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Estimation of the influx of pollutants to the territory of the South Pribaykalye

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ABSTRACT. We present the results of long-term (2010-2017) studies of chemical composition of snow cover from the area of the Baikal Region. There is significant pollution of snow cover in the industrial cities of Pribaykalye (Irkutsk and Shelekhov). Some areas are the most subjected to the atmospheric pollution. The emissions from Shelekhov enterprises influence the chemical composition of snow cover in Irkutsk. Transport, fuel and energy complex mainly influence the formation of the chemical composition in snow cover from Irkutsk. In Shelekhov, the source of contaminants is the aluminium smelter (RUSAL-IrkAZ) and power engineering. Based on the data on snow surveys, we estimated the influx of pollutants from the atmosphere to the underlying surface of industrial cities and the water area of South Baikal.

Keywords: industrial cities, Lake Baikal, snow cover, chemical element concentrations, accumulations, influx from the atmosphere

1. Introduction

According to Voeikov Main Geophysical Observatory, in 2018, the Priority list of Russian cities with the highest level of atmospheric air pollution contained seven cities of the Irkutsk Region, including Irkutsk and Shelekhov. A large amount of contaminants enters the atmosphere from heat power enterprises. Snow cover summarizes emissions for the entire winter period and is an integral indicator of atmospheric pollution (Vasilenko et al., 1985; Lomonosov et al., 1993; Chebykin et al., 2018; Paradina et al., 2019). After melting of the snow cover, the pollutants accumulated over the entire winter period enter the soil and water bodies. Assessment of their influx from the atmosphere with precipitation and dry deposition allows for the calculation of ecological risks to the environment. Under the decision of the UN Economic Commission for Europe, the group of the most hazardous heavy metals includes mercury, lead, cadmium, chromium, manganese, nickel, cobalt, vanadium, copper, iron, zinc, antimony as well as typical metalloids: arsenic and selenium. Of special hazard are metals that are not part of biomolecules, i.e. xenobiotics: mercury, cadmium and lead (Bashkin and Kasimov, 2004).

Almost all industrial production, fossil fuel combustion, transport and other human activities cause anthropogenic dispersion of elements and heavy metals in the environment (Kleeman et al., 2008; Wiseman and Zereini, 2009). Fuel combustion leads to the atmospheric pollution by As, Cr, Cu, Mn, Ni, Sb, Se, V, and Zn; non-ferrous metallurgy – by Al, Ag, As, Cd, Cu, Ni, Pb, Sb, and Zn; ferrous metallurgy – by Cd, Fe, Mn, Ni, Pb, and V (Saet et al., 1990; Berg et al., 2008; Pacyna et al., 2009).

Studies of the chemistry of snow cover in the Irkutsk Region enabled evaluation of the element fluxes from the atmosphere in the Baikal Natural Territory (Anokhin et al., 1981; Kokorin and Politov, 1991; Vetrov and Kuznetsova, 1997; Koroleva et al., 1999; Obolkin et al., 2004; Khodzher, 2005; Onischuk et al., 2012). Geochemical assessment of the state of snow cover in the cities of the Irkutsk Region revealed areas that are most subjected to atmospheric pollution (Lomonosov et al., 1993; Belozertseva, 1999).

The aim of the work is to assess the annual influx of pollutants from the atmosphere to the underlying surface in the cities of Irkutsk and Shelekhov as well as to the water area of Southern Baikal in the modern period.

2. Materials and methods

Snow cover was sampled for its entire height at the end of winter, in February and March. In the water area of Lake Baikal, snow was sampled from the ice in Southern Baikal, being the most subjected to the anthropogenic impact (Fig. 1, Table 1). Fluxes at the

