pollutant spreading (low negative temperatures, gentle wind, complex orography, etc.), smog situations are often observed in these cities, which is of high potential hazard to human health. This causes the need in studying the composition of pollutants, as well as the regularities of their dispersal in the urban air basin.[1–4]

The heat and power production system of Novosibirsk includes several large heat and power plants (HPP), as well as about 300 small and medium boiler houses. Aerosol emissions from these objects contribute significantly to atmospheric pollution in the city and its suburbs.

The main components of emissions are coal ash, heavy metals, sulfur and nitrogen oxides, polyaromatic hydrocarbons, etc.[5,6]

Normally, HPP use coal of Kuznetsk basin and brown coal from Kansk-Achinsk coalfield. In spite of relatively efficient fuel combustion and fume cleaning, HPP emit the majority of pollutants to the city atmosphere.[5,6] It is just this circumstance that has determined the priorities in choosing the objects for our study in Novosibirsk.

This paper analyzes the of experimental data on snow cover pollution with emissions from TETs-2, TETs-3, and TETs-5 heat and power production plants. Combustion products are emitted through 100 to 260-m tall stacks, which leads to pollutant spreading over a large territory in the city and its suburbs.

1. Characterization of objects of the study and sampling routes

All the studied HPP are located in the city. The plants TETs-2 and TETs-3 are close to each other in the northwestern part of Novosibirsk and the zone of their effect covers the major part of the city. Gas-air mixtures at TETs-2 are mostly emitted through two closely spaced smoke stacks 100 and 120 m-tall and 5.1 and 8 m in diameter, respectively. Kuznetskii coal is predominantly used as a fuel. TETs-3 is located several hundreds of meters to the north from TETs-2. The gas-air mixtures here are mostly emitted through 120-m-tall and 8.2 m in diameter smoke stack; the predominant fuel at this station is brown coal from the Kansk-Achinsk coalfield.

TETs-5 is located in the eastern suburbs. The fuel for its boilers is Kuznetskii coal of different kinds. The exhaust gas from boilers is emitted through a 260-m-tall and 10.8 m in diameter smoke stack.

Route snow sampling was carried out along the directions of dominant winds in late winter of 2001–2002 on March 1 in the vicinity of TETs-2 and TETs-3 and on March 15 in the vicinity of TETs-5. The data of snow sampling are given in Table 1 and Fig. 1.

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Table 1. Characteristics of the results of snow sampling near Novosibirsk HPP

Point	Distance, km	Sample mass, g	BP concentration, ng/kg
TETs-2, north-north-east direction			
1	0.9	740	68.4
2	1.6	510	44.0
3	2.3	705	42.2
4	3.2	680	18.0
TETs-3, northern direction			
1	0.5	650	16.0
2	1.9	780	16.1
TETs-5			
1	1.3	860	14.0
2	2.2	870	501
3	3.4	800	28.0
4	7.2	1080	22.0
5	16	920	8.0
6	11	1200	20.0
7	3.3	810	26.0
8	4.8	720	31.0

Note. Sampling area for TETs-2 and TETs-3 was 0.98 dm^2 and for TETs-5 it was 1.37 dm^2.

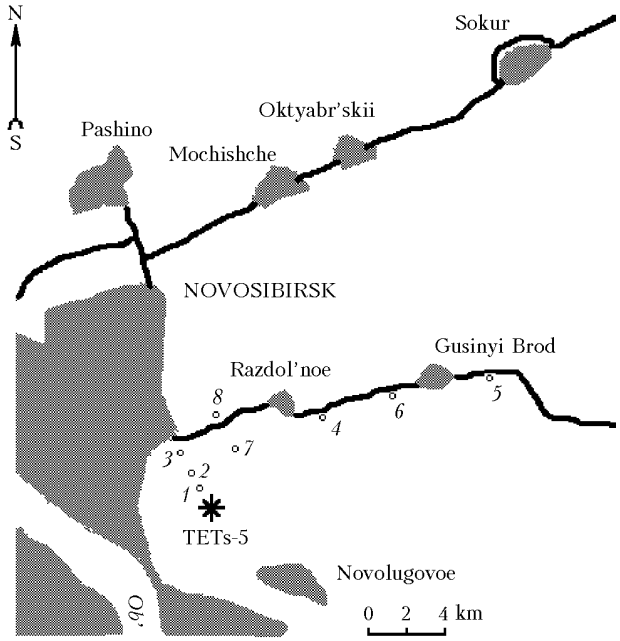

Fig. 1. Schematic of the route snow samplings in the vicinity of TETs-5; sampling site (○).

