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DETECTION OF ATMOSPHERIC POLLUTION SOURCES BY USING CROSS-PLUME SCANNING METHOD AND MOBILE RAILWAY LABORATORY

ABSTRACT. In this study the power of the sulfur dioxide emissions from the Mid-Urals copper-smelting enterprise (MUCE) was estimated by using plume cross-scanning. The combination of the observational data obtained by the TROICA experiments and information obtained by satellite photos of the Earth's surface together with the ISCST3 dispersion model is promising for studies of the short-range atmospheric transport of chemically inactive pollutants. The results of ISCT3 model simulations indicate that the SO_2 emissions in terms of sulfur make up about 3–4% of the plant sulfuric acid production. Also the cross validation between ISCST3 and NOAA HYSPLIT dispersion models was carried out. The emission rate obtained at the NOAA HYSPLIT model simulation is 1.5 times higher than the emission rate calculated at the ISCST3 simulation. It was emphasized, that the using of mobile platforms on electric traction has advantages in studying the environmental situation in comparison with the measurement system, constructed on the stationary Environmental Protection Stations. The cross-plume scanning method to a lesser degree depends on the wind rose, the features of the landscape and a relative location of emission sources and sensors.

KEY WORDS: atmospheric pollution, mobile railroad laboratory, TROICA experiments, quasi-stationary plume, cross-plume scanning

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INTRODUCTION

The TROICA international experiments (TRanscontinental Observations Into the Chemistry of Atmosphere) performed since 1995 have worldwide reputation (Crutzen et al. 1998), (Oberlander et al. 2002), (Tarasova et al. 2005), (Elansky et al. 2009).

The modern TROICA mobile railroad laboratory, which was manufactured in 2004 after a special design, is equipped with high-precision instruments intended for monitoring the gas and aerosol composition of the atmosphere and the radiative and meteorological characteristics and also for monitoring the soil, water, and vegetation pollution. At present, the laboratory is able of continuous real-time monitoring the concentrations of ozone (O_3), nitrogen oxides (NO and NO_2), carbon oxides (CO and CO_2), methane (CH_4) and a sum of

non-methane hydrocarbons (NMHC), sulfur dioxide (SO_2), ammonia (NH_3), about 30 volatile organic compounds (VOCs), and radon (^{222}Rn) and its decay products, the mass and number concentrations of aerosol particles (including soot) in the size range from 2 nm to 15 μm, and the entire spectrum of meteorological parameters and solar-radiation characteristics with a high temporal resolution (from 10 s to 20 min), see (Berezina et al. 2013), (Skorokhod et al. 2017), (Vasileva et al. 2017). As a result of cooperation between a number of Russian and world scientific centers, a series of 15 international experiments on observations of the state of the atmosphere over vast Russian regions, from Moscow to Vladivostok and from Murmansk to Kislovodsk, was performed. The peculiar features of the spatial and temporal variations in atmospheric greenhouse gases and in chemically active substances, including ozone-destroying ones, were first obtained. The main factors determining the air quality were first revealed and systemized for different cities against their population, infrastructure, and character of the surrounding country. The causes of formation of extreme ecological situations, such as forest fires, methane leakages from the systems of natural gas transportation, transport of polluted air from neighboring countries, and increases in the oxidative properties of the atmosphere near electric power lines and other objects of power industry, are established.

The TROICA experiments along the Moscow circuit railroad gave unique information on the megapolis effect on the state of the environment; see (Elansky et al. 2010). Such information cannot be obtained on the basis of the traditional means of observations. The discussion about quality of the emission databases available for Russia, in particular EDGAR v4.2, and the comparison of these databases with the measurements carried out by our laboratory can be found in (Elansky et al. 2016) and (Eansky et al. 2018).

One of the important possibilities of this laboratory is the control over the implementation of International ecological agreements (the Kyoto and Montreal Protocoles, the Convention on the transboundary transport of pollutants, desertification of territories, etc.), substantiation of the distribution of quotas for emissions of greenhouse gases, and arbitration in the course of revealing the sources of atmospheric pollutants.

In the present work, an example of using of the TROICA mobile laboratory for revealing the sources of atmospheric pollutants is presented. The work includes the detailed study of the space–time distribution of the concentrations of pollutants in plumes under quasi-stationary conditions by the example of the studies of the sulfur dioxide concentration distribution near the city of Pervoural'sk.

