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Articles

Sulfur content in needles of cedar (Pinus sibirica Du Tour) and siberian fir (Abies sibirica Ledeb.) of the Southern Baikal Region: Influence of industrial emissions

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The paper presents the results of studying the sulfur content in the needles of cedar and Siberian fir of the Southern Baikal Region, carried out in the 2014–2015 period. The research aims to determine whether Baikal Pulp and Paper Mill (BPPM) are the primary stress factor of abiotic nature, which leads to the degradation of cedar and Siberian fir stands in the Southern Baikal Region. The authors have determined that the primary factor is the long-term impact of the emission of pollutants from heat power companies in the Irkutsk Region, as indicated by numerous studies using mathematical modeling methods. The research demonstrates that the assimilation organs of the cedar are more sensitive to the effects of sulfur dioxide compared to the needles of Siberian fir. Therefore, cedar needles can be considered as a more sensitive indicator of atmospheric pollution with sulfur dioxide. The authors have established that the primary source of contamination of Siberian fir needles is the SO_2 emission from the heat and power enterprises of the Irkutsk Region and not the emission of sulfur-containing compounds of the BPPM. The authors found that in the period from 2010 to 2019, the total SO_2 emissions from large industrial companies were an order of magnitude lower than the volumes of SO_2 emissions from heat power companies in the Irkutsk Region. These emissions will provoke a further deterioration in the state of the boreal forests of the Southern Baikal Region.

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Keywords

emission of pollutants, cedar needles, Siberian fir, sulfur dioxide, Southern Baikal Region, heat power companies

Introduction

Environmental pollution is one of the most discussed issues of humanity. Over the past few decades, human activity has led to an increase in the concentration of carbon dioxide in the atmosphere and pollutants such as sulfur dioxide, nitrogen oxides, and heavy metals. This factor has caused the deterioration of the forests, including boreal ones. Gradual changes in the structure of forests and their species composition are recorded, as well as an increase in the mortality of forest stands (Etzold et al. 2019), their desiccation, defoliation (Augustaitis, Augustaitiene and Deltuvas 2007), and a decrease in radial growth (Savva and Berninger 2010). The authors believe that this condition is caused by the impact of a combination of various stress factors of abiotic and biotic nature.

Intense drying of Siberian fir is also observed in the mountain forests of Siberia, for example, in the Khamar-Daban Mountains, which belong to the Baikal Natural Territory (BNT) (Ayurzhanaev et al. 2016). Part of the BNT is a UNESCO World Natural Heritage Site and is protected by the Federal Law No. 94-FZ “On the protection of Lake Baikal” (Russian Federation 1999) (Fig. 1).

Since the mid-1970s, the area of damaged forests on the Eastern Khamar-Daban Ridge (Central Zone – I, Fig. 1) has been 55%, 25% of which are Siberian fir forests (Bazhina 2010; Voronin 1989).

The authors assume that the primary factor of these changes is atmospheric pollution by emissions from the BPPM (indicated by 4 in Fig. 1). It was put into operation in 1966. The primary components of air pollutants are sulfur dioxide (63%), nitrogen oxides (12%), carbon compounds (22.4%), as well as hydrogen sulfide, methyl mercaptan, furfural, methanol, ammonia, heavy metals (Tret'yakova, Zubareva and Bazhina 1996; Voronin 1989; Voronin and Sokov 2005).

The production activity of the BPPM was halted entirely in 2013. The environmental situation in the suburbs of Baykalsk has slightly improved. Nonetheless, a coal-fired heating station of the former BPPM, operating on coal and fuel oil, continues to function, polluting Baykalsk and the suburban area.

Studies in this area, which have been carried out after the closure of the BPPM (Belykh et al. 2015; Kharuk, Im, Petrov and Yagunov 2016), note a deterioration in the state of forests – there has been mass desiccation of dark coniferous forests in the mountains of Eastern Khamar-Daban. The etiology has not yet been established. One can assume that a decrease in atmospheric moisture level in recent years may cause an increase in the adverse effect of insect pests, micromycete fungi, and bacteria in weakened forest areas (Voronin 2018).

