

A Method to Estimate the Emission Rate from Remote Industrial Sources

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Abstract

A method is proposed for the determination of the emission rate of various components by industrial enterprises using the data on spatial-temporal variability of the concentrations of chemical elements or compounds in observation sites remote from sources.

INTRODUCTION

Enormous amount of various hazardous substances enters the atmosphere with industrial emissions every year. The major pollutants in the emission are solid particles (dust, ash, metals) and gaseous substances (carbon monoxide, sulphur dioxide, nitrogen oxides) [1]. The statistical data on the emission of pollutants into the atmosphere are regularly published in the annual Year-Book of the emission of pollutants into the atmosphere of cities and regions of the Russian Federation by the State Committee for Ecology RF, and in the reports of the State Committee of Statistics (Goskomstat) concerning protection of the atmosphere. Unfortunately, the data on the industrial emission of such species as heavy metals, organic compounds, hydrogen sulphide, *etc.*, are insufficient to make estimations of their effect on the level of atmospheric pollution.

Hazardous pollutants that enter the atmosphere from anthropogenic sources are scattered near a factory chimney (are deposited onto buildings, plants, soil, are washed with atmospheric precipitation) or transported at a distance of several thousand kilometers from the emission site. The transport of impurities into the upper layers of the atmosphere is determined by the vertical temperature dis-

tribution in the atmosphere, *i. e.*, the stability of atmosphere. Stable stratification (with the vertical temperature gradient less than 0.6° per 100 m) prevents the impurities from spreading, while unstable one (with the vertical temperature gradient more than 0.6° per 100 m) helps raising the impurities into the upper layers, which provides their long-range transport.

In the present investigation, an attempt is made to estimate the content of several chemical components (the sum of ions $\text{Ca}^{2+} + \text{Mg}^{2+}$, and elements Cu, Ni, Ca, Se) in the industrial emission using the ratio of their concentrations, determining in the atmospheric aerosol near the ground at a long distance from the emission source, to SO_4^{2-} concentration (the trace species).

EXPERIMENTAL

Since 1991, within the Project “Aerosols of Siberia” [2, 3, 5], systematic investigation of atmospheric aerosols (AA) was launched in the Siberian region. The goals and objectives of the Project are published in [3]. Results of the experimental investigation of the major characteristics of AA of Siberia, such as particle size distribution and chemical composition, as well as their spatial and temporal characteristics are described in [2, 4–6]. At present, AA is

sampled in every season (for one month during each season) in the south and in the north of West Siberia. Sampling sites are located in large settlements that possess local powerful sources of air pollution (plants, heat and power stations, automobiles), and in remote small settlements having no powerful sources of emission and located at a distance of 200 to 500 km from the large settlements.

The AA is sampled with the help of a special aspirator set-up in which an AFA-ChA type filter is fixed. About 200 m³ of air is drawn through it during 24 h. After that, the filter is divided into two parts. One of them is used to determine the multielement composition (up to 30 elements) by X-ray fluorescent analysis with the synchrotron radiation. The second half of the filter is situated into the container with deionized water for 24 h; after that, the ion composition is determined in water extract by means of ion chromatography and conductometric titration.

One of the goals of the Aerosol of Siberia Project is the investigation of the effect of long-range transport of industrial emission on the level of regional and global contamination of remote territories. At present, much attention is paid to long-range transport of the continental AA into polar regions. Investigations in Norway and in Alaska show that the western and central parts of Siberia can be the important sources of air pollution in Arctic [7]. The atmosphere of cities and many regions of the south of Siberia is extremely highly polluted with heavy metals and with gaseous emission. The level of such a contamination is much higher than that in the industrially developed countries of Europe and America. It is necessary to pay attention to the following features of the investigation of AA in Siberia. There are areas at a long distance from large industrial centres. According to the generally accepted opinion, AA in these regions is considered to be background, *i. e.* aerosol that is formed by natural processes and contains only small amount of contaminants.

The investigations of AA characteristics in many sites of the globe, performed for many years, show that the major part of atmospheric pollutants is comprised of the particles formed by wind erosion of the soil and ocea-

nic surface. These are the so-called dust and the sea salts particles. As a rule, the content of other particles of natural and anthropogenic origin in AA is much lower. Central Siberia lies at a distance of several thousand kilometers from powerful sources of soil-erosion aerosols. In winter soil is coated with snow in Siberia, while water surface, including seas and oceans, are covered with ice. So, this season provides extremely favourable conditions for the investigation of long-range transport of industrial emissions into the northern regions of Siberia.

According to the data of the global background monitoring established in the northern hemisphere, the annual emission of SO₂ from industrial regions is, Mt: USA and Canada, 23; Europe, 30; the area of the former Soviet Union, 23 [8]. Among the total emission of SO₂ from the area of the former Soviet Union, Siberian region accounts for 30 % (6.9 Mt). According to the statistical data [9], annual emission from the Noril'sk Mining and Metallurgical Plant (NMMP) and the industrial Urals is about 3.5 Mt. This makes a half of total emission in Siberia. Because of this, SO₂ can be used as tracer in the investigation of long-range transport of anthropogenic emission into background regions.

