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Ionic Composition of Winter Atmospheric Precipitation in the Urban Area (South of West Siberia, Russia)

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ABSTRACT

The work evaluates the urban snow cover pollution and determines the level of the city influence on the pollution of the urban atmosphere with major ions (ammonium, nitrite, nitrate, chloride, sulfate, phosphate, sodium, potassium, magnesium, calcium) during the winter period (on the case study of Barnaul city, 2014-2019). The priority ions that determine the high pollution of the urban atmosphere in winter are (nitrite, chloride, sodium), the sources of which are the exhaust of motor vehicles (nitrite) and the using of anti-ice reagents (chloride, sodium). The study showed an increase of the major ions in the urban snow cover (with the exception of nitrate ion) by more than two times compared with the regional and more than six times with the global natural background. To study the spatial features of the snow cover pollution interpolation surfaces of the spatial distribution of priority ions in the study area were constructed.

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INTRODUCTION

Climate, hydrological cycle, transport of transboundary pollutants, and air quality are considerably affected by atmospheric aerosols (Sakerin et al., 2012; Kuzu et al., 2020). According to many studies, the atmosphere is much polluted metropolitan areas. Barnaul - the capital and the largest city of Altai region is located at the forest-steppe natural zone of the south of West Siberia in the centre of Eurasia. Belonging to the warm summer subtype of humid continental climate (classification by climatologist Wladimir Köppen (Peel et al., 2007) this territory has significant differs between cold and warm periods and characterized by a stable snow cover from early November to late March. In the city of Barnaul, air pollution was assessed as high for many years and nitrogen dioxide is a priority pollutant (Report, 2019). Atmospheric precipitation cleans air and deposits pollutants, that contaminating affects aquatic and terrestrial ecosystems. As a result, the study of atmospheric precipitation chemistry is of great scientific interest (Migliavacca et al., 2005; Tao Sun et al., 2021; Yavuz et al., 2021). In regions with a cold and long winter period, the snow cover can be an integral indicator of the input of substances from the atmosphere to the underlying surface and can indirectly characterize the level of surface air pollution averaged over the whole cold period of the year. Therefore, the results of the chemical analysis of the snow cover sampling at the period of maximum snow storage can be used as a suitable integral indicator

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of atmospheric air pollution (Kuoppamaki et al., 2014; Mashkin et al., 2016). Having a high sorption capacity, due to their porosity and a slower sedimentation rate, snowflakes (in contrast to rain drops) more effectively grab atmospheric aerosols and adsorb chemical compounds from the gas phase (Paramonov et al., 2011; Filimonova et al., 2015). In practice, air pollution can be characterized as background (on global or regional scale) and local (Han et al., 2015). The background air pollution is caused by the transport of pollutants over long distances and on the global scale determined by the interaction of the general circulation of the atmosphere with the earth's surface. The local pollution is a halo formed around local sources of pollutant emission. In this case urban area can be taken as a single source of anthropogenic emission of pollutants into the atmosphere (Alekseev, 2013). It was noted that the urban snow cover contains high concentrations of mineral and organic pollutants in contrast to the background areas (Adamiec et al., 2013; Prokacheva & Usachev, 2013; Kuoppamaki et al., 2014; Mihailovic et al., 2014). For spatial analysis of the distribution of snow cover pollution over the area, it is necessary to build a continuous surface using a limited set of sampling points. The construction of a continuous surface is based on the use of various methods of spatial interpolation, namely, the calculation of an unknown value based on known values in nearby control points. The ArcGIS Desktop 10.2 software product with ArcGIS Geostatistical Analyst extension usedin our work has a set of tools designed to solve this problem.

This study aims to evaluate the influence of Barnaul City on the pollution of the urban airspace with major ions on the basis of snow cover monitoring, and using of geoinformation systems for visualization and processing the obtained data.

