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Study of transboundary transport of atmospheric pollutants in the region of Belarus

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The lidar and radiometric complexes for measurement of aerosol and ozone contents in the atmosphere are constructed. An algorithm is developed for joint processing of lidar and sun photometer data. To obtain information from adjacent regions, we used the data of global and regional observation networks of atmospheric monitoring. Data on ozone layer in the region of Belarus are presented. Development of ozone anomalies is explained by the process of migration of "ozone holes." The program for calculation of three-dimensional air mass trajectories in the free atmosphere and atmospheric boundary layer is developed. The created local and interregional models of aerosol and gas transport on the scales from a few tens to hundreds of kilometers are adapted to conditions of Belarus. The influence of big industrial centers on pollution of Belarus territory by aerosols and gases is estimated.

Study of transboundary and interregional transport of pollutants in the atmosphere caused by natural phenomena and anthropogenic catastrophes, and estimation of its influence on ecosystems and climate are the priority problem of environmental protection. An important stage in construction of the interregional transport monitoring system in Europe became the activity in the framework of the Cooperative program for monitoring and evaluation of the long range transmission of air pollutants in Europe (EMEP) launched in 1980s.

As a part of EMEP, monitoring stations in coordination measured atmospheric admixtures. However, the measurements mostly are conducted by local methods in the near-ground laver and the data are collected for relatively large time interval. This precludes from efficient control for pollutant fluxes in the atmosphere because the large-scale transport proceeds largely outside the near-ground layer. Consequently, this complicates the direct use of the measurement results for estimation and correction of model calculations of pollutant transport.

Presently, the European Union forms the monitoring programs with the use of recent achievements in the field of remote atmospheric sensing, able to give real-time information. Just this approach is implemented in the Governmental program of applied scientific studies in Republic Belarus "Creation of instrumentation complex and methods of monitoring of transboundary and interregional pollutant transport in the atmosphere with use of remote and local measurement systems" ("Transboundary monitoring"). The work involved 15 organizations of National

Academy of Sciences of Belarus, Ministry of Education, and Ministry of Nature. Below, we overview the results obtained under the auspices of this program in 2004-2005.

In a number of cases, for illustration of certain phenomena, the results of other years are given as well.

Instrumentation

For the field studies of the processes of largescale transport, instrumental complex of atmospheric remote sensing was organized. At Institute of Physics NAS of Belarus (IP), the panoramic and stationary multiwavelength lidar systems for aerosol sensing, as well as lidar for sensing of ozone in the stratosphere were constructed.¹ Intercalibration of instruments and processing algorithms was performed within the European lidar network EARLINET.² At the National Scientific Research Center of Monitoring of Ozonosphere at Byelorussian State University, the device for measurement of the total ozone content in the atmosphere was produced.³⁻⁵ The results of satellite observations were used to predict air pollution by forest fire products. In the processing of lidar, radiometric, and satellite observational data, both new algorithms and well-known methods were used.

The algorithm is developed for estimation of the vertical distribution of concentrations of coarse and fine fractions in the troposphere aerosol, based on the data of correlation sensing of aerosol layer by multiwave lidar and sun radiometer CIMEL.6,7 The a priori aerosol model assumed that the aerosol consisted of two fractions (coarse and fine with inter-

fraction border about 0.5 µm) with parameters, coinciding with modes of altitudinally integrated particle size distribution, reconstructed from CIMEL data. At the same time, the concentrations of aerosol fractions could vary with altitude. Base equation system, determining the relationship between measured optical signals and parameters of the aerosol layer, was constructed. It included lidar equations for all working wavelengths; equations for photometric quantities, integrated over the layer (spectral optical depth or altitudinally integrated concentrations of fine and coarse fractions, etc.); and limitations on the smoothness of the profiles of aerosol fraction concentrations. Likelihood function for the model was constructed, and then the altitude profiles of concentrations of fine and coarse fractions, ensuring maximum of likelihood function, were determined.

An algorithm is suggested allowing to detect small fires and estimate their areas in pixels in the presence of false alarm factor (clouds or flare from the water surface) by separating the contributions from different regions of the Earth's surface to the recorded radiation. The algorithm uses the local *a priori* information on the spectral behaviors of responses from different pixel components, as well as parameters of the current state of the surface and the atmosphere (temperature, humidity, meteorological visibility range, etc.). As a result, the probability of fire detection increased from 60 to 80%. The instrumentation has been developed allowing the accuracy increase in determination of coordinates of detected temperature anomalies.