In addition to an increase in water stress and an increase in average annual temperatures, the anthropogenic load on BNT continues to increase from the Irkutsk industrial hub: Usolye-Sibirskoye, Angarsk, Shelekhov, Irkutsk (Fig. 1, III — zone of atmospheric influence, marked in red) (Vologzhina and Akhtimankina 2013). The transport of pollutants is primarily since industrial centers are located on the line of prevailing winds directed towards Lake Baikal (Arguchintsev and Arguchintseva 2007).

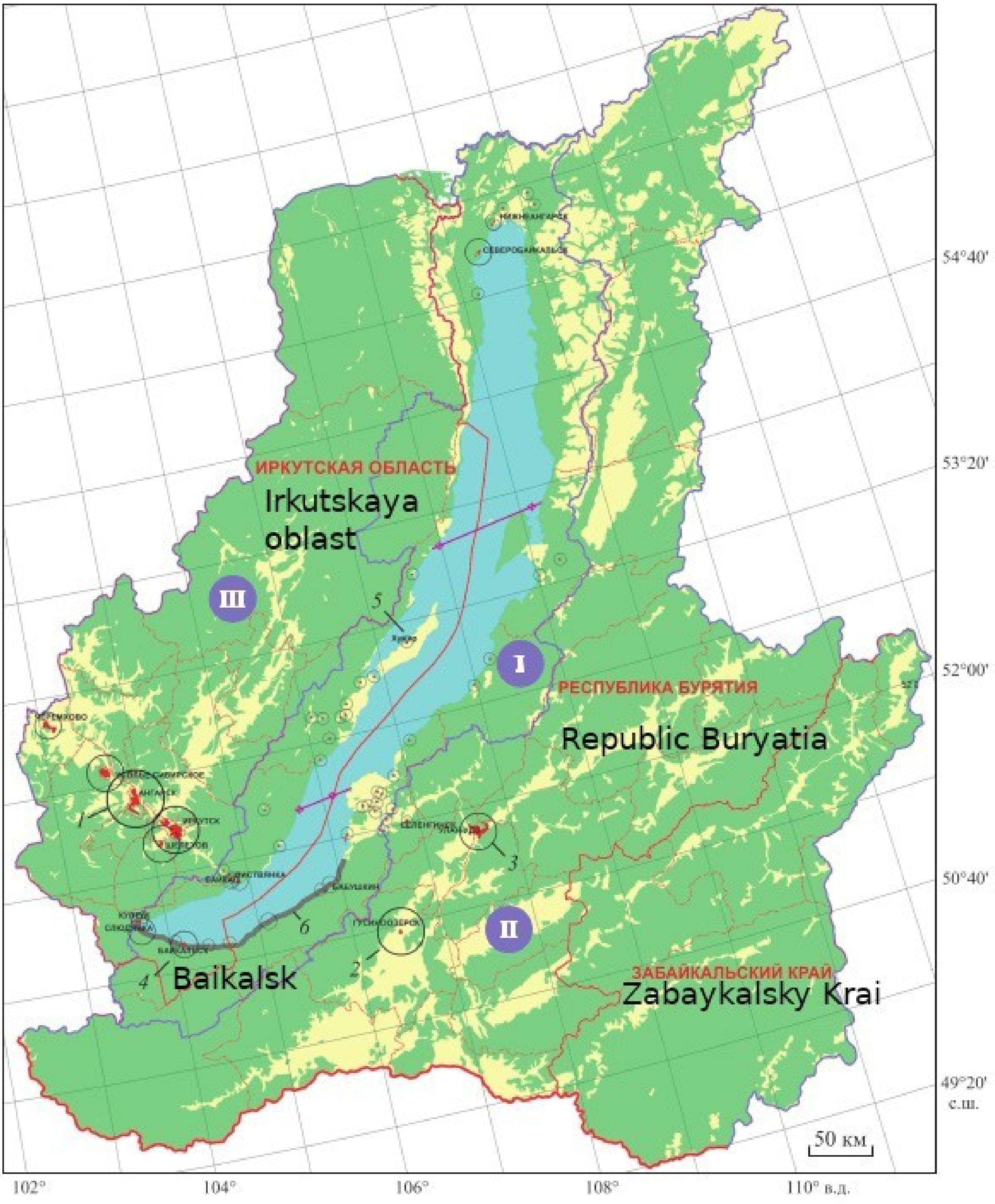

Figure 1. *The Baikal Natural Territory. I – Central Zone, II – Buffer Zone, III – Atmospheric Influence Zone. Sources of pollutant emissions with a volume of: 1 – more than 70 thousand tons/year; 2 – from 35 to 70 thousand tons/year; 3 – from 6 to 35 thousand tons/ year; 4 – from 0.5 to 6.5 thousand tons/year; 6 – section of the federal highway Irkutsk– Ulan-Ude. Source: Ayurzhanaev et al. 2016.*

Recent research (Safarov, Verkhozina and Makukhin 2013; Makukhin, Obolkin, Potemkin, Latysheva and Khodzher 2016; Obolkin, Potemkin, Makukhin, Khodzher and Chipanina 2017) is increasingly persuading scientists to believe that the atmospheric pollution of Lake Baikal may be associated with the periodic influence of plumes of atmospheric emissions from combined heat and power (CHP) plants located around large cities in the valleys of the Angara and Selenga rivers

(Irkutsk, Angarsk, Ulan-Ude, Gusinoozersk). The primary pollutants are sulfur and nitrogen dioxides.

Obolkin V. A. demonstrates that plumes of atmospheric emissions from CHP can be captured by jet currents at an altitude of 200–600 m above and transported hundreds of kilometers without scattering (Obolkin, Shamanskii, Khodzher and Falits 2019). Simultaneously, SO_2 is transported almost without chemical transformation to Southern Baikal and can have a toxic effect on the boreal forests of the northern slope of the Khamar-Daban Ridge. The SO_2 content in the ambient air may exceed the maximum permissible concentration. For example, during the winter of 2014, SO_2 concentrations exceeded 1000 micrograms/m^3 (Obolkin et al. 2019).