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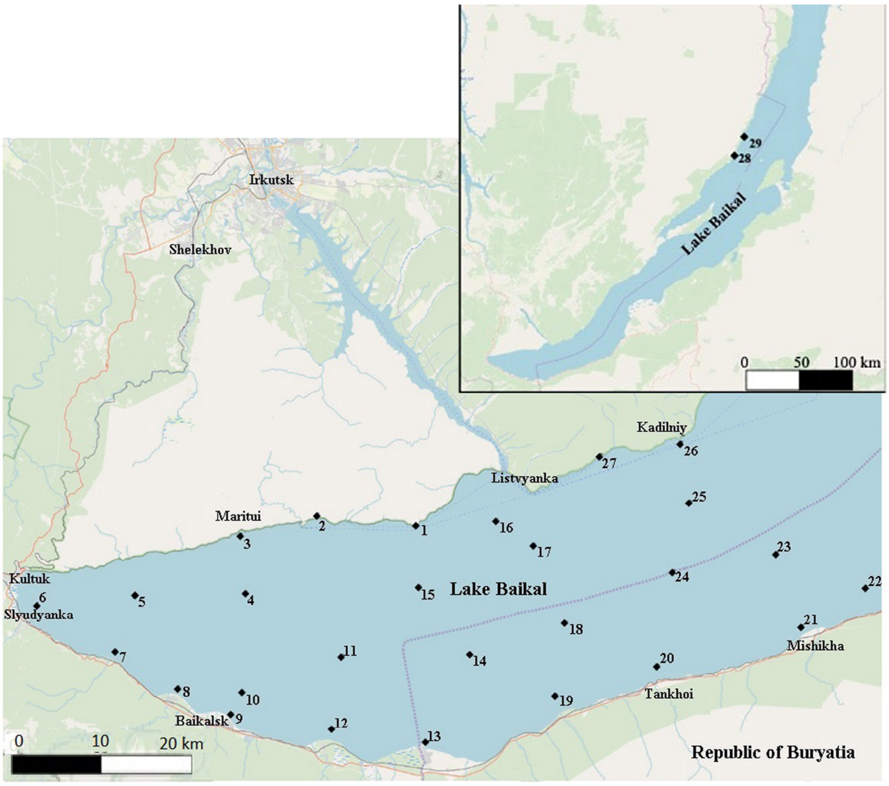

Fig. 1. Map of the sampling locations.

underlying surface were calculated from the averaged values between 2010 and 2015. In the industrial centres, Irkutsk and Shelekhov, located 70-80 km from the Baikal coast, samples were taken episodically from 2000 to 2013 at 20 sampling sites in each city. Snow cover samples were taken at a distance 500 m from the main highways. We took 99 snow cover samples from Shelekhov, 111 samples from Irkutsk and 90 samples from Southern Baikal. The chemical composition of snow cover sampled from the ice of Lake Baikal near the capes Onkholoy and Bolshoi Solontsovy (Baikal-Lena Nature Reserve (BLR), northwest coast of the lake) served as the background. To calculate the accumulation, we used averaged concentrations and moisture content in the snow cover. The moisture content (cm) was calculated as the ratio of the volume of the melted sample (cm^3) of the snow cover to the area (cm^2) from which it was collected.

Chemical analysis of snow water was carried out in the Laboratory of Hydrochemistry and Atmosphere Chemistry (Accreditation certificate No. ROSS RU.0001.513855) at Limnological Institute SB RAS. Concentrations of sulphates and nitrates were determined by ion chromatography (ICS-3000, Dionex, USA) and high-performance liquid chromatograph with UV detection (Milichrome A-02, Ekonova, Russia) (Khodzher et al., 2016). We used standard solutions from the "Kanto Chemical Co" (Japan) and "Ultra Scientific" (USA). Solids of snow water were investigated by the X-ray fluorescence technique using synchrotron radiation (SXRF) at the Institute of Nuclear Physics SB RAS. The element concentrations of water phase were determined on an Agilent 7500 ce quadrupole mass spectrometer in the Ultramicroanalysis Center at the Institute of Limnology SB RAS. The samples were supplied using a microflow sprayer (0.3 mL/min). The instrument was calibrated using a high purity standard solutions ICP-MS-68A-A-100 and ICP-MS-68A-B-100 (1, 2, 5, 10, 25 ppb of each element). The drift of the instrument was monitored using an In internal standard and a control sample (a standard solution with 5 ppb of each element), which was measured in ten sample intervals. Based on the geometric estimation of the spatial distribution of elements, we constructed maps illustrating accumulation of some metals in snow cover. The maps show the total flux of soluble and insoluble forms of metals.

