Variability of the Chemical Composition of the Atmospheric Aerosol in the Coastal Zone of the Southern Basin of Lake Baikal (East Siberia, Russia)

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Abstract: The role of the atmosphere in the formation of the chemical composition and quality of water in Lake Baikal and its tributaries has been increasing in recent years. In this regard, studies of the chemical composition of the constituents of the atmosphere have an important practical application. In 2020 and 2021, we studied the chemical composition of atmospheric aerosol, one of the indicators of air pollution, in the atmosphere of the coastal zone of the southern basin of Lake Baikal compared to the data from previous years. The studies were carried out in the summer on the southwestern (Bolshiye Koty) and southeastern coast (Boyarsky). In the absence of smoke in the aerosol on the southwest coast, the concentrations of NH₄⁺, NO₃⁻ and SO₄²⁻ ions prevailed. The mean total concentration of ions at the Bolshiye Koty research station was 2.08 ± 1.26 µg/m³. The appearance of smog contributed to the growth of the total ionic concentration in the aerosol on the southwest coast to 6.4 µg/m³ in 2020 and to 17.6 µg/m³ in 2021. On the southeast coast, the minimum concentration of the total amount of ions was 3.3 µg/m³. The concentrations of Ca²⁺, Na⁺, K⁺, Cl⁻, and SO₄²⁻ ions prevailed in the aerosol. Under the influence of smog, the total amount of ions increased to 34.1 µg/m³ in 2020 and to 18.6 µg/m³ in 2021. In periods of intense smoke, NH₄⁺ and SO₄²⁻ became the dominant ions in the aerosols at both stations. The contribution of NO₃⁻ ions increased. Although the effect of natural factors is periodic, they contribute significantly to the change in the chemical composition of atmospheric aerosol.

Keywords: aerosol; Lake Baikal; Bolshiye Koty; Boyarsky; wildfires; ions; tourism

1. Introduction

Lake Baikal is the world’s largest reservoir of fresh water. Its protection has become increasingly important in recent years. The atmosphere is one of the important channels of influence on the ecosystem condition of the lake. The physicochemical properties of aerosol and trace gaseous impurities above Lake Baikal were mainly studied in summer. This is associated with a specific hydrophysical regime of the lake [1], the inaccessibility of the region and the lack of technical capabilities, which do not allow for year-round observations. Analysis of individual particles revealed a large amount of soil dust in the Baikal aerosol. The contribution of gypsum particles was great above the northern and central basins of Lake Baikal (13 and 14% of the total number of particles, respectively) and amounted to 4.7% above the southern basin. For the southern part of the lake, the sulfur-enriched group of particles (6.4%) was the most representative. The iron-containing group of particles was evenly distributed above the entire area of Lake Baikal. The group of quartz particles was detected only above the southern part of the lake [2]. The results of the
2006 to 2008 studies revealed that the highest number of pollutants in the atmosphere in the central and northern basins of Lake Baikal was above the Selenga shallow water, Barguzin and Chivyrkuiisky bays, and the northern margin of the lake. Forests largely occupy the coast of the northern basin. The central basin is characterized by a dry climate, and steppe and semi-desert landscapes represent a significant part of its coasts. Sulfates, calcium, and ammonium prevail in the chemical composition of aerosols above the northern and central basins [2]. In the southern basin, elevated ionic concentrations (1.3–2.6 µg/m³) were recorded in the aerosol near the Listvyanka settlement and along the Listvyanka-Tanhkoy transect. The maximum total concentrations of ions (4.6 µg/m³) were detected in the atmosphere near the Baikalsk town [3]. In the southern basin, substances from the industrial areas of the Southern Baikal region entered the lake along the Angara River valley in the form of poorly dispersed plumes during mesoscale jet stream transport in the atmospheric boundary layer [4]. In 2019, atmospheric measurements above Lake Baikal confirmed previous studies. The most pollutant-free atmosphere above the water surface of Lake Baikal was located above the central deep part of the lake. Heterogeneity in the distribution of pollutants above the surface of Lake Baikal was mainly observed in the estuarine areas [5].

At present, tourism is developing in the southern basin [6]. Recreational resources are concentrated primarily along the coast of the lake. With the development of tourism, questions arise related to the sphere of tourist services, which are an expansion of transport and road infrastructure as well as the construction of hotels, ports with berths, cottage settlements, and other objects. The distribution of mass concentrations of PM₁₀ and soot over the Baikal water area indicate the effect of the emissions transported from industrial areas along the valleys of the Angara and Selenga rivers as well as from numerous tourist sites on the coast [7]. In this regard, the problem of the preservation of the environment on the coast of the lake becomes relevant.

The first regular stationary studies of the atmosphere were started in 1982 on the northeast coast of the lake in the Barguzinsky Nature Reserve (Davsha settlement) and stopped in 1998 due to a reduction in funding. The work program included regular monitoring of the concentrations of priority pollutants (lead, cadmium, mercury, arsenic, organochlorine pesticides, 3,4-benzpyrene, and sulfates). An analysis of the material led to the conclusion that the current levels of pollutants in the environment of the background areas of the Baikal region were extremely low [8].

The areas in the southern part of the lake are the most accessible for tourism development because they are conveniently located along transport routes near the coastal zone. Tourist priorities are given to the coastal administrative areas of the Republic of Buryatia and the Irkutsk Region situated in the central ecological zone of the Baikal natural territory, and specially protected natural zones are involved [6,9].

