Some Results of Experimental Observations and Mathematical Simulation of Distribution of Acidifying Atmospheric Admixtures in the Region of Southern Baikal

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Abstract

Experimental data on annual variability of 7–10 day mean concentrations of the main ions in the soluble fraction of atmospheric aerosols (1999–2002) and in precipitation (1999–2001) at acid precipitation monitoring stations in Listvyanka and Irkutsk have been obtained. The characteristics of atmospheric aerosol of these stations as continental one have been confirmed. The existence of various natural and technogenous factors that form the ionic composition of aerosol at the Irkutsk station in various seasons has been demonstrated. Influence of substance transfer along the route Irkutsk – Listvyanka on the chemical composition of the water-soluble fraction of aerosol at the Listvyanka station has been found. It is confirmed that winter precipitation at the Irkutsk station are considerably more mineralized than summer precipitation. Mineralization of precipitation at the Listvyanka station has no significant seasonal differences. Dominant ions in atmospheric precipitation throughout the year are SO_4^{2-} and Ca^{2+}. Acid deposition have been observed mainly at the Listvyanka station. Calculations have been carried out with the help of a mathematical model based on a numerical solution of a spatial non-linear non-stationary semi-empirical equation of turbulent diffusion of admixture. Spatial fields of month average concentration of sulphur and nitrogen compounds and minor gaseous components of the atmosphere in the region of southern Baikal have been calculated. The density of mass expenditure of sulphates and nitrates on the ground surface of the region under study for a year has been estimated

INTRODUCTION

Lately, in some sites of the Baikal region a regular monitoring of chemical composition of the atmospheric aerosol (AA), minor gaseous admixtures and atmospheric precipitation (AP) has been carried out. The results of such all-year-round observations may be useful in estimation of efficiency of mathematical models for transfer of atmospheric admixtures in the region under study. In this regard, two sites are of special interest: Irkutsk – one of the largest regional sources of anthropogenous emissions, and the settlement Listvyanka situated on the southwestern bank of Lake Baikal at 70 km from Irkutsk, *i. e.* leeward of it. Such a location of observation sites gives certain possibilities for following up the atmospheric transfer and transformation of admixtures along the route Irkutsk – Listvyanka.

As a result of 3 year long observations at these points, experimental data on annual variation of concentrations of the main ions in the soluble fraction of AA and AP have been obtained. In addition to the experimental studies, calculations were carried out with the help of a mathematical model [1] for the purpose of studying the processes of diffusion and transformation of sulphur and nitrogen compounds over the valley of the Angara river and southern Baikal.

RESULTS AND DISCUSSION

Atmospheric aerosol

Irkutsk Station. The seasonal dynamics of pH and of the total ion content of the water-soluble part of AA are shown in Fig. 1, *a*. The mean pH values throughout the observation period at the Irkutsk station vary from 5.21 to 6.28 and are close to the equilibrium pH value of distilled water. This, on the whole, witnesses to the absence of constant factors that create conditions for excessive acidity or alkalization of AA components. As to single aerosol samples, the pH varies from 3.72 to 7.48, which gives us reasons to speak of short-term acidification or alkalization situations. The occurrence of such situations is confirmed by the correlation between the ion content in winter and in summer separately in different years. In summer, the concentration of H^+ ions correlates well with that of ions NO_3^-, HCO_3^- in 2000 and SO_4^{2-} and Cl^- in 2001. A high correlation was found between the contents of ions Ca^{2+} and Cl^-, Na^+ and HCO_3^- in 2000, NH_4^+ and NO_3^-, Ca^{2+} and SO_4^{2-} in 2001. This witnesses to the presence of various factors influencing the chemical composition of AA of Irkutsk in winter.