3. Results and discussion

3.1 The Irkutsk city

The study of the chemical composition of snow cover in Irkutsk indicated the highest values of all analysed components near Glazkovo bridge, the Eternal Flame (the Kirov Square), the Paris Commune Park, the Ushakovka River (Rabocheye suburb), Novo-Lenino (area of Cinema college), and Marata suburb.

The most part of these areas is located in low reliefs along the valleys of the rivers Irkut and Angara where meteorological conditions are extremely unfavourable for the dispersion of air pollutants due to low wind speeds typical of lowlands. The above areas experience significant anthropogenic pressure from the industrial objects and roads located here (Novikova, 2015). They are also affected by impurities coming with airflow, which is due to local circulation processes, from Shelekhov along the valley of the Irkut River as well as from Angarsk along the valley of the Angara River.

Table 1. The sampling stations on South Baikal

№	Sampling stations	The coordinates	
		northern latitude	eastern longitude
1	Near the Cape Tolstiy	51°47'36,0"	104°36'62,0"
2	Near the Cape Polovinniy	51°97'85,0"	104°22'87,9"
3	Opposite the Maritui settlment	51°46'44,1"	104°12'56,7"
4	Center of the section Marituy-Solzan	51°41'40,0"	103°57'87,3"
5	Between Slyudyanka town and Maritui settlment	51°41'40,0"	103°57'87,3"
6	Opposite Slyudyanka town	51°40'48,9"	103°44'53,6"
7	Near the Cape Telegraphiy	51°36'49,6"	103°55'39,2"
8	Opposite the Utulik River	51°33'37,8"	104°04'16,1"
9	Opposite the Baikalsk city	51°31'28,4"	104°11'37,6"
10	3 km from the Baikalsk city	51°31'74,9"	104°12'96,2"
11	Opposite the Khara-Murin River	51°36'22,6"	104°26'46,7"
12	Near the Khara -Murin River	51°30'12,1"	104,24'90,6"
13	Opposite the Snezhnaya River	51°29'05,7"	104°37'83,7"
14	From Snezhnaya River to Listvyanka settlment	51°35'97,0"	104°43'87,6"
15	Section the Snezhnaya - Tolstiy	51°41'80,2"	104°36'87,1"
16	Between the Tolsty and Berezovy capes	51°48'00,1"	104°47'63,9"
17	Section Listvyanka-Tankhoy	51°45'53,6"	104°52'70,7"
18	Listvyanka- Tankhoy section	51°39'16,5"	104°57'33,6"
19	Near the Cape Kedroviy	51°32'64,0"	104°56'09,3"
20	The Pereemnaya River, 1 km from the shore	51°34'93,2"	105°10'11,8"
21	The Ushakovka River, 0.5 km from the shore	51°38'55,3"	105°29'67,1"
22	Opposite the River Klyuevka	51°42'17,1"	105°38'55,6"
23	Section the Kadilniy-Mishikha, 10-15 km from Mishikha River	51°44'68,1"	105°26'34,1"
24	Section the Kadilniy-Mishikha, the center	51°43'34,6"	105°12'25,3"
25	Section the Kadilniy-Mishikha, 10-15 km from the cape Kadilniy	51°48'90,0"	105°13'97,6"
26	Near the Cape Kadilniy	51°54'33,2"	105°13'28,5"
27	Opposite the Fall Chernaya	51°53'26,4"	105°02'16,7"
28	Near the cape Onkholoy (Baikal-Lena Nature Reserve)	53°47'46,7"	107°57'32,5"
29	Near the cape Bolshoi Solontsovy (Baikal-Lena Nature Reserve)	54°10'05,7"	108°21'40,2"

The microdistricts located on elevations, such as Zelyony, Raduzhny, Topkinsky, and Solnechny, showed the lowest levels of chemical ingredients in snow cover. These areas are characterized by both high wind speeds, which is normal for elevated surface, and the absence of large sources of anthropogenic impact. From the obtained concentrations, we calculated accumulation of pollutants in snow cover during the cold season. Figures 2 and 3 show the distribution of total accumulation (soluble and suspended forms) of metals in snow cover from Irkutsk.