DESCRIPTION OF THE WORK AND RESULTS

Quasi-stationary plumes

As is known, mobile laboratories performing studies of the atmospheric surface layer can be arranged on motor-car, aircraft, and railroad platforms. Due to the use of internal-combustion engines, the laboratories equipped on motor-car platforms provide no possibility of measurements under background conditions.

The aircraft laboratories have advantages and disadvantages. On the one hand, as a rule, they are capable of performing measurements of the concentrations of pollutants at heights exceeding 4000–5000 m, i.e., above the atmospheric mixing layer. On the other hand, their spatial resolution is low, about 1.6–2.2 km, due to high velocities of the flights (the cruising speed of the majority of aircraft laboratories is 600–800 km/h); this estimated resolution relates to the situation when the instrumentation installed in the aircraft laboratories is the same as that used now in the TROICA experiments and allows measurements with the 10-s intervals.

Notice that the resolution of the instrumentation installed at the mobile railroad laboratory is about 110–160 m at a mean train velocity of 40–60 km/h. Therefore, the TROICA experiments give the

unique possibility of studying the dispersion scattering with a spatial resolution of about 100 m.

Thus, the TROICA mobile railroad laboratory has evident advantages over the mobile laboratories installed on motor-car and aircraft platforms. Let us compare the possibilities of the stationary observation stations and of the TROICA mobile railroad laboratory as applied to studies of the atmospheric plumes.

In the case of the cross wind, the TROICA measurements allow for obtaining the cross-section of the distribution of pollutants from an emission source. The situation when the time of one measurement in a plume is much less than both the time of the variations in the plume concentrations and the time of the variations in the atmospheric properties is typical.

$$\begin{cases} t_0 \ll \tau_{em} \\ t_0 \ll t_{atm} \end{cases}$$

where t_0 is the time period of one registration of an enhanced concentration in a plume; τ_{em} is the characteristic time of the variations in the emission power; t_{atm} is the time of the variations in the atmospheric properties

Hereafter, such plumes are termed the quasi-stationary ones. The authors make a special emphasis on the principal impossibility of obtaining the space–time distribution of pollutants and the spatial dynamics of the variations in the chemical composition inside quasi-stationary plumes on the basis of the network of stationary ground-based observation stations. The system of stationary observations is based on the passive downwind scanning. To obtain the spatial distribution of pollutants in a plume, the record time at the observation station should exceed the characteristic time of variations in the atmospheric parameters. However, for this time period, the power of the emission source and the chemical composition of the plume may change. Notice that each of the approaches, i.e., monitoring with the network of ground-based observation stations and monitoring with mobile laboratories, has its advantages and disadvantages with a view to studies of dispersion processes.

The disadvantage of the stationary observation systems is a strong effect of the local conditions in the areas of location of stations, including the wind rose, lay of land, and local windward emission sources.

At the same time, the platform vibrations and excesses in the "background" characteristics near motorways and railroads over the background ones in remote areas should be attributed to disadvantages of mobile laboratories intended for monitoring the atmospheric state. However, for the TROICA expeditions, in the case when the mobile laboratory moves along electrified railroads, the excesses in the concentrations of the main pollutants under study are insignificant and do not influence the general pattern of distributions of the pollutants.

When the TROICA carriage-laboratory moves along an electrified railroad and is coupled just after the locomotive, it allows for obtaining information on the undisturbed atmospheric layer not affected by the train pollutants, because the constructional features of the laboratory provide air sampling from the counter airflow through the air inlets located at the top of the carriage-laboratory over the top of the majority of the Russian electric locomotives.

Summing up the aforesaid, we can state that the TROICA mobile railroad laboratory gives a unique possibility for studying the processes of transport of atmospheric pollutants.

General characteristic of the emission source

Along the Trans-Siberian Railroad, the industrial zones and large-scale complexes of most cities are located in the immediate vicinity of the railroad. Therefore, it is of interest to solve the "inverse" problem of determination of the location and power for an emission source from the data on the cross-sections of the plume. We solved this problem by using the standard Gaussian methods. Notice that the Gaussian

methods (OND-86, ISCST3) are applicable for distances up to 30–50 km from an emission source and for weakly-uneven territories with dispersed buildings.