Therefore, one should continue studying the mechanisms and effects of toxic effects of pollutant emissions on the boreal forests of the Southern Baikal Region.

The research aims at determining whether the primary stress factor of the abiotic nature for the forests of the Southern Baikal Region is the aero-industrial emissions directly from the BPPM, as many scientists have repeatedly stated, or the primary factor in the suppression of conifers is the long-term impact of the emission of pollutants by industrial companies of the Irkutsk Region, which are in the zone of atmospheric influence III (Fig. 1).

Therefore, the paper identifies the following tasks: assessing the level of contamination of the gray needles of cedar and fir in the Southern Baikal Region after the BPPM shutdown and the volume of sulfur dioxide emissions by industrial companies in the Irkutsk Region.

Materials and methods

The research has been carried out on the territory of the Lake Baikal basin in dark coniferous forest biocenoses of the Southern Baikal Region (Eastern Sayan) at an altitude of 400–1000 m above sea level at a different distance from the BPPM: from 0.6 to 20 km (Fig. 2).

The climate is extremely continental. Mountain dark coniferous forests of the Northern macroslope of the Khamar-Daban are not currently experiencing operational loads.

The research object is the needles of Siberian cedar (*Pinus Sibirica* Du Tour) and Siberian fir (*Abies sibirica* Ledeb.).

Research samples include eight test sites (Fig. 2). The authors have collected 2-year-old cedar needles from healthy trees from the lower part of the crown (vegetative layer). Needle samples were collected in the period from June 20–26 and October 10, 2014; again – June 22–26, 2015.

The analysis material has been taken from the lower part of the crown in four to five whorls. Needles of the second year of life are used for the analysis. Needle samples are not washed before the analysis. They are dried in a room without access to the sun to an air-dry state.

Samples of needles weighing 200-350 g are placed on a sheet of parchment paper, thoroughly mixed, and an average sample is taken by quartering. Next, the selected needles are crushed, and a fraction of fewer than 100 microns is selected.

