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REMOTE SENSING OF ATMOSPHERE, HYDROSPHERE, AND UNDERLYING SURFACE

Numerical Study of Gas and Aerosol Impurity Transfer and Transformation Processes in the Plume of the Norilsk Industrial Region

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Abstract—Models for estimating the average and maximum concentrations of chemically active impurities in cross sections of their plumes from remote stationary sources are proposed. The models were tested with the use of aircraft sounding data on the multicomponent contamination of the winter atmosphere in the Norilsk industrial region. The process of gas–particle active transition observed at a distance of 58–103 km from the emission source is numerically studied. The average and maximum sulfur oxidation rates in the winter polar atmosphere are estimated as 0.027–0.034 and 0.055–0.07 h^{-1}, respectively.

Keywords: atmosphere, pollution, model, parameter, estimate, impurity transformation

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INTRODUCTION

Atmospheric emissions of contaminants in the Norilsk industrial region have a considerable effect on the surrounding areas. Vegetation degradation is observed at distances of up to 200 km from the industrial zone. This is corroborated both by ground-based monitoring data and by aircraft and satellite observations [1–4].

Processes of transport and transformation of impurities from industrial sources can be described using different models of atmospheric hydrothermodynamics. Given the presence of necessary input information, they make it possible to numerically simulate fields of hydrometeorological elements and those of impurity concentrations [5–10]. Otherwise, these calculations acquire a scenario character and do not allow one to establish a one-to-one association with real processes of area contamination.

In this situation, it seems suitable to apply a give and take approach based on using model descriptions of impurity transport processes and observation data within the framework of formulating inverse problems of analysis and design of experiments [11–13]. Obtaining correct mathematical formulations of these problems needs an additional complex of analytical and numerical studies oriented to stylized descriptions of contamination processes according to available monitoring data and experimental possibilities.

Analysis of contamination processes in the atmospheric boundary layer at a considerable distance from impurity sources admits significant simplifications. The dependence of the impurity concentration on the distance is determined by a relatively small number of parameters. The most significant ones are the source intensity, height of the mixing layer and wind velocity in it, transverse dispersion, and coefficients of chemical transformation of impurities. Using methods of the similarity theory and asymptotic methods allows one to obtain effective models of estimating impurity concentration fields by observation data [12, 14, 15].

This work is aimed at determining quantitative characteristics of the transport and transformation of impurities from results of complex sounding of the winter atmosphere of the Norilsk industrial region using the Optik-E An-30 airborne laboratory [16].

SUBJECT AND METHODS OF THE STUDIES

The investigation was concerned with the total plume of aerosol–gas emissions of three main plants of the Norilsk Mining and Metallurgical Combine. Two of them are situated in the suburbs of Norilsk city and one in Kaierkan, a satellite town between Norilsk and Alykel Airport. The measurements of gas concentrations and sampling of aerosol admixtures were carried out November 10, 2002, in the main direction of the air mass drift at distances from 20 to 140 km to the west-southwest from Norilsk at several elevated sites from 400 to 1200 m above sea level along equidistant paths (from 3 to 6 at each elevated site) perpendicular to the main direction of the plume propagation. The study of sites at 600 m and higher showed a rapid approach of gas concentrations and aerosol admixtures to background values with the height. The main part of the plume was concentrated at a level of 400 m. The sounding route at this height horizon is depicted in Fig. 1. Gas components were measured continuously and aerosol samples were taken on AFA-KhP-20 filters, an individual sample on each rectilinear route portion. The first path (a rectilinear portion of the route) was normal to the average wind direction at a distance of 58 km from Norilsk; the last one (the sixth), at 103 km.

The experimental technique and characteristics of gas analyzers and methods of quantitative physicochemical analysis of the aerosol substance on filters were presented in detail in the pioneer description of this flight experiment [16]. It was also noted in that work that the main contribution to the contamination of air in the Norilsk industrial region is made by emissions from mining and processing of sulfide ores of nonferrous metals. Ore roasting leads to oxidation of sulfide sulfur to gaseous sulfur dioxide which constitutes 96% of the total emission mass. It means that acid sulfur oxides in the gaseous form are main components removed from the city by the atmospheric channel. According to the theory [17–19], processes of chemical transformation of sulfur occur by the scheme $H_2S \rightarrow SO_2 \rightarrow H_2SO_3 \rightarrow H_2SO_4 \rightarrow M(HSO_4)_m \rightarrow M_2(SO_4)_m$, where M^{m+} is a cation of a metal.