By using the SQL search system, the extreme value (35.1 ppb) for the sulfur dioxide concentration measured in the course of the TROICA-8 *East* expedition was taken from the TROICA-DB. The NOAA HYSPLIT model of the back trajectories for heights of 100, 500, 1000 m (a.g.l) was used in the framework of isobaric approximation of vertical mixing for the primary analysis of the transport of air masses enriched with sulfur dioxide near the city of Pervoural'sk. Details about NOAA HYSPLIT model see in (Draxler and Hess 1998), (Draxler 2006), (Stein et al. 2016), (HYSPLIT Model Guide). Basing on analyses of the back trajectories, of the pollutant concentration distribution along the mobile-laboratory route (Fig. 1), and of the data of video cameras installed in the laboratory, we concluded that the emissions of pollutants are not associated with the work of the Pervoural'sk industrial objects but are fully determined by emissions from industrial objects located to the south-west of the observation site. Fig. 1 presents the distributions of nitric oxide, nitrogen dioxide, and sulfur dioxide along the TROICA route; these distributions show that, at a point in time when the sulfur dioxide concentration in the plume was maximal. the plume concentrations of nitric oxide and nitrogen dioxide were already at the background level.

Cartographic studies of the expected emission sources located to the south of Pervoural'sk allowed for making the unambiguous conclusions that the observed anomalous increase in the sulfur dioxide concentration is associated with ejections of the Mid-Urals copper-smelting enterprise (MUCE) (town Revda, 56.85°N, 59.91°E). Notice that the main types of the MUCP products are black copper, sulfuric acid, oleum, tripoliphosphate, and xanthogenate.

The spatial distribution of pollutants by the Gaussian model of scattering

Let us use the *ISCST3* Gaussian model to calculate the spatial distribution of pollutants (ISCST3 1995). According to the model, the complete equation giving the spatial distribution can be expressed as follows (1)-(2):

$$C = \frac{QKV}{2\pi \cdot u_s \sigma_y \sigma_2} \times \exp\left(-0.5\frac{y^2}{\sigma_y^2}\right)$$

where: $C(x,y,z)$ (μg/m³) is the pollutant concentra-tion at a point with coordinates; (x,y,z) (g/s) is the ejection power for a substance; K=1·10⁶ is the scaling factor; V is the characteristic of vertical scattering; $ó_y ó_z$(m) are the vertical and horizontal standard deviations, м; u_s(m/s) is the wind velocity at the source effective height.

The *ox* axis is directed downwind from the emission source, the *oy* axis is perpendicular to the *ox* axis and lies in the horizontal plane, the *oz* axis is vertical. Under conditions with no dry deposition, the vertical-mixing coefficient is as follows:

$$V = \exp\left[-0.5\left(\frac{z-h_e}{\sigma_z}\right)^2\right] + \exp\left[-0.5\left(\frac{z+h_e}{\sigma_z}\right)^2\right]$$

$$+\sum_{k-1}^{\infty}\left(\begin{array}{l}\exp\left[-0.5\left(\frac{H_1}{\sigma_z}\right)^2\right]+\exp\left[-0.5\left(\frac{H_2}{\sigma_z}\right)^2\right]\\ +\exp\left[-0.5\left(\frac{H_3}{\sigma_z}\right)^2\right]+\exp\left[-0.5\left(\frac{H_4}{\sigma_z}\right)^2\right]\end{array}\right)$$

Here, $H_1 = z-\left(2kz_i - h_e\right)$,

$H_2 = z+\left(2kz_i - h_e\right), H_3 = z-\left(2kz_i + h_e\right)$,

and $H_4 = z+\left(2kz_i - h_e\right)$;

h_e is the emission-source effective height; z_i is the height of mixing.

The initial data necessary for the emission calculations, namely, the wind velocity and direction, temperature, and pressure, are obtained from the WMO#28440 (Ekaterinburg, meteo.infospace.ru) and

NOAA (www.arl.noaa.gov/ready/amet.html) meteorological stations and from the database on the temperature profiles measured with the MTP-5 instrument during the TROICA expeditions. The atmospheric boundary layer state was characterized by the vertical temperature profile. Fig. 2 gives the vertical temperature profile measured by the MTP-5 instrument near the point of maximal sulfur dioxide concentration. Fig. 2 also characterizes the height dependence of the atmospheric state (the stability class by Pasquill).