The sampling points were chosen located on open places with the allowance for buildings, neighboring local sources (highways, private sector, small boilers), forest and park zone. Fulfillment of the above conditions within a city is a difficult task, and the success achieved in its completion is usually determined at the last stages of the study when interpreting experimental findings. The observation conditions turned out most favorable in the zone of influence of TETs-5, and this allowed us to examine a significant territory up to 16 km far from the plant. This was favored by the following factors: high altitude of emission, suburban location of the plant, road system oriented sufficiently convenient for the sampling.

Before analyzing at the chemical laboratory, the snow samples were stored frozen. Melting was carried out at the temperature of 50–60°C. Two schemes were used for analysis. To determine inorganic components, a melted sample was filtered through a membrane filter (∅ 0.45 μm). The obtained sediments were dried in air and then analyzed along with the filtrate. The relative standard deviation S_r was 0.03–0.05 for the techniques of Ca, Mg, Na, and K determination; 0.04–0.06 for HCO_3^-; 0.05–0.10 for $NO_3^-$and Cl^-; 0.07–0.12 for NH_4^+ and SO_4^-; and 0.15–0.30 for Ba, Fe, Cd, Mn, Cu, Pb, Cr, Ni, and Be. Determination of PAH was carried out after prior extraction into the dichlorethane from the whole volume of a non-filtered sample.[7] The standard deviation S_r for PAH was 0.15–0.30. All the components determined in snow samples are tabulated in Table 2.

Table 2. Methods of analysis and components determined in snow samples

Components	Method
pH, HCO_3^-	Potentiometry (Anion-410)
SO_4^{2-}, Cl^-, NO_3^-	Ionic chromatography (Waters, U.S.)
Ca, Mg, Na, Zn, K	Flame atomic absorption spectrophotometry (Hitachi 8000, Japan)
Ba, Fe, Cd, Mn, Cu, Pb, Cr, Ni, Be	Atomic emission spectroscopy with arc excitation (PGS-2, Germany)
NH_4^+	Photocolorimetry (SQ-118, Merck, Germany)
Polyaromatic hydrocarbons (PAH)	Chromato-mass-spectrometry (Hewlett Packard, Germany)

2. Formulation of the inverse problems

The following equation is the basic one when developing the models for studying snow pollution reconstructed from the data on concentrations[8]:

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

This equation expresses the relation between the mean concentration $\bar{q}_{\bar{\tau}}$ for a long period $\bar{\tau}$ and snap sample concentrations q in the interval $\tau \ll \bar{\tau}$, $\rho_{\tau,\bar{\tau}}$ is the probability density for snap sample concentrations.

The q values can be found from solution of the stationary equation of turbulent diffusion[9,10]

$$u\cdot\frac{\partial q}{\partial x} - w\cdot\frac{\partial q}{\partial z} = \frac{\partial}{\partial y}K_y\frac{\partial q}{\partial y} +$$

$$+\frac{\partial}{\partial z}K_z\frac{\partial q}{\partial z} + Q\delta(x)\delta(y)\delta(z-H). \qquad (2)$$

Here the x axis is oriented along the wind direction averaged over the period τ; u is the wind speed; w is the rate of gravitational sedimentation of particles;

K_y and K_z are the exchange coefficients along the y and z axes, Q is the emission rate of a source located at the point $x = y = 0$, $z = H$, δ is the delta function.

(a) Local pollution

Frequently occurring meteorological conditions are of the primary importance in calculating the mean concentrations in the atmospheric surface layer. These conditions include the so-called normal meteorological conditions, to which the following power approximations of the wind velocity and the coefficients of vertical turbulent exchange are applicable[10]

$$u(z) = u_1\left(\frac{z}{z_1}\right)^n, \quad K_z = K_1\frac{z}{z_1}, \tag{3}$$

where u_1 and K_1 are the values of u and K_z at $z = z_1$.

