Experimental and theoretical investigations into the ion composition of atmospheric precipitation in the Southern Baikal region

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The results of 1998–2002 studies of the chemical composition of atmospheric precipitation sampled at monitoring sites in the Baikal region (Irkutsk, Listvyanka, and Mondy) are considered. It is shown that wet precipitation in Listvyanka is most acidified. Atmospheric fluxes of major ions on the underlying surface of the territory studied are calculated. In the southern region of Lake Baikal, the atmospheric fluxes of sulfates and nitrates are calculated using a mathematical model based on numerical solution of the spatial nonlinear non-steady-state semi-empirical equation of turbulent diffusion of admixtures. At the Irkutsk and Listvyanka sites, the order of magnitude of the computer-simulated flux estimate corresponds to that calculated from the experimental data.

Introduction

Instrumental investigations indicate that the problem of acid precipitation, which is quite urgent for a long time in industrially developed countries, is pressing for the Baikal region as well.[1,2] We have begun combined study of this problem in 1998 at three stations of round the year monitoring of acid precipitation.[3–7] To continue the study of the most important aspects of this problem, this paper considers the results of investigation of the acidity and ion composition of atmospheric precipitation and, in the first place, the compounds affecting their acidification: sulfates and nitrates, as well as ammonium ions as a source of potential acidity.[8]

Materials and methods

The material for this paper comprises the results of chemical analysis of about 700 atmospheric precipitation (AP) during the period since October 1998 until October 2002. At the stations Irkutsk and Mondy, precipitation in winter was sampled in plastic bags, which increased somewhat mineralization and pH in snow samples due to the contribution of dry deposition of aerosol particles. At the station Listvyanka, fresh snow was sampled from the surface of a rammed snow area, which should decrease the effect of dry deposition on the chemical composition of snow samples due adsorption of particles by the surface of this area. Wet precipitation in the warm season was mostly collected with automated precipitation collectors, which allowed reliable separation of dry and wet precipitation.

In AP, the following characteristics were determined: pH, electric conductivity, main cations, and anions. The pH and conductivity were measured with a Horiba instrumentation (Japan). Anions (HCO_3^-, SO_4^{2-}, NO_3^-, Cl^-) were determined by the method of ionic chromatography on a Milikhrom A-02 HPLC chromatograph (Eko-Nova, Novosibirsk).[9] Cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) were determined by the method of atomic absorption spectrophotometry on an AAS-30 (Carl Zeiss Yena, Germany). Ions NH_4^+ and NO_2^- were determined colorimetrically with Nessler and Griss reagents, respectively, on a Philips PU 8700 spectrophotometer.[10,11] The quality of the material obtained was checked through calculation of $R1$ – the error of balance between the equivalent forms of anion and cation sums, and $R2$ – the error obtained from comparison of the calculated and measured conductivity.[12] The total error of determination did not exceed 10%.

Discussion

The material obtained in 1998–2002 evidences that at the station Listvyanka there is no significant difference in the acidity of wet precipitation in winter and summer, as is observed at other stations. At the stations Mondy and Irkutsk, the seasonal behavior of pH in AP almost coincides (Fig. 1).

The pH peaks in the autumn-spring periods are connected with far smaller amounts of precipitation and long dry periods, during which great amounts of admixtures are accumulated in the atmosphere. Intensification of wind lifting dust particles into the atmosphere in the spring period, annual burning of garbage and forest fires also increase the concentration of all ions and pH value in AP. In winter months in Irkutsk, ash emissions from power plants form an additional source increasing mineralization and decreasing the acidity of wet precipitation. The lower pH values in summer at all the stations are caused by the larger amount of AP. The summer months from

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June to August in the region under study are responsible for 50–80% of the mean annual precipitation favoring purification of the atmosphere from admixtures of the natural and anthropogenic origin.

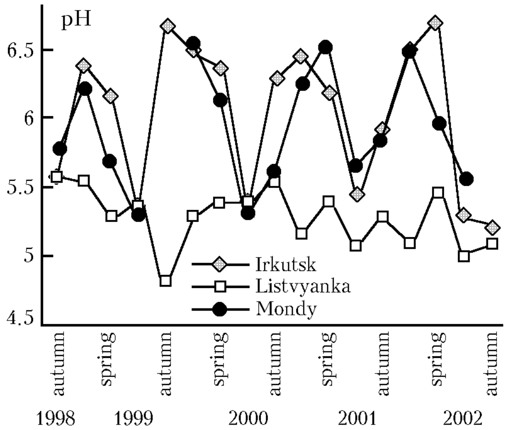

Fig. 1. Seasonal pH dynamics in AP at the monitoring stations of Baikal region.