MATERIALS AND METHODS

Sampling of snow cover in each point of the monitoring network in Barnaul (Fig. 1) was performed using the envelope method at the time of maximum snow accumulation (the first decade of March). Depending on the depth of the snow cover, a composite sample was formed from 5-10 single samples, which were taken over the entire depth of the snow cover with a plastic tube

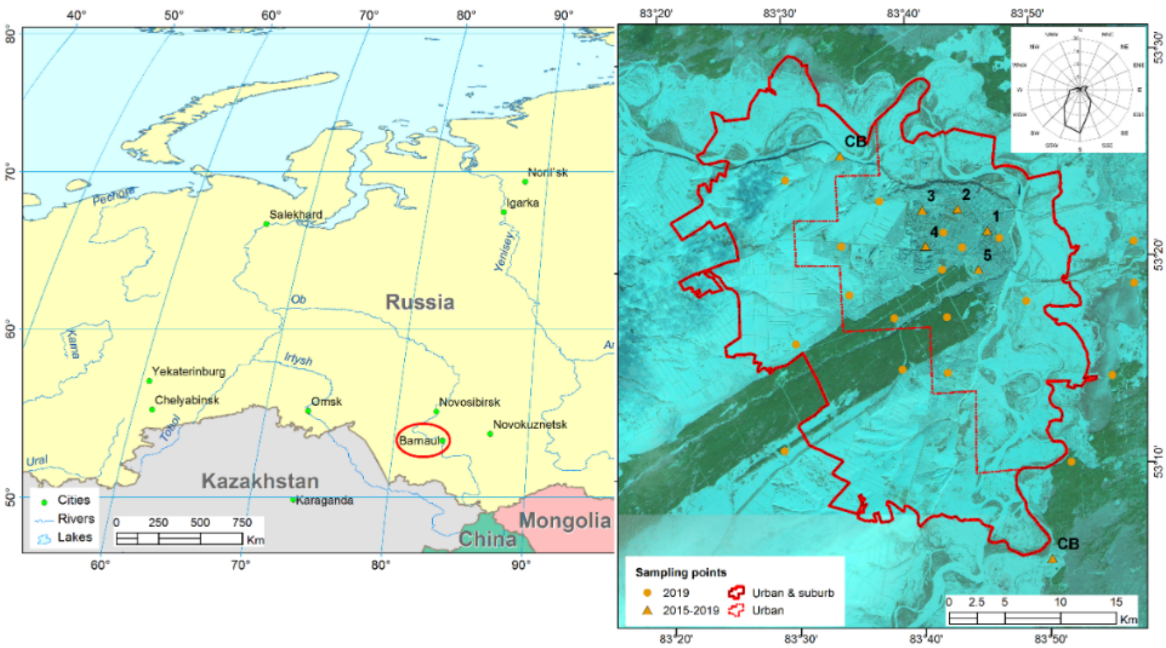


Fig. 1. The sampling points of the snow cover in and around of Barnaul in 2015-2019.The wind rose is based on wind direction data for the period 01.11.2018 - 31.03.2019).

(4.5 cm inner diameter). To exclude the influence of the underlying surface, the lower layer of the snow core (usually not more than 5 cm) was ejected. In 2015-2019 monitoring was carried out in 5 urban (No 1-5) and 2 "conditionally background" (CB) points. The urban points located in different zones of Barnaul City characterized by different anthropogenic impact. Thus, points 3 and 2 are located in the residential areas of the city, points 4 and 5 - in the zone of influence of industrial region. Points 1 and 4 are located in areas of heavy traffic. For comparison, sampling was also carried out at 2 "conditionally background" (CB) points located outside the zone of the predominant wind direction from the urban area and thus not affected by urban pollution. In 2019 the sampling area was expanded and sampling was carried out at 26 points in and around Barnaul City (including the previous 5 urban and 2 CB points) for geostatistical modeling of the spatial distribution of the main ions in the city and the adjacent territories snow cover.

In the winter 2018-2019 during the period of seasonal stable snow cover (the first decade of November 2018 to March 2019) atmospheric precipitation samples were continuously taken in the open area on the roof of IWEP SB RAS building (at a height of about 25 m from the earth's surface). After each snowfall a sample of precipitation was removed from the plastic container and stored frozen until analysis. In total, 54 samples of snow cover (7 samples per year in 2015-2018 and 26 samples in 2019) and 44 samples of wet atmospheric precipitation during the winter 2018-2019 were collected and analysed.

Before analysis samples of snow cover and atmospheric precipitation were melted in specially prepared closed plastic containers at room temperature (US EPA SESDPROC-206) and filtered through a 0.45 μm membrane filter. The quantitative determination of PO_4^{3-} was performed by photometric method with ammonium molybdate on the DR 2800 spectrophotometer (HACH Lange Company, Germany). Determination of other ions in the filtrate was performed by ion chromatography on a reagent-free ICS-3000 system (Dionex, USA) with a conductometric detector using the eluent generation technology. The method was developed and optimized for atmospheric precipitation analysis (Ovcharenko et al., 2015). For all ions the limit of detection (LoD) was calculated as 3 σ of 10 laboratory blanks.