Average accumulation of trace elements in the soluble fraction, the most aggressive for the environment, was 219 μg/m^2 for V, which is 161 times higher than in the background area (BLR); the accumulation of zinc, copper, arsenic, and barium was 1240, 290, 34, and 2260 μg/m^2, respectively, which is 42, 12, 1700, and 50 times higher than in BLR. The average excess for 20 elements was 220 times in comparison with BLR.

3.2 The Shelekhov city

In snow cover from Shelekhov, the maximum concentrations of the studied elements were detected in the industrial zone near the Thermal Power Plant - 5 (TPP-5) and the RUSAL-IrkAZ aluminium plant. Thus, high concentrations of Be, B, Ti, V, Ni, and Sr were determined in the southwest direction near TPP-5. The highest concentrations of most elements were identified in a small area near the smelter. The maximum concentration Al (27 mg/L) is recorded in the northwest of Shelekhov, near the fifth line of the smelter. This value is at the level determined in the snow cover of Bratsk, Irkutsk region, at a distance of

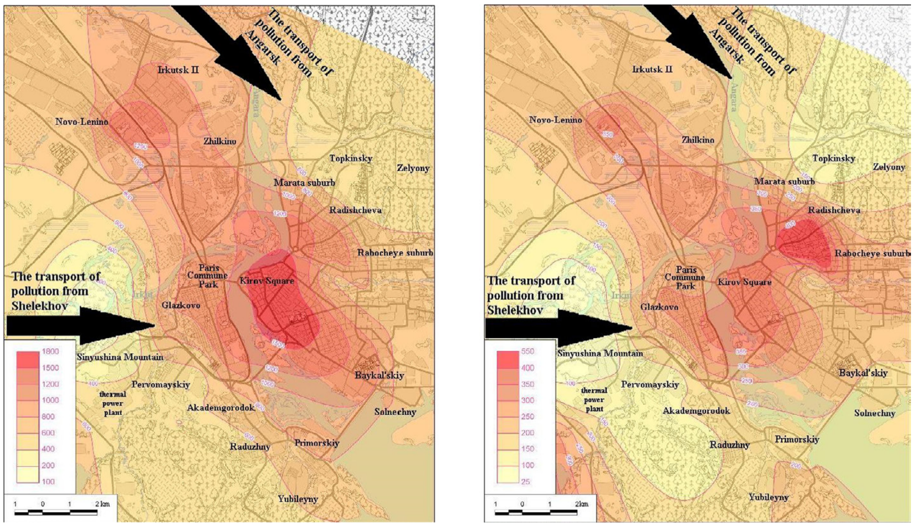

Fig. 2. Copper (left) and lead (right) accumulated in snow cover of Irkutsk, μg/m^2.

0.5 km from the aluminum smelter (20 mg/*L*) and 55 times higher than in Blagoveshchensk, the Far East (Ignatenko et al., 2007; Radomskaya et al., 2018).

Concentrations of Na and F, which are tracers in aluminium production, near the smelter were also high according to our 2013 data, up to 33 and 35 mg/L, respectively. According to (Belozertseva et al., 2015), F concentrations in snow cover were higher, up to 60 mg/L.

The accumulation of elements and their distribution on the earth surface in the modern period is close to that indicated in 1996 (Nechaeva et al., 2004). A large amount of contaminants enters the atmosphere from the TPP-5 located near the aluminium smelter. Figure 4 shows the distribution of copper and lead accumulated in snow cover. The maximums of these elements were recorded near the TPP-5. Atmospheric pollution halos from the emissions of the aluminium smelter and TPP-5 extend to the housing estate in Shelekhov. In the residential area of Shelekhov, aluminium concentrations decrease but 4500 times exceed background values (BLR). Industrial emissions from Shelekhov affect the state of the atmosphere in Irkutsk through the transport of contaminants along the valley of the Irkut River (Fig. 4, Fig. 5).

The average concentrations of all soluble elements, except for Al, are higher (up to 4.5 times) in Irkutsk compared to Shelekhov. Aluminium concentrations in Shelekhov is 60 times higher than in Irkutsk.