The non-destructive method of X-ray fluorescence analysis for plants has been developed at the Institute of Geochemistry of the Siberian Branch of the Russian Academy of Sciences (Irkutsk, Russia) (Chuparina and Martynov 2011). It does not require the processing of samples by chemical substances and temperature. Finely ground plant sample material (less than 100 microns) is pressed into a tablet emitter on a boric acid substrate. The needles sample mass is 1 g. The sulfur content in the needles is determined by X-ray fluorescence analysis on an S4 Pioneer wave spectrophotometer (Bruker AXS, Germany). The error characterizing the convergence of the results of X-ray fluorescence analysis does not exceed 5% rel.

Figure 2. *Sampling sites of the cedar needles: 1 – BPPM industrial zone; 2 – Baykalsk, the area between the railway and the park; 3 – Baykalsk, Yuzhny district, residential area; 4 – Mount Sobolinaya (foot); 5 – Mount Sobolinaya (peak); 6 – Overgrown with vegetation waste collection BPPM; 7 – Utulikskaya dacha; 8 – Areas for the reproduction of seeds of forest species.*

Results

Table 1 presents the total sulfur content in cedar and Siberian fir needles in the 2014–2015 period.

Sampling site number	Sampling site characteristics	Sampling point coordinates	Distance to BPPM, in km	Sulfur content, in mg·kg-1			
				Cedar		Siberian fir	
				2014	2015	2014	2015
(1)	BPPM industrial zone	N: 51° 52′07″, E: 104° 17′52″	0.6	1.570 ± 20	1.560 ± 20.0	n.d.	1.020 ± 15
(2)	Baykalsk, the area between the railway and the park	N: 51° 51′24″, E: 104° 14′12″	2.6	1.515 ± 25	1.630 ± 30.0	n.d.	n.d.
(3)	Baykalsk, Yuzhny district, residential area	N: 51° 43′24″, E: 104° 06′12″	3,0	1.520 ± 25 1.550 ± 30*	1.530 ± 20	n.d.	1.230±15
(4)	Mount Sobolinaya (foot)	N: 51° 29′56″, E: 104° 06′8″	4.3	1.513 ± 63 1.835 ± 75*	1.380 ± 20	1.450± 15	860 ± 10
(5)	Mount Sobolinaya (peak)	N: 51° 91′34″ E: 104° 06′27″	5.5	1.465 ± 25	1.330 ± 20	1.315 ± 20	1.010 ± 15
(6)	Overgrown lignin storage	N: 50° 31′15″, E: 104°	7.2	1.350 ± 20	n.d.	n.d.	n.d.

	cards of the BPPM (card number 12)	10′27″, 447 m					
(7)	Utulikskaya dacha, quarter 23	N: 51° 54′82″, E: 104° 07′76″	8.7	1.545 ± 19	1.530 ± 20	1.290 ± 20	1.180 ± 20
(8)	Areas for the reproduction of seeds of forest species	N: 51° 59′69″, E: 103° 91′28″	20.5	1.400 ± 20 1.410 ± 20*	n.d.	890 ± 10	710 ± 10

Table 1. *Total sulfur content in cedar and Siberian fir needles, calculated on dry matter.* Note*: The figure above the line is the sulfur content in cedar needles on June 20, 2014; below the line is the sulfur content in cedar needles on October 10, 2014.Source: Compiled by the authors.

The data analysis on the total sulfur content in cedar needles allows the authors to conclude that there is no evident relationship between the sampling sites distance from the BPPM and the concentration of sulfur in the assimilation organs.

One should note that the concentration of sulfur in Siberian fir needles at all test sites in 2014 and 2015 is significantly lower than in cedar needles. The range of differences is 0.0320–0.0540 mg.kg^{-1}. It indicates more powerful assimilation of sulfur from the atmosphere by cedar needles.