Geostatistical kriging models from ArcGIS Desktop 10.2 software product with ArcGIS Geostatistical Analyst extension were used to estimate continuous parameters for visualization of the obtained data distribution.

RESULTS AND DISCUSSION

Average and median concentration values of the major ions in the melt water of the snow cover in urban (1-5) and conditionally background (CB) sampling points for 2015-2019 (Fig. 1) are presented in Table 1. The background mineralization of atmospheric precipitation for the forest-steppe natural zones is 5-7 mg/L (Svistov, 2011). Variations in mineralization values at "conditionally background" points in Barnaul are within this range and they can be used as a regional natural background level. We used the major ions concentrations in the melt water of the Belukha ice core layers as a global background value for the influx of these substances into the Altai region. The Belukha ice core was recovered from Belukha glacier (4000 m above sea level; 400 km south-east of the sampling site) in 2001, time-dated and chemically analysed (Olivier et al., 2006). For comparison, we use the values of ice layers belong to the industrial period 1941-2001. Since the global background is conservative and changes over time relatively slow, we considered it possible to use the data of this time period to assess the current global background of the study area.

Analysis of the data (Table 1) shows that the regional (conditionally) background content of the studied substances in atmospheric precipitation (and, accordingly, in the atmospheric air as a whole) is an order of magnitude higher than their global background level for this region. At the same time, in the urban snow cover there is an additional increase in the content of the

Table 1. Concentration of major ions (median and mean values) in urban and conditional background (CB) points of snow cover in Barnaul (2015-2019) and layers of the Belukha ice core (1941-2001)

Major ions, mg /L	Urban points (n=25)		CB points (n=10)		Global background (Belukha ice core) (Olivier et al., 2006)
	median	mean ± CI	median	mean ± CI	
Ammonium	1.1	1.0±0.1	0.34	0.36±0.1	0.18
Nitrite	0.09	0.14±0.05	0.005	0.005±0.002	nd
Nitrate	1.6	1.6±0.2	2.0	2.0±0.4	0.22
Chloride	2.1	3±1	0.4	0.4±0.1	0.02
Sulfate	3.6	3.4±0.4	1.2	1.5±0.5	0.40
Phosphate	0.10	0.13±0.06	0.03	0.03±0.01	nd
Sodium	1.2	1.4±0.5	0.16	0.18±0.07	0.01
Potassium	0.6	1.0±0.5	0.16	0.16±0.04	0.01
Magnesium	0.3	0.5±0.2	0.12	0.12±0.04	0.01
Calcium	2.9	3.0±0.4	1.1	1.2±0.3	0.10

CI = 95% confidence interval; n – the number of samples; nd = no data

major ions from 3 to 18 times compared to the "conditionally background" sampling points, which indicates the predominant emission of these pollutants into the urban atmosphere from local sources. In this regard, ions NO_2^-, Cl^-, Na^+ make the largest contribution to the excess of the regional background. The exception is nitrate ion, its concentration in urban areas was slightly lower or did not statistically differ from the "conditionally background". The precursor of nitrate and nitrite ions is nitrogen dioxide, the content of which determines a high level of atmospheric pollution in Barnaul (Report, 2019). The source of nitrogen dioxide is not only industrial and automobile emissions, but also chemical reactions in the atmosphere.