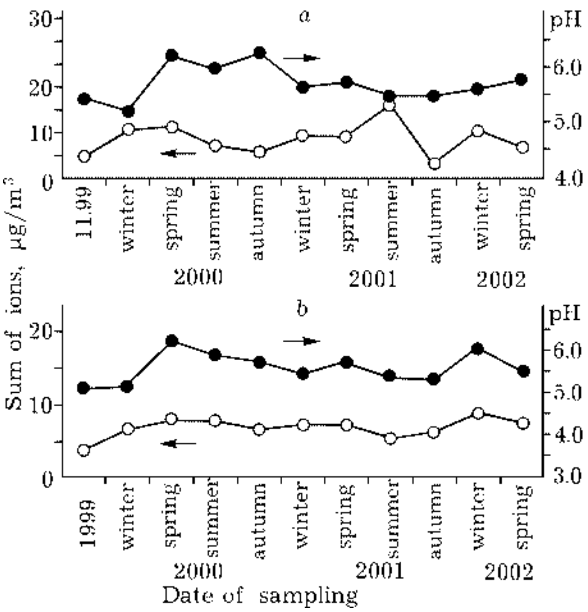

Fig. 1. Dynamics of pH value and of the total ion content in the water-soluble fraction of atmospheric aerosol: *a* – Irkutsk, *b* – Listvyanka (autumn 1999–spring 2002).

In winter, in AA a correlation between the content of H^+ ions with SO_4^{2-} in 2000, Cl^- in 2001 and 2002 was observed. Possibly, it is just the Cl^- ions in the presence of SO_4^{2-} and NO_3^- that contribute to occurrence of short-term acidification situations. The interaction of SO_4^{2-} and NO_3^- ions with the group of cations neutralizing these components is of undoubted interest. The analysis demonstrates a low degree of correlation of the content of NO_3^- ions and cations of this group throughout the years of observation. A correlation of SO_4^{2-} ions with Na^+ ions is manifested in 2001, and in 2002 such a correlation was observed between ions SO_4^{2-} and Na^+, K^+, Mg^{2+}, Ca^{2+}. One may hypothesize that it is just in this year that a constant factor mainly influenced the formation of the chemical composition of AA in winter.

Values of concentrations of ionic composition of the soluble part of AA in Irkutsk are presented in Table 1. Month average concentrations were calculated separately for each year of observation and averaged for seasons. In spring-winter period AA of Irkutsk contained more dissolved substances than in summer-autumn period. However, the maximum of the sum of ions was observed in summer 2001. The formation of the ionic composition of AA under the conditions of an industrial city in summer is influenced by atmospheric processes, soil erosion and automobile transport, in winter more by thermal power and transport discharges. Accordingly, we observe an increased concentration of NO_3^- and SO_4^{2-} ions in winter, whereas such ions of erosion processes as Na^+, K^+, Mg^{2+}, and Ca^{2+} have a higher concentration in spring-summer period. The largest amount of HCO_3^- ions is contained in summer aerosols. One may hypothesize that the hydrocarbonates in AA of Irkutsk are of terrestrial origin. This is confirmed by a comparison of calculated and experimental concentrations of HCO_3^- ions, discrepances between which are much more than 20–30 %. In autumn, the concentration of water-soluble salts of AA decreases. This may be linked with washing out of ions from the atmosphere by

TABLE 1

Ionic composition of the water-soluble fraction of atmospheric aerosol of Irkutsk (1999–2002), μg/m^3

Period	pH	HCO_3^-	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Mg^2	Ca^{2+}	H^+
						1999					
November	5.45	0.30	1.97	0.78	0.44	0.45	0.16	0.03	0.06	0.48	0.0019
						2000					
Winter	5.21	0.51	5.69	1.37	0.26	1.76	0.14	0.04	0.13	0.67	0.0045
Spring	6.23	3.53	2.78	1.08	0.23	1.28	0.18	0.30	0.26	1.26	0.0009
Summer	5.99	3.56	0.77	0.53	0.0	0.27	0.20	1.32	0.11	0.34	0.0020
Autumn	6.28	3.19	0.46	0.26	0.11	0.56	0.05	0.16	0.08	0.42	0.0011
						2001					
Winter	5.64	1.35	4.1	0.94	0.37	1.38	0.06	0.08	0.14	0.58	0.0175
Spring	5.73	0.77	4.36	0.79	0.38	1.4	0.09	0.10	0.12	0.67	0.0056
Summer	5.48	2.97	5.28	2.55	0.82	3.27	0.06	0.36	0.05	0.41	0.0034
Autumn	5.49	0.48	0.98	0.28	0.13	0.45	0.02	0.16	0.02	0.14	0.0025
						2002					
Winter	5.59	1.40	4.08	1.63	0.19	1.81	0.17	0.12	0.08	0.40	0.0011
Spring	5.76	1.25	2.12	1.02	0.24	1.18	0.06	0.05	0.04	0.30	0.0018

precipitation and by the diminution of the contribution of elements of soil origin.