Passing on to the polar coordinates in Eq. (2) and taking into account Eqs. (1) and (3), we obtain the following representation of the mean near-surface concentration[8]:

$$\bar{q}(r,\varphi) = \iint\limits_{\Omega} q(r,\varphi,K_1,u_1)\,P_1(K_1,u_1)\,\mathrm{d}K_1\,\mathrm{d}u_1\,, \tag{4}$$

where r and φ are the polar coordinates; $P_1(K_1,u_1)$ is the joint probability density of K_1 and u_1 for the period of averaging; Ω is the domain of actual variability of K_1 and u_1;

$$q(r,\varphi,K_1,u_1) = \frac{P(\varphi+180^\circ)\,q_\Lambda(r,K_1,u_1)}{r}. \tag{5}$$

Here $P(\varphi)$ is the near-surface wind rose; q_Λ is the snap sampling concentration from a linear source.

The use of power-law approximations (3) for the wind velocity and the turbulent exchange coefficient allows us to represent $q_\Lambda(r,K_1,u_1)$ in the following analytical form:

$$q_\Lambda = \frac{Q}{(1+n)K_1\varphi_0 r\sqrt{2\pi}}\exp\left(-u_1H^{1+n}/K_1(1+n)^2r\right). \tag{6}$$

It is rather difficult to perform calculations based on Eqs. (5) and (6) because they include a large number of parameters, which are either unknown or call for significant refinement. In this connection, it is quite useful to convert Eq. (5) into a more convenient form using the generalized integral mean value theorem,[11] which allows the mean concentration to be represented as:

$$\bar{q}(r,\varphi) = q\left(r,\varphi,\bar{K}_1,\bar{u}_1\right)\iint\limits_{\Omega} P_1(K_1,u_1)\mathrm{d}K_1\mathrm{d}u_1\,, \tag{7}$$

where $\bar{K}_1$, $\bar{u}_1$ are the mean values of the parameters in the domain Ω.

Assuming that the pollutant concentration in snow $\Phi(r,\varphi)$ is proportional to its concentration in the atmosphere and taking into account Eqs. (5)–(7), one obtains the following regression:

$$\Phi(r,\varphi,\boldsymbol{\theta}) = S(r,\boldsymbol{\theta})\,P(\varphi + 180^\circ), \tag{8}$$

where

$$S(r,\boldsymbol{\theta}) = \theta_1\,r^{\theta_2}\exp\,(-2r_m/r), \tag{9}$$

$$\theta_1 = \frac{cQB(2r_m)^{\varpi}}{2(1+n)\sqrt{\pi}\varphi_0\Gamma(1+\varpi)}, \quad \theta_2 = -\frac{\varpi}{K_1(1+n)} - 2\,, \tag{10}$$

c is the parameter characterizing pollutant sedimentation onto the snow surface, $\Gamma(1+\varpi)$ is the Euler gamma function.

The unknown parameters θ_1 and θ_2 can be estimated from experimental data using, for example, the least squares method.[12] The parameter r_m is determined by the source geometry.[9]

Regression (8) allows reconstructing aerosol fallout fields from the relatively small number of reference measurement points.[13] Other sampling points can be used to check the adequacy of the reconstruction technique proposed.

(b) Regional pollution

The process of a pollutant spread far (7–10 km) from a source can be described by the following dependence[14]:

$$Q(x,y) = F(x,y)\,P(\varphi + 180^\circ); \tag{11}$$

$$F(x,y) = \theta/\sqrt{(x-\lambda)^2 + (y-\mu)^2}\,; \tag{12}$$

$$\varphi(x,y) = \arctan\,(y-\mu)/(x-\lambda), \tag{13}$$

where $Q(x,y)$ is the pollutant concentration at the point (x,y); λ and μ are the coordinates of a point source; $P(\varphi)$ is the wind rose in the atmospheric boundary layer for the period considered; $\theta = (M/2\pi)\,uH$, M is the total pollutant income, u and H are the mean wind speed and the height of the mixed layer.

Regression (11) shows that to determine the function $Q(x,y)$ it is sufficient to estimate the unknown parameter θ using, for example, experimental data.

3. Numerical simulation

The experimental data obtained on the chemical composition of snow allow us to carry out their interpretation within the proposed models of local and regional transport of aerosol pollutants. Using of dependences (8) and (11), the data from Table 1, as well as the source characteristics, we can choose reference points for reconstruction of aerosol pollution fields of the snow cover in the vicinity of the HPP studied.