Atmospheric deposition causes the formation of multi-element pollution halos around cities in the south of the Irkutsk Region. Moreover, the distribution halos of some elements in snow cover from Irkutsk overlap with the distribution fields of elements coming from Shelekhov (Fig. 4, Fig. 5 right). Sources of pollutants typical of large cities, such as transport as well as fuel and energy complex, mainly influence the formation of the chemical composition in snow cover from Irkutsk. In the city of Shelekhov, the aluminium plant makes the main contribution to atmospheric pollution (77 %), the contribution of TPP-5 is much less - 19%.

In the snow cover from Shelekhov, the accumulation of V in the soluble fraction was 90 μg/m^2, which is 2.4 times lower than in the snow cover from Irkutsk and 70 times higher than in BLR. Accumulation

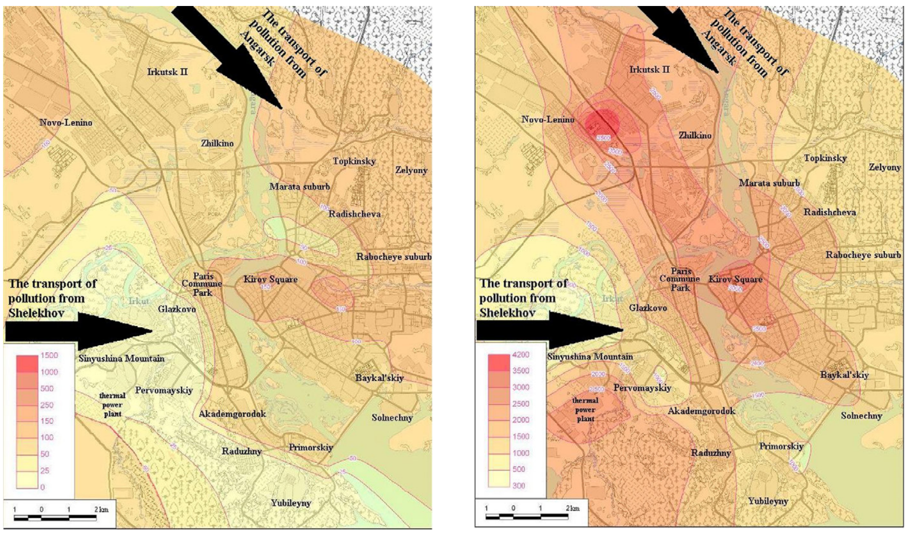

Fig. 3. Aluminium (left) and zinc (right) accumulated in snow cover of Irkutsk, μg/m^2.

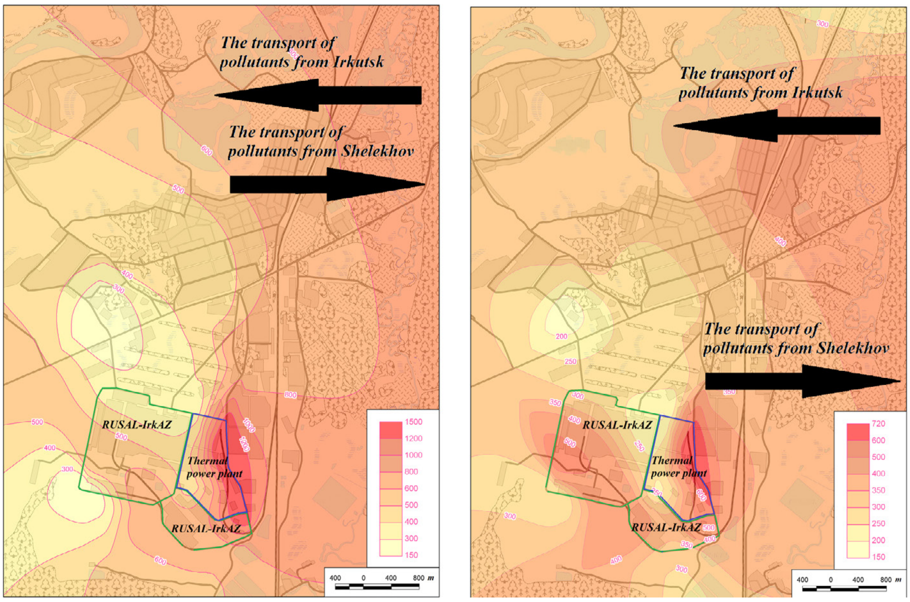

Fig. 4. Copper (left) and lead (right) accumulated in snow cover of Shelekhov, μg/m².