It is seen that, at low heights (up to 250 m), the atmosphere is unstable; at heights of 250–300 m, the atmospheric state is neutral (D); and, at heights above 550 m, the atmospheric is of low stability (E). Thus, the form of the plume depends strongly on the pollutant-emission effective height and on the height level of pollutant propagation. If the distance from the point of registration of an anomalously-high pollutant concentration to the pollutant source is not long (10–12 km), it can be considered that, during the pollutant propagation, the pollutant has no time for rising to a height of more than 250 m. Therefore, we take that the atmosphere within the plume is somewhat instable (class C according to Pasquill).

The subsequent calculations were performed by the *ISCST3* method. To calculate the emission-source effective height h_e, the Brigg coefficients (F_b and F_m), the distance (x_f) from the pollutant source to the point of the maximum pollutant concentration, and the wind velocity (u_s) at the effective height were calculated. The dispersions ($ó_y$) and ($ó_z$) along the axis oriented perpendicularly to the plume in the horizontal plane and along the axis oriented upward, respectively, were calculated by the Pasquill-Gifford formulas, see (Briggs 1971), (Briggs 1972).

In the calculations, the following additional parameters of an industrial pollutant source were a priori taken: the smokestack height (h_s) is about 120 m, the smokestack diameter (d_s) is 4.5 m, the gas temperature (T_s) is 150^{0}C, and the emission rate (v_s) is 10.0 m/s.

The percentage Δ of the sulfur emissions in the enterprise total sulfur production was calculated on the basis of the data for 2004 when 483 442 tons of sulfuric acid was produced. The results of calculations with the *ISCT3* model are presented in Table 1. It is seen that SO_2 emissions in terms of sulfur make up about 3–4% of the enterprise sulfuric acid production. The experimental data and the values calculated with the ISCT3 model for different classes of atmospheric stability were presented in Fig.3. Notice that the figure gives the transverse dispersion projected on the route trajectory, i.e., considering the angle between the wind and route directions.

Notice that the choice of the stability class C proved to be justified, because the dispersions for the stability classes D and E turned out to be smaller than those measured in the course of the TROICA expeditions. This means that, really, the upward flows of the warmed pollutant did not reach the height layers located above 350–400 m, where stable atmospheric conditions occur.

The spatial-temporal SO_2 distribution in the vicinity of town Pervoural'sk

In most cases, simple accumulation of information without consideration of its spatial distribution does not allow the performance of a sufficiently accurate analysis of ecological situation. A great body of data, which is typical for monitoring, is usually difficult for perception until data are visualized on the map. Mapped data allow for revealing the regularity in the distribution of objects or phenomena and their variations in space and time.

Therefore, one of the main aims of the modern geoinformatics is the creation of an integrated database that accumulates the advanced geoinformation technologies. The geoinformation system (GIS) applied in this work consists of the ArcInfo 9.3 together with the GoogleMap space photos and the raster regional itineraries related to reference marks by using the Reoreferencing/ArcInfo standard technology.