The calculations showed (Fig. 2) that the highly acidified rains (pH = 4.0–4.5) most often fall at the Irkutsk and Listvyanka stations. Rains with pH = = 4.5–5.0 are most characteristic of the station Listvyanka, while rains with pH = 5.0–5.5 are typical for the stations in Mondy and Irkutsk. In snow the following pH values occur most often: pH = 5.0–5.5 in Listvyanka, 6.0–6.5 in Mondy, and 6.5–7.0 in Irkutsk.

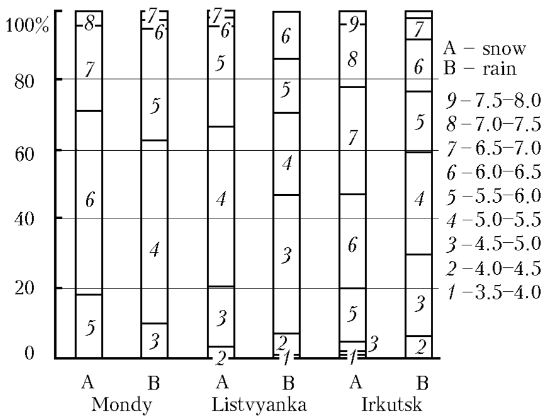

Fig. 2. Occurrence of different pH in AP at the monitoring stations in Baikal region (1998–2002).

Usually, the acidity of rains at the monitoring stations is not directly related to the concentration of anions in precipitation. The correlation coefficients between H^+ ions and anions of strong acids were negative in the majority of cases. The maximum value of the correlation coefficient (0.46) was calculated at the station Mondy in summer of year 2000 for the H^+ and NO_3^- ions. The rain acidity then was largely determined by nitrates (0.5–1.5 mg/l), since the concentrations of sulfates were very low (0–0.4 mg/l). However, in the general case the AP acidity is caused not only by the content of anions of strong acids, but also by the amount of cations capable of neutralizing the extra acidity. To reveal the efficiency of this process in AP, we have considered various cation/anion ratios in AP (Table 1). The data given in Table 1 are averaged over four years of the investigations discussed.

The contribution of the Na^+ and K^+ ions in the AP composition in the Baikal region is insignificant as compared with that from Ca^{2+} and NH_4^+, therefore they were ignored in calculations. As follows from Table 1, the concentration ratios of the main cations and anions in rain at the monitoring stations (version 4) is close to or slightly larger than unity, that is, these ratios do not allow us to speak about the problem of acid precipitation in Baikal region. However, if we consider every case separately, we can see that in 43, 65, and 71% cases of rains at the stations Irkutsk, Mondy, and Listvyanka, respectively, the cation/anion ratio is less than unity, and in 7, 15, and 18% cases it is ≤ 0.5. Hence, it follows that sometimes the AP acidification occurs at the monitoring stations.

Some deficit of cations in AP at Mondy station in summer occurs largely due to the deficit of Ca^{2+} and Mg^{2+} ions of the erosion origin, since NH_4^+ ions are in excess in rains ($NH_4^+/SO_4^{2-} > 1$). The most part of rains at this station fall in June–July, and it is just the precipitation of this period that are characterized by very low concentrations of cations. The decrease of Ca^{2+} content from 1.3 (maximum of the summer period) down to 0.01–0.25 mg/l and the content of all other cations down to analytical zero lead to an increase in the concentration of hydrogen ions, that is, to the pH decrease down to 4.7–5.2. Lower pH values were not observed at this station. In some cases (17%) hydrogen ions dominated among cations.