RESULTS AND DISCUSSION

Since 1996, systematic measurements of spatial-temporal variations in chemical composition and concentration of atmospheric aerosols in the boundary layer of atmosphere are performed in some sites in the north of Siberia. These sites are Samburg (67° N. L., 78° E. L.), Tarko-Sale (65° N. L., 78° E. L.), and Krasnosel'kup (66° N. L., 82° E. L.).

Some results of investigations allowing one to distinguish the effect of the sources of natural and man-made type are reported in [4, 8]. Measurements of the chemical composition of AA in Siberia show that the particles from anthropogenic sources (enrichment coefficients more than 10) are present in submicron fraction [12]. The particles of this fraction are transported over very long distance and form global background as revealed by the long-range

temporal averaging-out (more than 10–15 days). In particular, this is well confirmed by our observations of the variations of monthly averaged aerosol concentrations in observation sites located at a distance of more than 1500 km from each other [4]. Because of this, in [4] we estimated the type of source using the change of relative multielement composition of AA; in [10], to reveal a possible source of pollution, we used experimental data on temporal variations of the concentrations of separate components of the chemical composition of AA, and the data on the velocity and direction of wind in the observation site.

In [15] we proposed an estimation of the annual emission from powerful industrial plants or regions using temporal variations of the chemical composition of AA in observation site located at a distance of several hundred kilometers (regional scale) or more than thousand kilometers (global scale). Using the results of observations of these regularities, we estimated annual emission of Cu, Ni by the NMMP, as well as the emission of Ni, Ca, $\text{Ca}^{2+} + \text{Mg}^{2+}$ by the plants of the Urals. In the present communication, we report additional data concerning estimations of the annual emission of Se and $\text{Ca}^{2+} + \text{Mg}^{2+}$, as a development of our investigation of NMMP emission. In addition, probabilistic characteristics of the temporal

variability of concentration of the individual components of the AA chemical composition are shown, along with the probability of the synchronous appearance of the maximum concentration of a set of elements.

Diurnal mean concentrations of these components during observation periods from 9 January to 7 February (Tarko-Sale) and from 1 to 30 July 1999 (Krasnosel'kup) are shown in Figs. 1 and 2. One can see that in each observation site there are periods with sharp increase in the content of all the components under question. It can be assumed that such an increase in concentration is connected with simultaneous emission of these pollutants from the same source. This hypothesis is confirmed by the results of correlation and factor analysis. In the first case, in Tarko-Sale, on the 6th of February, we observed synchronous substantial increase in the concentrations of SO_4^{2-} , of the sum of ions $\text{Ca}^{2+} + \text{Mg}^{2+}$, Ni and Ca. Correlation coefficients between these components increased substantially (compared to the entire observation period) for the period from the 2nd to the 7th of February; the largest increase was observed for Ni and SO_4^{2-} (from 0.53 to 0.92). Factor analysis was performed for a complete series of observations; all the mentioned components got into the first factor with high weigh (Table 1).