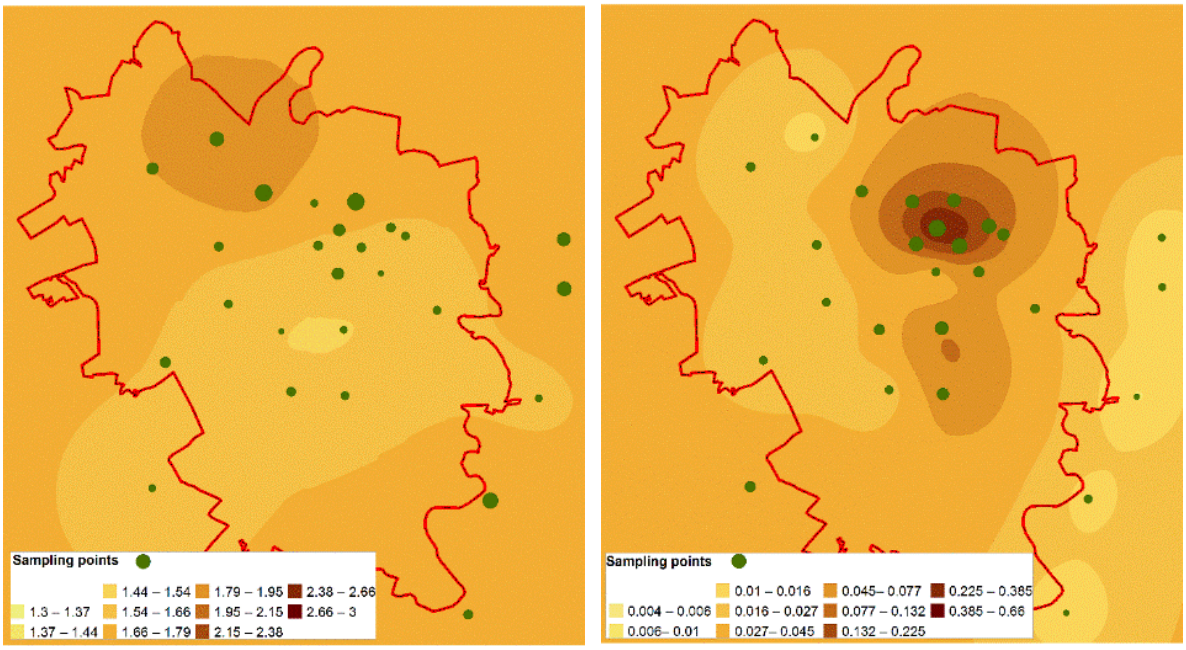
The meteorological potential and climatic features of the Asian part of Russia contribute to ground-level air pollution. As a result, the content of nitrogen dioxide increases by 50-60% compared to the European part of Russia (Bezuglaya et al., 2013). Thus, the increase of nitrate-ion concentrations relative to the global background level may be a regional feature of the studied region and, according to the geostatistical model, this ion has an almost homogeneous distribution over the study area (Fig. 2(a)).

To construct a geostatistical model of the distribution of mineral ions in the snow cover, the flux of the studied components from the atmosphere to the underlying surface during the winter period was calculated using the following formula:

$$P = \frac{C \cdot V}{S \cdot N}, \tag{1}$$

where P – the mass of the determined component per unit area during the period of snow cover, mg/dm^2; C – the concentration of the component in the melt water, mg/L; V – the volume of the melt water in the sample, L; S – the area of the internal transverse pipe cross-sections, dm^2; N – the number of snow cores taken at one point.

Nitrite is the most toxic of all ions studied in this work. The high concentrations of this ion characterize a significant anthropogenic load in Barnaul City and the greatest excess of the natural regional background (by 30 times). Spatial analysis of the nitrite ion distribution (Fig. 2(b)) showed that its highest concentrations are observed in the locations of large traffic intersections (points 1 and 4 Fig. 1). The maximum content of nitrite ions was noted at point 1



(a) (b)

Fig. 2. The spread of pollutants in the snow cover in and around of Barnaul in 2019, mg/gm^2: nitrate (a), nitrite (b). The point numbers correspond to Fig. 1.

Table 2. Comparison of median, mean and volume-weighted mean (vwm) values of the major ion concentrations in the wet atmospheric deposition with their content in the snow cover at the place closest to the sampling point (Barnaul City, winter period 2018-2019).

Major ions, mg/L	Atmospheric deposition			Snow cover
	median	mean ± CI	vwm	
Ammonium	1.4	1.5±0.3	1.2	1.0
Nitrite	0.028	0.037±0.009	0.028	0.078
Nitrate	2.0	2.7±0.6	2.0	1.8
Chloride	1.7	2.1±0.5	1.6	2.3
Sulfate	4.2	6.4±1.7	4.6	3.6
Phosphate	0.02	0.04±0.02	0.03	0.04
Sodium	1.0	1.2±0.2	1.0	1.2
Potassium	0.27	0.31±0.08	0.23	0.23
Magnesium	0.12	0.16±0.04	0.12	0.23
Calcium	2.6	3.4±0.7	2.7	3.2

CI = 95% confidence interval

located at the intersection of all transport arteries of the city and having main source of pollution associated with the intensity traffic and at point 4, which has a double load from both automotive and industrial pollution. However, at point 5, which is affected by the southern industrial zone of the city, the nitrite ion concentration was minimal. Thus, it can be assumed that in Barnaul City the main source of nitrite ion emission is motor transport.

