Numerical simulation of spread, transformation, and deposition of sulfur, nitrogen, and carbon compounds in the Baikal region

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We estimate the amount of acids deposited annually from the atmosphere onto the Lake Baikal surface and on the neighboring areas by solving numerically a semiempirical equation of turbulent diffusion.

One of the most urgent global problems nowadays is connected with the increase in acidity of atmospheric precipitation and topsoil. Acid rains result from human activity accompanied by emissions of sulfur, nitrogen, and carbon acids and combustion of oil, shale oil, coal, and gas. In the atmosphere these compounds are transported at long distances, interact with water, and transform into solutions of mixed organic and inorganic acids, which fall with precipitation and produce a harmful effect on the land and water ecosystems. Acid precipitations were observed in the USA, Great Britain, Germany, Netherlands, Denmark, Scandinavian countries, and on the territories of other European countries, as well as in Moscow region and in the northwestern Russia. In recent years, the problem of acid rain has become increasingly urgent for the Asian part of Russia as well, as indicated by the results of instrumental observations.[1,2]

This paper continues the investigation into the processes of spread, transformation, and deposition of sulfur and nitrogen compounds in the Lake Baikal region with the use of mathematical modeling that was started in Refs. 3–6. In this paper the emphasis is put on the analysis of the dependence of the mass flow density of acids deposited onto the lake surface on the atmospheric humidity. The sources of emissions of sulfur and nitrogen oxides were industrial plants of 25 cities and towns located on Lake Baikal sides (the largest of them are Slyudyanka, Baikal'sk, Nizhneangarsk, Severobaikal'sk), as well as at the distance up to 70 km from the lakeside (Irkutsk, Shelekhov, Selenginsk, Kamensk). The values of the mass flow of sulfur and nitrogen dioxides were determined based on the data from Refs. 7–11. The statistical characteristics of the wind field used in the calculations were obtained from processing the many-year observations of the wind velocity.[12,13]

The calculations were carried out for the region having the area of 250×700 km^2 and the height of 5 km above the Lake Baikal surface. The time and horizontal steps were, respectively, 150 s and 5 km, while the vertical step was specified in the following way: it was 50 m up the height of 300 m, and then 200, 1000, 1500, and 2000 m. The turbulent diffusion coefficients were calculated with the use of equations of the semiempiric theory of turbulence.[6] It was assumed that the gases emitted are distributed homogeneously in a spatial cell of a grid region. The initial concentrations were taken to be the following: 0.93 kg/m^3 for molecular nitrogen [N_2], 0.297 kg/m^3 for molecular oxygen [O_2], 10^{-9} kg/m^3 for [H_2O_2], and 10^{-7} kg/m^3 for molecular hydrogen [H_2]. The initial concentration of water vapor [H_2O] varied from $2.23{\cdot}10^{-6}$ to $2.23{\cdot}10^{-2}$ kg/m^3, that is, the H_2O mass used was from $1.7{\cdot}10^{-4}$ to 1.7% of the air mass. Molecular oxygen and water vapor were assumed to take part only in the transformation process.

Table 1 gives the calculated concentrations of sulfur and nitrogen compounds and minor atmospheric constituents in the surface atmosphere above Lake Baikal at the initial concentration [H_2O] = $2.23{\cdot}10^{-4}$ kg/m^3. The second and next columns present the same concentrations at the profile passing through St. Dulikha (near Khamar-Daban Ridge) and oriented from the southwest to the northeast at different distances from this station (distances are given in kilometers). The comparison of calculated concentrations of sulfates and nitrates with the data of instrumental measurements[4] showed a satisfactory qualitative and quantitative agreement between them. The concentrations of other minor gases are close, in the order of magnitude, to those observed in other regions.[14–26] It follows from the Table 1 that the southern part of Lake Baikal is much more polluted than the central one, and the concentrations in the southern part (columns 2, 3, and 4) exceed the corresponding values in the central part (columns 5, 6, and 7) by the order of magnitude and more.

Figures 1–4 depict the calculated mass flow densities of sulfuric, nitric, nitrous, and peroxynitric acids in the Baikal region. The strongest effect of these compounds was noticed in River Angara valley (emissions from Irkutsk and Shelekhov), over Southern Baikal, between Listvyanka and Vydrino due to emissions from industrial plants in Priangar'e, Slyudyanka, and Baikal'sk, in the region of emissions from Selenginsk and Kamensk, and in the northern Baikal region (emissions from Severobaikal'sk and Nizhneangarsk). It should be noted that the maximum in the southern part of Lake Baikal is shifted with respect to the location of pollution sources.