In the morning of November 10, 2002, weather near Norilsk was determined by the central part of a filling cyclone with surface fronts; then, a small-gradient low-pressure field was observed. The air mass was Arctic. The meteosynoptic description of the experiment was also presented in [16].

Results of measurements at a height of 400 m are presented in Table 1 and Fig. 2.

MODELS OF ESTIMATING CONCENTRATION FIELDS IN THE PLUME

The concentration fields were estimated by observation data according to models based on the following relation between the mass balance of a weakly settling passive and chemically active admixture coming from a stationary point source to the boundary layer of the atmosphere [14, 15]:

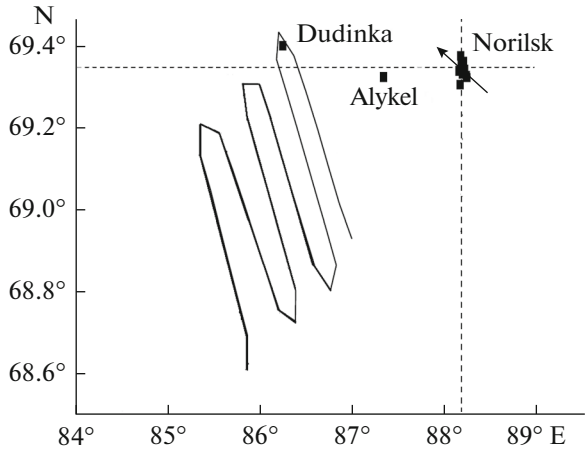

Fig. 1. Scheme of aircraft sounding of November 10, 2002, in the plume of impurities from the Norilsk industrial region at a height of 400 m.

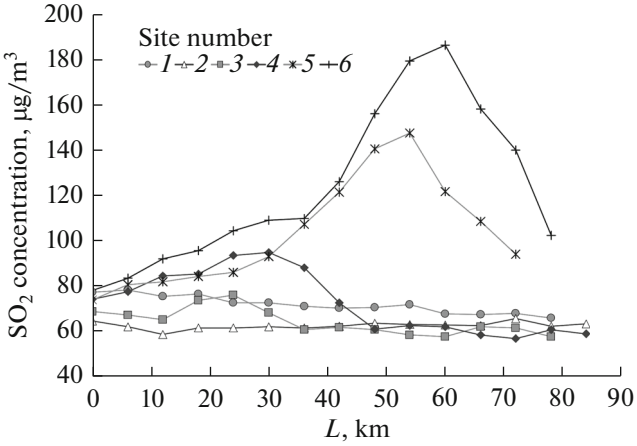

Fig. 2. Variation in the SO_2 concentration at sites *1–6* (distance L is shown from the instant of mounting or taking filtered samples for all sites during southward returns of the flying laboratory).

$$u(z)\frac{\partial q}{\partial x} = \frac{\partial}{\partial y}\mu(z)\frac{\partial q}{\partial y} + \frac{\partial}{\partial z}\nu(z)\frac{\partial q}{\partial z} - \alpha q \qquad (1)$$

with the following boundary conditions:

$$\nu(z)\frac{\partial q}{\partial z}\bigg|_{z=0,\ z=h} = 0, \quad q \to 0 \quad \text{as} \quad |y| \to \infty. \qquad (2)$$

$$u(z)q\big|_{x=0} = M\delta(y)\delta(z-H), \qquad (3)$$

where the x axis is oriented in the direction of the horizontal wind; the y axis, in the transverse direction; $u(z)$ is the wind speed; $\mu(z)$ and $\nu(z)$ are the coeffi-

Table 1. Average concentrations of individual chemical aerosol components, sulfur dioxide, and total concentrations of all ions and elements determined in aerosol at route sites near Norilsk