Table 1. Acidity and ratio of mean equivalent concentrations (μg-equ/liter) of main acidic and neutralizing ions in AP (1998–2002)

Station	AP	pH	NH_4^+/SO_4^{2-}	$\frac{Ca^{2+} + Mg^{2+}}{SO_4^{2-}}$	$\frac{Ca^{2+} + Mg^{2+}}{SO_4^{2-} + NO_3^-}$	$\frac{NH_4^+ + Ca^{2+} + Mg^{2+}}{SO_4^{2-} + NO_3^- + Cl^-}$
			1	2	3	4
Mondy	rain	5.45	1.1	0.9	0.5	0.9
	snow	6.27	0.5	2.7	1.9	1.7
Listvyanka	rain	5.12	0.7	0.8	0.5	0.9
	snow	5.35	0.4	1.3	0.7	0.9
Irkutsk	rain	5.43	0.7	1.1	0.7	1.1
	snow	6.48	0.4	1.9	1.3	1.4

In our opinion, rain acidity at Mondy station, in the majority of cases, is caused by natural causes, that is, by the natural acid components of the atmosphere. At thunderstorms a large amount of nitrogen oxides, which can quickly transform into nitric and nitrous oxides, is produced in the atmosphere.[14] Possibly, the maximum content of nitrates in rains at Mondy station (1.5 mg/l) is caused just in this way. In the most rains sampled at this station, the concentrations of nitrates were within 0.4–0.6 mg/l. The maximum content of sulfates in low-mineralized rains (0.3–2.5 mg/l) did not exceed 1.2 mg/l. The further increase in the concentration of this ion always led to a decrease in the AP acidity, since the content of cations increased simultaneously.

Thus, at Mondy station the rain acidity at low mineralization increased in most cases not because of the high content of anions of strong acids, but because of the deficit of erosion-origin cations capable of neutralizing them. The same features, but in the chemical composition of snow, were observed in northern regions of Western Siberia.[13] In snow at Mondy station, the acidity is completely neutralized mostly due to calcium and magnesium of natural origin, as evidenced by both high values of cation/anion ratios (version 4) and low acidity of snow (Table 1, Fig. 2).

At Listvyanka station almost all the considered ratios (versions 1–4) are less than unity for the whole year. This is indicative of continuous deficit of cations of both lithophile and atmospheric origin in the composition of rain and snow. The deficit of neutralizing cations manifests itself in prevalence of H^+ ion in 25% of rains. The decrease of the ratio (version 2) from 1.3 down to 0.7 (version 4) at summation of sulfates and NO_3^- ions points out the significant effect of this ion on the acidity of snow at Listvyanka station. This is also indicated by the correlation ($k_{cor} = 0.6$) between the H^+ and NO_3^- ions in winter of 1999 and 2001 and high values (1–5) of the NO_3^-/SO_4^{2-} ratio in December–February, which was not observed at other stations. In Listvyanka we can also see the gradual increase of the fraction of nitrates in the ion composition of snow (from 10% in 1999 to 23% in 2002). Besides, if in 1999 the NO_3^- ion prevailed in 21% of snow cases in Listvyanka, then in 2002 it prevailed already in 53% of cases. Correspondingly, the number of samples with prevailing sulfate decreased markedly. All these factors are evidence of the appearance of an additional local source of the NO_3^- ion. Most probably, it is a local boiler house, which replaced coal with fuel oil. Fuel oil burning, as any other high-temperature process, produces much more nitrogen oxides then coal burning does, and this leads to an increase in the nitrate fraction.

Thus, as compared with other monitoring stations, wet precipitation at Listvyanka is subject to stronger acidification, which occurs due to the transport of acidic substances from industrial centers of the Irkutsk region, the effect of local sources of acid gases with simultaneous deficit of cations capable of neutralizing excessive acidity in AP.

As follows from Table 1, in rains at Irkutsk station, acidity of anions of strong acids is neutralized due to the joint effect of all cations. In the industrial center, this process is most efficient due to high content of alkaline components in the atmosphere and, consequently, the high fraction of Ca^{2+} and Mg^{2+} ions in rains (Table 2).

However, as follows from Fig. 2, rains in Irkutsk sometimes have increased acidity, and the fraction of such rains in different years varies from 8 to 11% of the total number of wet precipitation, which is 2–3 times smaller than in Listvyanka.

Analysis of the back trajectories of air mass motion shows that the cases of most strongly acidified rains at the monitoring stations are accompanied by low rates of atmospheric transport, which is likely indicative of the main contribution of regional industrial centers to acidification of AP.