In 2014, the highest concentration of sulfur in cedar needles (from 0.1513 to 0.1570 mg.kg^{-1}) was observed at sites near the BPPM (1), in Baikalsk (2)-(3), at the foot of Mount Sobolinaya (4), and a distance of 8.7 km from the BPPM (7). The lowest concentration is found at 7.2 km from the BPPM on the overgrown lignin storage cards of the BPPM. Analysis of needle samples collected three months later, in October 2014, demonstrates that there has been an accumulation of sulfur in the cedar assimilation organs. The most significant increase in the sulfur content in the needles is observed at the site (4) at the foot of Mount Sobolinaya. This increase is 0.0322 mg.kg^{-1}.

In 2015, the sulfur content in the cedar assimilation organs (taking into account the estimation error) practically did not change at sites (1), (3), and (7). At sites (4) and (5), the sulfur content in needles decreased: by 0.0133 and 0.0135 mg.kg^{-1}, respectively. However, at the site adjacent to the railway (2), the total sulfur content in cedar needles increased by 0.0115 mg.kg^{-1}.

The highest concentration of sulfur in fir needles in 2014 was observed at the foot of Mount Sobolinaya — at the site (4), and the lowest — 20.5 km from the BPPM at the site (8).

In 2015, there was a decrease in the concentration of sulfur in fir needles by 0.0590, 0.0305, 0.0110, and 0.0180 mgkg^{-1} at sites (4), (5), (7) and (8), respectively.

The level of contamination of cedar needles under the influence of atmospheric emissions can be estimated using the sulfur concentration factor in the needles (K_c) according to the method (Saet et al. 1990). The coefficient (K_c) is calculated as the ratio of the element content in the studied object under technogenic load (C) to its background content (C_f). The element content in the sample is considered anomalous if K_c 1, with K_c = 0.8-1.2 (background level); K_c = 1.3–1.4 (weak level of contamination); K_c = 1.5–1.9 (average level of contamination); K_c = 2.0–2.8 (strong level of contamination).

Bazhina Yu. V., Storozhev V. P., Tretyakov I. N. (Bazhina, Storozhev and Tretyakov 2013) take the indicator 1100 mgkg^{-1} as the background value of sulfur concentration in cedar needles. (These data are obtained for cedar needles (Sanina, Filippova, Yurkova and Chuparina 2008) on the territory of the Pribaikalsky National Park (near Olkhon). According to various parameters, including the state of coniferous stands, this territory is characterized as background). Table 2 depicts the results of the K_c assessment.

Sampling site number	Distance to BPPM, in km	Value of Kc	
		2014	2015
(1)	0.6	1.42	1.42
(2)	2.6	1.38	1.48
(3)	3.0	1.38/1.40	1.39
(4)	4.3	1.38/1.67	1.25
(5)	5.5	1.33	1.21
(6)	7.2	1.23	n.d.
(7)	8.7	1.40	1.39
(8)	20.5	1.27/1.28	1.12

Table 2. *Value of the sulfur concentration coefficient in cedar needles.* Source: Compiled by the authors.

All values of K_c in cedar needles in 2014 ranged from 1.2 to 1.4. It indicates a low level of sulfur contamination of cedar needles. In October 2014, at the site (4), an average pollution level was observed (K_c = 1.67). In 2015, the K_c values remained practically unchanged. The background level of pollution was detected only at the site (8).

Therefore, if one judges by the results obtained for fir needles (Tab. 1), they, at first glance, indicate an improvement in the environmental situation in the Southern Baikal Region, which occurred two years after the termination of the BPPM. On the other hand, the results that have been obtained for cedar needles do not provide such a straightforward answer.

Discussion

The most significant number of studies of sulfur content in the coniferous trees of the Southern Baikal Region is devoted to Siberian fir. Several studies show that the most significant damage to Siberian fir occurs in dark coniferous forests of the north-eastern macroslope of the Khamar-Daban Ridge, where the proportion of weakened trees reaches 60%–100% (Massel, Shvets and Seredkova 1989; Tret’yakova et al. 1996; Voronin 1989; Voronin and Sokov 2005).