The fraction distribution of ions of the water-soluble fraction of AA also witnesses to the absence of constant factors of formation of aerosol chemical composition in Irkutsk (Fig. 2, *a*).

As one can see from Fig. 2, *a*, dominant AA ions are HCO_3^-, SO_4^{2-}, NH_4^+ and Ca^{2+}; however, their annual and seasonal dynamics is inconstant during the observation period. In winter and early spring 1999–2000, the prevalent contribution to the chemical composition was made by ions NH_4^+, Ca^{2+} and SO_4^{2-}. From April to December 2000, the proportion of ions HCO_3^- increases, and their contribution becomes prevalent. In the fraction distribution of cations at this period the main role is played, as before, by cations NH_4^+ and Ca^{2+}, although the contribution of K^+ in June and K^+ and Na^+ in July is noticeably increased. The fraction of SO_4^{2-} ions becomes insignificant and begins to increase only in December 2000, becoming dominant again till the end of the observation period. At the same period, the fraction of NO_3^- ions increases, too. In November 2001 NO_3^- ions make the main contribution to AA. It is noteworthy that in the proportion of cations in this month the contribution of ions NH_4^+ decreases, and that of H^+ increases, the Ca^{2+} ions being prevalent. Despite the redistribution of the fraction proportion of ions in November, it is the NH_4^+ ion that is prevalent in the chemical composition of the ionic part of AA from January 2001 to May 2002.

Listvyanka Station. As one can see in Fig. 1, *b*, the mean pH values at the Listvyanka Station vary from 3.99 to 7.95. Correlation analysis of averaged data failed to show any effect of the constant factor on the AA formation in the settlement Listvyanka; however, an analysis of the ionic composition of AA separately in winter and in summer lets suppose the existence of such a factor.

In the ionic composition of AA, a high degree of correlation between cation H^+ and ions SO_4^{2-} in 1999, ions SO_4^{2-} and NO_3^- in 2001, and ions SO_4^{2-} and HCO_3^- in 2002 is manifested. The fraction of ions SO_4^{2-} in winter 1999–2002 well correlates with such of ions Ca^{2+} and Mg^{2+}, and in separate years with that of ions K^+ and Na^+. Despite the fact that in the ionic composition of AA of Listvyanka the proportion of ions NH_4^+ and SO_4^{2-} is high,