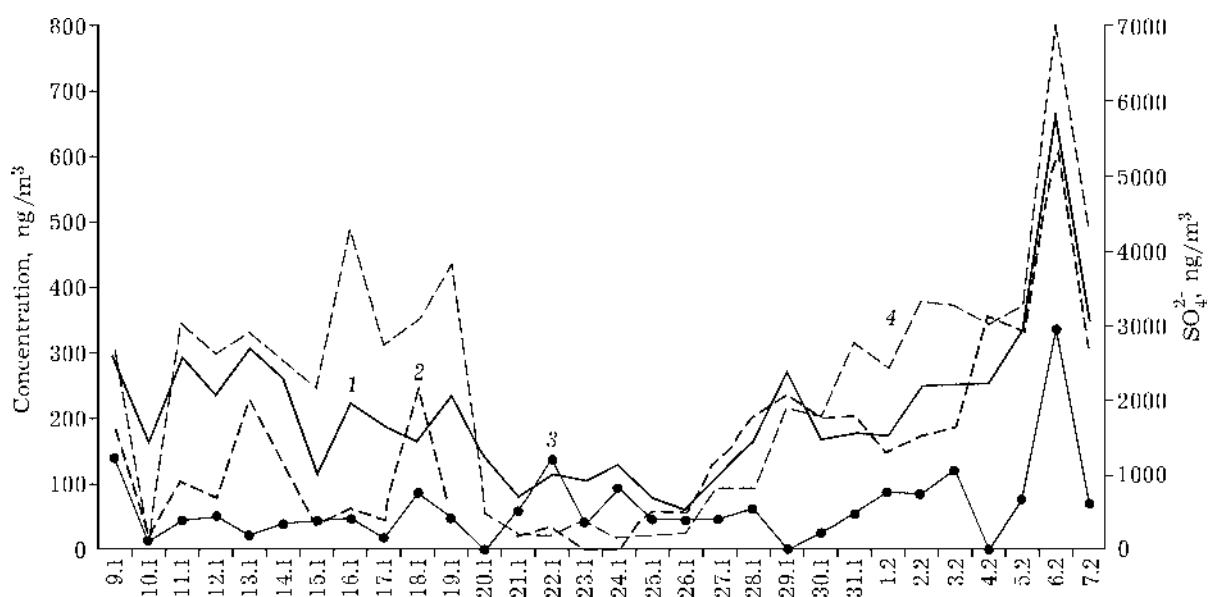


Fig. 1. Diurnal mean concentrations of chemical components in atmospheric aerosol (Tarko-Sale settlement, January 9–February 7, 1999): 1 - $\text{Ca}^{2+} + \text{Mg}^{2+}$, 2 - Ca, 3 - Ni ($\times 40$), 4 - SO_4^{2-} .

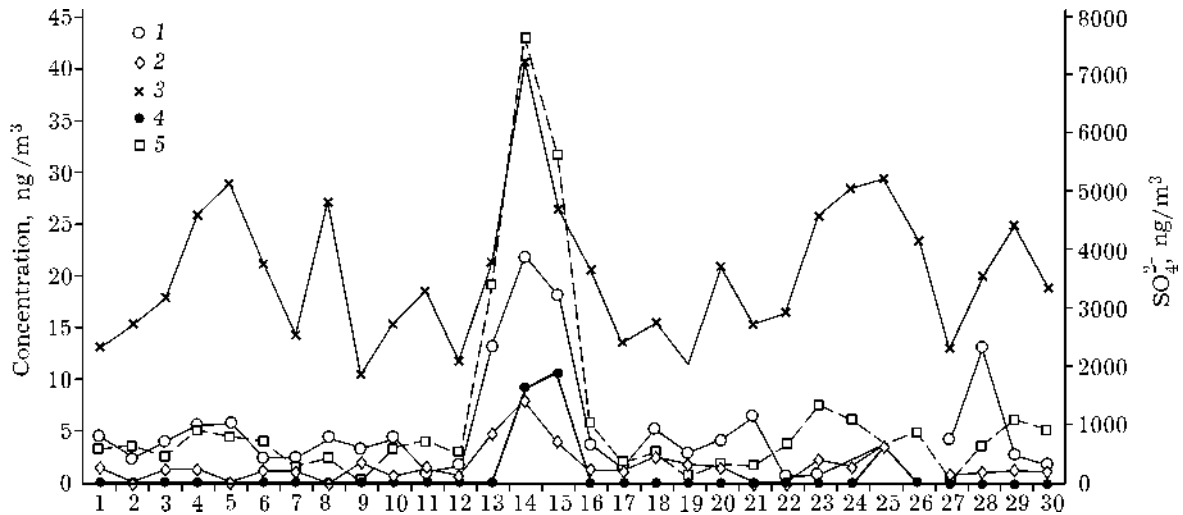


Fig. 2. Diurnal mean concentrations of chemical components in atmospheric aerosol (Krasnosel'kup settlement, July 1–30, 1999): 1 – Cu, 2 – Ni, 3 – $(\text{Ca}^{2+} + \text{Mg}^{2+})/10$, 4 – Se ($\times 10$), 5 – SO_4^{2-} .

In the second case, as one can see in Fig. 2, in Krasnosel'kup from the 13th to the 15th of July, we also observed a sharp increase in the concentrations in SO_4^{2-} , $\text{Ca}^{2+} + \text{Mg}^{2+}$, Ni, Cu, Se. The correlation dependence for them is sufficiently high when considered for the whole month and increases since the 10th to the 20th of July. The largest increase was observed for nickel and copper (from 0.72 to 0.93). Factor analysis isolated all the five components into one factor with high factor weigh, similarly to the previous case (Table 2). Attention should be paid to high correlation coefficients (more than 0.9) between the SO_4^{2-} concentration and the concentrations of other components in both cases of sharp concentration increase.

The AA was also sampled during this period in Tarko-Sale and in Samburg. The analysis of those samples shows that in the former site an increase of SO_4^{2-} content was observed on

the 14th and 15th of July, though this increase was not so large as in the latter site. Concentrations of other components did not increase in any of the two sites.

In order to determine a place from which these contaminants could be transported to observation sites, back trajectories of air masses at the height of 10, 100, 300, 500, and 1000 m were calculated employing the HYSPLIT software [11] (Fig. 3, a, b). One can see that the air mass that arrived into Tarko-Sale on the 7th of February had passed over the southern Urals; on the 13–15th of July, air mass was transported into the observation region from north-east, the trajectory passing near Noril'sk.

