Studies of the dispersal of sulfur dioxide and ozone in the south of Eastern Siberia

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Received June 27, 2005

We present analysis of data on sulfur dioxide concentrations in Irkutsk surroundings in winter–spring 2005 obtained with a gas-analyzer and juxtapose these data with meteorological conditions for the same period. The numerical model of dispersal and transformation of admixtures used helps to reveal the areas with increased level of pollution by sulfur compounds in the Lake Baikal region. Calculated and measurement data on sulfur dioxide concentrations are in a good agreement. Types of meteorological conditions favoring to the atmospheric pollution have been determined. Areas of maximum concentrations of ozone have been calculated. It has been revealed that the speed and direction of winds and concentration of water vapor influence the dispersal and transformation of the admixtures.

Introduction

In recent years, intense investigations into the development of models of transfer and transformation of pollutants in the atmosphere emitted by industrial enterprises have been carried out in our country and abroad. There are many different mathematical models both probabilistic-statistical and deterministic, such as Euler-type, Lagrange-type, and hybrid ones, i.e., combining the approaches of Euler and Lagrange. Transformation and transfer of sulfuric and nitric compounds were studied using the following analytical models: balance model with elements of statistics and two-dimensional stationary diffusion model.[1] The essential disadvantage of these models, which do not take into account the effect of relief, is the fact that vertical distribution of admixtures is assumed to be uniform. An attempt to estimate the effect of anthropogenic pollution by dust, sulfur and nitrogen oxides on Lake Baikal and surrounding territories in different meteorological situations[2] was undertaken using the model of Euler type taking into account evolution of atmospheric aerosols due to coagulation.[3] Let us note that all numerical experiments in these papers were carried out with the spatial step of 25 km, that is larger than the mean width of the ridges in the region of the lake.

Purpose and methods of investigations

The purpose of this study was to investigate the effect of meteorological conditions on the formation of fields of concentrations of pollutants, in particular, sulfur dioxide as one of the main pollutants, in cold season. This study continues our earlier investigations.[4,5] Measurements of sulfur dioxide were carried out using a C-310 chemiluminiscent gas analyzer (OPTEK company, Saint Petersburg, the measurement error does not exceed 25%) in Irkutsk Akademgorodok, in the valley of Angara river on its left bank. In addition to instrumental measurements, numerical experiments were carried out based on the mathematical model of dispersal and transformation of admixtures.[6] In contrast to previous papers, model calculations have been carried out with higher spatial resolution (the horizontal step was equal to 1 km).

Results and discussion

Time average concentrations of sulfur dioxide were determined using the measurement data. The corresponding hourly mean values for March 2005 are shown in Fig. 1.

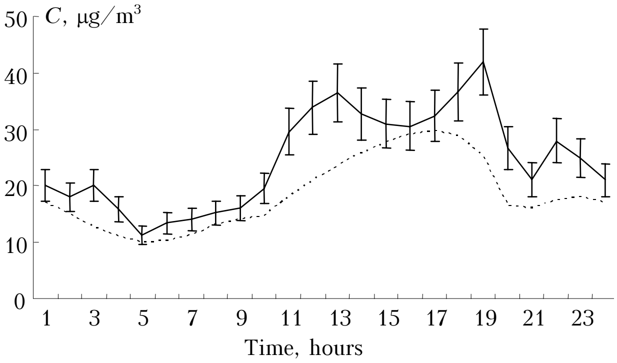

Fig. 1. Time average concentrations of sulfur dioxide in the atmosphere of Irkutsk in March. Solid line corresponds to measurements, dotted line is calculation.

Diurnal behavior is well pronounced with minimum in the morning and maximum in the afternoon. In discussing the data presented in Fig. 1, one should take into account the arrangement of the site where the sampling was done. It is in the upper part of northeast slope of the valley of Angara river. Local maximum in the evening near 9 p.m., also noted in model calculations, should be related to the local wind regime – weakening of the general circulation and formation of katabatic, down slope wind. Separation of two peaks in the daytime maximum (1 and 7 p.m.) is, most likely, explained by relatively short series of observation and strong variance of the values because of the destruction of Asian anticyclone and transformation of the pressure field occurred in March.