In the modern period, the state of the atmosphere in the southern basin is monitored by measurements of the composition of aerosol, precipitation, and gaseous impurities in summer and snow in winter, which are carried out at the research station of Limnological Institute, Siberian Branch of the Russian Academy of Sciences (SB RAS) (Irkutsk, Russia) near the Bolshiye Koty research station and at the research station of Institute of Physical Material Science SB RAS (Ulan-Ude, Russia) in the Boyarsky settlement (Figure 1).
Both research stations are tens of kilometers away from large industrial sources of pollution. In addition, the study of the atmosphere near in the area of the southern basin of Lake Baikal is important because of the greatest amount of precipitation there (up to 800 mm per year) [11]. Moisture is brought to this area mainly with the northwesterly air masses passing through the industrial complexes of East Siberia. As previously determined, the chemical composition of water in the lake tributaries that are supplied by 80% through the atmosphere has changed in recent decades, and forests have dried up. In the chemical composition of the tributaries, there was a decrease in the alkaline components such as calcium, magnesium, and bicarbonate ions, while the concentrations of acidic components such as sulfates and nitrates increased; the pH value decreased. The pH value of precipitation in this area ranged from 4.6 to 5.2 [12,13]. The infrastructure of the region and numerous tourist objects are the main anthropogenic sources of air pollution in the coastal zone of the southern basin of Lake Baikal, and wildfires are the natural ones. In recent decades, due to climate change in Siberia, the number of wildfires has been increasing (Figure 2). The smoke from wildfires with prevailing northwestern transport of air masses enters the watershed basin of Lake Baikal, as well as its water area, and has a significant effect on its physical characteristics and the chemical composition of aerosol above this area of the lake and adjacent areas [5,14]. In 2021, the area of wildfires in the Irkutsk Region was 407.25 thousand hectares, which is almost 1.5 times larger than in 2020. The greatest smoke pollution of the atmosphere in the southern basin of Lake Baikal was observed in August 2021.
According to the EANET, the samples were taken on filters of four types at a 2 m distance from the underlying surface and in the location free of obstruction [18]. The filters were arranged in successive order on a block that was connected to a pump and a gas meter. The aerosol substance was collected on an external (first) Whatman-41 filter, the analysis results of which were used in this study. The gaseous impurities were sorbed on four types of filters, respectively. The average daily value was used to interpret the results of the analysis. In the current study, the data for previous years were used to evaluate the actual state of the environment, served as a source material for this study.

2. Materials and Methods

Regular studies of the chemical composition of atmospheric aerosol have been carried out at the Bolshiye Koty research station (N 51°53′, E 105°03′) since 2002, and at the Boyarsky research station—since 2014. The research stations are located at the same latitude on both sides of Lake Baikal (Figure 1). The Bolshiye Koty station is located in a coastal area about 200 m wide. Aerosol samples were taken at the water edge. The research station in the Boyarsky settlement is situated at a 500 m distance from the coastline. At both research stations, aerosol sampling is carried out by a universal method applied by the EANET and EMEP international networks (European Monitoring and Evaluation Programme). According to the EANET, the samples were taken on filters of four types at a 2 m distance from the underlying surface and in the location free of obstruction [18]. The filters were arranged in successive order on a block that was connected to a pump and a gas meter. The aerosol substance was collected on an external (first) Whatman-41 filter, the analysis results of which were used in this study. The gaseous impurities were sorbed on filters 2–4. In this study, we did not use the analysis results from filters 2–4. Taking into account that the proportion of submicron aerosol fraction is predominant [5], we believe that the use of a universal method applied by the EANET and EMEP is adequate. Air was sampled in the daytime (from 7:00 a.m. to 9:00 p.m.) and at night (from 9:00 p.m. to 7:00 a.m.) by a Becker GmbH vacuum pump (Becker Vertriebs, Bremen, Germany) at the Bolshiye Koty research station and by a high-volume sampler General Metal Works model (Andersen Instrument Inc., New York, NY, USA) at the Boyarsky research station. The average volume of air drawn through each filter was 15–18 m$^3$ and 500–1000 m$^3$, respectively. The average daily value was used to interpret the results of the analysis. In the present study, the data for the 2020–2021 measurements were used (Table 1), and a comparison was made with the previous results.
Table 1. Atmospheric aerosol sampling periods at the Bolshiye Koty and Boyarsky research stations in 2020 and 2021 (the number of samples is given in parentheses).

| Year | Month | Bolshiye Koty | Boyarsky |
|------|-------|---------------|----------|
| 2020 | June  | 01/06–30/06 (41) | no measurements |
|      | July  | 01/07–31/07 (30) | 20/07–30/07 (7) |
|      | August | 01/08–31/08 (33) | 04/08–12/08 (17) |
|      | September | 01/09–30/09 (29) | no measurements |
|      | October | 13/10–23/10 (11) | no measurements |
| 2021 | July  | no measurements | 25/07–31/07 (8) |
|      | August | 03/08–12/08 (20) | 01/08–27/08 (14) |

To determine the ionic composition of aerosol, the exposed filter was placed in a polypropylene tube, and a known amount of double distilled water was added. The collected pollutants were extracted in an ultrasound bath for 30 minutes. In the extracts, after filtration through a membrane filter with a pore size of 0.2 µm, the Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$, NH$_4^+$, NO$_3^-$, Cl$^-$, and SO$_4^{2-}$ ions were determined, and pH values were measured. The ionic composition of aerosol was determined using an ICS-3000 ionic system ( Dionex, Sunnyvale, CA, USA) with an accuracy of up to 2–6%, which was approved by the US Environmental Protection [5]. An IonPac AS19 analytical column (2 × 250 mm) was used to determine anions, an IonPac CS12A (2 × 250 mm) for cations, potassium hydroxide and methanesulfonic acid eluents ( Dionex, Thermo Scientific (Thermo Fisher Scientific, Waltham, MA, USA) [19]. The determination of the level of fluctuation noises, the zero signal drift, and the deviation of the output signal of the device were controlled using the IC-SCS1 and IC-FAS-1A reference samples (Inorganic Ventures, Christiansburg, VA, USA).

The quality of the analyses has been approved by inter-laboratory comparative experiments performed within the framework of international programs Global Atmosphere Watch (GAW) under the aegis of the World Meteorological Organization (WMO) and Acid Deposition Monitoring Network in East Asia (EANET). The results of these analyses were included in the reports of GAW (QA/SAC-Americas) and EANET [20,21].

To identify the additional sources of ions entering the atmosphere, a quantitative assessment of the difference in the ratios of the concentrations of Ca$^{2+}$, K$^+$, Mg$^{2+}$, Cl$^-$, and SO$_4^{2-}$ relative to Na$^+$ was carried out according to the known formula [22]:

$$EF_i = \frac{\left(\frac{C_{i}}{Na^+}\right)_{aer}}{\left(\frac{C_{i}}{Na^+}\right)_{SW}}$$  \hspace{1cm} (1)

where $EF$ is the enrichment factor, and $\left(\frac{C_{i}}{Na^+}\right)_{aer}$ is the concentration of the $i$-th element relative to Na$^+$ in aerosol (aer) and seawater (SW) [23].