The forward trajectories built up for large industrial centres situated in the southern Urals (Perm', Ekaterinburg, Chelyabinsk) and in the north of East Siberia (Noril'sk) confirm the

TABLE 1

Correlation coefficients and factor weights of the concentrations of Ca, Ni, $\text{Ca}^{2+} + \text{Mg}^{2+}$, and SO_4^{2-} (Tarko-Sale, 1999)

Component	Factor weight	Correlation coefficient							
		January 9–February 7				February 2–7			
		Ca	Ni	$\text{Ca}^{2+} + \text{Mg}^{2+}$	SO_4^{2-}	Ca	Ni	$\text{Ca}^{2+} + \text{Mg}^{2+}$	SO_4^{2-}
Ca	0.81	1	0.72	0.83	0.69	1	0.87	0.99	0.93
Ni	0.79		1	0.69	0.53		1	0.91	0.92
$\text{Ca}^{2+} + \text{Mg}^{2+}$	0.87			1	0.86			1	0.97
SO_4^{2-}	0.88				1				1

TABLE 2

Correlation coefficients and factor weights of the concentrations of Ca, Ni, $\text{Ca}^{2+} + \text{Mg}^{2+}$, and SO_4^{2-} (Krasnosel'kup, 1999)

Component	Factor weight	Correlation coefficient									
		July 1-30					July 10-20				
		Ni	Cu	Se	$\text{Ca}^{2+} + \text{Mg}^{2+}$	SO_4^{2-}	Ni	Cu	Se	$\text{Ca}^{2+} + \text{Mg}^{2+}$	SO_4^{2-}
Ni	0.79	1	0.72	0.73	0.55	0.83	1	0.93	0.73	0.88	0.92
Cu	0.79		1	0.77	0.52	0.86		1	0.87	0.87	0.98
Se	0.84			1	0.57	0.87			1	0.79	0.90
$\text{Ca}^{2+} + \text{Mg}^{2+}$	0.77				1	0.66				1	0.91
SO_4^{2-}	0.92					1					1

assumption that pollutions is transported into Tarko-Sale from Ekaterinburg and Chelyabinsk, and into Krasnosel'kup from Noril'sk (Fig. 4, *a, b*).

For these towns, with the help of HYSPLIT model, we calculated the regions over which the emitted sulphur dioxide is spread, the emission being $17 \cdot 10^{13}$ $\mu\text{g}/\text{h}$ for the towns of the Urals (since the annual emission of SO_2 by the plants of the Urals is about 1.5 mln t), and $23 \cdot 10^{13}$ $\mu\text{g}/\text{h}$ for Noril'sk (since the annual emission of SO_2 from NMMP is 2 mln t) [9] (Fig. 5, *a, b*). The areas with different levels of the concentration of SO_2 transformed into SO_4^{2-} within the layer 0–10 m were revealed, with averaging from 00 A.M. of the 6th of February till 00 A.M. of the 7th of February, 1999, and from 00 A.M. of the 14th of July till 00 A.M. of the 15th of July. These data confirm the previous considerations concerning the assumed sources of the components under question. One can see that during the first period (see Fig. 5, *a*) Tarko-Sale is in the region of medium contamination by the pollutants coming from the plants of Chelyabinsk. Calculation for Ekaterinburg gives the same picture. During the second observation period, all the three observation sites were covered by the tail of pollution from Noril'sk (see Fig. 5, *b*); however, Krasnosel'kup got into a region with higher concentration of the impurity. One can also see in Fig. 4, *b* that the trajectories of air masses arriving from Noril'sk embrace Krasnosel'kup and turn to the east without touching Samburg and Tarko-Sale. This explains

why the content of components under consideration increased only in Krasnosel'kup.

It is necessary to note that in the first case (February 5–7, 1999) a vast deep cyclone occupied the territory of Europe; spreading to the east, it provided the transport of air mass from the southern Urals to the north of West Siberia. An elongated shape of the region of emission spreading (see Fig. 5, *a*) is explained by large wind velocity. In the second case (July 13–16, 1999), a low-gradient baric field with weak wind of the north-eastern direction was observed in the investigation area. Because of this, the emission scattering region is less elongated (see Fig. 5, *b*). In this case, an increase the concentrations of the components under consideration was observed during three days without a break, which can be explained by the fact that the distance from the source of emission to the sampling site was rather short (about 500 km). In the first case, the distance between observation sites and emission source was about 1500 km.