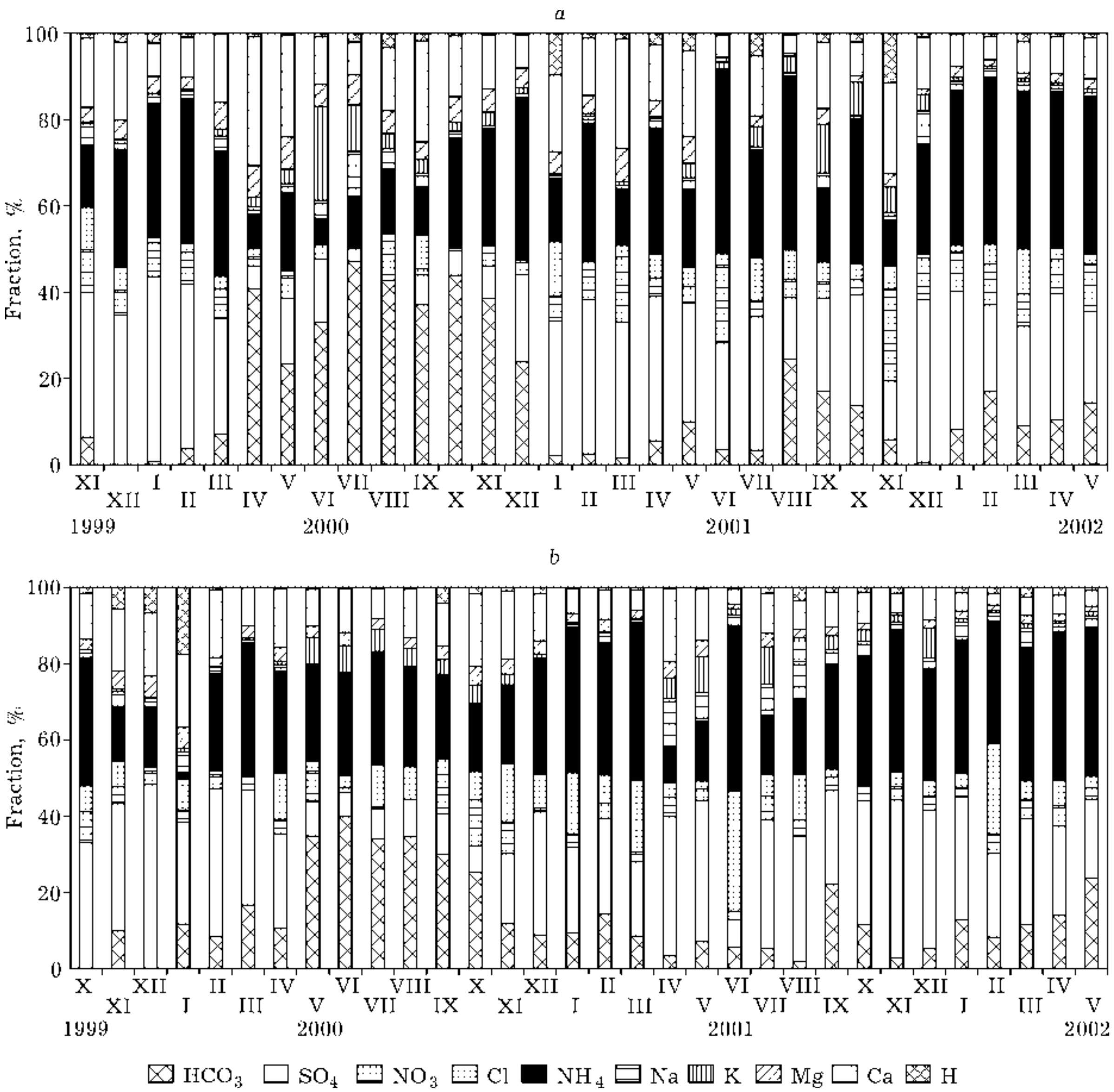

Fig. 2. Fraction distribution of equivalents of ions in the atmospheric aerosol: *a* – Irkutsk, *b* – Listvyanka (autumn 1999 – spring 2002).

there is no correlation between them. The content of ions NO_3^- correlates with the group of neutralizing cations only in 2000–2001; in 1999 and 2002 no such correlation in the ionic composition was found. As for the correlation of ions HCO_3^- with the group of cations, throughout the winter observation period their high correlation with NH_4^+ and K^+ ions was found (with the exception of 2000). In 2002, such a correlation was observed for the whole cation group. In summer, the correlation between anions and cations is also high. This is especially the case with anions SO_4^{2-} and NO_3^- and with the whole group of neutralizing cations. On the basis of the analysis, one may hypothesize that the ionic composition of AA is influenced by the constant factor, but that it has different origin in summer and in winter. This conclusion is corroborated by [2].

The ionic composition of the water-soluble part of AA of Listvyanka is presented in Table 2.

