Results of synchronous measurements of the content of nitrogen and sulfur oxides in the atmosphere of Southern Baikal in July 2023



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ABSTRACT. The paper presents the results of synchronous measurements of the content of small gas impurities at the three atmospheric monitoring stations located on the shore of Southern Baikal, July 2023. The measurements were carried out using automatic gas analyzers, which enabled to obtain data on the content of sulfur and nitrogen oxides in the atmospheric air with high temporal resolution. The studies showed that in spite of a small distance between the observation stations, the fluctuations of concentrations of small gas impurities on different shores of Southern Baikal occur independently of each other. We suggested that an increase of nitrogen and sulfur oxide concentrations on the western shore in summer is probably correlated more with the impact of regional thermal power plants (TPP) and on the eastern shore with the orographic features of the southern basin of the lake and local sources. The influence of meteorological conditions on the content of gas components in the atmosphere of Southern Baikal was estimated using multivariate statistical methods, namely, NWR and PSCF.

Keywords: air pollution, sulfur dioxide, nitrogen oxides, the Baikal natural territory, Siberia

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1. Introduction

of sulfur (SO_{2}) and nitrogen $(NO_v = NO + NO_2)$ are common atmospheric impurities that play a significant role in the troposphere and lead to a number of environmental problems such as acid rain, acidification of freshwater ecosystems (Moiseenko et al., 2018; 2022; Obolkin et al., 2016) and photochemical smog (He et al., 2007; Shon et al., 2011). Sulfur and nitrogen oxides in the atmosphere are known to contribute to the generation of aerosol particles with aerodynamic diameter less than 2.5 µm (Seinfeld and Pandis, 2016; Liu et al., 2019), which in turn have negative effects on human health (Lelieveld et al., 2015; Tiotiu et al., 2020; Southerland et al., 2022), contribute to a decrease in atmospheric transparency (Kovadlo et al., 2018; Taschilin et al., 2021), and lead to an enhanced greenhouse effect and climate change (Gharibzadeh et al., 2021; Liu et al., 2022). Besides, nitrogen oxides are precursors for the formation of ground-level ozone, which leads to deterioration of air quality and adversely affects human health.

Over the last four decades, there has been an increase in the content of small gas impurities in the atmosphere of reference areas worldwide, along with improvements in air quality in industrial cities (Fenger, 2009; Sillanpää et al., 2022) (Golobokova et al., 2018a; Sicard et al., 2023). This trend is also relevant for the Baikal region. The atmosphere in this region is showing an increase in nitrogen oxides (Golobokova et al., 2018b). It was previously found that Southern Baikal is subjected to significant atmospheric pollution as a result of high-altitude transport of sulfur and nitrogen oxides from remote regional TPP located to the northwest and southeast of the lake (Obolkin et al., 2014; Shikhovtsev et al., 2023) (Obolkin et al., 2017; Popovicheva et al., 2021). Emissions from sources of atmospheric pollution located within the Central Ecological Zone of the lake, as a rule, spread locally, and their contribution to the total level of atmospheric pollution over the lake in the cold period is insignificant (Molozhnikova and Kuchmenko, 2004; Molozhnikova et al., 2023).

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In summer, forest fires are a significant source determining the state of the atmosphere above the lake. Thus, according to the studies of (Marinaite et al., 2019; Golobokova et al., 2022; Khodzher et al., 2024), an increase in the concentration of oxides of nitrogen and sulfur, persistent organic pollutants, including polyaromatic hydrocarbons (PAHs), soot, nutrients (mineral nitrogen, potassium), trace elements (iron, manganese, vanadium, zinc, barium, lead) was recorded both in the atmosphere and in the surface waters of the lake. The long-term input of nitrogen- and sulfur-containing substances into the atmosphere over Southern Baikal resulted in a decrease in the pH level of atmospheric precipitation, which is the main source of supply for the lake's southeastern tributaries. This led to acidification of river waters (Tomberg et al., 2016).

Khodzher and Sorokovikova (2007) showed that from 2 to 6 per cent of soluble components and from 30 to 60 per cent of nutrients enter the catchment area of Lake Baikal from the atmosphere, providing an extra inflow of nutrients. Precipitation with an increased nitrogen content may be one of the key factors contributing to the mass development of the under-ice dinoflagellate community Gymnodinium baicalense var. minor Antipova in Listvenichnichny Bay (Southern Baikal) in 2018 (Obolkin et al., 2019). In Bay of Bolshiye Koty, located on the southern shore of Lake Baikal, similar phenomena have been repeatedly observed. In July 2019, mass reproduction of cyanobacteria occurred after four days of nitrogen- and phosphorus-enriched rains (Bondarenko et al., 2021). In June 2022, heavy rainfall (34 mm) caused an increase in fecal indicator bacteria in the same area (Malnik et al., 2024).