Taking into account that sulphur dioxide SO_2 is transformed by chemical reactions into the sulphate ion SO_4^{2-} rather rapidly (within several hours), and the latter enters solid aerosol particles, we obtain a connection between mass concentrations of SO_2 and SO_4^{2-} :

$$C_{\text{SO}_4^{2-}} = 1.5C_{\text{SO}_2} \quad (1)$$

After that, we assume that the relative content of components in emission is conserved during spreading, and the maxima of concentrations are observed when a cloud containing a pollutant from a source covers observation

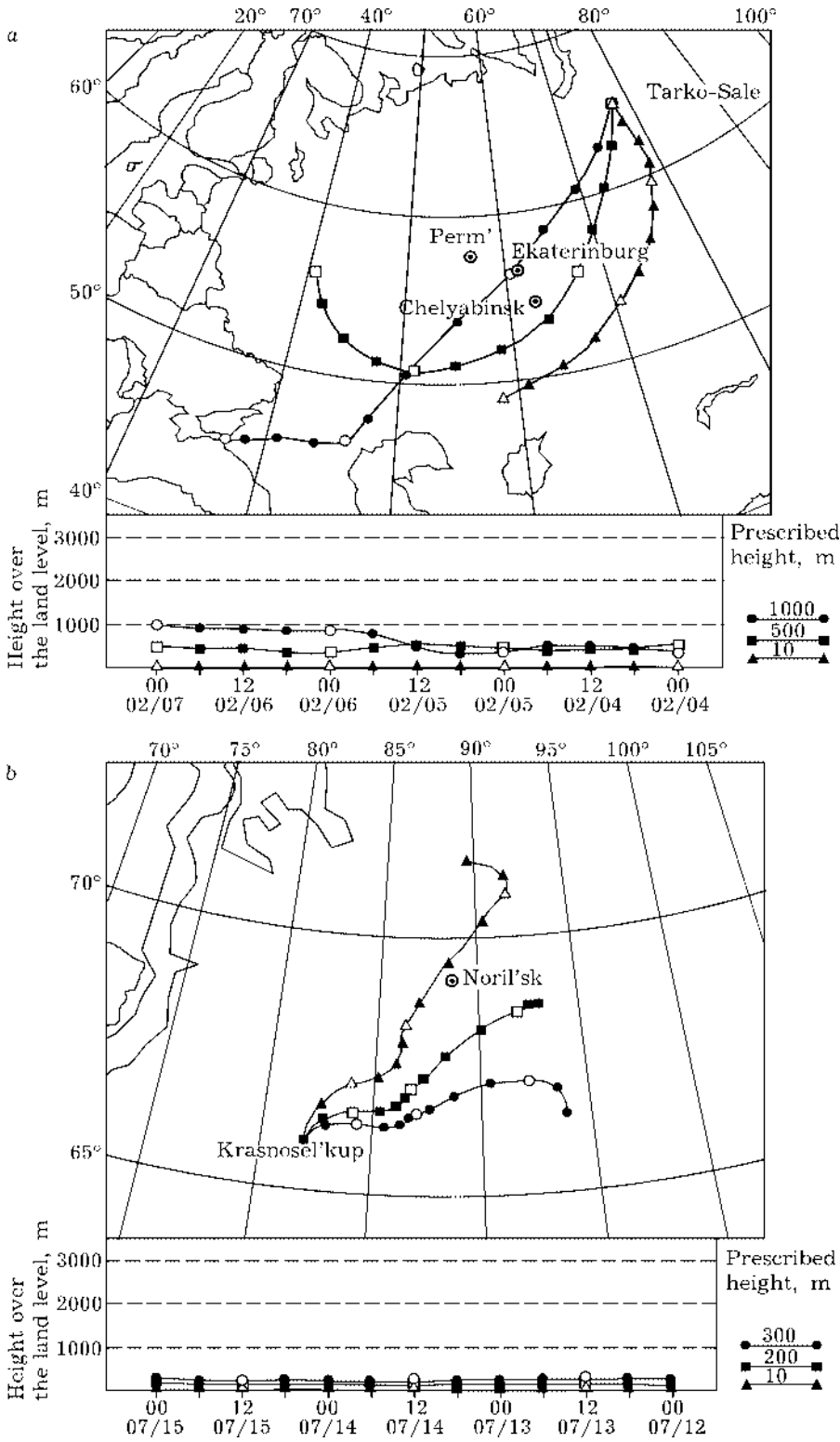


Fig. 3. Back trajectories of air mass transport: *a* - from Tarko-Sale, final time: 00 AM February 7, 1999; *b* - from Krasnosel'kup, 12 AM July 15, 1999.