Comparison of the fractional distribution of ions in rains at the monitoring stations shows that the total contribution of nitrates and sulfates is somewhat higher at Listvyanka station (38.6% equ. vs. 37.1% equ. at Irkutsk and Mondy stations) with the lower fraction of calcium, magnesium, and ammonium (Table 2). Some decrease of the contribution of hydrogen ions here as compared to rains at Mondy station is connected with higher fraction of potassium and sodium ions. Comparison of the seasonal distribution of ions in AP reveals the effect of various factors on the formation of ion composition of snow and rain water. Thus, at Mondy station the prevalence of the HCO_3^- ion among anions of the snow water points out the large contribution of the erosion sources to its content in the atmosphere and in snow. This is also confirmed by the high correlation (0.92) between the HCO_3^- and Ca^{2+} ions.

Table 2. Fractional distribution of main ions in AP at the monitoring stations in Baikal region, % equ. (1998–2002)

AP	HCO_3^-	SO_4^{2-}	NO_3^-	Cl^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	NH_4^+	H^+
					Mondy					
snow	20.3	16.8	6.5	5.7	3.6	1.9	29.9	6.0	8.2	0.9
rain	4.4	22.1	15.0	5.9	2.7	1.4	12.4	3.5	21.0	10.4
					Listvyanka					
snow	2.8	24.9	17.9	4.2	4.2	2.5	22.6	5.6	10.6	4.7
rain	4	26.6	12.0	5.7	3.4	2.7	15.6	4.2	16.5	9.3
					Irkutsk					
snow	14.3	21.8	7.4	5.4	4.8	1.3	30.4	4.9	8.8	0.7
rain	5.6	27.3	9.8	5.3	2.2	1.3	20.2	4.5	17.8	5.9

The dominating ions in rain are NH_4^+ and SO_4^{2-}, but the close mutual dynamic of these ions was observed only in summer of 2001–2002. In all the years, except for 2001, the SO_4^{2-} ion correlated well with lithophilic cations, thus indicating the existence of not only atmospheric, but also aeolian source of sulfates in the composition of rain.

In snows sampled at Listvyanka and Irkutsk stations the fraction of sulfate and nitrate (at domination of the SO_4^{2-} and Ca^{2+} ions) is higher in Listvyanka, while that of calcium and hydrocarbonate is higher in Irkutsk. This is quite natural, since the maximum transformation of the acid gases SO_2 and NO_x into the corresponding acids and, consequently, their effect on the AP composition is more pronounced at some distance from a source of emission then in its immediate proximity. The calcium ions in the composition of coarse $CaSO_4$ and $Ca(HCO_3)_2$ particles faster deposit near a source, which always affects the snow composition. The close correlation between these ions was observed in Irkutsk during the whole period of observations.

It is worthy to note the continuous co-dynamics of Na^+ and Cl^- ions in snow in Irkutsk. It can hardly be connected with the marine factor in formation of the snow composition. The source of anthropogenic origin, namely, transport of polluted air masses from Usol'e-Sibirskoe, where a salt production plant is located, is more probable in this case. In rains at Listvyanka and Irkutsk stations the fraction of ammonium ions is somewhat higher (at the prevalence of the SO_4^{2-} anion) than that of calcium ions in Listvyanka, while in Irkutsk the situation is quite opposite. From 1999 to 2002 we can see the increase in the fraction of the ammonium and hydrogen ions and the decrease in the fraction of calcium in the ion composition of rains in Listvyanka. Here the correlation between the NH_4^+ and SO_4^{2-} ions was observed only in 2000–2001, which is likely connected with the large amounts of rains in these years as compared to 1999 and 2002 and, thus, with more regular purification of the atmosphere from pollutants.

It is known that ammonium sulfate is the main soluble component of continental aerosol only in regions far from pollution sources.[14] At Irkutsk station close correlation between the SO_4^{2-} and Ca^{2+} ions was observed in all years except for 2002.

Based on the experimentally obtained concentrations of the main ions and the precipitation amounts, we have calculated the flows of the main ions by the commonly accepted method.[15] As follows from Table 3, the major part of the annual sum of ions at Listvyanka and Mondy stations falls with rains. This is quite natural, since the maximum amount of AP at these stations falls in warm seasons. In Irkutsk the flows of some ions (HCO_3^-, Cl^-, Na^+, Ca^{2+}, Mg^{2+}) in snow are higher than in rain due to their higher snow content. This is connected with significant pollution of the atmosphere in the industrial center in winter with anthropogenic products, in particular, with solid emissions of heat and power plants. As to sulfates, nitrates, ammonium, and hydrogen ions, their amount is much higher in warm seasons at all the stations.