Information was used from the database server of the National Oceanic and Atmospheric Administration, USA (https://www.arl.noaa.gov/, accessed on 20 May 2022) as source data to calculate the backward trajectories [24]. To carry out calculations, we used the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT model).

**Study Area**

The Baikal depression is surrounded by high mountain ranges. The south coast of Lake Baikal is framed by the Hamar-Daban mountain range that extends from the valley of the Selenga River and borders the southern part of Lake Baikal. Its heights range from 1000 to 2370 m [25]. This complicates the dispersal of pollutants in the atmosphere in the southern part of the lake. Previously, Khodzher and coauthors [26] showed that a significant amount of air pollution from regional industrial centres located to the west and northwest entered through the southwestern border of the southern basin of Lake Baikal along the valley of the Angara River, which is open to Lake Baikal. Small settlements on the
coast do not contribute significantly to air pollution of the lake due to the small amounts of emissions [26].

Long-term observations of the change in the chemical composition of atmospheric aerosols on the southwest coast of Lake Baikal are carried out at the research station of Limnological Institute SB RAS located near the Bolshiye Koty settlement (N 51°91’, E 105°07’) with a population of about 50 people. This settlement is 24 km away from the large tourist site, the Listvyanka settlement, situated on the southwest coast of the lake (population about 2000 people) and more accessible to tourists. In summer, intensive navigation takes place on Lake Baikal (see Figure 1).

On the opposite southeast coast, the atmosphere is studied at the Boyarsky research station of Institute of Physical Material Science SB RAS (N 51°43’, E 105°52’) located near the Boyarsky settlement. The Trans-Siberian Railway passes through this settlement (population about 150 people), and the federal highway runs there.

3. Results
3.1. Atmospheric Aerosol at the Bolshiye Koty Research Station (Southwest Coast of Lake Baikal)

Studies carried out at the Bolshiye Koty research station reflect the condition of the chemical composition of aerosol in the background sparsely populated areas on the west coast of Lake Baikal. Wildfires significantly influenced the change in physical and chemical properties of the near-water atmosphere during summers [5].

3.1.1. Studies at the Bolshiye Koty Research Station in 2020

The most detailed study of the change in the ionic composition of aerosol was carried out in 2020 when aerosol was sampled daily for five months, from June to October. In June, predominantly calm and sunny weather was observed. Air temperature varied within 11–17 °C, with an increase up to 19 °C at the end of the month. In some periods, the speed of northwesterly wind increased to 3–4 m/s. In the third decade of the month, there were short-term rains. In July, air temperature increased to 19–22 °C, and precipitation became more frequent. From 5 to 13 July, the smog from long-range wildfires and the smell of burning occurred.

In August, daytime temperatures decreased to 17–19 °C; the southerly quarter winds intensified, and the periods of sunny and cloudy weather with precipitation alternated. September and October were characterized by the subsequent decrease in the air temperature to 1 °C, and there were snowfalls and an increase in wind regime.

Figure 3 shows the variability of the total ionic concentration in aerosol during samplings in 2020.

A detailed analysis of the variability of the total ionic concentration in aerosol over this period revealed the following. The sum of ions in the aerosol composition varied from 0.4 µg/m³ to 6.4 µg/m³ (mean value 2.1 ± 1.3 µg/m³). The lowest concentrations were observed in early June 0.39–0.50 µg/m³ (from 01 to 08 June). From July to the second ten days of September, the ionic concentrations in aerosol significantly increased. On 6 to 13 July, a smoke aerosol contributed to an increase in the ionic concentrations to 6.4 µg/m³. The accumulation of pollutants in the atmosphere was also observed on weekends when the flow of tourists arriving at the settlement by water transport increased (see Figure 3, brown columns). There was a smell of smoke from campfire. Periodic bursts of the total ionic concentrations from 2.4 µg/m³ to 4.0 µg/m³ occurred when the ships were mooring at the pier of the research station. Notably, during calm weather conditions above the lake, high ionic concentrations in the aerosol persisted for one to two days under the influence of the emissions from ships. From 16 to 21 August, an increase in the northwesterly quarter wind with gusts up to 15 m/s and precipitation contributed to the purification of the atmosphere and a decrease in the total amount of ions in the aerosol up to 0.6–0.9 µg/m³ [27]. Therefore, in the absence of such external effects as ship moorings at the pier with running engines, an increase in water transport units in the water area of the lake and smoke from campfires,
the total amount of ions in the aerosol composition at the Bolshiye Koty research station, like in other background areas of Lake Baikal, is much lower than 1.0 μg/m³ [5].

Figure 3. Dynamics of the total ionic concentration in aerosol at the Bolshiye Koty research station in 2020, μg/m³. Brown columns—Saturday, Sunday.

The extremely high variability in the concentrations of some ions is noteworthy. Figure 4 shows the fluctuations in the average monthly concentrations of some ions in aerosol.

Figure 4. Variability of the cations (A) and anions (B) average monthly concentrations in atmospheric aerosol in the air at the Bolshiye Koty research station in 2020, μg/m³.

The main ions in the aerosol composition were NH₄⁺, Cl⁻ and SO₄²⁻. Their dynamics were similar, indicating the identity of the formation sources of these ions. When the sunny weather predominated, these ions were most likely the products of gas-phase transformations. There was some difference in the dynamics of the concentrations of terrigenous ions (Na⁺, K⁺, Mg²⁺, and Ca²⁺). To determine a relationship between the ionic concentrations in aerosol, we carried out a correlation analysis. The data were divided...
into an array with relatively high ionic concentrations totaling more than 2.0 $\mu$g/m$^3$ and an array with the total ionic concentration of less than 1.5 $\mu$g/m$^3$. Table 2 shows the result of the correlation analysis with low concentrations. For low concentrations, the correlation analysis revealed a good relationship between the concentrations of all ions $(r = 0.65 \div 0.99)$. In the absence of the external anthropogenic sources of pollution, the composition of aerosol at the Bolshiye Koty research station resulted from the activity of natural sources.