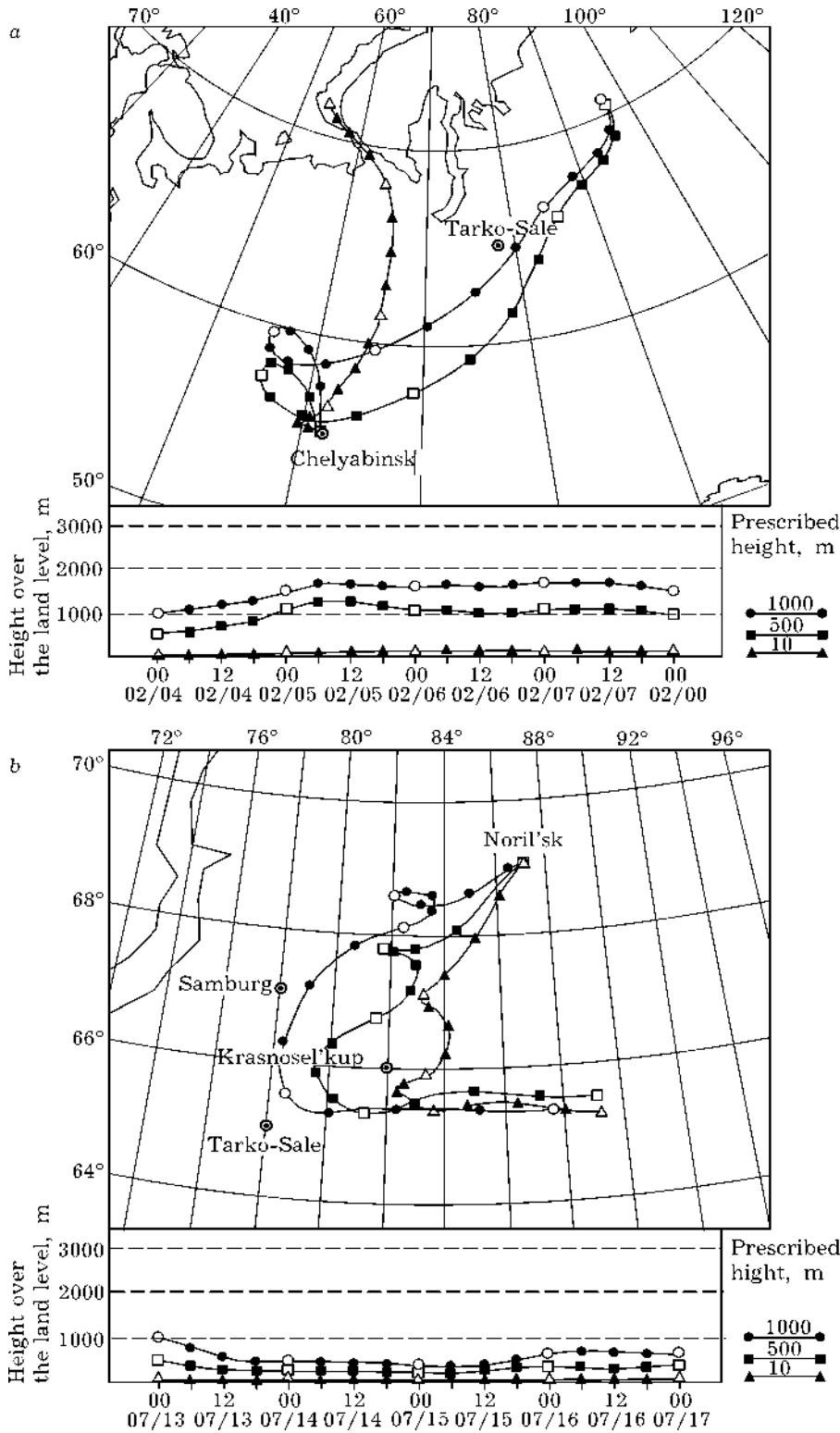


Fig. 4. Forward trajectories of air mass transport: a - from Chelyabinsk, final time: 00 AM February 4, 1999; b - from Noril'sk, 00 AM July 13, 1999.

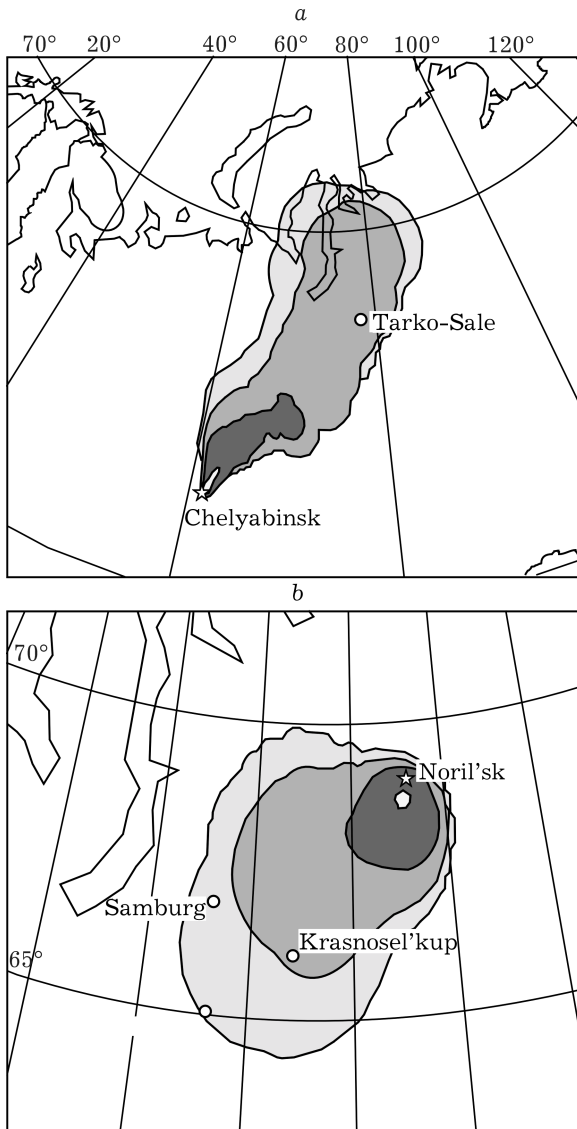


Fig. 5. The spreading regions of SO_2 emission by industrial plants: a - Chelyabinsk, February 6, 1999; b - Noril'sk, July 14, 1999.