The annual variation of the sum of ions during the period under study points to a tendency to gradual increase in the total concentration of the water-soluble part of AA of

TABLE 2

Ionic composition of the water-soluble fraction of atmospheric aerosol of the settlement Listvyanka (1999–2003), μg/m^3

Period	pH	HCO_3^-	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Mg^2	Ca^{2+}	H^+
1999											
November	5.13	0.40	1.75	0.40	0.26	0.43	0.80	0.02	0.05	0.32	0.0044
2000											
Winter	5.17	0.67	3.80	0.42	0.23	0.51	0.13	0.04	0.12	0.71	0.0172
Spring	6.26	2.91	2.00	0.71	0.26	1.08	0.64	0.27	0.08	0.45	0.0008
Summer	5.91	4.42	0.72	0.09	0.48	0.98	0.00	0.48	0.07	0.41	0.0009
Autumn	5.74	2.19	1.10	1.19	0.52	0.63	0.01	0.25	0.09	0.55	0.0038
2001											
Winter	5.46	1.49	2.49	0.45	0.81	1.33	0.08	0.04	0.07	0.32	0.0016
Spring	5.72	0.94	2.72	0.42	0.97	1.16	0.22	0.26	0.08	0.42	0.0014
Summer	5.37	0.46	1.39	0.35	1.35	1.00	0.20	0.25	0.04	0.19	0.0024
Autumn	5.33	1.22	2.70	0.38	0.14	1.00	0.13	0.18	0.03	0.25	0.0024
2002											
Winter	6.05	1.35	3.42	0.63	1.12	1.45	0.17	0.24	0.05	0.25	0.0031
Spring	5.45	2.12	2.36	0.57	0.35	1.42	0.16	0.08	0.04	0.19	0.0037

Listvyanka. The seasonal dynamics of the sum of ions is clearly expressed. In winter and spring, the ion concentration is higher as compared to that in summer and autumn. The dynamics of content of separate ions at various periods of observation is different. The greatest changes among anions concern SO_4^{2-} ions. The maximal content of these ions is observed in winter, and the minimal one in summer 2002. In the rest of years, a similar dynamics is also observed with respect to SO_4^{2-} ions, but with smaller variations of concentration. The content of Cl^- ions gradually increases from October 1999 to August 2001, whereupon in autumn 2001 a sharp decrease in the concentration of this component takes place. In 2002, the concentration of Cl^- ions increases again. The content of NO_3^- ions has been stable during the last two years. The maximal amount of HCO_3^- ions is contained in AA in summer 2001. Among the cations, an increase in NH_4^+ ion concentration is observed with its maximal value also in summer 2001.

In Fig. 2, *b*, the proportion of ions in the water-soluble fraction of AA of Listvyanka is presented. One can see that the annual and the seasonal dynamics is on the whole similar to that of the relative proportion of AA ions in Irkutsk. In this regard, one may hypothesize that the determinant factors in the formation of the ionic composition of AA in Listvyanka are the transfer and transformation of admixtures on the route Irkutsk – Listvyanka.

In this way, comparison of the data on the ionic composition of the water-soluble part of AA of Irkutsk, taking into account the relative proportion and correlation of ions, permits concluding that with respect to the chemical composition of AA in which the fractions of SO_4^{2-} and NH_4^+ ions prevail, may be mainly characterized as continental [2]. The nonuniform proportion of ions across seasons and the difference in their correlations in average data point to the absence of constant sources taking part in the formation of the chemical composition of AA. Several different factors of both natural and technogenous origin make their contribution to the change of the ionic composition of AA in Irkutsk. There are no constant factors influencing acidification or alkalization of the atmosphere, although one may not rule out the occurrence of such short-term situations. The formation of winter AA in 2002 was influenced by a constantly acting factor.

The proportion of ions of the water-soluble part and the correlation analysis of relations between ions confirm the influence of transfer of substances in the route Irkutsk – Listvyanka on the formation of AA in Listvyanka.

Atmospheric precipitation

Studies of the chemical composition of AP at the Irkutsk and Listvyanka stations in 1999–2001 have demonstrated that at the Irkutsk station AP are more mineralized. Maximally, the total ion content (sum of ions) in snow samples for the observation period amounted to 188–190 mg/l, the minimal one was 2.5–2.6 mg/l; in rain samples it was 72–77 mg/l and 1.2–1.8 mg/l, respectively. The average sum of basic ions (22–26 mg/l) in the AP of Irkutsk in winter was more than in summer by 10 mg/l. This is associated both with the influence of emissions from thermal power production and with a poor dissipation of industrial discharges as a result of high repeatability of weak winds, still weather and temperature inversions which are so characteristic of Irkutsk in winter [3]. In addition, during snowfalls the most efficient washout of contaminants takes place, because snow flakes trap much more aerosol particles than raindrops do, since they have a lower falling velocity and a larger trapping surface [4].