Earlier we have shown (Korobka et al., 2014) that the content of the major ions in atmospheric precipitation falling on the territory of Barnaul during the cold season varies widely. The highest concentrations were observed for the nitrate ion.

Table 2 shows a comparison of the median, mean and volume-weighted mean concentrations of analytes in precipitation taken on the roof of IWEP SB RAS building during the winter period 2018-2019 with their concentrations in the integral sample of snow cover taken near this building in March 2019. As can be seen from Table 2, the median and volume-weighted mean concentrations of the mineral substances in atmospheric precipitation are well comparable with each other, but have some differences from their concentrations in the snow cover. In particular, minor differences are observed for Ca^{2+}, Cl^{-} and significant differences for Mg^{2+}, NO_2^{-}. It is possible the underlying soil surface can affect the contamination of the lower layers of snow cover with soil particles during the initial stage of snow cover formation (Papina et al., 2018).

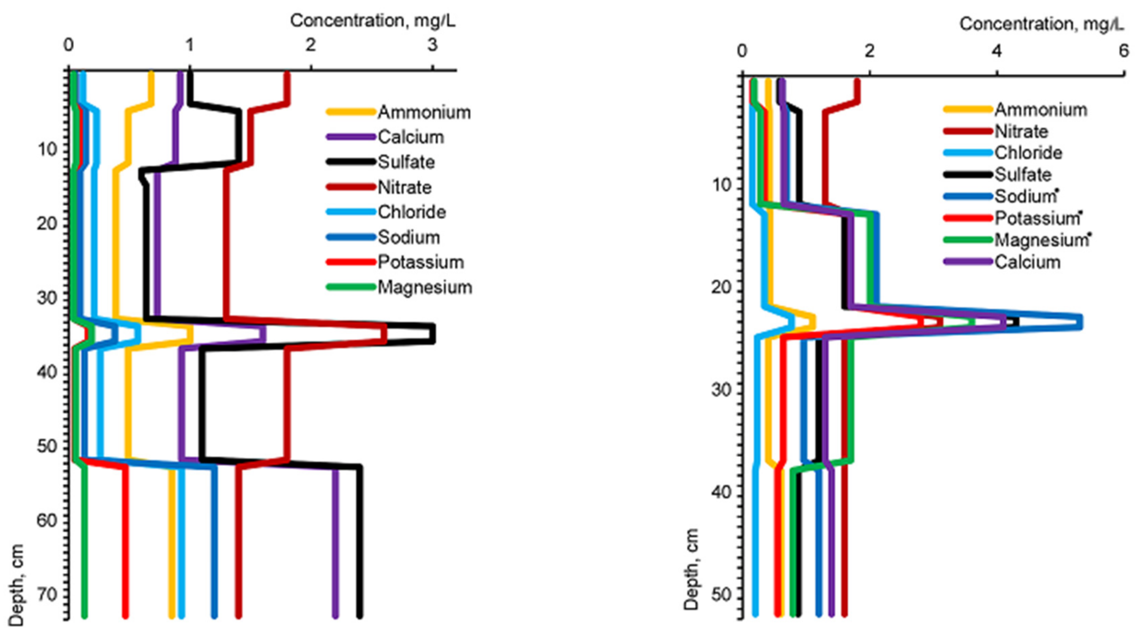
The volume-weighted mean concentration of the major ions in atmospheric precipitation for the winter period was calculated by the formula 2:

$$C_{vwm} = \frac{\Sigma(C_j \cdot Q_j)}{Q}, \qquad (2)$$

where C_{vwm} – the seasonally volume-weighted mean concentration, mg/gm³; C_j – the concentration in the j-th precipitation sample, mg/gm³; Q_j – the amount of the j-th precipitation sample, mm weq.; Q – the total precipitation amount for the winter period, mm weq.

As a result, the increased ions concentration (Mg^{2+}, Ca^{2+}, Na^{+}, Cl^{-}), first of all, may be associated as a pollution of the snow cover with local particles of soil erosion origin (Ermolov & Smolentsev, 2020). Significant differences are observed for NO_2^{-} (higher contents in the snow cover). In our opinion, a noticeable increase in NO_2^{-} may occur due to photolysis of nitrates (Dubowski et al., 2002).