Study of transboundary transport, in addition to information on Belarus atmosphere state, requires the similar data in adjacent regions. Since 2000 in Europe, the regular measurements of aerosol parameters are conducting by the lidar network EARLINET.² At present time, the network incorporates more than 20 stations in Europe, including the lidar station of the Institute of Physics of NAS of Belarus. Since 2005, in NIS the coordinated observations are conducted at the lidar network CIS-LiNet.¹

One of the important purposes of construction of the CIS-LiNet is organization of coordinated lidar observations at the Eurasian continent through integration of EARLINET, CIS-LiNet, and East-Asian lidar network (AD-Net).⁸

Detailed studies of aerosol parameters by means of sun sky-scanning radiometers CIMEL are made within worldwide network AERONET (Fig. 1), while ozone content is studied by different methods in the NDSC network.

The results of observations in above-mentioned monitoring networks were used in solution of the program "Transboundary monitoring".

Ozone and aerosol in the upper atmospheric layers

Ozone is one of the most variable constituents of the earth's atmosphere. Total ozone (TO) content in the atmosphere above each particular region is determined by photochemical processes of its formation and destruction in the upper stratosphere and by the processes of ozone transport by air masses in the lower layers. At the middle to high latitudes, ozone mostly is accumulated in the lower and middle stratosphere, i.e., at the altitudes, where a major role in ozone budget is played by the transport processes. Therefore, the factor of large-scale ozone transport with air masses plays an important role in ozone formation over the territory of the Republic in all seasons.

General pattern of TO variations in Belarus during last years is presented in Fig. 2 according to data of lidar measurements, TOMS satellite data (TOMS, http://toms.gsfc.nasa.gov/teacher/ozone_ overhead.html), and results of the measurements at the National Scientific Research Center of Monitoring of Ozonosphere (NSRCMO) (http://www.tm.basnet. by/ru/index.htm). The TO content varied in the range 220-450 DU.

The TO estimates, obtained on the basis of lidar and TOMS measurements, are characterized by the correlation coefficient equal to 0.8. Of note is the frequent occurrence of negative ozone anomalies, i.e., phenomena representing rapid, short-term and considerable (in absolute value) decrease of TO in a local region of the atmosphere. An example of migration of such anomalous regions is presented in Fig. 3.

The ozone anomalies develop mainly in North Atlantic and arrive at the region of Europe by transporting with air masses from these local ozonerich or ozone-poor regions. The TO-deficient regions migrate eastward, changing in shape and size. The depth of the anomalies first increases, and then decreases and vanishes. Mostly, there are the final stage of anomalies above Belarus. The TO content deficiency is 20–30% in this case.

The aerosol in the upper troposphere and stratosphere was monitored simultaneously with measurements of ozone concentration profiles. Main information source for retrieval of aerosol parameters is the 532 nm channel.

Based on the results of lidar sensing, the altitude profiles of aerosol backscattering coefficient $\beta_a(h)$ were reconstructed. The main sources of aerosol appearance in the stratosphere are troposphere–stratosphere exchange and powerful volcanic eruptions, whose emissions may "break through" the tropopause and deliver aerosol substance to the stratospheric layer. Large volcanic eruptions exert powerful pulsed impact on the stratospheric aerosol layer. Subsequently, due to troposphere–stratosphere exchange, the stratosphere layer returns to the equilibrium (background) state.

The last eruption, most powerful over the observation period, was explosion of Pinatubo volcano on June 15–16, 1991. As a result of the eruption, huge amount of gases and aerosols was injected to the atmosphere. By satellite-based estimates,⁹ the mass of injected sulfur dioxide was of the order of 20 Mt, three times larger than SO_2 emissions produced by preceding powerful eruption of El Chichon volcano in 1982.



* — EARLINET, $\mathbf{0}$ — CIS-LiNet и \blacksquare — AD-Net





Fig. 1. Worldwide networks of the atmosphere state monitoring: lidar networks of Europe and NIS and East Asian countries (a); worldwide photometer network AERONET, designed for monitoring of the spectral atmospheric transparency and aerosol composition (b); ozonometer network NDSC (c).