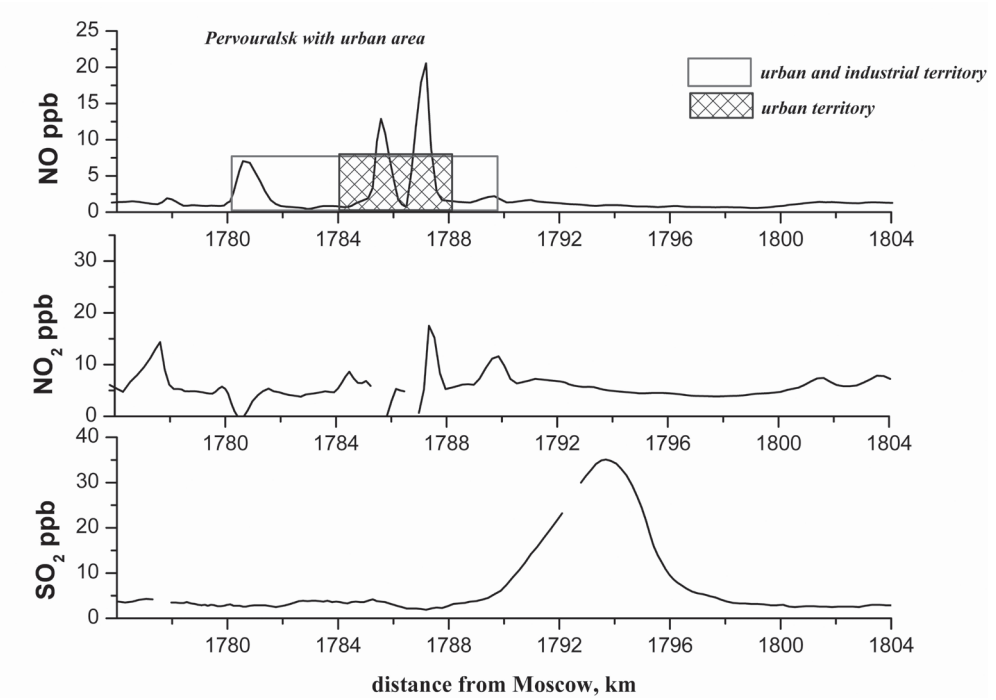

Fig. 1. NO, NO_2 and SO_2 surface concentrations along Trans-Siberian railway in the vicinity of town Pervoural'sk according to mobile laboratory measurements in March, 2004

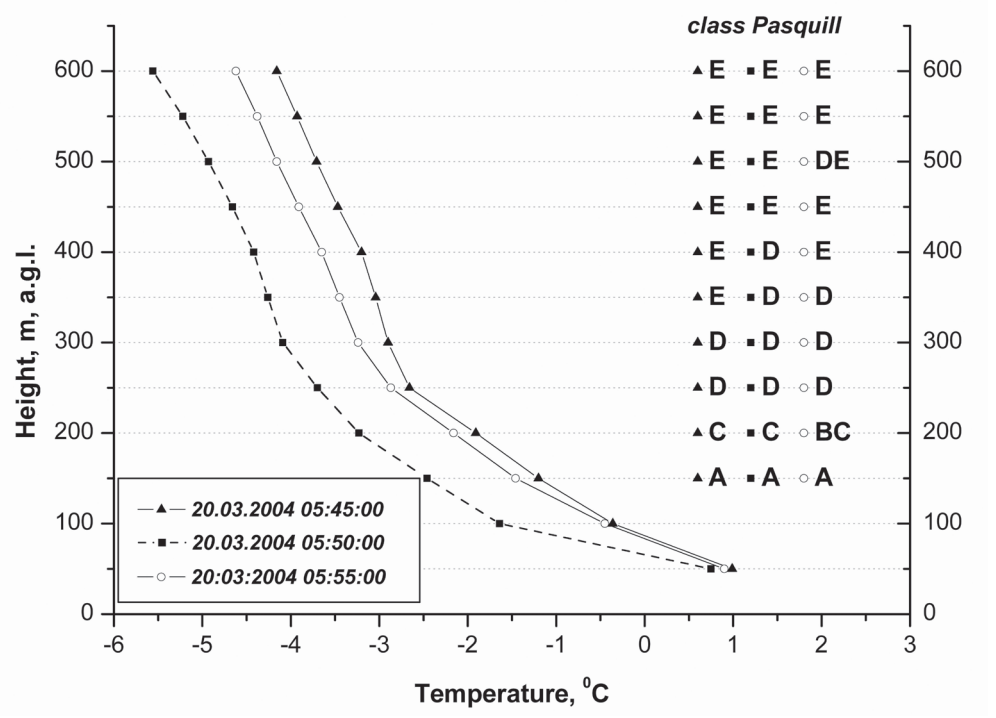

Fig. 2. Temperature vertical profile according to MTP-5 temperature profiler data near sulfur dioxide maximum point