In addition to experimental investigations, we have calculated the flows of sulfates and nitrates onto the surface at the territory under study using the mathematical model based on numerical solution of the spatial nonlinear non-steady-state semi-empirical equation of turbulent diffusion.[16] We have studied the processes of spread, transformation, and deposition with precipitation for sulfur and nitrogen compounds in the Angara river valley and southern Baikal. Stationary and nonstationary sources of emissions of sulfur and nitrogen oxides situated near Angara river, in the southern Baikal region, and in the Selenga river valley were taken into account. The mass flows of sulfur and nitrogen dioxides were determined following the technique from Refs. 17 and 18. The statistical characteristics of the wind field used in the calculations were obtained from processing the data of many-year observations of the wind velocity.[19]

The calculations were performed for a region with the area of 500×250 km and the height of 5 km above the surface of Lake Baikal. The main characteristics of emission sources and the grid parameters are given in Ref. 5. Since the processes of formation of sulfuric and nitric acids, sulfates and nitrates in droplets are much more intense than in air, for pollutant wash-out by precipitation we used the rate constants of oxidation reactions of sulfur and nitrogen dioxides for the water medium. Gravitational sedimentation of water droplets was taken into consideration.

Using the simulated spatial concentration fields of sulfur and nitrogen compounds, we have determined the mass flow rates of sulfates and nitrates with AP onto the surface in the region under study for a day (Fig. 3) and for a year (Fig. 4). The largest precipitation amount in the studied region falls at passage of Atlantic cyclones, therefore we have performed the calculations at the northwestern flow with the constant speed equal to 5 m/s (Fig. 3*a*).

Table 3. Mean flows of the main ions with atmospheric precipitation in 2000–2002, mg/m²

Precipitation	HCO_3^-	SO_4^{2-}	NO_3^-	Cl^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	NH_4^+	H^+
					Irkutsk					
rain	233	845	367	71	26	32	281	32	177	3.06
snow	809	685	193	102	65	31	455	37	91	0.13
					Listvyanka					
rain	102	456	266	52	22	30	101	15	112	3.64
snow	24	218	201	26	17	25	85	14	26	0.85
					Mondy					
rain	83	220	175	31	11	15	59	8	93	1.13
snow	43	36	14	5	2	3	20	2	9	0.02

The results showed that the total amount of sulfates and nitrates fallen onto the lake surface with AP is 50 ton. The largest contribution to lake pollution in this case is due to plants and motor vehicles of the Irkutsk–Cheremkhovo industrial center – 92%; the contribution due to Slyudyanka and Baikalsk towns is 8%; the effect of pollution sources located in the Selenga river valley is insignificant. Atmospheric precipitations at southern cyclones in Baikal region are much more rare. Figure 3*b* depicts the calculated results on wet depositions of sulfates and nitrates at the southeastern wind with the speed of 5 m/s. For a day, 20 ton of sulfates and nitrates fall on the Lake Baikal surface. The largest contribution to lake pollution in this case comes from the emission sources located in the Selenga river valley – 90%; the contribution of Slyudyanka and Baikalsk is 10%.