site. Knowing the amount of SO_2 emission and calculating the ratios of measured concentrations of Ni, Cu, Ca, Se, and $\text{Ca}^{2+} + \text{Mg}^{2+}$ to SO_4^{2-} concentration in the maximum, we estimate the annual emission of these components:

$$Q_i = \frac{C_i^{\max}}{C_{\text{SO}_4^{2-}}^{\max}} Q_{\text{SO}_4^{2-}} = 1.5 \frac{C_i^{\max}}{C_{\text{SO}_4^{2-}}^{\max}} Q_{\text{SO}_2} \quad (2)$$

Here Q_i is the annual emission of an i pollutant, Q_{SO_2} is SO_2 annual emission rate, C_i^{\max} is maximal diurnal averaged concentration of i pollutant (Ni, Cu, Ca, Se, and $\text{Ca}^{2+} + \text{Mg}^{2+}$), $C_{\text{SO}_4^{2-}}^{\max}$ is maximal diurnal concentration of sul-

TABLE 3

Estimation of annual emission rate of some components by the plants of the Urals and of Noril'sk, t/y

The Urals		Noril'sk	
Ni	2600	Ni	3000
Ca	193 000	Cu	8250
$\text{Ca}^{2+} + \text{Mg}^{2+}$	225 000	Se	400
		$\text{Ca}^{2+} + \text{Mg}^{2+}$	160 000

phate ion. Using the eq. (2) we obtain an estimation of the annual emission (in tons) of the components under consideration by the plants of the Ural region and Noril'sk (Table 3).

Some additional confirmations in favour of the correctness of our conclusions can be listed. The question can be solved more unambiguously for the Noril'sk Mining and Metallurgical Plant (NMMP). This is a point source with the known rate of annual SO_2 emission. In addition, it is situated at a distance from observation site approximately three times shorter than that of the second source. For the NMMP, the concentrations of some pollutants registered near Noril'sk are listed in [13]. For example, the concentrations of SO_4^{2-} , Ni, Cu in snow cover are reported. Let us compare relative concentrations $\text{Ni}/\text{SO}_4^{2-}$ and $\text{Cu}/\text{SO}_4^{2-}$ in our experiments with the experimental data on the presence of these impurities in snow cover near Noril'sk (Table 4).

One can see that our results coincide with the known literature data at an accuracy of the factor of 2–5. Thus, our assumption concerning the constancy of concentrations of the impurities of interest in the emission from NMMP is in agreement with the data available from literature.

Now let us consider the reasonability of our assumptions, starting from the analysis of temporal variability of the concentrations of components under question. One can see in Fig. 2

TABLE 4

Comparison of relative concentrations of $\text{Ni}/\text{SO}_4^{2-}$ and $\text{Cu}/\text{SO}_4^{2-}$ with the data reported in [13]

Ratio	Our data	The data of [13]
$[\text{Ni}]/[\text{SO}_4^{2-}]$	10^{-3}	$4.6 \cdot 10^{-3}$
$[\text{Cu}]/[\text{SO}_4^{2-}]$	$2.8 \cdot 10^{-3}$	$5.7 \cdot 10^{-3}$

TABLE 5

Comparative characteristics of components during different observation periods

Component	Period without concentration increase			Period with concentration increase			t	Probability
	x_{50} , ng/m ³	σ_{ig}	n	x_{50} , ng/m ³	σ_{ig}	n		
<i>Tarko-Sale, January–February, 1999</i>								
Ca	103	2.19	23	390	1.38	3	5.43	0.043
Ni	132	1.74	24	306	2.05	3	1.94	0.093
Ca ²⁺ + Mg ²⁺	170	1.5	26	430	1.4	3	4.34	0.045
SO ₄ ²⁻	1220	3.1	26	4600	1.4	3	5.12	0.11
<i>Krasnosel'kup, July, 1999</i>								
Cu	301	1.9	21	175	1.2	3	10.8	0.001
Ni	136	1.6	21	533	1.3	3	6.85	0.007
Se	036	1	1	10	1.1	2	13.4	0.003
Ca ²⁺ + Mg ²⁺	184	1.4	27	284	1.3	3	3.17	0.14
SO ₄ ²⁻	470	2.6	21	5300	1.4	3	6.6	>0.001

that diurnal concentrations of impurities vary substantially during an observation period of one month. Because of this, one cannot exclude the possibility of the influence of nearby sources, including local ones. It can be demonstrated that the probability of the change of diurnal average concentrations is well described by the lognormal distribution. The experimental data on the changes of diurnal concentrations for Ni, Cu, Ca²⁺ + Mg²⁺ are presented in Fig. 6 in lognormal anamorphism. The horizontal axis represents logarithms of the concentrations of different components ($y = \ln x$), the vertical axis represents the values of func-