Thus, gas impurities can influence the formation of the chemical composition of the Baikal water and determine its quality. Thanks to the intra-basin circulation of air (Arshinov et al., 2001), impurities entering the atmosphere can be transported throughout the water area, contributing to the pollution of hydrosphere

of the region. Moreover, the mountain basin can accumulate pollutants not only from local sources but also from vast territories of Siberia, China, and Mongolia (Mashyanov et al., 2022; Nasonov et al., 2023). It is essential to study environmental changes in the region in terms of negative impacts on unique natural objects.

The study aims to determine how small atmospheric impurities are distributed in the atmosphere of Southern Baikal and to estimate the location of remote sources during the period of minimum impact of regional TPP. For this purpose, data of continuous automatic registration of the concentration of small gas impurities in the atmospheric air of Southern Baikal with a high frequency of measurements were used.

2. Materials and methods2.1. Location of stations and equipment used

The measurements were carried out simultaneously at the three stations: Listvyanka (51.84 N, 104.89 E), Bolshiye Koty (51.89 N, 105.06 E), and Boyarsky (51.84 N, 106.07 E). The location of observation stations, which represent the largest stationary sources of atmospheric pollution, and a map-scheme of the study area are presented in Figure 1. The measurements were performed using chemiluminescent gas analyzers manufactured by OPTEC (St. Petersburg, Russia): SV-320 and SV-320A (SO₂); PA-310A (NO₂ and NO); with a detection limit of 0.001 mg/m³, a reduced error of \pm 25% in the range from 0 mg/m³to 0.05 mg/m³ and a relative error of \pm 25% in the range from 0.05 mg/ m³to 2.0 mg/m³. These devices are verified annually in the laboratory of OPTEC. Data on the direction and speed of the wind were obtained using the AMK-11 acoustic meteorological system (Boyarsky station) and the Sokol-M meteorological complex (Listvyanka and Bolshiye Koty stations).

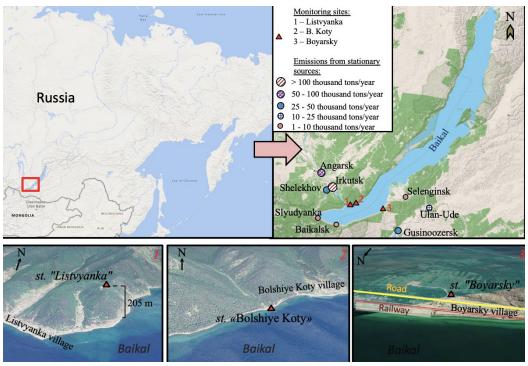


Fig. 1. Location of the sampling station: 1 - "Listvyanka"; 2 - "Bolshiye Koty"; 3 - "Boyarsky" and map of the studied area

2.2. Non-parametric wind regression (NWR)

A receptor-based non-parametric wind regression (NWR) model was used to identify potential local sources of atmospheric pollution using meteorological data (Henry et al., 2009). In this study, the openair package (R language) was used to analyze the NWR, and the Gaussian kernel smoothing method was applied to the resulting wind direction and speed (Carslaw and Ropkins, 2012). The NWR analysis is defined as follows (1) and (2):

$$E(C|\theta,u) = \frac{\sum_{i=1}^{N} K_1 \left(\frac{(\theta - W_i)}{\sigma}\right) K_2 \left(\frac{(u - U_i)}{h}\right) C_i W_i}{\sum_{i=1}^{N} K_1 \left(\frac{(\theta - W_i)}{\sigma}\right) K_2 \left(\frac{(u - U_i)}{h}\right)}$$
(1)
$$K(x) = \frac{1}{\sqrt{2\pi}} \cdot e^{-0.5x^2}$$
(2)

where Ci is measured pollutant concentration, Ui and Wi are resulting wind speed and standard deviation of wind direction for the i-th observation. N is total number of observations; K_1 and K_2 are smoothing kernels; θ is wind direction; u is wind speed; and σ and h are smoothing parameters for wind direction and wind speed, respectively.