Table 1. Results of calculations on model ISCST3

Model ISCST3		Stability class according to *Pasquill*					
parameter	dimension	A	B	C	D	E	F
u_s	m	3.3	3.3	3.6	4.1	6.7	11
$Ó_y$	m	1527	1163	813	539	403	269
$Ó_z$	m	5000	1385	508	136	79	47
F_b	m^4/s^3	178	178	178	178	178	178
F_m	m^4/s^2	324	324	324	324	324	324
x_f	m	946	946	946	946	515	639
h_e	m	380	380	362	333	206	175
V	–	38.33	10.98	4.24	0.40	0.07	0.00
Q	g/s	384	282	205	435	1707	35152
	t/year	12064	8855	6460	13673	53702	1105514
$Δ_{\%}$	%	3.8	2.8	2.1	4.3	17	350

As a cartographic basis of Fig. 4, Google Map satellite-borne photography of the Earth's surface is taken. The Mid-Urals copper-smelting enterprise (MUCE) plume of sulfur dioxide concentration distribution calculated with the above-mentioned *ISCST3* model was mapped. The histogram of Fig. 4 designed on the basis the TROICA-8 *East* expedition data gives additional information on the SO2 distribution. It shows that the *ISCST3* model allows a good spatial description of observational data for the 10-km vicinities of pollutant sources.

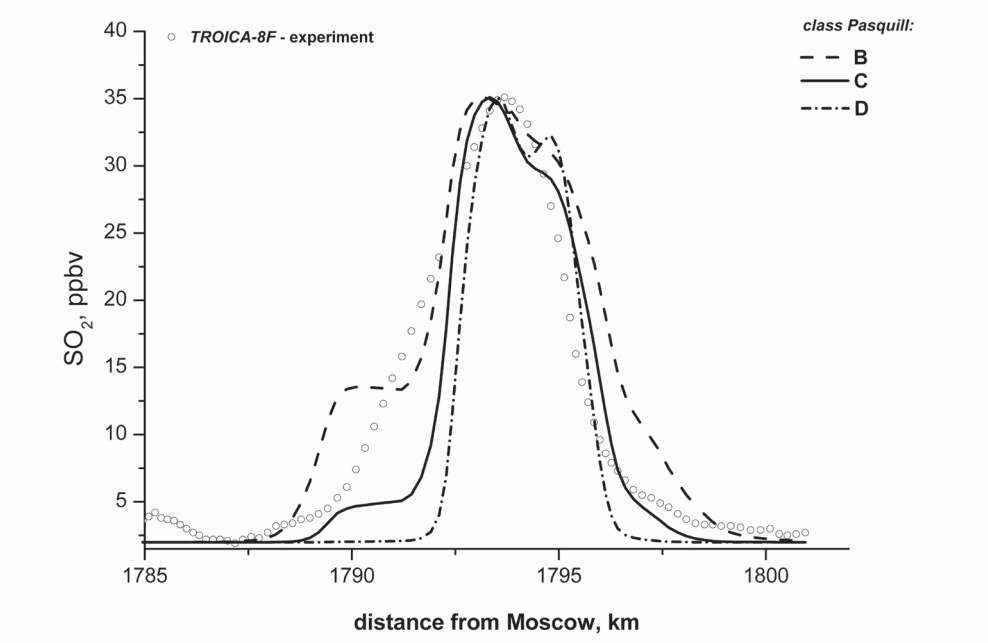

Fig. 3. Experimental data and values calculated on *ISCST3* model for different atmosphere stability classes