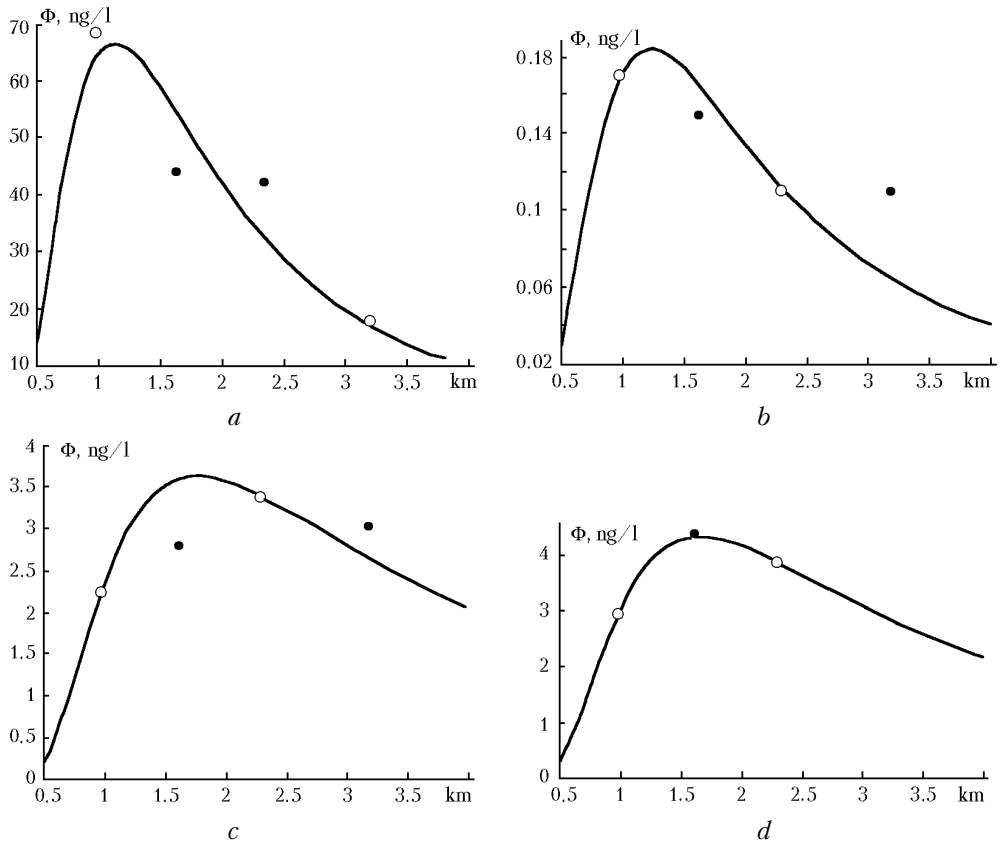

Fig. 2. Reconstructed and measured content of benzapilene (*a*), dust (*b*), water-soluble calcium (*c*), and coarse-disperse part of zinc (*d*) in the north-north-east direction from TETs-2: reference (o) and control (•) points.

Figure 2 depicts the specific content of benzapilene (BP), dust, and water-soluble calcium and zinc as estimated by model (8) for TETs-2 in the direction of the chosen sampling route. For the source under consideration, r_m was 1.8 km. Analysis of data presented in Fig. 2 shows quite a satisfactory agreement between the reconstructed and measured concentrations and allows us to draw some tentative conclusions on the relative characteristics of pollutant sedimentation. Unlike calcium and zinc, the peaks in the surface concentrations of dust and BP are markedly closer to the source of emission. This means that these components are transported as rather heavy aerosol fractions.

Figure 3 depicts the relative mass content (in %) of carcinogenic PAH to the total PAH mass in snow samples collected on the territories neighboring TETs-2 and TETs-3. The BP content is given separately among carcinogenic PAH. Comparison of data presented in Figs. 3*a* and 3*b* shows that the relative emission of carcinogenic PAH, including BP, is markedly higher at TETs-2. This circumstance is likely connected with both the combustion conditions and the features of the fossil fuel used.