Fig. 3 shows the vertical distribution of the major ion's concentrations in the layers of the snow cover on the different underlying surfaces at the same time. In the first case (Fig. 3(a)), when the sampling of the snow cover was carried out directly from the underlying surface of



(a) (b)

Fig. 3. The vertical profile of the snow cover taken in 2017: (a) – directly from the soil surface, (b) – from the ice surface of Ob River (*mg/L·10).

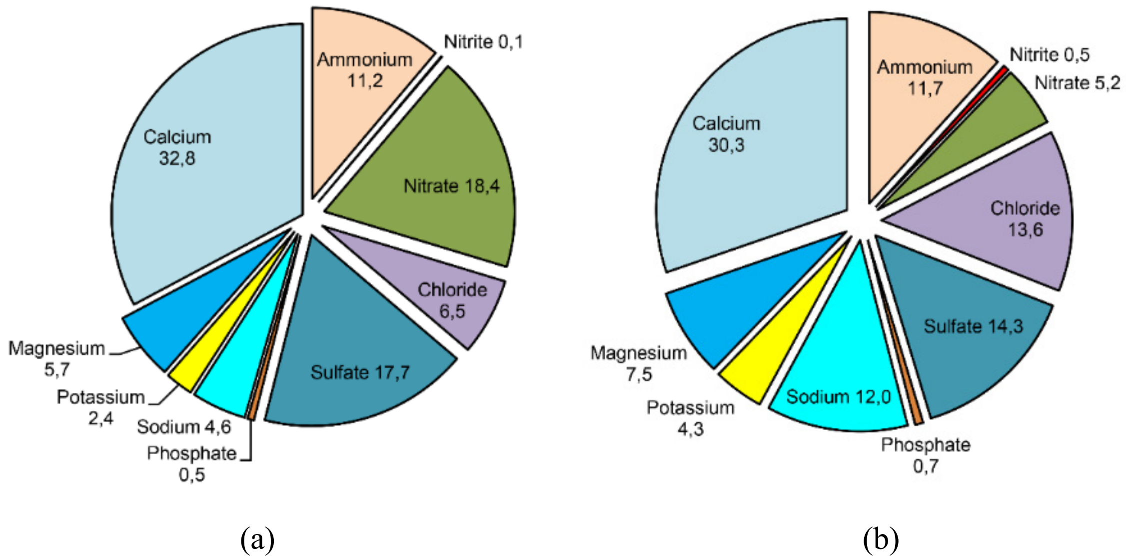


Fig. 4. Percentage of the major ions in conditional background (a) and the urban (b) snow cover from the sum of their equivalents.

Table 3. Coefficients of pair correlation of major ions in urban snow cover.

	NH_4^+	NO_2^-	NO_3^-	Cl^-	SO_4^{2-}	PO_4^{3-}	Na^+	K^+	Mg^{2+}	Ca^{2+}
NH_4^+	1									
NO_2^-	0.35	1								
NO_3^-	0.25	0.03	1							
Cl^-	0.37	0.35	0.39	1						
SO_4^{2-}	0.66	0.42	0.57	*0.58*	1					
PO_4^{3-}	0.56	0.09	0.09	0.29	0.39	1				
Na^+	0.50	0.53	0.37	***0.94***	0.61	0.32	1			
K^+	0.43	0.44	0.27	0.35	0.43	***0.78***	0.42	1		
Mg^{2+}	0.24	0.07	0.33	***0.89***	0.50	0.31	***0.73***	0.27	1	
Ca^{2+}	0.32	0.12	0.63	0.42	***0.74***	0.29	0.34	0.39	0.51	1

the earth, an increased content of the studied components was observed in the lower layer of snow. In the second one (Fig. 3(b)), when the sampling was carried out from the ice of the river channel and the possibility of the snow contamination with soil particles during the initial stage of snow cover formation was excluded, the increased concentrations of the ions under study was no longer observed in the lower layer of snow.