2.3. Potential Source Contribution Function (PSCF)

The location of remote sources of the analyzed pollutants was determined using the potential source contribution function (PSCF). The method is based on the analysis of inverse trajectories of air masses. The 72-hour back trajectories required for the analysis were generated using the HYSPLIT model developed by the National Oceanic and Atmospheric Administration (NOAA) (Draxler and Hess, 1998; Stein et al., 2015). The PSCF is calculated on the basis of the probability

of occurrence of i concentration in each grid cell, as specified in equation (3):

$$PCSF_{i,j} = \frac{m_{ij}}{n_{ii}}$$
 (3)

where m_{ij} is number of points whose concentration exceeds the limit value, and n_{ij} is number of endpoints of the return paths that pass through each grid cell (Karnae and John, 2011; Liu et al., 2024).

3. Results and discussion

3.1. Content of gas impurities in the atmosphere of Southern Baikal

The study considers the results of an analysis of data on the concentrations of SO₂, NO, and NO₂ in the surface layer of the atmosphere in the area of Southern Baikal, obtained between 17 and 28 July 2023. Figure 2 shows the results of measurements of surface concentrations of nitrogen and sulfur oxides at the three monitoring stations. Figure 2 shows that the highest variability was recorded for sulfur dioxide. The hourly mean concentrations of SO2 at the Boyarsky station ranged from 5 to 30 $\mu g/m^3$, while at Listvyanka station, they varied from 0 to 28 μ g/m³. On average during the observation period, concentrations of SO₂ on the southeastern shore were 15 μ g/m³, on the southwestern shore were 5 μg/m³, which corresponded well with measurements made both during ship expeditions in 2020-2022 (Zhamsueva et al., 2020) and groundbased measurements conducted at the Boyarsky station in 2015 (Zayakhanov et al., 2019).

The highest recorded concentration of nitrogen oxide was also observed at the Boyarsky station. During the observation period, concentrations of nitrogen dioxide increased up to 33 $\mu g/m^3$, while nitrogen monoxide concentrations reached 7 $\mu g/m^3$. Figure 2-B illustrates that short-term spikes in nitrogen dioxide levels above 20 $\mu g/m^3$ were periodically observed at the Boyarsky station.

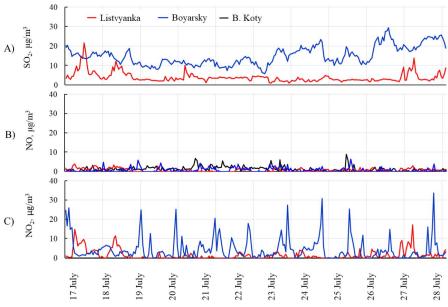


Fig.2. Temporal variability of hourly average values of: A) sulfur dioxide; B) nitrogen oxide; C) nitrogen dioxide at «Listvyanka», «Bolshiye Koty», «Boyarsky» stations on 17-28 July, 2023.

3.2. Identification of local sources using the NWR model

We used a receptor model of non-parametric wind regression (NWR) to assess the influence of meteorological parameters on the atmospheric concentration of impurities for all stations. The 10-minute concentration of impurities (NO, NO₂, SO₂) was selected as a dependent variable, and the mean wind direction and wind speed were selected as predictor variables (Fig. 3-8).

The analysis (Fig. 3-5) revealed that the highest content of SO₂, NO and NO₂ in the surface atmosphere of Listvyanka station was recorded in the night and morning hours (from 12 a.m. to 8 a.m.), at winds from the north and north-west with velocities from 1 to 4 m/ sec. This is caused by the fact that emissions of nitrogen and sulfur oxides spreading at high altitudes (with TPP plumes), due to a decrease in the thickness of the atmospheric boundary layer and weakening of thermal convection can go lower. During the daytime hours, the heating of the Earth's surface and the atmospheric boundary layer cause turbulence to develop, creating ideal conditions for the dispersion of impurities. The wind regime becomes more diverse, which leads to a decrease in concentrations of sulfur and nitrogen oxides to the summer background values of Southern Baikal (Zhamsueva et al., 2022; Shikhovtsev et al., 2024). In the daily dynamics of nitrogen oxides, the second increase from 12 p.m. to 15 p.m. was observed. This increase occurred at winds of south-east, south, and south-west directions, which corresponded to the location of Listvyanka settlement and Listvennichny Bay. The probable sources of these gaseous impurities may be road and water transport.

The highest concentrations of SO_2 and NO_2 were recorded at the Boyarsky station at winds of south and south-west directions. These directions corresponded to the position of local sources of atmospheric pollution, which are situated on the south-eastern coast of Lake Baikal (Fig. 6-7). According to the NWR analysis,

individual episodes were identified where air masses enriched with nitrogen and sulfur oxides are transported from the south along the Manturikha River valley. The calculation of direct trajectories of air mass movement (Fig. 9) showed that an increase in the content of oxides of sulfur and nitrogen at the Boyarsky station can be attributed to the influence of remote sources located in the town of Gusinoozersk.