of Zn, Cu and Ba was 570, 100 and 790 μg/m², respectively, which is lower than in the snow cover from Irkutsk. These values are much higher than in the background area, from 5 to 20 times. Accumulation of As in Shelekhov and Irkutsk was the same, with an average of 34 μg/m².

3.3 Snow cover of Lake Baikal

Water in Lake Baikal is low-mineralized, and the content of trace elements in this water is very low (Kulikova et al., 2017). The assessment of the balance of trace elements in the lake should take into account that the significant part comes from the atmosphere (Khodzher, 2005). Due to the prevalence of north-western winds in the region, some part of atmospheric pollution of Irkutsk and Shelekhov can enter the water area of Southern Baikal along the valley of the Angara River. Table 2 shows the concentrations of trace elements, sulphates, nitrates and the total concentration of ions (Σ_i) in snow cover of Irkutsk and Shelekhov as well as on the ice of Southern Baikal. The concentrations of trace elements in snow cover of the cities are one-two orders of magnitude higher than in the lake snow cover.

On Southern Baikal, the maximum concentrations of the studied elements were determined in the sections Kadilny-Mishikha and Listvyanka-Tankhoy. At the central point of the Kadilny-Mishikha section, we determined the maximums of aluminium, iron, nickel, copper, strontium, and barium: 326, 48, 1.6, 3.8, 44, and 20 μg/L, respectively. At the Listvyanka-Tankhoy section, element concentrations were higher than the average value for Southern Baikal (Table 2) and were 203 μg/L for aluminium, 0.87 μg/L for vanadium, 10 μg/L for manganese, 40 μg/L for iron,

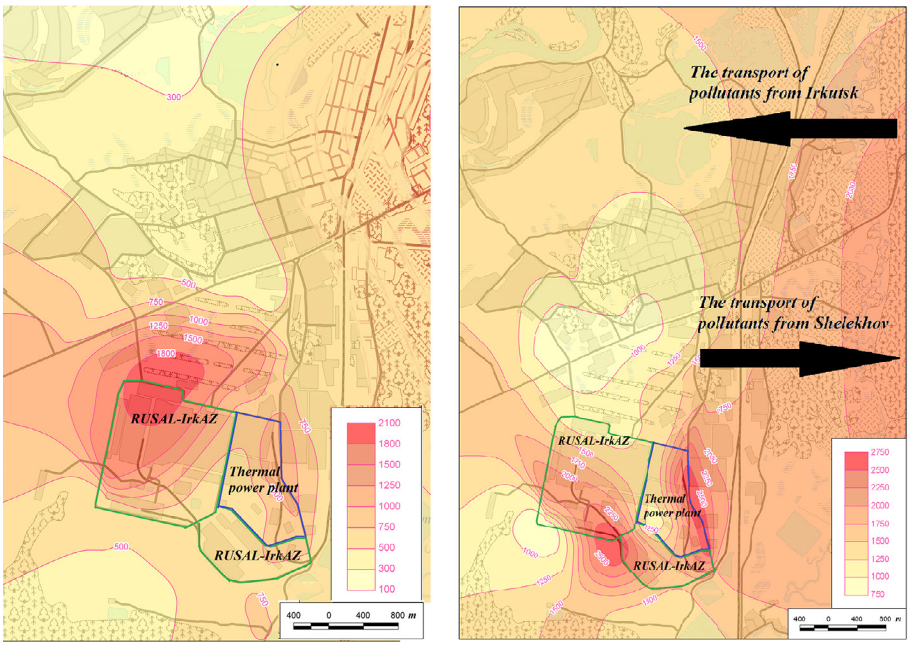

Fig. 5. Aluminium (left) and zinc (right) accumulated in snow cover of Shelekhov, μg/m².