According to Voronin V., air pollutants under the influence of diffuse processes and local circulations concentrate in the foothills of the northern macroslope of the Khamar-Daban Ridge (Voronin 1989). Fogs form under these conditions, which are a source of condensation of toxic substances, the primary component of which is considered to be sulfur dioxide. In places where the weakening and drying out of Siberian fir forests is observed, samples of Siberian fir needles are taken for chemical analysis (Voronin and Sokov 2005). The assessment demonstrates the sulfur accumulation in the amount of 2400–5220 $mg.kg^{-1}$ of the arid mass. It has significantly exceeded its natural content in gymnosperm needles (650–1800 $mg.kg^{-1}$). Batraeva A. L. (Batraeva 1990) believes that the sulfur content in the needles of Siberian fir is more than 1200 $mg.kg^{-1}$ is an indicator that shows air pollution with sulfur dioxide.

Studies present the analysis of sulfur content in the needles of Siberian fir during the period of BPPM activity from 1991 to 1993 at an altitude of 400–500 m above sea level near the shores of Lake Baikal at different distances from the BPPM: 3–5 km, 10–15 km, as well as at a distance of 70–75 km (valley of the Pereemnaya River), 115–120 km (valley of the Bolshaya Ivanovka River), and 125-130 km (near Babushkin) (Tret’yakova et al.1996). For research, the needles of the upper, middle, and lower parts of the crown are taken. The mentioned data suggests that the highest sulfur content is found in the needles of trees growing in the highlands of Khamar-Daban at a distance of 130–135 km from BPPM. Variations in sulfur concentration in fir needles on average over the crown in the high mountains are 1020–1510 $mg.kg^{-1}$ and on the Baikal coast (low mountains) — from 730 to 1160 $mg.kg^{-1}$.

Table 3 depicts the studies of Siberian fir needles in the lower part of the crown, carried out by the authors in 2014–2015 and the data obtained 23–25 years ago (Tret'yakova et.al. 1996).

Distance to BPPM, in km	Total sulfur content calculated on dry matter, in mg.kg-1	
	1990–1993*	2014–2015 (Table 1)**
0.6	n.d.	n.d. / 1.020 ± 15
3–5	1.000 ± 801.160 ± 20910 ± 20	1.450 ± 15 / 1.230 ±151.315 ± 20 / 860 ± 10n.d. / 1.010 ± 15
10–15	980 ± 30890 ± 40	1.290 ± 20 / 1.180 ± 20
70–75	1.140 ± 101910 ± 30	n.d.

Table 3. *Total sulfur content in Siberian fir needles in the lower part of the crown (research results for 1991–1993 and 2014–2015).* *Source: Tret'yakova et.al. 1996.**Source: Compiled by the authors

The sulfur content in the needles of Siberian fir growing in 1991–1993 at different distances from the BPPM near the coast of the Lake Baikal fluctuated within 890–1160 mg.kg^{-1} (Table 3). According to Batraeva A. L., this amount does not exceed the background level of sulfur in the needles (Batraeva 1990).

The sulfur concentration in Siberian fir needles in 2014 at a distance of 3 to 15 km exceeds the background level in some areas by 20.8%, and in 2015 — by 2.5%. If one compared the maximum sulfur concentration in fir needles in 1991–1993 (1160 mg.kg^{-1}) with its maximum values in 2014–2015, one could note that in 2014 it increased by 25%, and two years after the BPPM shutdown decreased slightly and amounted to 6%.

The K_c value for the maximum sulfur concentration in Siberian fir needles (1450 mg.kg^{-1}) in 2014 does not exceed 1.2. This factor indicates the background level of atmospheric pollution on the Baikal coast. Nevertheless, the study demonstrates that under severe conditions of Siberian fir growth on the Khamar-Daban Ridge, even a slight excess of sulfur concentration in conifers above acceptable values contributes to a decrease in the quality of pollen grains (Tret'yakova et.al. 1996). High sulfur content in Siberian fir needles may have led to irreversible changes in the morphostructure of trees and the drying out of fir stands in the mountains (Massel et al. 1989; Voronin 1989).