Fig. 2. TO transformation according to data of lidar, TOMS, and NSRCMO measurements.



Fig. 3. Negative ozone anomaly on January, 07–10, 2004 (deviation from multiyear average ozone content is shown).

In the stratosphere, this has led to formation of powerful aerosol clouds consisting of droplets of water solution of sulfuric acid with total mass of 20–30 Mt.¹⁰ Such, the stay of the stratospheric aerosol layer at near-background state was interrupted. Since this moment, the process of formation of stratospheric aerosol clouds and their subsequent dissipation had begun.

Figure 4 presents the time transformation of integrated aerosol backscattering coefficient of stratospheric layer $B = \int_{h_1}^{h_2} \beta_a(h') dh'$ in the region

of Minsk for period from eruption of Pinatubo volcano to late 2005.



rig. 4. Time transformation of integrated aerosol backscattering coefficient in the stratosphere (above 13 km) at the wavelength 532 nm in the region of Belarus for period 1990–2005.

It follows from these data that the integrated parameter B after Mt. Pinatubo eruption increased by two orders of magnitude for a half-year. Subsequent process of relaxation is characterized by two timescales.¹¹ The first timescale, characteristic for the entire period 1992–1994 and associated with the removal of the coarse fraction, had a length of 280 days. The length of the second one, since 1995, was about 730 days.

Presently, the mean value of the parameter B is approximately 0.0001 sr⁻¹. The dynamics of B variations in recent years gives grounds to believe that it will stabilize near this level in the future. Today, the content of aerosol substance in the stratosphere is the least since 1970s–1980s, when regular lidar observations of the stratosphere began. Thus, there has been no confirmation for concerns, claimed in the scientific literature, about a considerable growth of the background level of stratospheric aerosol substance due to anthropogenic pollution, and for predictions of substantial influence of this mechanism on radiation processes in the atmosphere and possible climatic consequences.

The measurements show that the aerosol scattering coefficient, which can be considered proportional to aerosol mass content, decreases with height in the interval 10–30 km, according to the near-exponential law.

Aerosol pollution caused by anomalous natural phenomena

Measurements in the framework of the EARLINET network show that the dust storms in the northern part of Africa, and primarily in Sahara desert, as well on Arabian peninsula, lead to systematic outbreaks of dust particles not only to south but also to central and north regions of Europe including Belarus. During the year, 8–10 significant dust outbreak episodes are recorded in the region of Belarus. The dust is transported predominately at heights 3–7 km. The elaborated observation procedure includes the analysis of results of the calculations, based on prognostic NAAPS, SKIRIN, DREAM models, and special series of field lidar and radiometric measurements at the stations EARLINET and CIS-LiNet.

An efficient method of diagnostics of dust particles in the atmosphere is the polarization sensing. The degree of depolarization of backscattering by dust particles is approximately 0.1-0.2.

The maximum degree of atmospheric pollution by dust particles, wind blown from Sahara desert, was observed in Belarus in early August 2001, when the optical depth of the dust particles reached 0.6 level at $\lambda = 532$ nm.

In the summer season in Belarus, one of the main sources of atmospheric pollution are the forest and peat fires in the west regions of Russia, Ukraine, and Belarus. The lidar and radiometer measurements make it possible to detect and carefully analyze the smoke spread. Most strong smoke pollution in Minsk was observed in September 2002 (Fig. 5).



Fig. 5. Aerosol optical depth of the atmosphere in the first half of September at $\lambda = 440$ nm.

It is seen, that the air turbidity increased at the beginning of the month, reached the maximum on September 10, and in the next two days the air has been very clean. The lidar measurements have shown that the smog was lifted up to a height of 1-1.5 km. The CIMEL radiometer data indicated the presence of large amount of finely dispersed nonabsorbing aerosol fraction in the smoggy days. This signified that the main pollution sources were fires in the smoldering phase. The calculated trajectories of air mass transport showed that the main pollution sources for Minsk were located on the territory of Belarus. On September 11, the air flows from Arctic regions descended from high altitudes to the near-ground layers in Belarus and expelled the smog.