Unlike the Irkutsk station, AP mineralization at the Listvyanka station does not vary considerably throughout the year. The variation limits of the sum of basic ions in winter were 2.1–18.1 mg/l, and in summer 0.9–20 mg/l. The average sum of ions in AF during the cold period varied within the range of 5.6–7.2 mg/l, and during the cold period from 4.7 to 7.7 mg/l.

Consideration of equivalent relations between ions in AP at monitoring stations permits speaking of dominance of ions SO_4^{2-} and Ca^{2+} during the year. In the ionic composition of rains, the contribution of ammonium ions increases; their appearance at the warm time of the year in the atmosphere and precipitation is associated mainly with natural sources and with agricultural activity.

It is noteworthy that at the Listvyanka station in 11 out of 25 rain samples taken from May to September 2001 the prevalent cation was H^+. Therein, pH value was within the range of 4.30–5.13. Rains with a similar and even higher acidity were observed by us at this station also in 1999 [3]. At the Irkutsk station, there have also been cases of acid AP with prevalence of ions forming the sulphuric and hydrochloric acids. However, in the majority of rain samples calcium and ammonium remained the dominant cations. A peculiarity of the ionic composition of snow samples at the Listvyanka station is the considerable increase, and in some cases prevalence over sulphate, of nitrate ions, which seems to be associated with the use of mazut in the settlement, because its combustion is accompanied by release of a considerable amount of nitrogen oxides [4]. This is especially characteristic of snow samples from November to February 2001. Our results on the Listvyanka station confirm also the hypothesis of the author of [4] that snowfall eliminates nitrogen compounds from the atmosphere efficiently, and therefore one may expect a higher nitrate content than in rain.

MODEL STUDIES

In addition to experimental studies, calculations were carried out with the help of a mathematical model based on a numerical solution of a spatial non-linear semi-empirical equation of turbulent diffusion of admixture [1]. The model was verified earlier using the data of expedition measurements [5, 6]. When studying the processes of diffusion and transformation of aerosol and gas admixtures in the Angara and Lake Baikal regions, the stationary and non-stationary sources of sulphur and nitrogen oxide discharges were taken into account. The mass expenditure of sulphur and nitrogen dioxides was estimated on the basis of [7, 8]. The statistical characteristics of the wind field used in the calculations had been obtained from the treatment of data of long-term observations on the wind velocity vector [9].

Calculations were carried out in a site with the area of 500×250 km and an altitude of 5 km above the surface level of Lake Baikal. Steps with respect to time and horizontal were