2.1 μg/L for copper, and 41 μg/L for strontium. These are the most likely pathways of transporting air masses from anthropogenic sources located in the influence zone of the Irkutsk-Cheremkhovo industrial hub. High concentrations of the studied elements were also determined on the snow of the lake towns of Baikalsk and Slyudyanka. Currently (the 2017 data), the concentration of sulphates in snow from Baikalsk does not exceed 1.8 mg/L, and Na – 0.18 mg/L, whereas, in 2011, these values were respectively 5 and 15 times higher. The pH values also decreased significantly in the snow cover of the town. In 2011, the maximum pH value reached 8.7 units, and in 2017 – only 6.2 units. At present, Baikalsk and its surroundings show pH values typical of snow cover in the southeast coast of the lake, which is not affected by the large anthropogenic sources, 4.7-5.1 units. However, after the closure of the Baikalsk Pulp and Paper Mill (BPPM) in 2013, the concentrations and accumulation of heavy metals and major ions in snow cover decreased significantly. This indicates a decrease in the anthropogenic pressure on the southeast coast of Lake Baikal.

Comparison of concentrations of the studied elements in snow cover of Southern Baikal and BLR indicated that in the southern basin, element concentrations are six times higher than the background, while Al and Sr show the highest excess in 17 and 14 times, respectively. Considering the wind rose, the snow mostly accumulates on the east coast of the lake; the western part of Lake Baikal is less covered with snow. Therefore, the impact of pollutants from the atmosphere to the east coast is much higher than in the western part of Southern Baikal.

To calculate the influx of elements to surface, we used the data on their average concentrations in snow cover and long-term average annual (2010-2016) amount of atmospheric precipitation in the area of Southern Baikal (the Listvyanka settlement) as well as in Irkutsk and Shelekhov (https://ru.climate-data.org). For Southern Baikal, we calculated fluxes without taking into account the concentrations of sulphates and nitrates in snow cover from Baikalsk and Slyudyanka (Table 3). Comparison of the obtained values showed that amount of sulphates and nitrates entering on the Southern Baikal are respectively 5.5 and 1.5 times lower than ones in the studied industrial cities of the Angara region. However, element fluxes are up to 150 (Al) times higher in the cities. An exception is Pb, whose influx in the area of the Southern Baikal is at the same level as in Irkutsk. Most likely, lead enters the area of Southern Baikal on small aerosol particles due to distant transport from industrial sites. High concentrations of lead were also recorded in other areas of the Earth remote from industrial areas. For example, studies of lacustrine deposits in Antarctica revealed that lead concentrations (obtained from the burning of fossil fuels) have increased significantly, especially over the past 50 years. Alaska shows the same pattern, where lead deposition has become approximately three times higher over the same period (Polar Lakes…, 2008).

Table 3 shows amount of sulphates, nitrates and some trace elements in the water column of Lake Baikal. Estimates of fluxes were carried out based on the concentrations in the cold season; however, in recent years, due to wildfires, a large amount of aerosol matter has been released in the warm season (Khodzher et al., 2019). Therefore, during wildfires accompanied by atmospheric precipitation and dry deposition, more pollutants can enter the water area of the lake.

Table 2. Concentrations of pollutants in the soluble fraction of snow cover in 2013, μg/L