Therefore, the authors make the following conclusion: during the activity of the BPPM, the level of toxicant accumulation in Siberian fir needles is significantly lower than after 21–24 years.

Since the sulfur content in the cedar needles in the southern Baikal Region has not been studied, the authors cannot make such comparisons. However, the Table 1 and Table 2 data indicate a greater degree of sulfur assimilation by the needles of the cedar in comparison with the Siberian fir needles. Unlike the value K_c = 1.2 for Siberian fir with the highest level of sulfur concentration in 2014, the K_c values for cedar needles indicate not background but weak and medium levels of sulfur contamination of cedar needles. Consequently, cedar needles can be considered as a more sensitive indicator of atmospheric pollution with sulfur dioxide.

In the last decade, in the Region of South Baikal, there have been massive infections by bacterial dropsy, drying out, and death of cedar forests, and the Siberian fir has been simultaneously affected (Cherpakov 2015). Due to poor knowledge and the practical absence of phytobacteriological studies in the forestry of Russia, it is difficult to give the real reasons for these processes in the coniferous forests of the Southern Baikal Region. Nevertheless, one should consider the presence of a possible relationship between the accumulation of pollutants in the needles of cedar and Siberian fir and a decrease in the resistance of conifers to diseases, for example, bacterial dropsy.

To analyze the possible mechanisms and sources of sulfur dioxide transport from the primary regional sources to the Southern Baikal Region, scientists have carried out numerous experiments using a mathematical model (Arguchintsev and Makukhin 1996). Studies demonstrate that the primary contribution to the pollution of the South Baikal territory makes CHP of Irkutsk and Angarsk, whereas the influence of local CHP (Slyudyanka, Baikalsk) is limited to the effect on adjacent areas and do not apply to the entire southern part of Lake Baikal. Therefore, the paper serves as a direct empirical confirmation of the research (Arguchintsev and Makukhin 1996; Makukhin et al. 2016; Obolkin et al. 2016; Obolkin et al. 2014; Obolkin et al. 2017; Obolkin et al. 2019; Safarov et al. 2013;).

Fig. 3 depicts the dynamics of sulfur dioxide emissions for the 2010–2019 period.

Recently, the Irkutsk Region has been an outsider in the environmental rating of Russian regions (Green Patrol 2020). A comprehensive air pollution index is utilized as one of the criteria for compiling the ranking. According to the value of this index, the Irkutsk Region ranks at the bottom of this rating.

In the 2010–2019 period, the total emissions of sulfur dioxide from heat power companies in the Irkutsk Region remain at a very high level (Fig. 3) and, on average, amount to 188.4 thousand tons per year. The total emission of other large industrial companies of the Irkutsk Region, on average, is 18.12 thousand tons. It is an order of magnitude lower than the volume of emissions from heat power companies.