Distance from the city, km	Average concentration, μg/m^3							
	Pb	Ni	Cl^-	Br^-	Ca	SO_4^{2-}	SO_2	Sum of all ions and elements
58	1.42	–	2.14	0.71	45.59	2.85	71.9	71.2
65	1.53	–	4.08	0.51	31.75	7.53	62.5	51.0
73	–	1.25	1.99	0.99	12.56	6.60	64.3	24.9
82	0.83	0.62	1.87	0.83	5.93	8.97	72.8	20.8
92	0.68	0.68	1.60	0.68	1.03	5.75	103.4	11.4
103	–	–	–	–	16.86	5.69	123.2	25.75

cients of the horizontal and vertical turbulent exchanges; α is the coefficient of chemical transformation; and M and H are the power and height of the admixture source. Setting

$$\mu(z) = k_0 u(z), \tag{4}$$

one can demonstrate [15] that

$$q(x, y, z) = p(x, y)\, q'(x, z), \tag{5}$$

where $p(x, y) = \dfrac{1}{2\sqrt{\pi k_0 x}} \exp\left(-\dfrac{y^2}{4k_0 x}\right)$, and the function $q'(x, z)$ is a solution of the equation

$$u(z)\frac{\partial q'}{\partial x} = \frac{\partial}{\partial z} k(z) \frac{\partial q'}{\partial z} - \alpha q' \tag{6}$$

with the corresponding boundary conditions. Integrating Eq. (6) with respect to z from 0 to h with allowance for boundary conditions (2), we obtain the relation

$$\frac{d}{dx}\int_0^h u(z) q'(x, z) dz = -\alpha \int_0^h q'(x, z) dz. \tag{7}$$

Applying the generalized integral mean value theorem [20] to both sides of Eq. (7), we come to the relation

$$\frac{d}{dx}\left\{ q'(x, \xi_1) \int_0^h u(z) dz \right\} = -\alpha h q'(x, \xi_2), \tag{8}$$

where $\xi_1, \xi_2 \in (0, h)$. With allowance for the fact that the concentration of a light admixture in the mixing layer becomes equal with respect to height at a sufficiently long distance from the source (7–10 km) [14], the following asymptotic formula follows from (8):

$$q'(x, \xi) \to C e^{-\frac{\alpha x}{\overline{U}}} \quad \text{as} \quad x \to \infty, \quad \xi \in (0, h), \tag{9}$$

where C is the integration constant and $\overline{U} = \dfrac{1}{h}\int_0^h u(z) dz$ is the average wind speed in the mixing layer. Then, taking into account (5) and (9), we obtain the following relation for estimating the integral concentration in the direction transverse to the admixture plume:

$$P(x, \boldsymbol{\theta}) = C e^{-\frac{\alpha x}{\overline{U}}} \int_{-\infty}^{\infty} p(x, y) dy = \theta_1 e^{\theta_2 x}, \tag{10}$$

where $\theta_1 = C$ and $\theta_2 = -\dfrac{\alpha}{\overline{U}}$.

Note. With allowance for (5) and (6), the relation for estimating maximum concentrations in directions transverse to the admixture plume has the form

$$q(x, \mathbf{S}) = \frac{S_1}{x} e^{S_2 x}. \tag{11}$$

The parameters θ_1, θ_2, S_1, and S_2 in regression dependences (10) and (11) are estimated by the least squares method [21].

RESULTS AND DISCUSSION

Analysis of measurement data presented in Table 1 testifies about directed changes in concentrations of calcium, total aerosol, and sulfur dioxide in the considered ranges of distances from the source. These changes are related to the process of the gas–particle active transition. It is evident that the passive impurity propagation model is unsuitable for describing concentration fields of these components in this case.

For other components of the chemical composition, the changes are less noticeable. In general, concentrations of chlorides, bromides, and sulfates at sites *1–6* change relatively slightly. The dynamics of the decrease in lead and nickel concentrations with the distance from the source is less considerable as compared to calcium.