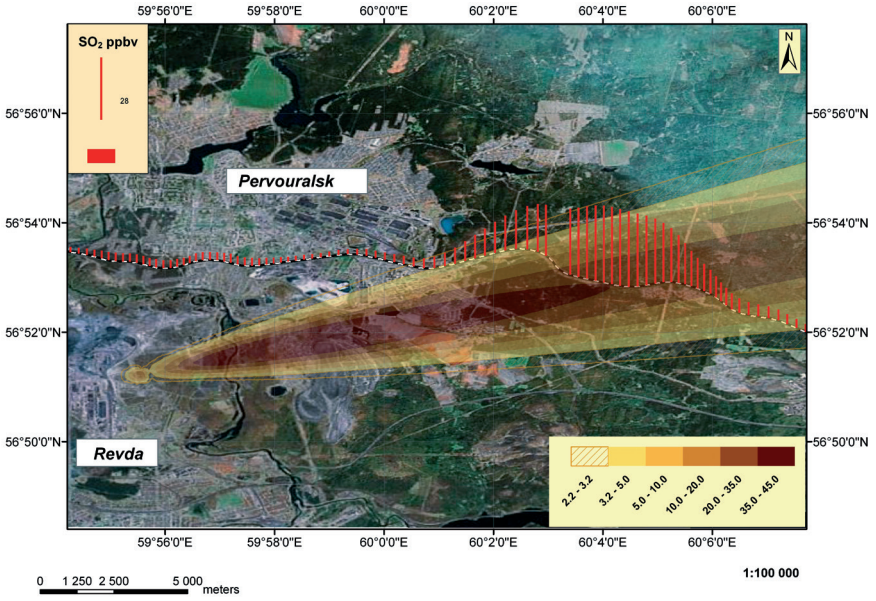

Fig. 4. The SO_2 plume from от Mid-Urals copper-smelting enterprise calculated on *ISCST3* model and shown on satellite image (GoogleMap)

For comparison, the result of the dispersion NOAA HYSPLIT model application to calculations of the spatial distribution of emitted pollutants is given in Fig. 5. It is seen that significant atmospheric pollution expends over a distance of 100–160 km. The estimation of the pollutant-emission power with the dispersion NOAA hysplit model on the basis of the hourly-mean pollutant concentration measured in the 100–200 m (a.g.l.) atmospheric layer leads to a value of 350 g/s. This value is somewhat higher than the value 205 g/s calculated with the ISCST3 model. The difference between these results is possibly caused by the application of the hybrid dispersion TH-part model (the Top Head and Lagrange approximations for the horizontal and vertical directions, respectively) to the calculations with the NOAA HYSPLIT model. The TH-part model underestimates the vertical mixing at small distances from pollutant-emission sources.

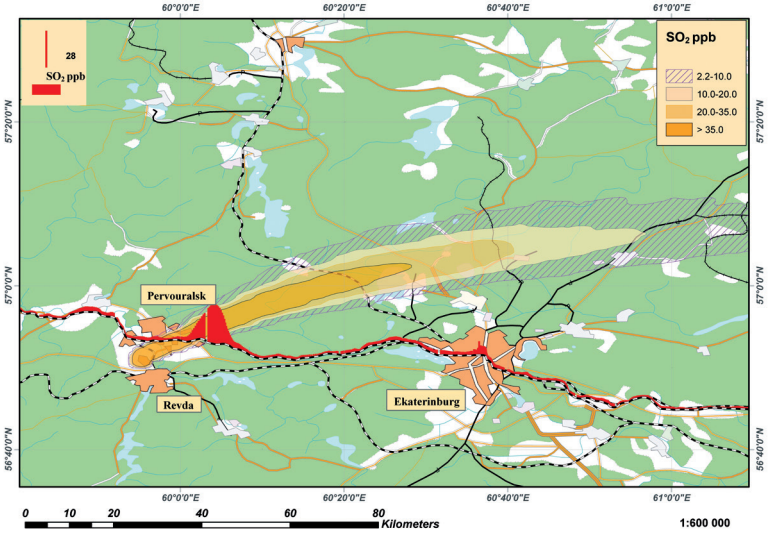

Fig. 5. SO_2 carrying-out spatial distribution calculated by NOAA HYSPLIT dispersion model

CONCLUSIONS

A procedure allowing the retrieval of the locations and powers of sources of atmospheric pollutants from the concentrations and directions of the atmospheric short-range plumes of chemically inactive pollutants monitored by the mobile railroad laboratory is developed. As an example, the power of the sulfur dioxide emissions from the Mid-Urals copper-smelting enterprise (MUCE) is estimated. It is shown that, for estimation of the powers of harmful emissions from the polluting sources located in flat regions at a distance up to 50 km from the Trans-Siberian Railroad, the ISCST3 computational model combined with the detailed satellite-borne photography of the Earth's surface can be successfully applied. It is noted that a combination of the observational data obtained by the TROICA experiments and taken from the geoinformation system (ArcInfo) and of the satellite-borne photos of the Earth's surface (GoogleMap) with the ISCST3 dispersion model is promising for studies of the short-range atmospheric transport of chemically inactive pollutants.
The cross-plume scanning method by using a mobile platform provides a number of advantages over the standard method of assessing of the environmental situation by means of a network of stationary stations. This method to a lesser degree depends on the wind rose, the features of the landscape and a relative location of emission sources and sensors.

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