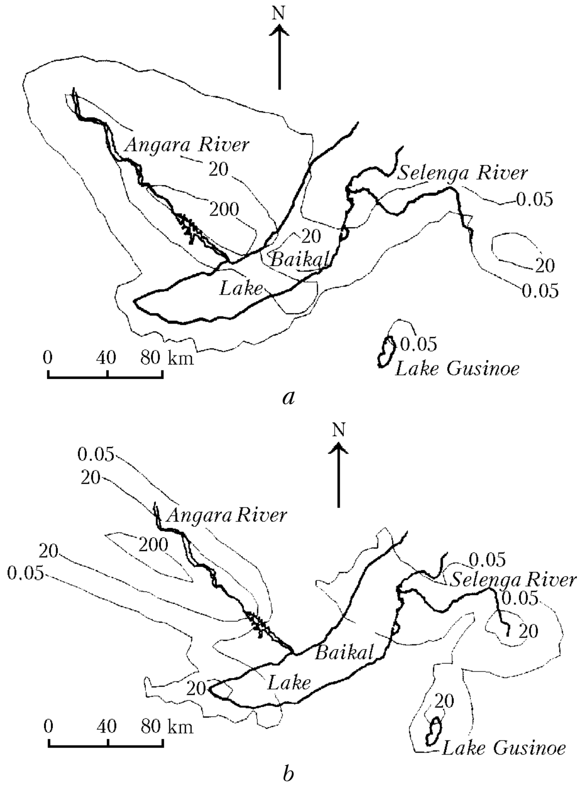

Fig. 3. Isolines of the calculated mass flow rate of the sum of nitrates and sulfates with AP near the surface in the southern Baikal region, mg/(m^2 · day): at the northwestern (*a*) and southeastern (*b*) wind.

In estimating the annual wet deposition of sulfur and nitrogen compounds, we took into account the information about the amount of AP in different regions of the territory under study.[20] Figure 4 illustrates the density distribution of the mass flow of the sum of sulfates and nitrates in the considered region. Comparison of the calculated results with estimates of the annual wet deposition in the studied region showed that the model calculations of the mass flow rate for a year in Irkutsk and Listvyanka have the same order of magnitude as the values obtained from analysis of the measurement data (Table 4).

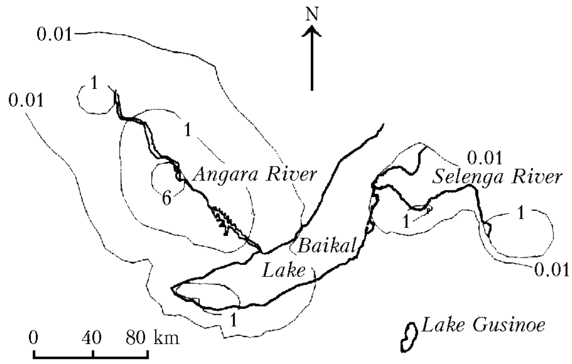

Fig. 4. Isolines of the calculated average mass flow rate of the sum of sulfates and nitrates with AP near the surface in the Southern Baikal region, g/(m^2 · year).

Comparison of simulated values of nitrate and sulfate deposition for a day with the actual flows of these ions with some AP in Irkutsk and Listvyanka also demonstrates their satisfactory agreement. The mean flows of the sum of sulfates and nitrates with some AP in Irkutsk and Listvyanka are equal to 25 and 20 mg/m^2, respectively. The corresponding simulated values obtained taking into account the actual number of days with AP at the stations are, on the average, 32 and 15 mg/(m^2 · day). In comparing these values, one should keep in mind that the calculations for Listvyanka station ignored pollution sources located in this site itself, and that the sampling point in Irkutsk is located at some distance from the downtown area, where air is most polluted. We hope that as the parameters of the pollution sources will be refined at their inventorization and the spatial characteristics of wind flow in the region under consideration for the studied period will be determined more accurately, results of model calculations should become closer to the actual processes.

Conclusion

Investigations of the chemical composition of some AP at the monitoring stations in the Baikal region showed that AP at Listvyanka station is subject to acidification during the whole year while at Irkutsk and Mondy stations – mostly in summer.

Table 4. Annual flows of sulfates and nitrates onto the surface, mg/m^2

Station	Sulfates		Nitrates	
	Experiment	Calculation	Experiment	Calculation
Irkutsk	1336–1702	1800	398–706	1200
Listvyanka	593–744	500	427–494	400

Acid rains in Listvyanka and Irkutsk are connected with transport of polluted air masses from industrial areas of Irkutsk Region in the system of small slow-moving cyclones. Snow at Listvyanka station is acidified due to the effect of local sources. The acidity of AP at Mondy station is caused by natural factors what confirms that this station can be considered as a background continental one.

The contribution of emission sources in Slyudyanka and Baikalsk to pollution of the southern part of Lake Baikal with sulfates and nitrates fallen with AP is twice as large as the contribution coming from Irkutsk–Cheremkhovo industrial complex. Comparison of the experimental and first calculated data on fallouts of sulfates and nitrates with AP at Listvyanka and Irkutsk stations has demonstrated quite good agreement between them.

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