Table 1. Calculated concentrations, in μg/m^3, of sulfur and nitrogen compounds and minor atmospheric constituents in the surface atmosphere above Lake Baikal

Ingredient	Distance from St. Dulikha, in km							
	20	70	120	160	200	320	375	435
1	2	3	4	5	6	7	8	9
SO_2	9	0.08	0.01	0.5	0.2	0.008	$2\cdot10^{-6}$	0.0005
H_2SO_4	1	0.1	0.02	0.2	0.3	0.01	$3\cdot10^{-6}$	0.002
HSO_3, $\cdot\ 10^{11}$	2	0.05	0.005	0.3	0.2	0.004	$9\cdot10^{-7}$	0.0003
SO_3, $\cdot\ 10^{9}$	5	0.2	0.02	0.9	0.6	0.01	$3\cdot10^{-5}$	0.001
SO_2^*, $\cdot\ 10^{11}$	4	0.046	0.005	0.2	0.1	0.004	10^{-6}	0.0002
SO, $\cdot\ 10^{17}$	4	0.0003	$5\cdot10^{-6}$	0.01	0.003	$3\cdot10^{-6}$	$2\cdot10^{-13}$	10^{-8}
NO_2, $\cdot\ 10^{2}$	4	0.03	0.003	0.2	0.2	0.005	$6\cdot10^{-7}$	0.0003
HNO_3, $\cdot\ 10$	2	0.1	0.02	0.3	0.6	0.03	10^{-5}	0.002
HNO_4, $\cdot\ 10^{7}$	2	0.08	0.009	0.4	0.7	0.03	10^{-6}	0.01
NO	2	0.02	0.003	0.2	0.1	0.003	10^{-6}	$4\cdot10^{-5}$
NO_3, $\cdot\ 10^{7}$	1	0.3	0.05	0.7	2	0.07	$2\cdot10^{-5}$	0.006
HNO_2, $\cdot\ 10$	2	0.07	0.01	0.6	0.5	0.01	$2\cdot10^{-6}$	0.0001
N_2O_5, $\cdot\ 10^{11}$	9	0.01	0.0003	0.2	0.6	0.0008	$2\cdot10^{-11}$	$3\cdot10^{-6}$
OH, $\cdot\ 10^{3}$	0.6	2	2	2	3	2	1	2
HO_2, $\cdot\ 10^{3}$	0.6	3	3	2	4	5	2	31
$O(^1D)$, $\cdot\ 10^{13}$	1	0.05	0.01	0.2	0.2	0.01	0.0001	0.002
O_3	5	0.2	0.04	0.7	0.9	0.06	0.0005	0.009
$O(^3P)$, $\cdot\ 10^{8}$	2	0.08	0.02	0.3	0.4	0.02	0.0002	0.004
H, $\cdot\ 10^{18}$	2	0.04	0.005	0.3	0.6	0.02	$4\cdot10^{-5}$	0.02
H_2O_2, $\cdot\ 10$	4	1	1	3	4	2	0.6	15

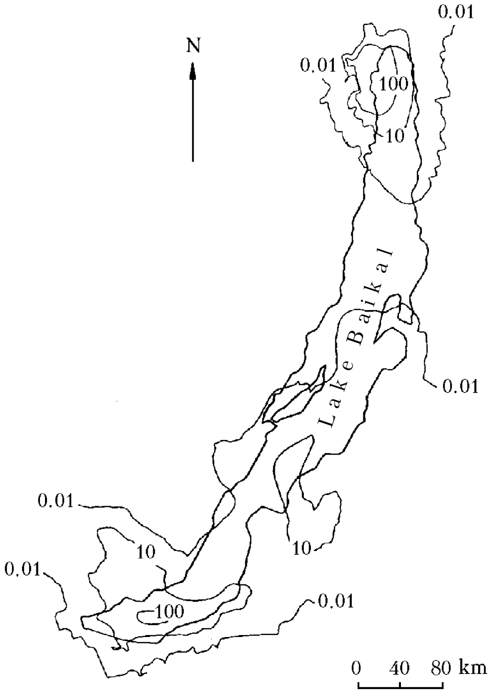

Fig. 1. Isoline of calculated mass flow density of H_2SO_4 in the surface atmosphere of the Baikal region, in kg/(km^2·year).

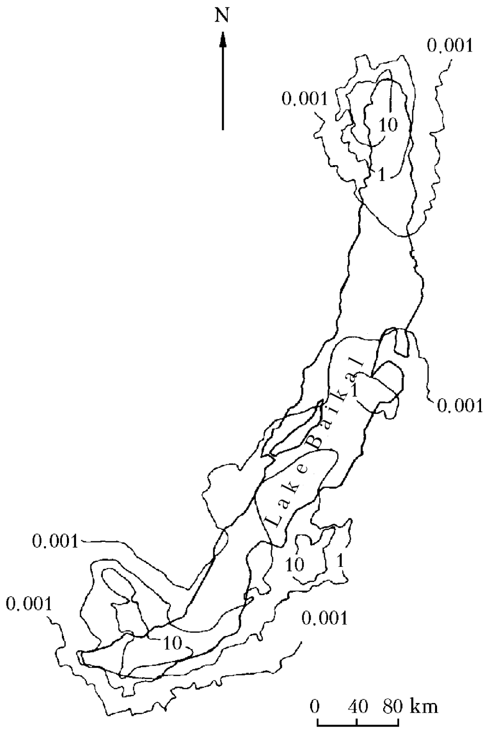

Fig. 2. Isoline of calculated mass flow density of HNO_3 in the surface atmosphere of the Baikal region, in kg/(km^2·year).