Table 2. Correlation matrix of ionic for low ion concentrations (<1.5 $\mu$g/m$^3$) in the atmospheric aerosol of the Bolshiye Koty research station in 2020.

| Ion     | H$^+$ | Na$^+$ | NH$_4^+$ | K$^+$ | Mg$^{2+}$ | Ca$^{2+}$ | Cl$^-$ | NO$_3^-$ | SO$_4^{2-}$ |
|---------|-------|--------|----------|-------|-----------|-----------|--------|----------|------------|
| H$^+$   | 1     | 0.89   | 0.70     | 0.78  | 0.86      | 0.88      | 0.88   | 0.81     | 0.87       |
| Na$^+$  | 1     | 0.73   | 0.87     | 0.75  | 0.79      | 0.93      | 0.74   | 0.79     |            |
| NH$_4^+$| 1     | 0.75   | 0.65     | 0.68  | 0.71      | 0.81      | 0.73   |          |            |
| K$^+$   | 1     | 0.67   | 0.71     | 0.75  | 0.69      | 0.70      |        |          |            |
| Mg$^{2+}$| 1    | 0.99   | 0.76     | 0.75  | 0.75      | 0.79      |        |          |            |
| Ca$^{2+}$| 1    | 0.78   | 0.74     | 0.80  |          |           |        |          |            |
| Cl$^-$  | 1     | 0.71   |          | 0.75  |          |           |        |          |            |
| NO$_3^-$|       |        |          |       |          |           |        | 1        | 0.86       |
| SO$_4^{2-}$|     |        |          |       |          |           |        | 1        |            |

Table 3 shows the result of the correlation analysis with high concentrations. Like for ions with low values, there was a predominantly high correlation of ionic concentrations $(r = 0.62 \div 0.98)$. The weaker correlation was observed for the K$^+$ and Na$^+$ concentrations with other ions, which indicated the additional source of this ion entering the atmosphere (bold type).

Table 3. Correlation matrix of ionic for high ion concentrations (>2.0 $\mu$g/m$^3$) in the atmospheric aerosol of the Bolshiye Koty research station in 2020.

| Ion     | H$^+$ | Na$^+$ | NH$_4^+$ | K$^+$ | Mg$^{2+}$ | Ca$^{2+}$ | Cl$^-$ | NO$_3^-$ | SO$_4^{2-}$ |
|---------|-------|--------|----------|-------|-----------|-----------|--------|----------|------------|
| H$^+$   | 1     | 0.47   | 0.65     | 0.44  | 0.62      | 0.64      | 0.73   | 0.64     | 0.71       |
| Na$^+$  | 1     | 0.59   | 0.49     | 0.55  | 0.57      | 0.64      | 0.56   | 0.57     |            |
| NH$_4^+$| 1     | 0.54   | 0.88     | 0.87  | 0.85      | 0.93      | 0.93   |          |            |
| K$^+$   | 1     | 0.7    | 0.78     | 0.5   | 0.44      | 0.46      |        |          |            |
| Mg$^{2+}$| 1    | 0.98   | 0.82     | 0.83  | 0.83      | 0.83      |        |          |            |
| Ca$^{2+}$| 1    | 0.8    | 0.81     | 0.82  | 0.82      | 0.82      |        |          |            |
| Cl$^-$  | 1     | 0.86   | 0.85     |       |           |           |        |          |            |
| NO$_3^-$|       |        |          |       |           |           |        | 1        | 0.88       |
| SO$_4^{2-}$|     |        |          |       |           |           |        | 1        |            |

Enrichment factors of ions were calculated according to Formula 1. The highest values of correlation coefficients were obtained for potassium ions $(EF_{(K^+ / Na^+)}$, sulfate ions $(EF_{SO_4^{2-} / Na^+})$ and calcium $EF_{(Ca^{2+} / Na^+)}$, the mean values of which were 20, 25 and 38, respectively (Figure 5).

The increase in the concentrations of K$^+$ and SO$_4^{2-}$, as well as enrichment factors of these ions, respectively, occurred mainly in the same periods when the influence of anthropogenic pollution sources was evident. The appearance of smog from wildfires above the lake from 5 to 13 July 2020 also contributed to the growth of concentrations and
enrichment factors of K⁺ and Ca²⁺ ions. The enrichment of the aerosol substance in SO₄²⁻ was two days later. An increase in the enrichment factors for K⁺ ions indicated its emission due to biomass combustion, including fuel. It is widely acknowledged that, depending on the type of biomass burnt, Si, Ca, S, K, Al, Fe, Mg, and Cl are inorganic components of ash as well as carbon, nitrogen, sulfur, and ammonia oxides—among gases. During ageing, volatile inorganic components condense in the form of chlorides and sulfates in a group of particles enriched in potassium [28–30].

It is known that the most realistic conversion of sulfur oxides into sulfates is a period of 37 h. The maximum amount of H₂SO₄ is formed within five to seven h with an almost five-fold decrease in the amount of SO₂. After 30 h, sulfates begin to predominate [31].

Dynamics of the concentrations of chloride ions in aerosol were different. The chloride concentration increased with the intensification of northwesterly and northerly winds. Figure 6 shows the increase in the concentrations of Cl⁻ ions during the last ten days of August (from 22 to 29 August) after a long period of influx of the northwesterly quarter air masses from industrial areas of the Irkutsk Region.

3.1.2. Studies at the Bolshiye Koty Research Station in 2021

The study of the aerosol composition in 2021 at the Bolshiye Koty research station was continued from 3 to 12 August when most of the Baikal region was covered with smog from wildfires that took place in Yakutia, Krasnoyarsk Krai and the north of the Irkutsk Region. The sum of ions in the aerosol composition varied from 1.6 μg/m³ to 17.6 μg/m³ (mean value 9.0 ± 5.0 μg/m³). Visual atmospheric contamination by smoke was observed at the station from 6 August, with an increase during the following days and dispersal by 11 August. Comparison of the total ionic concentration in aerosol collected during the day and at night revealed their higher values only on the first day of observations, an additional source of which was a ship located near the station. On subsequent days, the total amount of ions in aerosol in both periods of the day was practically the same; therefore, their average daily values were analyzed. Although at the beginning of the study on 3 to 5 August, the visual presence of smoke was not traced, the ionic concentrations in aerosols were elevated and amounted, on average, to 3.8 μg/m³. During the days when there was the highest atmospheric contamination by smoke (7–9 August), the total amount of ions in aerosol became more than 3.5 times higher than at the beginning of the study, reaching the maximum value of 17.6 μg/m³ (Figure 7). The main ions in the aerosol composition were
NH₄⁺ and SO₄²⁻; during the period of atmospheric smoke, the aerosol composition changed, and the main ones were NH₄⁺, SO₄²⁻, and NO₃⁻.