Pollutions caused by anthropogenic activity

The anthropogenic pollution is an important factor, determining air quality in the region of Belarus.^{6,7,12,13} Study of the effect of large-scale transport on the aerosol characteristics in European region was performed within the framework of the European lidar network EARLINET. To study the altitude transformation of aerosol parameters, we selected the data of lidar observations in the EARLINET network at the stations:

- Aberystwys (52°N, 4°W), which is the edge north-western station of EARLINET network on the coast of England. In the case of westward transport, it is characterized by the minimum degree of the presence of anthropogenic effects. The results of the measurements at the station are considered as background-reference ones and used for comparison with the measurements in other regions.

- Palaiseau (49°N, 2°E), located south-west of Paris. Given westward transport, the station is located before the first industrial Paris region.

- Belsk (51°N, 20°E), located south to Warsaw in forested zone, free of sources of industrial emissions to surrounding regions.

- Minsk (53°N, 27°E), the site of Institute of Physics of NAS of Belarus.

Figure 6 presents the results of lidar EARLINET observations; they are performed in the summer period at a pronounced westward transport, encompassing the European region.¹⁴



Fig. 6. Mean vertical profiles of backscattering coefficient in summer for a westward transport across the stretches of Europe at the stations: Aberystwys, Palaiseau, Belsk, and Minsk. The aerosol optical depth (AOD) and the number of measurement days (in parentheses) are given (data of EARLINET studies¹⁴).

The turbidity parameters were found to increase when passing from west to east regions. Only in Belarus the turbidity somewhat decreased. The minimum aerosol loading at all altitudes was observed in the region of reference station Aberystwys. In Palaiseau, in the layer up to 2 km, the aerosol content somewhat increased. The aerosol loading and aerosol optical depth of atmospheric masses substantially increased in Belsk and Minsk (by five times) after crossing the Europe.

Study of influence of the trajectory of air mass transport on characteristics of aerosol component in the region-recipient was based on determination of correlations between the measured characteristics of aerosol fields and type of the trajectories realized during measurements.

The initial information for statistical analysis were the data of two-year cycle of radiometer measurements of atmospheric aerosol parameters in Minsk, 2002–2004. In the course of data processing, the arrays of aerosol optical depths at $\lambda = 440$ nm, as well as the total contents of small and large particles were compiled. The radius separating the two fractions was chosen at 0.5 µm. The "backward" trajectories were selected for each measurement, along which the admixture was delivered to the measurement point. Initial data for analysis were 5-day backward trajectories for the levels 950, 850, and 700 hPa.

The backward trajectories of air mass transport are sets of coordinates in four-dimensional space (threedimensional space plus time), which characterize the prehistory of air mass migrated, having arrived at the observation point with the given coordinates at a given height within a fixed time interval. Array of data of backward trajectory calculations for Minsk is available from database of AERONET radiometric network.

The method of cluster analysis was used to form into seven trajectory classes with the following properties from the whole array of backward trajectories:

- the back trajectories correspond to the time of measurements of aerosol particle parameters;

- for each situation, the trajectories have close geographic coordinates at three levels, i.e., the air mass transport is similar throughout the layer 950-700 hPa;

- the transfer trajectories inside a given trajectory class are close in Euclidean metrics.

The number of backward trajectory classes was chosen in such a way that the representation of aerosol transport in the region of Belarus was achieved in detail. At the same time, the number of observations of aerosol parameters, corresponding to each class, still remained statistically significant. Based on these criteria, 126 measurements and corresponding sets of backward trajectories were selected.

Figure 7*a* presents the mean values of backward trajectories for each of the constructed classes. It is seen that the 5-day trajectories cover the most part of the European territory.

In accordance with division of backward trajectories into 7 classes, the data arrays of total content of small and large aerosol particles, as well as AODs, were split into 7 subsets. The concluding stage of the data processing was estimation of mean values of the suspended particle parameters in subsets, corresponding to different classes of backward trajectories.



Fig. 7. Dependence of aerosol particle parameters on air mass transport trajectory: average trajectories in seven backward trajectory classes, constructed using the method of cluster analysis (*a*); and distribution of the mean value of the total content of small (*F*) and large (*C*) particles, their ratio (F/C), and AOD at the wavelength 440 nm as functions of the air mass transport trajectory type (*b*).

Figure 7b presents the mean optical and microphysical aerosol parameters, realized in the atmosphere of Minsk, as functions of the type of air mass transport trajectories, along which the admixture was delivered to the observation point.