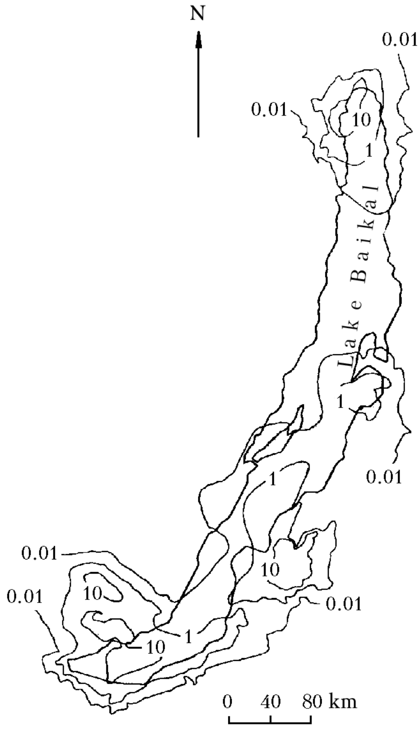

Fig. 3. Isoline of calculated mass flow density of HNO_2 in the surface atmosphere of the Baikal region, in kg/(km²·year).

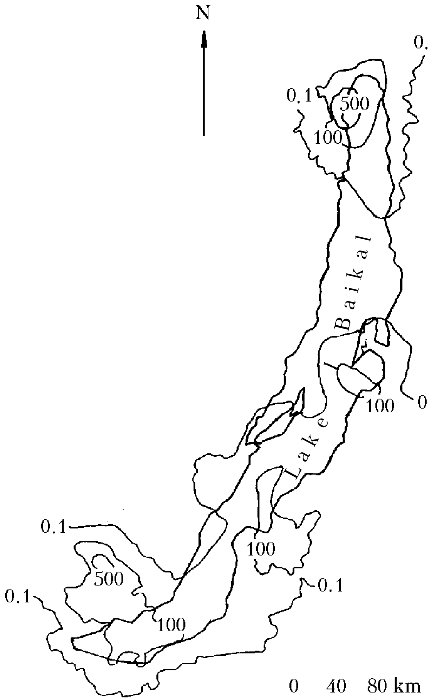

Fig. 4. Isoline of calculated mass flow density of HNO_4 in the surface atmosphere of the Baikal region, in kg/(km²·year).

To study the effect of air humidity, we conducted numerical experiments with different initial water vapor concentration. The results of these experiments are given in Table 2.

Table 2. Calculated masses of main organic acids deposited onto Lake Baikal, in kg, a year

Initial concentration of $[H_2O]$, kg/m³	Mass, kg			
	H_2SO_4	HNO_3	HNO_2	HNO_4
$2.23 \cdot 10^{-6}$	7680	1120	1030	0.017
2.23^{-4}	7780	1140	1050	0.017
$2.23 \cdot 10^{-2}$	14080	2360	2160	0.022

The calculations showed that the mass of the acids deposited depends considerably on the air humidity: as the water content in the atmosphere increases from 0.017 to 1.7% of the air mass, the mass of sulfuric, nitric, and nitrous acids deposited onto the surface is doubled.

It should be noted that the amount of the acids deposited depends on the presence of other atmospheric constituents as well. As the initial concentration of H_2O_2 increases by two orders of magnitude (from 10^{-10} to 10^{-8} kg/m³), the mass of sulfuric, nitric, and nitrous acids deposited onto the Lake Baikal surface a year increases by 7 (from 2.5 to 18 t), 40 (from 0.2 to 6.6 t), and 4 times (from 0.4 to 1.5 t), respectively. As the initial concentration of atomic oxygen $O(^3P)$ increases by two orders of magnitude (from 10^{-14} to 10^{-12} kg/m³), the mass of the sulfuric acid increases from 0.3 to 1.6 t, the mass of the nitric acid increases from 0.02 to 0.5 t, and that of the nitrous acid increases from 0.05 to 0.2 t. Thus, the initial concentrations of minor atmospheric constituents considerably affect pollution of Lake Baikal.

The results obtained can be used to estimate the contribution of acids falling with precipitation to the total mass of acids deposited onto Lake Baikal through wet and dry deposition.

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