The NWR analysis performed for NO (Fig. 8) demonstrates that concentration of nitrogen monoxide increases between 7 a.m. and 11 p.m. local time regardless of wind direction. This may indicate that local sources of atmospheric pollution, such as motor transport, affect the Boyarsky station. During night hours, concentrations of NO decrease to values close to the detection limit of the device.

3.3. Identification of remote sources using PSCF analysis

We used the method of multivariate statistics techniques to determine the areas of potential sources of gas impurities (NO_2 and SO_2) in the atmosphere of Southern Baikal. This involved a joint analysis of inverse trajectories calculated with the HYSPLIT model and measurements of surface concentrations of small gas impurities. To estimate remote sources, the duration of the return trajectories was accepted as 72 hours. Calculations were carried out for 1,000 meters above ground level (AGL) for the period from 17 to 28 July 2023.

The results of the PSCF analysis are presented in Figure 10. The calculations demonstrate that the probabilities of remote source locations for NO_2 and SO_2 (greater than 0.5) are similar for the two stations. The first maximum is defined to the north, north-west of the observation stations. The air masses entering this sector are characterized as arctic and temperate, formed over the North Atlantic and transformed as they move over the industrial areas of Western and Eastern Siberia, where the major sources of nitrogen and sul-

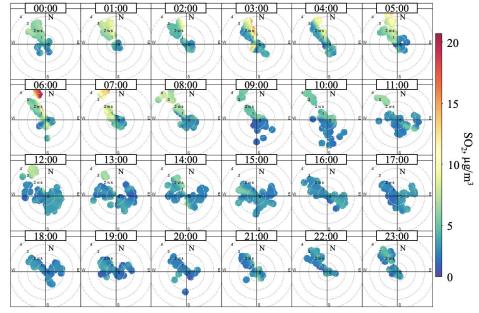


Fig. 3. NWR analysis for ten-minute concentrations of SO₂ in the polar coordinate system at «Listvyanka» station, July 2023.

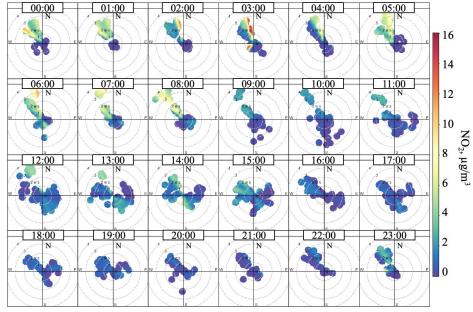


Fig.4. NWR analysis for ten-minute concentrations of NO₂ in the polar coordinate system at «Listvyanka» station, July 2023.

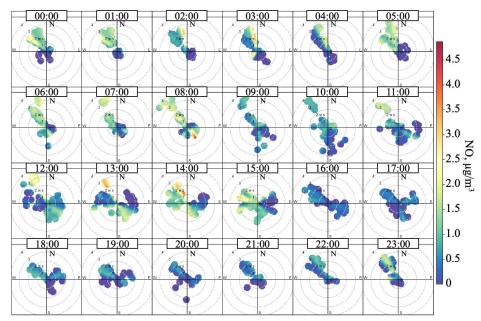


Fig.5. NWR analysis for ten-minute concentrations of NO in the polar coordinate system at «Listvyanka» station, July 2023.

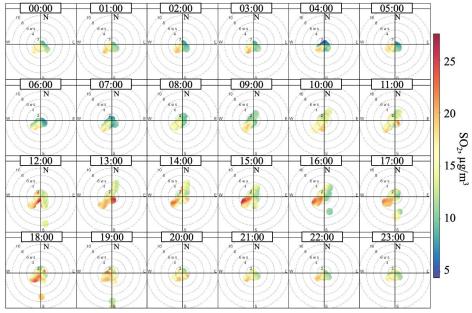


Fig. 6. NWR analysis for ten-minute concentrations of SO₂ in the polar coordinate system at the «Boyarsky» station, July 2023.

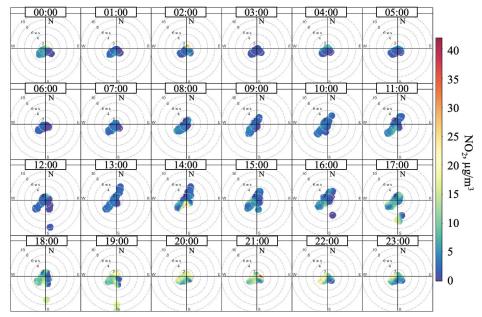


Fig. 7. NWR analysis for ten-minute concentrations of NO₂ in the polar coordinate system at the «Boyarsky» station, July 2023.