Figure 7 suggests that the air masses, arriving from the regions located southward to Minsk, bring more particles than the air masses from other regions. At the same, the relative content of small particles increases and the aerosol optical depth is maximal.

The chemical composition of the transported aerosol is very diversified.¹⁵⁻¹⁸ In many cases, the particles may contain heavy metals in amounts, harmful for living organisms. Based on the estimates of Meteorological Synthesizing Center "Vostok" of the EMEP program, the territory of Belarus receives yearly more than 100 t of lead, about 7 t of cadmium, and 2.1 t of mercury. At the same time, the transboundary fraction is more than 80% for lead, more than 70% for cadmium; 99% of mercury sources

are outside the Belarus territory. The territorial structure of fallout levels is presented in Fig. 8.



Fig. 8. Fallout levels for lead (*a*) and cadmium (*b*) on the territory of Belarus (zone is bordered with solid closed line; lines inside this zone define the borders of the regions) according to the data of calculations in the framework of EMEP program, $g/(km^2 \cdot yr)$.

It is seen that the main gradient of fallout levels is from south to north for lead, and from west to east for cadmium. Thus, the maximum calculated fallout levels are characteristic of south (marshy woodland) regions of Belarus for lead and of west regions for cadmium. In calculation with the use of the unchanged approach, surpluses of permissible fallout levels of lead are 14% of areas of all natural plantations of the country, averaging 3.7 g/ha/yr.

The high density of soil pollution by radionuclides after Chernobyl disaster in the alienable zone is potentially dangerous source of air pollution not only in this zone, but also in other regions of the Republic. Presently, the radioactive air pollution is produced under influence of the secondary windinduced lifting (re-suspension) and transport of radioactive particles, which depend on many factors of natural and anthropogenic origin. Experimental studies have shown that in 2004 and 2005 no significant transport of radioactive aerosols, caused by soil erosion and forest fires, was observed. Possibly, those years were relatively quiet relative to fires and atmospheric anomalies.

Simulation of the processes of pollutant transport in the atmosphere

Analysis of pollution transfer requires development of the corresponding transfer models. Therefore, modelers have developed (or adapted to conditions of Belarus) the models of aerosols and gases transfer (and, in particular, radioactive gases), which describe the spread of anthropogenic pollutants from anthropogenic sources (boiler smoke stacks, cities), forest fires, wind erosion of soil with coverage from a few tens to hundreds of kilometers (the local and interregional models). The basis information is data on concentration of polluting admixtures at different heights and amount of components deposited on the earth's surface.

For analysis and prediction of the processes of pollutant transport, a program was developed to three-dimensional air mass transport calculate trajectories in the free atmosphere and atmospheric boundary layer. The program is based on the meteorological data, provided by the Republic Hydrometeorological Center. Since the meteorological centers do not routinely measure the vertical wind speed, the wind value was calculated by meteorological models. Comparison of wind speeds, calculated from three models,¹⁹ made it possible to choose as a preferable tool the model of calculation of vertical adiabatic wind speed from the temperature measurements.

For determination of the velocity field inside the atmospheric boundary layer, we used the literatureavailable parameterization methods of atmospheric boundary layer characteristics. Comparison of backward trajectory calculations by the developed program and using NASA data, with calculations of other authors²⁰ have shown a satisfactory agreement.

In 1986, the explosion and subsequent wind flows caused by the Chernobyl plant reactor disaster have led to radioactive pollution of large territories. The initial information on emissions and meteorological situation in those days was used in numerical simulation of the spread of polydisperse polluting particles to the distance up to one thousand km in variable wind field from nonstationary source. Nonstationary trace of the pollution was calculated, as well as the procedure was constructed to estimate the amount of the deposited admixture versus time having elapsed since disaster at different points of the polluted region. Figure 9a presents the positions of the pollution flows at different times from the emission start. They reflect the fact that the south-east wind was blowing for several first hours after disaster.



Fig. 9. Positions of admixture flow at different times elapsed since the beginning of Chernobyl plant reactor disaster (*a*); and admixture concentrations in the middle polluted zone (without taking into account the pollution of near zone by large particles) (*b*). Contour values of concentrations are 0.01; 0.03,..., 3.0 Ci/km².