Fig. 4 (according to the data in Table 1) shows the contribution of the major ions to the total mineral composition of the snow cover in the urban (Barnaul City) and "conditionally background" (suburb) points. It is known that natural factors play a major role in the formation of the ionic composition of atmospheric precipitation in the south of Western Siberia, and the main source of water-soluble mineral salts is particles of continental aerosol (Smolyakov et al., 2006). The main ions in the snow cover of the "conditionally background" points near Barnaul are Ca^{2+}, NH_4^+ (cations) and SO_4^{2-}, NO_3^- (anions), which also make up the mineral bulk of the Belukha ice core.

According to the correlation analysis of the major ions in the snow cover of Barnaul (Table 3), sodium and chloride ions have the highest direct dependence (0.94), which confirms their combined input from the source of pollution. In addition, a significant correlation (> 0.70) is observed between the following pairs: Mg^{2+} and Cl^{-} and SO_4^{2-}, K^{+} and PO_4^{3-}, Na^{+} and Mg^{2+}.

The sea salt fraction (*% SSF*) was calculated according (2008; Alves, 2018; Meng, 2019) and used to estimate the contribution of various ion sources in snow cover. The sodium ion was used as a reference for these calculations that carried out according to formula 3:

$$\%SSF = \frac{100\cdot\left(\frac{X}{Na}\right)marine}{\left(\frac{X}{Na}\right)snow}, \tag{3}$$

where *%SSF* –sea salt fraction; (*X/Na*) *marine* – ratio of ions in the sea; (*X/Na*) *snow* - ratio of ions in the snow cover. No sea salt fraction (*%NSSF*) identified by the formula 4:

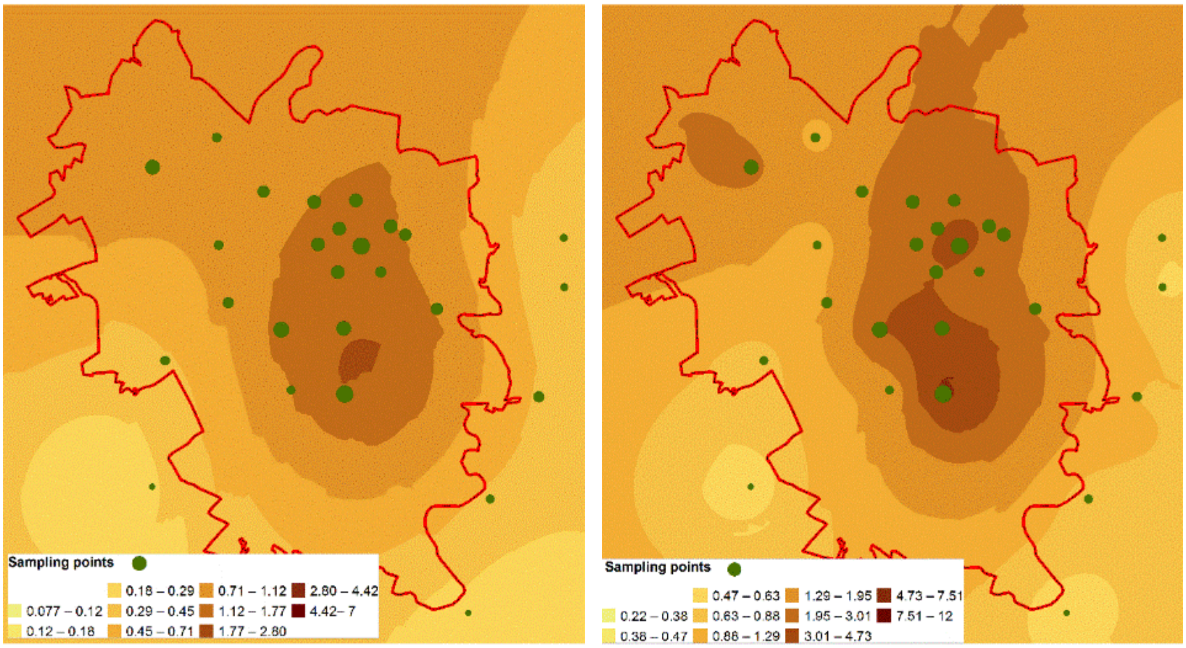
$$\%NSSF = 100 - \%SSF, \tag{4}$$

The sea salt fraction (% SSF) and was calculated according formula 3 and used to estimate the contribution of various sources of ions entering the snow cover (Table 4).