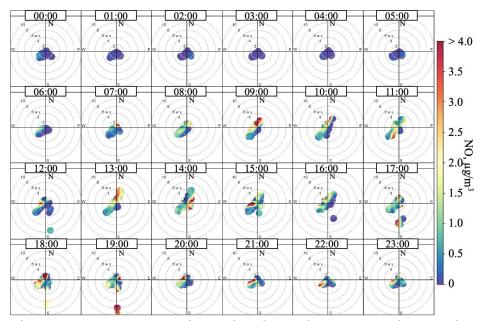


Fig.8. NWR analysis for ten-minute concentrations of NO in the polar coordinate system at the «Boyarsky» station, July 2023.

fur oxides are enterprises of the fuel and energy complex (FEC) (Maysyuk, 2017). Therefore, we can assume that at the beginning of the study (17-23 July), when a stable atmosphere was observed in the surface layer with insignificant northwestern flows directed towards Southern Baikal, the contribution to air pollution in the southern basin of the lake was influenced not only by sources of the Irutsk-Cheremkhovo industrial hub but also by industrial towns of Novosibirsk Region, Krasnoyarsk Krai, and the north of Irkutsk Region. This is consistent with the data of surface measurements demonstrated in Figures 2-5.

The second maximum probability of remote source locations is visualized to the south, south-east of the lake. It is correlated with the change of meteorological situation in the region on July 23. The monitoring stations were subjected to a low-gradient field of atmospheric pressure with light local winds of different directions (Fig. 3-8). There was a slight predominance of southern and southeastern winds at the

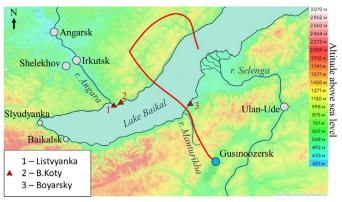


Fig.9. Topographic map of the study region (https://ru-ru.topographic-map.com) and direct trajectory of air mass movement from stationary sources of Gusinoozersk at an altitude of 500 m AGL, 20/07/2023 7a.m. MT.

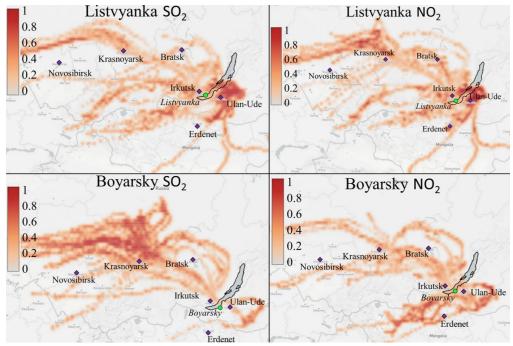


Fig.10. Spatial distribution of the probability of SO₂ and NO₂ source location at the «Listvyanka» and «Boyarsky» stations, which was performed using PSCF analysis from 17 to 28 July.

"Listvyanka" station and south-western, southern winds at the "Boyarsky" station. The state of the Southern Baikal air basin was impacted by the air masses that developed over Kazakhstan and Mongolian territory. The Republic of Buryatia's industrial towns – Ulan-Ude, Gusinoozersk, and the settlements of Kamensk and Selenginsk – are the primary sources, as Figure 10 makes evident. Additionally, the town of Erdenet (Mongolia) has a slight effect.

4. Conclusion

We examined the hourly average concentrations of small gas pollutants, such as sulfur and nitrogen oxides, as well as meteorological factors, such as wind direction and speed, at the three South Baikal atmospheric monitoring stations during the summer of 2023.

The study results demonstrated that, despite the proximity of the observation stations, changes in the concentration of small gas impurities on the different shores of Southern Baikal occur independently of each other. The location of the stations, peculiarities of the intra-basin circulation of air and location with respect to anthropogenic sources – all explain the reasons for such changes.

The estimates of the influence of meteorological parameters on the content of gas impurities in the atmosphere of Southern Baikal using the NWR model showed that the maximum concentrations on the western coast of the lake were recorded at winds of a northwest direction with a speed of 1 to 4 m/s. The eastern shore of the lake is characterized by a gradual increase in concentration, with the southwestern direction in the same velocity range.

The potential source contribution function (PSCF) was used to determine the locations of remote sources that influenced the atmospheric air content in

the southern basin of the lake in summer 2023. The most probable contribution was made by the sources located in the Republic of Buryatia and Krasnoyarsk Krai.

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Conflict of interest

The authors declare no conflicts of interest.

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