For five days, the wind direction slowly changed so that the admixture flow gradually veered by almost 360° . Figure 9b shows contour lines of pollution in the middle zone with respect to the station on the fifth day after disaster.

Local model of admixture transport in the atmosphere with use of the method of emissions was constructed and tested by the example of forest fires.²¹ The model traces the wind transport of two admixtures: small particles of ash and unburned material (with particle size of $0-100 \mu m$), as well as solid smoke particles. It was assumed that the mediumand large-sized particles of ash and unburned material did not enter the atmosphere and remained on the underlying surface in the immediate vicinity of the fire. Input data were satellite images of forest fires, meteorological conditions reported by the meteorological station nearest to the fire, and digital maps of the territory. Figure 10 illustrates the aerosol pollution of air and underlying surface in 18 h after detection of forest fire in the region of Polotsk.

For Belarus, located at the center of Europe, and having large industrial centers both inside Republic

and in surrounding regions, it is very important to determine the impact zones, whose emission of polluting admixtures initiate the transfer. To do this, we developed a regional model of transport of nine polluting components, produced by atmospheric emissions from 15 Byelorussian cities, each with emission rate more than 2000 t/yr. Wind field was taken from statistical data for a long period of time. Figure 11 shows the field of normalized concentration of carbon monoxide at a height of 500 m after 9 h of model time for June.



Fig. 10. Forest fire in the region of Polotsk after 18 h of model time: volume concentration of aerosol at height 20 m (mg/m³) (*a*); and density of pollution of the underlying surface, mg/m² (*b*).

The prevailing transfer is seen in west and southwest directions and the long plumes are characteristic for Minsk, Novopolotsk, and Gomel. The conservation zones and chases, situated along the Sozh river and in Polesye turned out to be the vulnerable regions.

To the right of the map, the main numerical characteristics of the considered process are presented: the admixtures, which can be analyzed; a particular considered admixture; date, start time of counting,



Fig. 11. Concentration of carbon monoxide over the territory of Belarus after 9 h of model time.

current time; height (it can vary) of the measurement; maximum admixture concentration at this height (for CO, it is $6.8 \ \mu\text{g/m}^3$ at a height of 500 m); maximum permissible concentration (it is 3000 $\ \mu\text{g/m}^3$ for CO); mass of emitted admixture (41.53 t by 9 h of model time in this case); total mass of admixture residing the atmosphere (28.48 t of CO); and emission rate (110.14 t/day for CO), etc.

The results of the performed studies can be accessed at the information site of the program "Transboundary monitoring" (http://www.tm.basnet.by/ru/index.htm).

Main conclusions

The result of the complex program is the creation of the technical and methodic basis for study of the processes of transboundary monitoring with use of the systems of remote sensing, integrated into the international measurement networks EARLINET, AERONET, and GAW.

The lidar and radiometric complexes for measurements of the atmospheric constituent parameters are created. An experimental version of instrumentation is developed for receiving and thematic processing of satellite information from NOAA spacecrafts, as well as transfer of results to users. The complex application for identification of thermal anomalies ensures as high as 80% detection probability.

We developed and used in practice the algorithm for estimation of the vertical distribution of fine and coarse troposphere aerosol concentrations, based on the data of coordinated sensing of aerosol layer by multiwavelength lidar and sun radiometer CIMEL.

The processes of development of ozone anomalies in the region of Belarus have been studied; they occur mostly in North Atlantic and are transferred in the eastward direction. In recent years, a certain increase in the frequency of occurrence of negative ozone anomalies is observed.

The influence of the processes of large-scale transport on the content of suspended particles in the atmosphere in the region of Belarus was estimated. The air masses from territories located to the south from Belarus turned out to be more polluted, on the average, by the suspended species.

The local and interregional models of aerosol and gas transport on scales from a few tens to hundreds of kilometers are developed and adapted to conditions of Belarus.

The contributions from different regions to the total pollution pattern of Belarus are quantitatively estimated. It is found that the long pollution plumes, characteristic for Minsk, Novopolotsk, and Gomel, exert an adverse influence on the forest reserve zones and chases, situated along Sozh river and in Polesye.

The integrated site is created and the database of parameters of polluting atmospheric constituents is compiled, oriented to solution of problems of largescale transport monitoring.

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