The experimental data available allowed us to perform more detailed numerical analysis of the pattern of both local and regional pollution of the territory by emissions from TETs-5. Figure 4*a* depicts the results of reconstruction of the BP concentration in snow normalized to the winter wind occurrence. The concentration was estimated using dependence (9) and the experimental data divided by their weight fractions in the total occurrence of wind directions in the winter season of 2001/02 in the atmospheric surface layer.

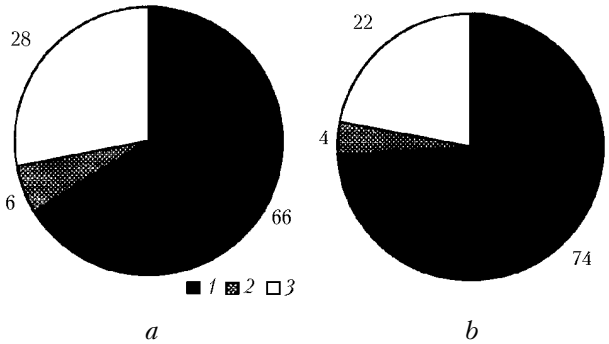

Fig. 3. Relative mass content (in %) of PAH components in snow samples collected near TETs-2 (*a*) and TETs-3 (*b*): non-carcinogenic PAH (*1*), benzapilene (*2*); other carcinogenic PAH (*3*).

Analysis of data shown in Fig. 4*a* shows quite satisfactory agreement between the calculated and experimental data at distances up to 6 to 8 km far

from the source. The peak of aerosol BP fallouts is markedly shifted to the source as compared to r_m = 3.5 km for the peak of the surface concentration of a weakly depositing admixture. The presence of this shift means that the BP is deposited as large aerosol fractions at relatively short distances from TETs-5 and to find the regularities of this deposition it is needed to conduct further experimental studies in the close zone of the source. Significant differences between the calculated and measured concentrations at long distances from HPP are mostly explained by the inadequacy of model (9) used and differences in the occurrence of wind directions in the atmospheric surface and boundary layers.[15]

For analysis of findings in the far zone (points *4*, *5*, *6*, *8*) it is worth using model (11)–(13). The results of this estimation are shown in Fig. 4*b*. For this purpose, we used regression (12) and the measured BP concentrations divided by their weight fractions in the total winter occurrence of wind directions in the atmospheric boundary layer.[15] Analysis of Fig. 4 shows that the model developed rather well agrees with the observations.

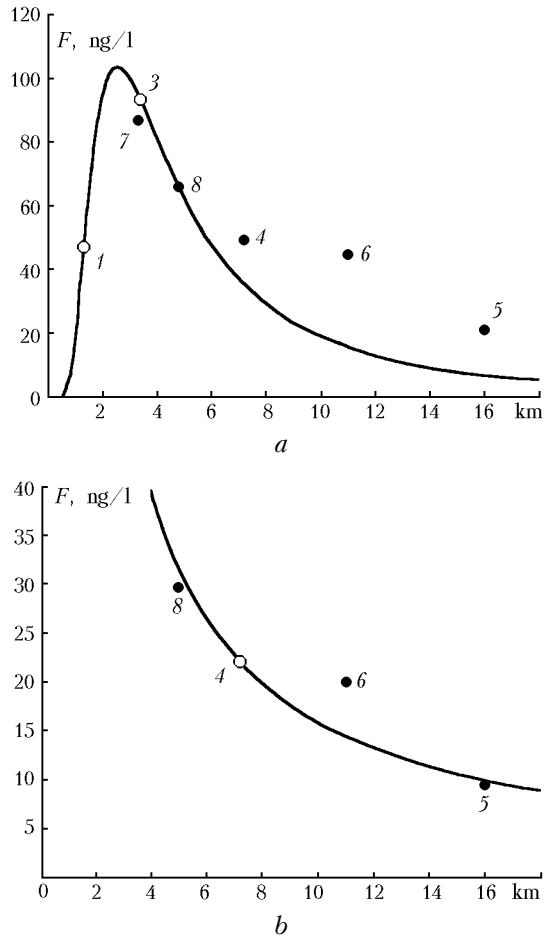

Fig. 4. Calculated and measured benzapilene content normalized to the occurrence of wind directions near TETs-5: reconstructed based on dependence (9) (*a*) and using Eq. (12) (*b*).