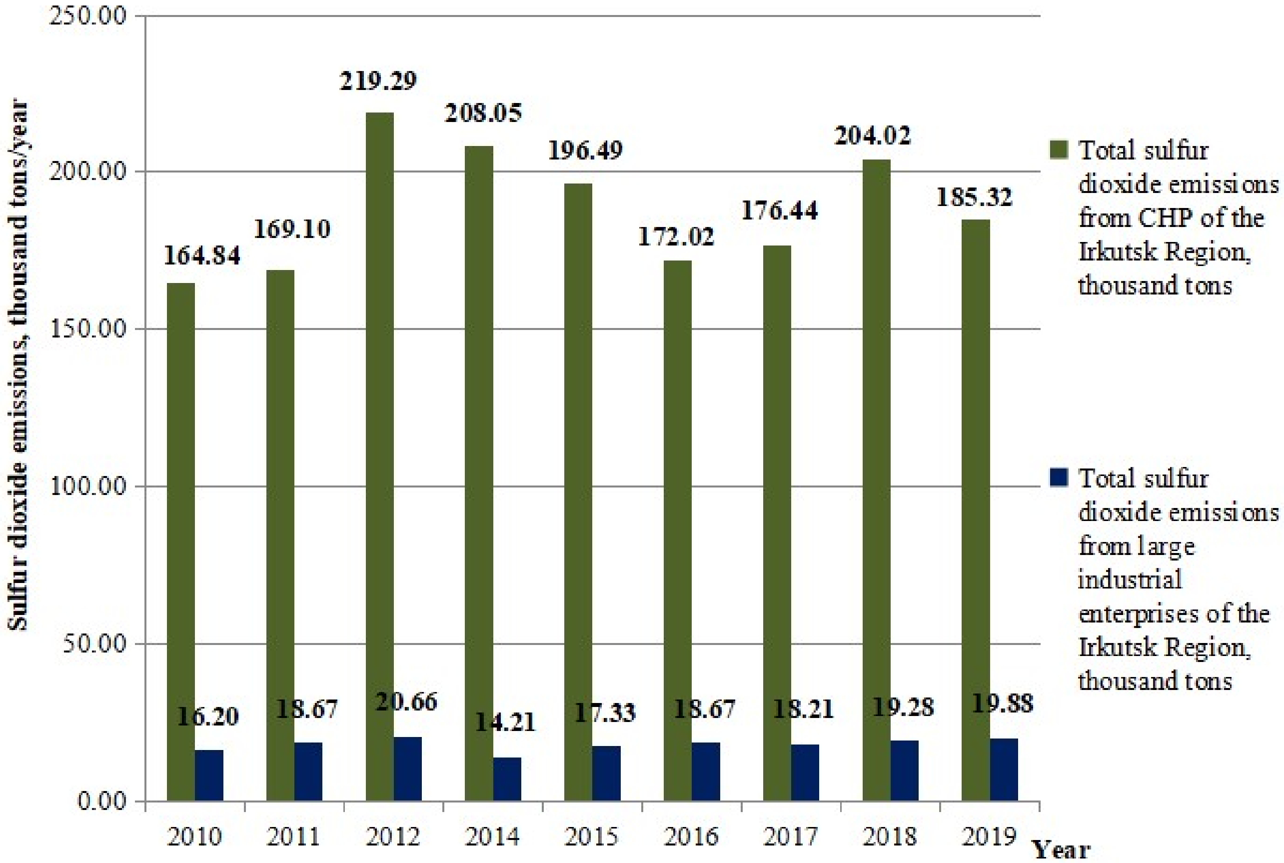

Figure 3. *Dynamics of sulfur dioxide (SO_2) emissions into the atmosphere from CHP and other industrial facilities of the Irkutsk Region.*

Therefore, based on the data on the total sulfur content in the needles of cedar and Siberian fir, one can assume that the primary air pollutant is not so much the emissions of BPPM as the emissions of pollutants from industrial companies that are located in the zone of atmospheric influence (III) (Fig. 1). These are large companies in the heat power industry.

Conclusion

Studies of the sulfur content in the needles of the cedar and Siberian fir on the territory of the Southern Baikal Region show that the cedar assimilation organs are more sensitive to the effects of sulfur dioxide in comparison with the Siberian fir needles. It can be used to study the ecological state of the stand of boreal forests. Cedar needles can be considered a more sensitive indicator of sulfur dioxide pollution in the atmosphere.

The authors find that during the period of BPPM activity, the level of sulfur accumulation in Siberian fir needles is significantly lower than after more than 20 years. This factor suggests that the pollution of the atmosphere of Lake Baikal may be associated with the periodic influence of plumes of atmospheric emissions of SO_2 from heat power plants located around prominent cities in the valleys of Angara and Selenga rivers. It is confirmed by the fact that in the period from 2010 to 2019, the total emissions of sulfur dioxide from substantial industrial companies are an order of magnitude lower than the volumes of SO_2 emissions from heat and power companies in the Irkutsk Region.

One should investigate the presence of the relationship between the effect of sulfur compounds accumulating in the needles of cedar and fir and the resistance of conifers to bacterial diseases.

Therefore, one of the possible solutions to the issue of reducing the environmental burden and preserving the boreal forests of the Southern Baikal region should be considered the transition to other heat carriers of heat power plants, such as natural gas.

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