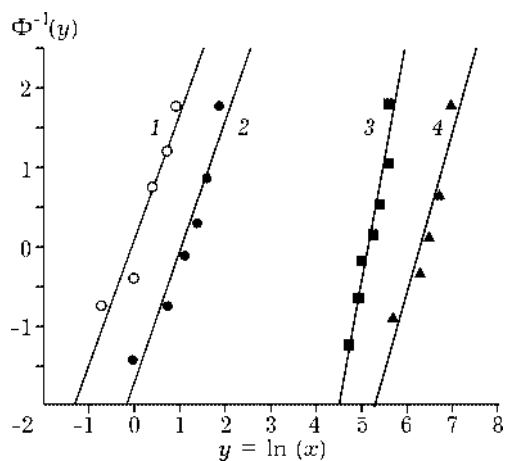


Fig. 6. The regularity of the variability of diurnal concentrations: 1 - Ni, 2 - Cu, 3 - Ca²⁺ + Mg²⁺, 4 - SO₄²⁻.

tion reverse to the integral of normal distribution ($\Phi^{-1}(y)$). Points show experimental results. The straight line is drawn by means of least squares. One can see a good correspondence between the experimental data and their approximation by the lognormal distribution; points fit the straight line.

Table 5 shows the characteristics of the content of the mentioned impurities in AA during the period containing no episodes of increased concentrations, and the period with increased concentrations. These characteristics include the geometric mean concentration x_{50} , standard geometric deviation σ_{ig} , and the number of days n for which the parameters of distribution were calculated. The value of t -criterion demonstrating the difference between geometric mean concentrations during peak-bearing period and during the rest time is also shown.

The last column shows the probability for the geometric mean concentration peak to appear if the regularities of temporal variability corresponded to the period having no peak values. If we assume the appearance of maximal concentrations to be random, then, the probability for the appearance of all the elements in one and the same moment (period) will be equal to the product of the probabilities for each component. Then such a probability will not exceed $1.9 \cdot 10^{-5}$ for the episode in Tarko-Sale, while in

Krasnosel'kup it will be $2.9 \cdot 10^{-12}$. It is clear that in the case of such a low probability the appearance of such an event is not chancy.

If we assume that the observed sharp increase in concentrations is connected with the appearance of emission from some local source in the sampling site, this will be in contradiction to the results reported in [14]. The rate of SO_2 emission from the most powerful local sources was determined in this work using the content of this ion in snow cover in the region near the burning oil and gas torch. For example, the rate of emission within the whole snow cover period (about 8–9 months a year) is about 300 kg. The radius of torch influence is about 1 km. At a large distance, SO_4^{2-} content in snow is close to the background level. The maximum content is only 2–3 times higher than the background one; however, the background level of SO_4^{2-} in snow is determined by global pollution, its effect being spread over hundreds kilometers.

For submicron particles, the velocity of their dry deposition usually varies from 0.5 to 1–2 cm/s. If we accept that the background concentration is determined by the geometric mean during a period without any episodes of sharp increase of SO_4^{2-} , then, according to our data, for Tarko-Sale, the value is $1.2 \mu\text{g}/\text{m}^3$; for Krasnosel'kup, it is $0.5 \mu\text{g}/\text{m}^3$. Accepting the rate of dry deposition to be equal to 0.5 cm/s, we obtain the density deposition of the "background" during the whole snow cover period (8 months) in Tarko-Sale to be $120 \text{mg}/\text{m}^2$, and in Krasnosel'kup $50 \text{mg}/\text{m}^2$. According to the data of [14], in the area under the influence of oil and gas torch in Tarko-Sale, the mean density of deposition is about $100 \text{mg}/\text{m}^2$. In Krasnosel'kup, there are no such powerful anthropogenic sources, so the appearance of a sharp increase in concentration, several times as much as the background value, cannot be explained by the effect of local sources.

CONCLUSIONS

A method is proposed that allows one to determine the location of the sources of pol-

lution on local and regional scales, and to estimate the emission rate of these sources using the experimental data on spatial-temporal variability of the content of some components of AA and the trajectories of air mass transport.

The annual amount of Ni, Cu, Se, $\text{Ca}^{2+} + \text{Mg}^{2+}$ emission by the Noril'sk Mining and Metallurgical Plant, and Ni, Ca, $\text{Ca}^{2+} + \text{Mg}^{2+}$ emission of the plants of the South Urals is estimated.

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