The measurement data were complemented with the results calculated using the model of dispersal and transformation of admixtures. As known, time variations of the concentrations of the emitted admixtures are caused by variations of the intensity of emissions by industrial enterprises and by the meteorological processes. The sources of emissions of sulfur dioxide were industrial objects of the cities Usol'e-Sibirskoe, Angarsk, Shelekhov, Irkutsk, Slyudyanka, Baikalsk. The data on the power of the sources were taken from Refs. 7 and 8. Meteorological parameters (wind speed and direction, temperature and humidity of air, and pressure) were measured by a portable meteorological stations situated near the sampling site.

Simulation of the processes of the admixture dispersal was carried out for the region of 200×200 km area and high 4-km height over the surface of the Lake Baikal. Time and horizontal steps were 150 s and 1 km, respectively, and the vertical step was 50 m up to the height of 350 m and then 150, 500, 1000 and 2000 m. The initial concentration of molecular nitrogen N_2 was taken 0.93 kg/m^3, molecular oxygen O_2 – 0.297, water vapor H_2O – $2.23 \cdot 10^{-4}$, molecular hydrogen H_2 – 10^{-7}, and ozone O_3 – $6 \cdot 10^{-8}$ kg/m^3. It was assumed that hydrogen peroxide H_2O_2 permanently exists in the atmosphere, and its concentration equals to 10^{-9} kg/m^3 and does not change in space and time. The turbulent diffusion coefficients were calculated using the relationships of semi-empiric theory of turbulence.[6]

To confirm reliability of the results obtained by means of the model of transformation and transfer of admixtures, the first part of numerical experiments was carried out. The results of comparison of the model calculations of concentration of sulfur dioxide with the measurement data obtained using the gas analyzer are shown in Fig. 1. The correlation coefficient between the calculated and measured concentrations is equal to 0.8. The relative errors in calculations do not exceed 33%, and rms deviation of these errors is 14%.

The next series of numerical experiments was carried out in order to study the effect of meteorological parameters on the dispersal and transformation of the admixtures. Wind speed and direction were varied, as well as the initial value of the concentration of water vapor (the value of 22 g/m^3 characteristic of tropics was chosen for demonstration of the capabilities of the model). Some of the results obtained are shown in the Table.

Calculated values of the concentrations of sulfur and nitrogen compounds and trace gases in the atmosphere over Irkutsk

Ingredient	Concentration, μg/m^3				
	Direction (wind speed, m/s)				
	NW (5)			NW (3)	SW (5)
	Concentration of water vapor, g/m^3				
	0.22	2.2	22	2.2	2.2
SO_2	48	48	43	62	0.5
H_2SO_4	0.54	1.5	7.4	2.3	0.04
NO_2	5.3	5	5.7	6.6	0.06
HNO_3	0.74	1.9	7.1	2.7	0.07
$HNO_4 \cdot 10^4$	0.21	0.7	4.7	1	0.23
NO	2.1	2	1.4	2.2	0.04
$NO_3 \cdot 10^4$	1.4	1.4	1.4	2	0.04
HNO_2	0.04	0.13	0.61	0.13	0.02
$N_2O_5 \cdot 10^4$	1.1	1.1	0.78	2	0.0004
$OH^- \cdot 10^4$	0.11	0.36	2.7	0.33	3
$HO_2^- \cdot 10^3$	0.05	0.16	1.5	0.17	4.6
O_3	55	55	55	65	68
$H^+ \cdot 10^{16}$	1.1	1	0.9	1.2	0.52

The calculations have shown the significant effect of the change of the initial concentration of water vapor on the concentration of atmospheric admixtures. If the concentration of water vapor in the atmosphere changes by an order of magnitude, the concentrations of OH^- and HO_2^- also change by an order of magnitude, the concentration of HNO_4 by 7 times, the concentrations of H_2SO_4 and HNO_2 by 5 times, and the concentration of HNO_3 – by 4 times. The decrease of northwesterly wind (to 3 m/s) leads to an increase of the concentrations of the majority of atmospheric admixtures. Southwesterly and southeasterly winds favor cleaning of the atmosphere over Irkutsk, they cause the decrease of the concentrations of the main admixtures; on the contrary, the concentrations of the main hydrogen radicals OH^- and HO_2^- have recovered to the background values.