**Figure 6.** Variability of the Cl⁻ concentrations in the atmospheric aerosol in the air at the Bolshiye Koty research station in 2020, μg/m³.

Under the influence of smoke aerosol, the concentrations of NH₄⁺, K⁺, NO₃⁻, and SO₄²⁻ changed the most. The concentrations of these ions became 2.9–5.1 times, 1.8–4.6 times, 4.4–13 times, and 2.5–3.8 times higher, respectively. The values of \( EF_{(K^+/Na^+)} \) and \( EF_{(SO_4^{2−}/Na^+)} \) had a 3.6-fold and 2.2-fold increase, respectively, during the atmospheric smoke.

**Figure 7.** Satellite images of Lake Baikal with smoke plume from wildfires (A) 9 August 2021 [24]. Variability of the cation (B) and anion (C) concentrations in atmospheric aerosol in the air at the Bolshiye Koty research station in 2021, μg/m³.

3.1.3. Comparison of Long-Term Results of the Study of the Ionic Composition of Aerosol on the Southwest Coast of Lake Baikal

Table 4 shows the mean average concentrations of ions in the aerosol collected during the summers of different years at the Bolshiye Koty research station.
Overall, there was a qualitative similarity in the ionic composition of aerosol, although the quantitative characteristics of ions varied in different years. The highest total amounts of ions were determined in 2015 and 2021. In 2015 and 2021, samplings were carried out under conditions of smoke-contaminated atmosphere. In 2015, wildfires were observed near the research station, while, in 2021, smoke came from remote areas. In the aerosol of 2015, the concentrations of Na\(^+\), Ca\(^{2+}\) and K\(^+\) were elevated compared to other study years. In 2021, NH\(_4\)\(^+\) prevailed among the cations; the concentrations of other cations were comparable with the data from previous years. Previously, we have shown that air carrying mainly small combustion particles enters the lake basin under the influence of long-range wildfires. The proportion of submicron aerosol fraction was predominant and averaged 99.1 to 99.5 % of the total particle number concentration above the lake [5]. We revealed that NH\(_4\)\(^+\) and K\(^+\) were adsorbed on small particles. At the same time, there was a weak correlation of the concentrations of Ca\(^{2+}\) and NO\(_3\)\(^-\) with particles sized 5.0 µm. Hence, under the conditions of nearby wildfire, large particles could bring Na\(^+\), Ca\(^{2+}\), and K\(^+\) to the atmosphere of the coastal settlements and the water area of the lake. Among anions, SO\(_4\)\(^{2-}\) predominated in the aerosol composition almost throughout the entire study period. The results obtained indicate that, in the absence of wildfires, the intensity of which has been increasing in recent years (Figure 2), in 2002, 2014, 2017, and 2018, the total ionic concentration in aerosol at the Bolshiye Koty station was close to the background level for clean areas of Lake Baikal (<1.0 µg/m\(^3\)). In 2020, the mean total concentration of ions in aerosol almost doubled.

### 3.2. Atmospheric Aerosol at the Boyarsky Research Station (Southeast Coast of Lake Baikal)

#### 3.2.1. Studies at the Boyarsky Research Station in 2020 and 2021

The aerosol at the Boyarsky research station was sampled at different times of the day from 27 July to 12 August 2020. The total ionic concentrations in aerosol varied greatly from 1.7 µg/m\(^3\) to 34.1 µg/m\(^3\) (mean value for the entire period is 6.6 µg/m\(^3\)). The lowest concentrations were determined on 12 August during the daytime. The highest

| Period (Number of Samples) | H\(^+\) | Na\(^+\) | NH\(_4\)\(^+\) | K\(^+\) | Mg\(^{2+}\) | Ca\(^{2+}\) | Cl\(^-\) | NO\(_3\)\(^-\) | SO\(_4\)\(^{2-}\) | The Sum of Ions |
|-----------------------------|-------|--------|------------|-------|-----------|-----------|--------|-----------|-----------|-------------|
| 01/06-08/02, 2002 (18)      | 0.00  | 0.09 ± 0.05 | 0.10 ± 0.05 | 0.11 ± 0.10 | 0.02 ± 0.02 | 0.08 ± 0.05 | 0.05 ± 0.04 | 0.23 ± 0.25 | 0.54 ± 0.40 | 1.21 ± 0.77 |
| 08/07-17/07, 2007 (21)      | 0.01 ± 0.00 | 0.11 ± 0.15 | 0.09 ± 0.01 | 0.10 ± 0.01 | 0.01 ± 0.01 | 0.15 ± 0.01 | 0.04 ± 0.22 | 0.18 ± 0.02 | 1.01 ± 0.05 | 1.72 ± 0.46 |
| 20/12-30/01, 2014 (9)       | 0.00  | 0.02 ± 0.00 | 0.01 ± 0.00 | 0.01 ± 0.00 | 0.02 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 |
| 09/01-09/15, 2015 (11)      | 0.00  | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 |
| 10/04-20/07, 2017 (17)      | 0.00  | 0.02 ± 0.02 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 | 0.01 ± 0.01 |
| 15/05-25/07, 2018 (20)      | 0.01 ± 0.05 | 0.03 ± 0.08 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.04 ± 0.91 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 | 0.00 ± 0.00 |
| 24/07-08/08, 2019 (18)      | 0.00  | 0.01 ± 0.01 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 | 0.02 ± 0.02 |
| 01/06-25/10, 2020 (144)     | 0.03 ± 0.02 | 0.10 ± 0.13 | 0.26 ± 0.24 | 0.08 ± 0.32 | 0.01 ± 0.08 | 0.09 ± 0.38 | 0.32 ± 0.46 | 0.29 ± 0.23 | 0.91 ± 0.36 | 2.08 ± 0.92 |
| 03/08-12/08, 2020 (23)      | 0.02 ± 0.01 | 0.13 ± 0.04 | 2.04 ± 1.22 | 0.32 ± 0.19 | 0.08 ± 0.05 | 0.38 ± 0.20 | 0.46 ± 0.09 | 2.23 ± 0.23 | 3.36 ± 1.71 | 9.02 ± 5.03 |

#### Table 4. Ionic composition of the atmospheric aerosol at the Bolshiye Koty research station from 2002 to 2021, µg/m\(^3\) (the upper line shows the arithmetic mean ± standard deviation; the lower line shows the range).
total amount of ions was detected in the aerosol collected at nighttime on 27–28 July and 9 August. The trajectory analysis of air mass transport indicated that, during 9 August, air masses entered the atmosphere on the east coast of the southern basin of Lake Baikal with smoke aerosol from the areas engulfed by wildfires [24]. As an example, Figure 8 shows the influx of air masses from Yakutia.