Element	Irkutsk	Shelekhov	Southern Baikal	Baikal-Lena Nature Reserve, background
SO_4^{2-}*	8.0-28 / 13	2.6-27 / 11	0.85-18 / 4.83	0.74
$N\text{-}NO_3^-$*	0.49-0.63 / 0.56	0.38-0.82 / 0.51	0.35-2.7 / 0.85	0.19
Σ_i*	21-124 / 44	9.0-104 / 42	4.3-41 / 12.2	2.0
Al	67-251 / 144	18-26627 / 8446	6.9-326 / 101	5.9
V	3.2-13 / 6.9	0.03-4.2 / 1.6	0.10-1.3 / 0.55	0.08
Cr	1.3-3.7 / 1.9	0.01-16 / 1.3	0.06-1.7 / 0.28	0.08
Mn	27-92 / 62.8	6.86-162 / 36.9	2.6-24 / 6.3	1.2
Co	0.4-2.5 / 1.3	0.01-3.8 / 0.49	0.02-0.67 / 0.24	0.04
Ni	2.3-8.8 / 5.2	0.39-25 / 3.4	0.29-1.6 / 0.64	0.11
Cu	1.6-10 / 6.7	0.25-13 / 1.8	0.39-3.8 / 1.8	0.62
Zn	3.2-32 / 22	0.67-80 / 11	4.2-35 / 13	1.79
As	0.26-1.4 / 0.79	0.06-1.5 / 0.56	0.01-0.19 / 0.02	0.001
Se	0.75-1.5 / 1.2	0.01-2.1 / 0.36	0.01-0.10 / 0.04	0.03
Sr	34-108 / 58	3.0-86 / 21	3.7-44 / 14	0.97
Cd	0.08-0.38 / 0.18	0.001-0.66 / 0.17	0.001-0.16 / 0.03	-
Ba	25-60 / 39	0.14-51 / 11	5.1-20 / 8.3	1.88
Pb	0.12-1.0 / 0.38	0.14-0.98 / 0.29	0.05-1.7 / 0.70	0.17

Note - * mg/L; numerator - the minimum and maximum concentrations, denominator - the average concentration

4. Conclusion

Long-term studies of element composition of snow cover in large industrial cities of the Baikal region (Irkutsk and Shelekhov) revealed that atmospheric anthropogenic deposition leads to the formation of multi-element pollution halos around these cities. The distribution halos of some elements in snow cover

Table 3. The influx of pollutants from the atmosphere on the South Baikal, Irkutsk and Shelekhov in 2010-2016

Element	Southern basin 7381 km^2		Irkutsk	Shelekhov
	mg/m^2/year	tons	mg/m^2/year	mg/m^2/year
SO_4^{2-}	971±199	7165±1469	5124±787	5363±583
NO_3^-	823±72	6073±531	1287±188	1247±97
Al	29±4	221±30	59±13	4294±233
Cr	0.08±0.02	0.6±0.2	0.55±0.36	0.79±0,55
Mn	1.88±0.16	14±1	20±6	21±1
Ni	0.19±0.02	1.4±0.2	2.8±0.7	1.8±2.8
Cu	0.53±0.04	3.9±0.3	2.8±1.0	0.85±0.64
Zn	6.12±0.32	45±2	12±3	3.8±2.4
Pb	0.21±0.02	1.6±0.2	0.20±0.08	0.14±0.13
Cd	0.009±0.002	0.10±0.02	0.06±0.02	0.08±0.03
Ba	2.50±0.20	19±2	21±5	6.2±8.2
V	0.17±0.02	1.2±0.2	2.0±0.8	0.78±0.18
As	0.006±0.003	0.05±0.02	0.30±0.08	0.27±0.05

from Irkutsk overlap with the distribution fields of elements coming from Shelekhov. Transport, as well as enterprises of fuel and energy complex, mainly influences the formation of the chemical composition in snow cover from Irkutsk. The concentrations of most soluble elements, except for aluminium are higher here than in Shelekhov where the main sources of air pollutants are the aluminium smelter and TPP-5. In comparison with the BLR snow cover, the industrial centres show a significant excess (more than 100 times) of the concentrations and accumulation of such elements as aluminium, vanadium, manganese, nickel, arsenic, and strontium.

Concentrations of most trace elements in snow cover from the southern basin of Lake Baikal are on average six times higher than the regional background values. Anthropogenic impurities enter the water area of Southern Baikal from sources located in the area of the Irkutsk-Cheremkhovo industrial hub as well as from local sources located on the coast of Southern Baikal. After the closure of BPPM, the substantial source of pollutants in the atmosphere, there is a tendency to a gradual decrease in concentrations and accumulation of chemical elements in snow cover from the southern part of the lake. Pollutants entering the water area of Southern Baikal are from 1.5 (nitrates) to 150 (Al) times lower than those in Irkutsk and Shelekhov. An exception is a lead, whose fluxes in the area of Southern Baikal and industrial centres are close.

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