Figure 3 presents results of numerical reconstruction of calcium and sulfur dioxide concentrations based on dependences (10) and (11) according to measurements at two reference sites. In the distance range under consideration, the estimation models quite adequately describe processes of transformations of the

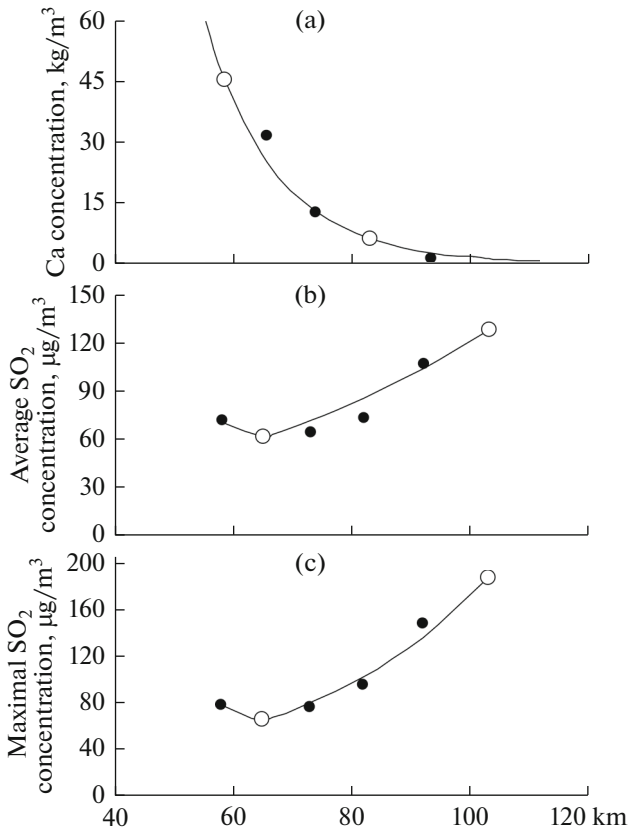

Fig. 3. (a) Measured and reconstructed Ca concentrations and (b) average and (c) maximum SO_2 concentrations at sites *1–6* in the plume; ○ and ● are the reference and test measurement points, respectively.

studied admixtures in the atmosphere. According to Figs. 3b and 3c, the sulfur dioxide concentration reached the minimal value due to chemical transformation at a distance of 65 km. At the same time, the calcium concentration rather rapidly decreased before the distance of 92 km (Fig. 3a). The estimates for the parameters θ_2 and S_2 by models (10) and (11) for average and maximal concentrations of sulfur dioxide were equal to 0.019 and 0.039 km^{-1}, respectively. According to literature data [18], the average rate of sulfur oxidation in the atmosphere is 0.027 h^{-1}.

Comparing our estimates with literature data requires taking into account the speeds of the air mass (AM) transport and aircraft along the flow. Proceeding from the meteosynoptic description of the experiment [16], the time of AM transport from Norilsk emission sources to the measurement square was 25–30 h, i.e., the transport speed of AM with the plumes lay in the range of 1.9–2.3 km/h. With allowance for the fact that the average difference of the sample exposure and measurements between paths reached 14 min, the real AM transport speed in the inertial frame of sounding (relative to the aircraft position at medians of the paths) was 1.4–1.8 km/h. In this case, sulfur dioxide oxidation rates in the atmosphere calculated by estimates of the parameters θ_2 and S_2 of models (10) and (11) lie in the range of 0.027–0.034 and 0.055–0.07 h^{-1}, respectively.

Thus, the average sulfur dioxide oxidation rate obtained for the region of average concentrations based on aircraft sounding in the real atmosphere satisfactorily agrees with the estimate established using the kinetic model [18].

CONCLUSIONS

Based on stationary models for estimating fields of regional transport and transformation of impurities, numerical analysis of aircraft sounding data on the contaminated winter atmosphere of the Norilsk industrial region has been carried out. It has been shown that the constructed two-parametric model of transformation quite adequately describes the change in fields of average concentrations of calcium, as well as average and maximum concentrations of sulfur dioxide, in directions transverse to impurity plumes at certain distances from sources.

Being a base of the aerosol mass, human-made calcium oxide is the main counterpart of SO_2 fixation in processes of dry heterogeneous condensation. Its presence accelerates the process of transformation and elimination of sulfur dioxide from the polar atmosphere.

Under conditions of the performed investigations, the gas–particle active transition was experimentally recorded at a distance of 58–103 km from the emission source. The average and maximum estimates of the sulfur oxidation rate in the winter polar atmosphere have been obtained: 0.027–0.034 and 0.055–0.07 h^{-1}, respectively.

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