Distributions of the concentrations of sulfur dioxide and ozone at northwesterly wind with the velocity of 5 m/s and the initial value of the concentration of water vapor of 2.2 g/m^3 are shown in Figs. 2 and 3. The highest values of the concentration of SO_2 are observed in the valley of Angara river, where the main sources of emission of admixtures have been situated. The maximum concentrations of O_3 are observed over the water area of Lake Baikal, slopes of the ridge Khamar-Daban, and Olkhinskoe plateau, i.e., in the regions, which undergo the effect of atmospheric emissions in the least degree.

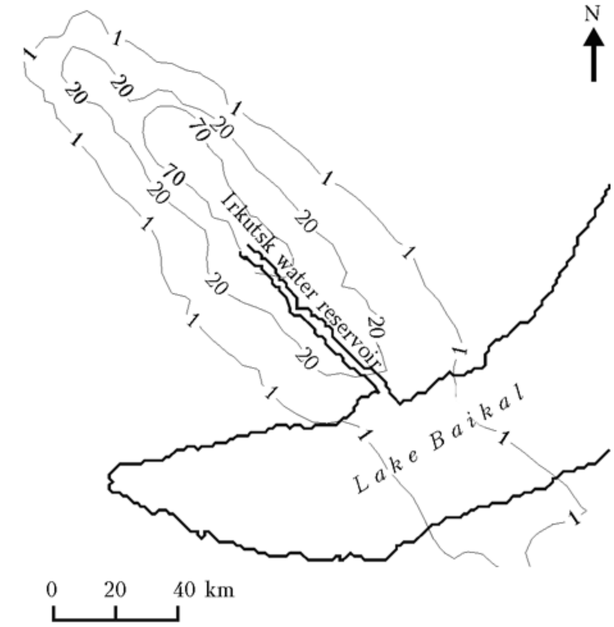

Fig. 2. Isolines of the calculated near-ground concentrations of sulfur dioxide in the region of Southern Baikal at northwesterly wind, μg/m^3.

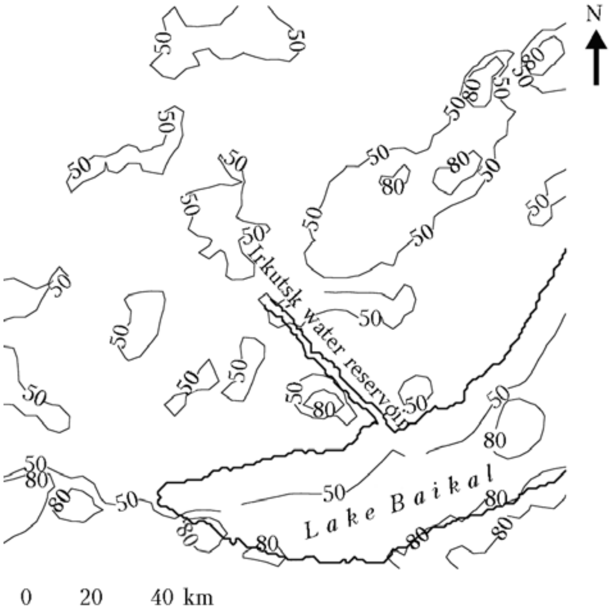

Fig. 3. Isolines of the calculated near-ground concentrations of ozone in the region of Southern Baikal at northwesterly wind, μg/m^3.

Conclusions

Thus, the well-pronounced diurnal behavior of the concentration of sulfur dioxide with minimum in the morning and maximum in the afternoon is observed in Irkutsk Akademgorodok. It is supposed that the local maximum in the evening, near 9 p.m., is caused by weakening of the general circulation and formation of katabatic down slope wind. Comparison of the data of observations of sulfur dioxide and the calculated results shows their good agreement (R = 0.8). The numerical experiments enable one to determine the quantitative dependence of the concentrations of atmospheric admixtures on the initial concentration of water vapor. When changing the concentration of water vapor in the atmosphere by an order of magnitude, the concentrations of some individual components of the atmosphere change by several times: 5 times for sulfuric compounds, and 4–7 times for nitric compounds. Wind speed and direction also essentially affect the dispersal of admixtures.

Acknowledgments

The study was supported in part by the Russian Foundation for Basic Research (grant No. 05–05–97232–r).

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