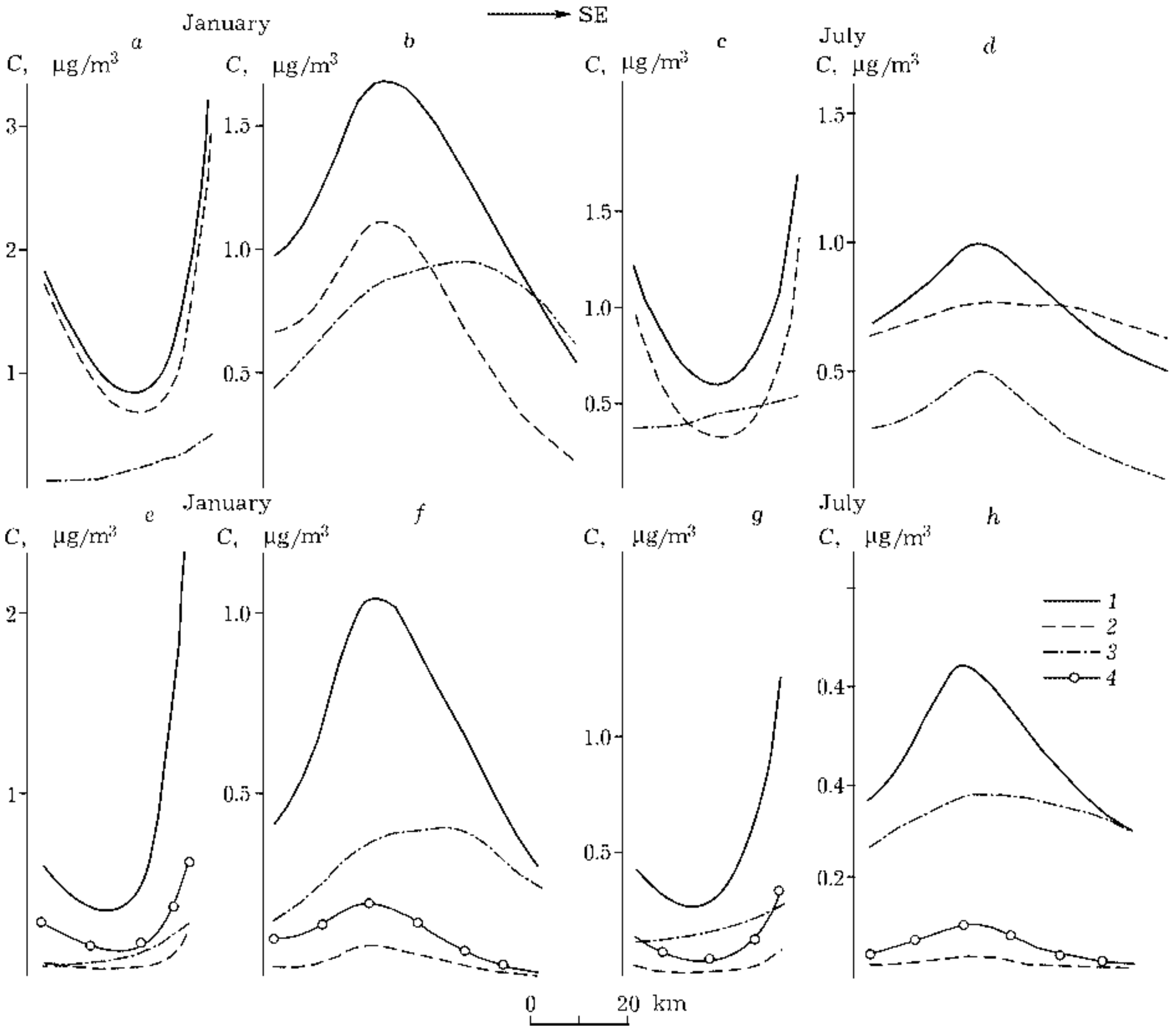

Fig. 3. Calculated concentrations of sulphur (*a*–*d*) and nitrogen (*e*–*h*) compounds near the water surface on sections Kultuk – Baikalsk (*a, c, e, f*) and Sharazhalgay – Vydrino (*b, d, f, h*): *1* – SO_2 and NO_2 concentrations without taking into account the transformations; *2* – the same, taking into account the transformations; *3* – H_2SO_4 and HNO_3 concentrations; *4* – NO concentration (according to [7]).

150 s and 5 km, respectively; step with respect to vertical was set as follows: up to the height of 350 m, it was 50 m, whereupon it was 150, 1000, 1500 and 2000 m. The initial concentration of molecular nitrogen $[N_2]$ was assumed to be equal to 0.93 kg/m^3, that of molecular oxygen $[O_2]$ equal to 0.297 kg/m^3, that of water $[H_2O]$ – 2.23 10^{-4} kg/m^3, $[H_2]$ – 10^{-7} kg/m^3. It was assumed that in the air atomic hydrogen was continuously present, and its concentration, which is equal to 5 10^{-22} kg/m^3, was not changed in space and in time. Coefficients of turbulent diffusion were calculated using the equations of semi-empirical theory of turbulence [1].