The estimates of the parameters of models (9) and (12) as applied to TETs-5 obtained with the use of the corresponding winter wind roses of 2001–2002 allows the BP concentration fields in the near and far vicinities of the power plant to be determined. The fallout fields shown in Fig. 5 have a pronounced trace character, which is the consequence of rather specific wind roses in this winter season. The occurrence of surface winds of southern directions in the considered winter season was about 90%.

The large height of the source of emission provides for relatively moderate levels of BP content in snow. However, on the other hand, it should be noted that the zone of HPP influence covers a vast territory, at which hazardous ecological situations may arise due to additional effect of other sources.

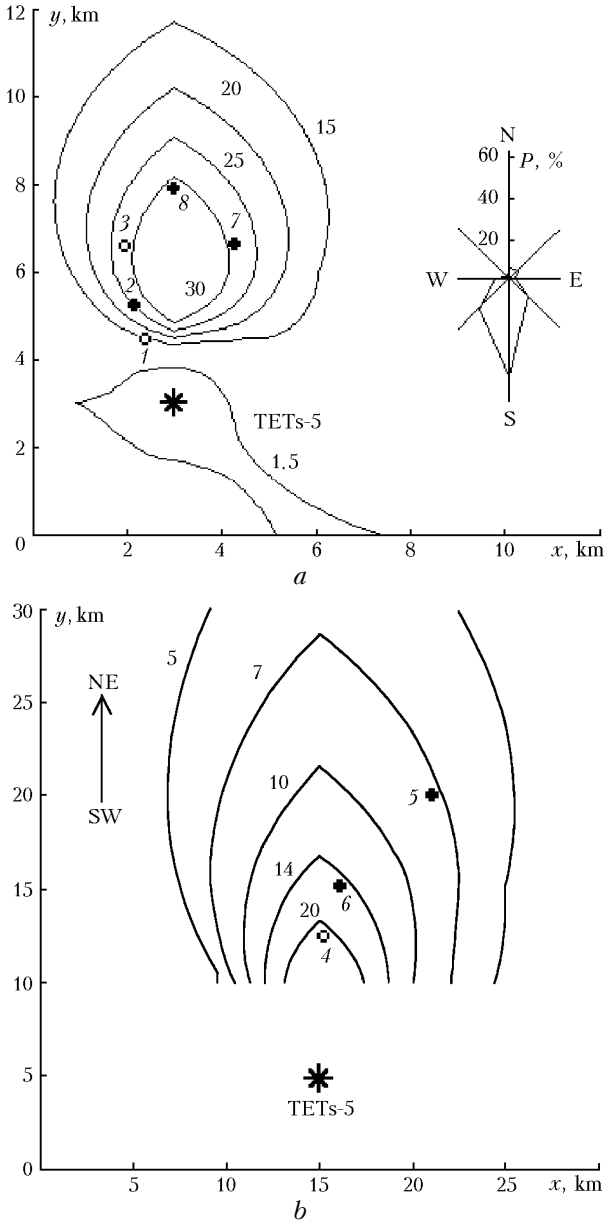

Fig. 5. Reconstructed concentration of benzapilene (ng/l) in the near (*a*) and far (*b*) vicinities of TETs-5.

Conclusion

Numerical analysis of the data of snow cover monitoring near Novosibirsk HPP shows the existence of rather simple regularities in formation of fields of long-term pollution of the studied territory. The procedure of clustering the unknown parameters significantly improves the efficiency of solving inverse problems of pollutant transport. With a small number of measurement points and limited input information, we have demonstrated the possibility of developing quantitative models of long aerosol pollution of a territory.

Based on the inverse problems of pollutant transport formulated in the atmospheric surface and boundary layers and the experimental findings, the fields of dust, macro- and micro-components, and PAH deposited in the snow cover were reconstructed. Comparison of the calculated concentrations with the measurements at the control points showed their good agreement. Analysis of the results of numerical simulation suggests that PAH are transported with both light and relatively heavy aerosol fractions and the zone of significant HPP influence covers a vast territory.

The studies reported in this paper form only one stage in investigation of the sources of aerosol emissions and the degree of their effect on Novosibirsk territory. The further development of this study assumes consideration of other sources (highways, boilers, private sector, etc.), extension of the number of chemical parameters to be determined, and optimization of observations.

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