![Figure 8. Seven-day back trajectories of the air mass transport to the Boyarsky settlement during the aerosol sampling on 9 August 2020, which were brought at heights of 100 m (1), 1000 m (2), and 1500 m (3) [24].](image)

The greatest daily variability in the ionic composition of aerosol was observed in the first two days. During the following days, the differences between daytime and nighttime were lower than the standard deviations; therefore, the mean values can be considered typical for this observation period. Like the aerosol at the Bolshiye Koty research station, the measurement data were averaged geometrically due to high positive coefficients of skewness and kurtosis.

Figure 9 shows the distribution of ionic concentrations in the aerosol composition in July and August 2020. The concentrations of Ca$^{2+}$, Na$^+$, K$^+$, Cl$^-$, and SO$_4^{2-}$ predominated in the aerosol. In continental regions, salt deposits in the soil become sources of chlorides in the absence of a marine component. In the Republic of Buryatia, approximately 210 thousand hectares of saline lands were recorded, including 27.6 thousand hectares of solonchaks; 182.4 thousand hectares were classified as medium or highly saline. Saline soil was mostly widespread in the southern, southeastern, and southwestern parts of Buryatia. Ca$^{2+}$ and Mg$^{2+}$ prevailed in the exchangeable cations. Soil erosion in this region has become widespread, resulting in a serious environmental problem [25,26]. Based on the calculation of back trajectories, we identified a periodic flow of air masses to the observation station both from these areas and from the deserted areas of Mongolia.

The chemical composition of the aerosol in July and August 2021 at the Boyarsky research station differed from the aerosol in the previous year. Despite intense atmospheric smoke, the total amount of ions varied from 1.3 μg/m$^3$ to 18.6 μg/m$^3$ (mean value 5.5 ± 4.5 μg/m$^3$). As on the west coast, the highest ionic concentrations were determined from 6 to 11 August 2021. NH$_4^+$ and SO$_4^{2-}$ ions predominated in the aerosol composition, while the contribution from NO$_3^-$ ions increased during the fire danger period. We calculated the enrichment factors of ions. In 2020, the highest enrichment was identified for potassium, calcium, and sulfate ions. In 2021, the mean values of the enrichment factors for these ions were 2.6, 1.7 and 8.5 times higher. During atmospheric smoke, there were 5.8- and 18-fold increases in $EF_{(K^+/Na^+)}$ and $EF_{(SO_4^{2-}/Na^+)}$ respectively.
Ionic composition of the atmospheric aerosol at the Boyarsky research station from 2014 to 2020, µg/m³.

3.2.2. Comparison of Long-Term Results of the Study of the Ionic Composition of Aerosol on the Southeast Coast of Lake Baikal

Table 5 shows the mean concentrations of ions in the aerosol collected during the summers of different years at the Boyarsky research station.

Table 5. Ionic composition of the atmospheric aerosol at the Boyarsky research station from 2014 to 2021, µg/m³ (the upper line shows the arithmetic mean ± standard deviation; the lower line shows the range).

| Period (Number of Samples) | H⁺ | Na⁺ | NH₄⁺ | K⁺ | Mg²⁺ | Ca²⁺ | Cl⁻ | NO₃⁻ | SO₄²⁻ | The Sum of Ions |
|---------------------------|----|-----|------|----|------|------|-----|------|-------|---------------|
| 20/07–15/08, 2014 (45)    | 0.02 ± 0.01 | 0.77 ± 1.73 | 1.72 ± 7.19 | 0.27 ± 1.02 | 0.40 ± 0.50 | 1.62 ± 5.32 | 2.93 ± 1.49 | 4.46 ± 3.02 | 4.36 ± 11.96 | 16.55 ± 32.24 |
| 02/08–18/08, 2015 (36)    | 0.01 ± 0.05 | 0.32 ± 1.70 | 0.56 ± 4.03 | 0.07 ± 0.88 | 0.12 ± 1.01 | 0.74 ± 3.99 | 1.17 ± 5.77 | 0.54 ± 9.45 | 1.37 ± 9.36 | 7.11 ± 34.34 |
| 30/07–18/08, 2016 (40)    | 0.00 ± 0.10 | 0.71 ± 4.61 | 1.16 ± 29.28 | 0.31 ± 5.32 | 0.05 ± 1.29 | 0.41 ± 11.37 | 0.51 ± 4.06 | 0.65 ± 16.31 | 4.66 ± 26.68 | 12.10 ± 96.45 |
| 27/07–15/08, 2017 (50)    | 0.04 ± 0.13 | 0.62 ± 0.24 | 1.29 ± 1.11 | 0.45 ± 0.29 | 0.26 ± 0.18 | 0.45 ± 0.11 | 3.99 ± 1.25 | 0.28 ± 0.28 | 1.63 ± 0.79 | 9.00 ± 2.37 |
| 01/06–05/06, 14/07–28/08, 2018 (35) | 0.01 ± 0.01 | 0.42 ± 0.99 | 0.29 ± 0.39 | 1.43 ± 1.22 | 0.07 ± 0.06 | 0.69 ± 0.52 | 1.94 ± 1.43 | 0.15 ± 0.20 | 0.88 ± 1.96 | 5.88 ± 4.86 |
| 12/07–08/08, 2019 (52)    | 0.00 ± 0.02 | 0.04 ± 5.81 | 0.01 ± 1.82 | 0.07 ± 5.12 | 0.02 ± 0.25 | 0.17 ± 2.56 | 0.27 ± 5.54 | 0.00 ± 1.05 | 0.12 ± 11.89 | 1.61 ± 24.23 |
| 25/07–27/08, 2020 (24)    | 0.08 ± 0.02 | 0.81 ± 0.10 | 0.82 ± 1.15 | 0.60 ± 0.19 | 0.09 ± 0.03 | 1.20 ± 0.23 | 1.90 ± 0.40 | 0.70 ± 11.2 | 2.48 ± 2.21 | 8.71 ± 5.45 |
| 25/07–27/08, 2021 (22)    | 0.02 ± 0.02 | 0.10 ± 0.06 | 0.15 ± 1.00 | 0.19 ± 0.14 | 0.03 ± 0.02 | 0.23 ± 0.17 | 0.40 ± 0.31 | 1.12 ± 1.96 | 2.21 ± 1.58 | 5.45 ± 4.53 |