The ratio of ions (X/Na) in the snow cover is much higher than in seawater, therefore the main contribution to the content of ions is made by other land sources (soil and anthropogenic), which is quite typical for the continental territory. In this case, the ratio Cl^{-} / Na^{+} = 1.14, which is practically equal to the ratio of ions in sea salt and may indicate the marine origin of ions, which is an incorrect statement. The anthropogenic source is responsible for the high NaCl content of the snow cover in Barnaul City. This was confirmed by a geostatistical model of their spatial distribution in the urban agglomeration (Fig. 5). The main localization of sodium and chloride ions is observed in the densely populated part of the city, where the main transport lines are located and the roads and sidewalks are intensively treated with anti-icing reagents. The change of the ionic composition in the urban snow cover (the increase of a sea ions Cl^{-} and Na^{+} in the proportion) is associated with anthropogenic influence, namely, with the use of sodium chloride as the main component of the anti-ice reagent. The chemical composition of atmospheric precipitation falling on the territory of Barnaul in winter period is formed under the influence of regional and local natural and anthropogenic factors. The underlying surface has a great influence on the mineral composition of snow covers at the initial stage of their formation. The impact of urban anthropogenic activity is manifested not only in an increase of the natural regional background by more than two times, but also in an increase of the percentages of sea ions (Cl^{-}, Na^{+}), which is not typical for the global and regional backgrounds for the study area. The highest emission of pollutants into the atmosphere of Barnaul, exceeding the natural regional background by 7, 8 and 30 times, is observed, respectively, for Cl^{-}, Na^{+}, NO_2^{-} ions, the main sources of which are associated with motor vehicle emissions (NO_2^{-}) and

Table 4. Ratio ions, sea salt fraction, no sea salt fraction in urban (Barnaul city) snow cover.

Ions	Ratio(X/Na) marine (Keene et al., 1986)	Urban snow cover		
		Ratio (X/Na)snow	%SSF	%NSSF
Nitrate	0.00002	2.528	0.005	99.995
Chloride	1.161	1.136	≈ 100	≈ 0
Sulfate	0.121	1.194	10.1	89.9
Potassium	0.022	0.356	6.2	93.8
Magnesium	0.227	0.626	36.2	63.8
Calcium	0.044	2.528	1.7	98.3



(a) (b)

Fig. 5. The spread of pollutants in the snow cover in and around of Barnaul in 2019, mg/gm^2: sodium (a), chloride (b). The point numbers correspond to Fig. 1.

intensive use of anti-ice reagents (Cl^-, Na^+), in the winter period. High concentrations of these ions worsen the quality of urban atmospheric air and can have an adverse effect on public health and the ecological state of urban soils and water bodies. Geostatistical models of the spatial distribution of pollutants clearly demonstrate the main localization and distribution of the studied ions relatively the sources of urban air pollution.

According to the schematic maps (Fig. 3(b) and 5(a), 5(b)), pollutants spread far beyond the urban agglomeration in the northern and northeaster directions from Barnaul City in accordance with the prevailing wind direction in winter (Fig. 1). On the leeward side, the contents of the studied ions at a distance of 20-25 km from the contaminated centre part of the city decrease to the regional background concentrations. An exception is nitrate ion, the concentrations of which in the urban area were somewhat lower than the "conditionally background", but were well comparable with the previously obtained results for Barnaul City (Noskova et al. 2014) and other adjacent regions of Western Siberia (Kokovkin et al. 2011; Zharnikov et al. 2019). While the contents of other nitrogen-containing compounds in the snow of the urban area were noticeably higher (3 times for NH_4^+ or significantly higher (30 times for NO_2^-) relative to the natural regional background. In this regard, the transformation and distribution of mineral forms of nitrogen in the atmosphere of Barnaul City requires a more detailed study, which will be the subject of our further research.

CONCLUSIONS

The study of the ionic composition of the snow cover of Barnaul City for 5 years (2014-2019) showed:

– pollution of the snow cover with major ions occurs mainly from local natural (soil erosion) and anthropogenic (vehicle exhaust and anti-ice reagents) sources;

– geostatistical models of the spatial distribution of pollutants, clearly showed that the localization of ions occurs in the densely populated and a heavy traffic part of the city

– the content of the major ions in the urban atmosphere is more than 2 times higher than the regional natural background and 6 times than the global one. The priority ions that determine the high pollution of the urban winter atmosphere are (Cl^-, Na^+, NO_2^-). For the study area are characteristic of high concentrations of nitrate ion, whose content is the same in urban points and regional background, but at least ten times more which global background.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy have been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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