As a result of numerical experiments on the model, spatial fields of month average concentrations of sulphur and nitrogen compounds and of minor gaseous atmosphere components in the region under consideration were obtained. Maximal values of the calculated concentration of sulphur and nitrogen oxides were found near the sources of discharges: on Lake Baikal, these were the sites of Slyudyanka and Baikalsk. In the water surface layer of the Lake Baikal, maximal concentrations of sulphates and nitrates were observed in the regions of Murino – Vydrino – Tankhoy stations, *i. e.* at some distance from the emission sources. This can be accounted for by the relief configuration and

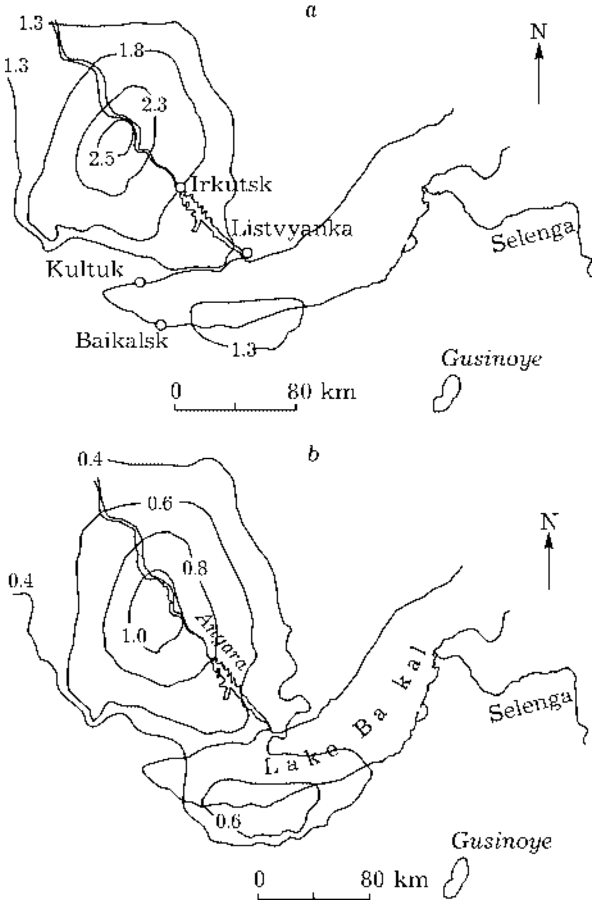

Fig. 4. Isolines of density of mass expenditure of H_2SO_4 (*a*) and HNO_3 (*b*) on the ground surface of southern Lake Baikal region, t/(km^2 year).

by the longer duration of transformation processes as compared to processes of diffusion and transfer. In Fig. 3, the values of concentrations of some admixtures near the water surface of Lake Baikal on sections Kultuk – Baikalsk and Sharazhalgay – Vydrino are presented.

A comparison of model estimates of the concentration of sulphates and nitrates with the results of instrumental measurements described in [7] has demonstrated their qualitative agreement. The concentrations of other minor components of the atmosphere correspond, with respect to their magnitude, to those observed in other regions [4, 10–21].

The density of the mass expenditure of sulphates and nitrates on the ground surface of the research location throughout the year has been estimated (Fig. 4). Maximal values of the mass expenditure density in this region have been obtained for the Murino – Vydrino – Tankhoy region.

The results obtained may be used for estimation of the contribution of dry and moist settling of acids to the total mass of their deposition onto Lake Baikal.

In conclusion, let us note that in future, for a more reliable verification of models, it is necessary to carry out special experiments with more detailed meteorological measurements.

CONCLUSIONS

1. The relative proportion of ions of the water-soluble part of AA in Irkutsk across seasons and the different correlation of their proportions point to contribution of various factors to formation of the ionic composition of aerosol. The formation of AA of in winter 2002 was influenced by a constantly acting factor.

2. The formation of the ionic composition of AA in the settlement Listvyanka was influenced by the transport of substances along the route Irkutsk – Listvyanka.

3. AP at the Irkutsk station are influenced more by the discharges of contaminant substances than at the Listvyanka station.

4. Acid deposition at monitoring stations falls out mainly in summer.

5. According to calculations, the regions of local maxima of primary and secondary admixtures on Lake Baikal do not coincide. Elevated sulphur and nitrogen oxide concentrations have been noted above the emission sources – in the regions of Slyudyanka and Baikalsk, and those of sulphates and nitrates in the region Murino – Vydrino – Tankhoy.

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