Despite a slight increase in the total amount of ions in 2020 and 2021, there was a trend towards a decrease in the ionic concentrations in atmospheric aerosol on the southeast coast of Lake Baikal. No general pattern could be traced in the variability of the concentrations of some ions. If the dominance of the concentrations of Na⁺, K⁺ and Ca²⁺ could be observed among cations throughout the entire study period, among anions, there was no such a distribution.
3.3. Comparison of the Ionic Composition in Aerosols on the West and East Coasts of the Southern Basin of Lake Baikal

On the east coast of Lake Baikal, the number of local anthropogenic sources of air pollutants was greater than on the west coast. Here, as mentioned above, the Trans-Siberian Railway and federal highway pass together with transport infrastructure (see Figure 1). Autonomous coal-fired and electric boilers heat public and administrative buildings; residential buildings have stove heating. The largest settlements, Slyudyanka (population about 18,200 people) and Baikalsk (population about 12,500 people), are located in the south of the lake (see Figure 1). The mining industry in Slyudyanka and the pulp and paper mill in Baikalsk ceased operations more than ten years ago. The influence of local thermal power plants is limited to the surrounding areas and does not extend to the rest of the southern basin of Lake Baikal. According to Obolkin et al. [4,32], the transport of air pollution to the southern basin from Buryatia occurs from large thermal power plants (Ulan-Ude, Selenginsk and Gusinozersk) and does not reach the west coast of the lake. The frequency of transport is associated with the direction of southeasterly and southerly winds, accounting for 13% per year [32].

Less than 3000 people live on the southwest coast of the southern basin. The largest settlement is Listvyanka. Atmospheric pollution is transported to the southern basin from the enterprises of the Irkutsk-Cheremkhovo industrial hub along the Angara River valley during mesoscale jet transport in the boundary atmospheric layer with northwesterly flows (30% per year, [32]). The chemical composition of aerosol on the southeast coast, which is different from the aerosol composition on the southwest coast both qualitatively and quantitatively.

The concentrations of NH$_4^+$, Cl$^-$, and SO$_4^{2-}$ ions predominated in the aerosol at the Bolshiye Koty research station. On the east coast, Ca$^{2+}$, Na$^+$, K$^+$, Cl$^-$, and SO$_4^{2-}$ were major ions. To trace the similarity and difference of the aerosol composition at the investigated stations, we chose the dates when the sampling was simultaneous. The comparison revealed the following (Table 6). The concentrations of K$^+$, Ca$^{2+}$, and Cl$^-$ ions, which prevailed in the aerosol from the Boyarsky research station in 2020, were higher than in the aerosol from the Bolshiye Koty research station. The concentrations of ammonium ions that were among the major ions in the aerosol from the Bolshiye Koty research station exceeded the same values in the aerosol from the Boyarsky research station. The greatest heterogeneity was observed in the distribution of sulfates. There were days when the concentrations of these ions were elevated in the aerosols from both research stations. Nevertheless, the comparison of the results over the entire observation period in 2020 indicated that the mean concentrations of cations in the aerosol on the southeast coast were from 1.6 to 13.7 times higher than in the aerosol on the southwest coast, and the mean concentrations of anions—from 1.5 to 5.4 times higher. Despite the variations in the concentrations of some ions, the total concentration of ions in the aerosol of the Boyarsky research station exceeded this value by a factor of four at the Bolshiye Koty research station. At both study sites, the Ca$^{2+}$, K$^+$, and SO$_4^{2-}$ enrichment factors of aerosol particles were high.

In 2021, in the quantitative comparison of ions in the aerosol on the southeast and southwest costs, the situation was opposite to the previous study year. A similar comparison revealed (see Table 6) that the ionic concentrations in the aerosol on the southwest coast, except for one day, were higher than in the aerosol on the opposite coast. This was most likely caused by the more intense smoke in the atmosphere on the west coast (see Figure 6). Figure 10 shows the prevailing wind direction above the ground surface at the moment of the influx of air masses from the northwestern areas. Probably, in the atmosphere of the Boyarsky research station, the southerly quarter winds contributed to the distribution of the entering pollutants, while at the Bolshiye Koty research station, the northwesterly and northerly winds prevailed [27,33], bringing smoke aerosol with them. In 2021, the total concentration of ions on the southwest coast had a 2.7-fold decrease compared to the same
observation period in 2020 (from 3 to 12 August), while it did not change on the southeast coast. At both research stations, the ratios of ionic concentrations in the aerosols changed, and NH$_4^+$ and SO$_4^{2-}$ became major ions.

Table 6. The ionic composition of the aerosols from the Bolshiye Koty and Boyarsky research stations in August 2020 and 2021, µg/m$^3$.

| Date     | Bolshiye Koty | Boyarsky |
|----------|--------------|----------|
|          | NH$_4^+$ K$^+$ Ca$^{2+}$ Cl$^-$ SO$_4^{2-}$ NH$_4^+$ K$^+$ Ca$^{2+}$ Cl$^-$ SO$_4^{2-}$ |
| 04/08/2020 | 0.52 0.13 0.14 0.36 1.40 0.50 0.34 0.96 1.31 1.10 |
| 05/08/2020 | 0.26 0.16 0.11 0.25 0.89 0.12 0.28 0.64 1.10 1.14 |
| 06/08/2020 | 0.30 0.07 0.08 0.34 1.05 0.02 0.27 0.59 0.89 1.45 |
| 10/08/2020 | 0.66 0.21 0.21 0.29 1.51 0.12 0.63 0.49 1.43 0.71 |
| 11/08/2020 | 0.65 0.13 0.21 0.39 1.37 0.08 0.37 0.82 1.45 1.27 |
| 12/08/2020 | 0.18 0.07 0.06 0.26 0.43 0.25 0.42 0.57 1.26 0.83 |
| 04/08/2021 | 0.77 0.11 0.36 0.43 1.51 0.503 0.081 0.101 0.286 0.949 |
| 05/08/2021 | 1.07 0.12 0.46 0.44 3.01 0.734 0.115 0.147 0.340 1.739 |
| 06/08/2021 | 1.69 0.24 0.51 0.50 3.61 0.920 0.104 0.135 0.388 2.136 |
| 08/08/2021 | 4.06 0.58 0.64 0.56 5.87 3.960 0.400 0.476 0.368 3.639 |
| 10/08/2021 | 2.96 0.47 0.22 0.48 4.55 3.808 0.555 0.288 0.545 5.136 |
| 11/08/2021 | 1.98 0.36 0.21 0.44 2.95 1.026 0.292 0.123 0.291 4.373 |
| 12/08/2021 | 1.36 0.24 0.28 0.45 2.23 0.853 0.274 0.341 0.252 1.487 |

Figure 10. Prevailing wind directions at the Bolshiye Koty (A) and Boyarsky (B) research stations on 6 to 11 August 2021 [27,33].

Overall, the state of the atmospheric aerosol in the southwest coast in the absence of smoke aerosol was less affected by the anthropogenic factor than in the southeastern part of the lake. Its chemical composition, even during the increased recreational activity, was close to the aerosol composition in clean areas of Lake Baikal. On the southeast coast, the infrastructure of the region influenced the chemical composition of aerosol. The atmosphere of the coastal areas of the lake, as well as above its surface, was exposed to natural phenomena. Despite the episodic effect of natural factors, there was a growth and change in the ionic concentrations in the atmospheric aerosol.

4. Conclusions

In 2020 and 2021, we studied the chemical composition of atmospheric aerosols at the Bolshiye Koty and Boyarsky research stations located at a 50 km distance from each other, on the opposite shores in the southern basin of Lake Baikal, which characterize
mainly the background atmospheric conditions above this part of the lake. The study aimed at clarifying the real condition of the atmospheric air in summer above this area that periodically experiences the anthropogenic impact of industrial complexes in the Baikal region with the prevailing northwesterly air mass transport to the lake. The condition of the atmosphere in winter is monitored in this area by the chemical composition of snow.

A detailed analysis of major ions at the stations revealed differences in their composition and mass concentration in the summer of the compared years, which was due to the different influence of local natural and anthropogenic sources of pollution. Atmospheric aerosols were studied for two years, both under the influence of local natural (wildfires) and anthropogenic sources of pollution (emissions from operating water transport engines and smoke from campfires in tourist zones) and in their absence. In 2020, when large sources of air pollution of the lake water area were absent, the mean total concentration of ions at the Bolshiye Koty research station was 2.08 ± 1.26 µg/m³, which two times exceeded the natural background level obtained for other clean areas of the lake (≤1.0 µg/m³). The total concentration of ions in the aerosol from the Boyarsky research station in the summer of 2020 was 4.7 times greater than at the Bolshiye Koty research station. NH₄⁺, Cl⁻, and SO₄²⁻ were major ions in the aerosol on the southwest coast (Bolshiye Koty), and NH₄⁺, K⁺, NO₃⁻, and SO₄²⁻—on the southeast coast (Boyarsky). The mean concentrations of cations in the aerosol on the southeast coast were 3.1 to 15.6 times higher than on the southwest coast, and those of anions—3.1 to 7.2 times higher. The more diverse composition of the aerosol on the southeast coast was due to the transport of pollutants from large thermal power plants in Buryatia as well as to the development of soil erosion processes in this area.

The smoke aerosols from wildfires, the number of which has been increasing in recent years, contribute the most to the air pollution in summer. In 2021, during wildfires in Siberia, there were significant changes in the ionic composition of aerosols and their total mass. The mean total amount of ions under the influence of smoke from wildfires near the Bolshiye Koty research station increased to 9.0 ± 5.0 µg/m³ and was 5.5 ± 4.5 µg/m³ at the Boyarsky research station. NH₄⁺ and SO₄²⁻ became major ions in the aerosols at both stations. In periods of intense smoke, the contribution of NO₃⁻ ions increased.

This study is relevant and of practical importance, based on which it is possible to assess the supply of chemical substances from the atmosphere during dry deposition on the water surface and adjacent areas of Lake Baikal. These estimates are important in the analysis of surface water quality in Lake Baikal and rivers flowing into the southern basin of the lake, for which the atmospheric supply is the main, as shown previously.

**Author Contributions:** Conceptualization, L.P.G.; methodology, L.P.G., A.S.Z., O.I.K. and G.S.Z.; software, A.S.Z.; validation, A.S.Z., O.I.K. and A.S.; formal analysis, L.P.G.; investigation, L.P.G., A.S.Z., O.I.K., G.S.Z. and T.V.K.; resources, T.V.K., O.I.K. and A.S.Z.; writing—original draft preparation, L.P.G.; writing—review and editing, L.P.G., T.V.K. and G.S.Z.; visualization, T.V.K.; supervision, L.P.G., T.V.K. and G.S.Z.; project administration, T.V.K.; funding acquisition, T.V.K. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Acknowledgments:** This study was supported by the project of the Russian Science Foundation (RSF) No. 19-77-20058 «Studies of the composition and spatiotemporal distribution of the aerosol-gas impurities in the Lake Baikal atmosphere». The investigations were carried out using the equipment of the center of shared facilities «Atmosphere», Institute of Atmospheric Optics Siberian Branch of the Russian Academy of Sciences. The authors thank Valentina M. Domysheva, Natalia A. Onischuk, and Olga G. Netsvetaeva from Laboratory of Hydrochemistry and Atmosphere Chemistry at Limnological Institute SB RAS for assistance in sampling the atmospheric aerosol in 2020.

**Conflicts of Interest:** The authors declare no conflict of interest.
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