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CHANGES IN SNOWFALL CHEMICAL COMPOSITION IN BASTAK NATURE RESERVE

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Abstract

The content of a wide range of chemical elements in snowfall in Bastak Nature Reserve in winter 2020-2021 is defined. The amount of precipitation and density of elements in snowmelt is compared with the previous wintertime. Snow sampling is carried out at 6 stations with different ecological conditions located on the territory of Bastak Nature Reserve. The following methods are used in the study: direct potentiometry for pH measurements; titrimetry for chlorides, total hardness and hydrocarbonates; photometry for nitrate, sulphate, nitrite, ammonium ions and phosphates concentration; and atomic absorption spectroscopy to specify heavy metal concentration (content). It was found that the winter of 2020-2021 was snowier and characterized by the air streams coming from the ocean, which is confirmed by the most abundant ions such as sulphates, chlorides and ammonium in the samples. In March, there was a change in the direction of air mass flows from the continent to the ocean, which caused decreasing concentrations of 'oceanic' ions. Tracers of terrigenous influence of iron and manganese were found in all snow samples, as well as tracer material of anthropogenic influence - zinc and copper, which was probably dependent on the location of the reserve near an urbanized area.

Keywords: Air pollution, heavy metals, nature reserve, snow cover

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1. Introduction

The atmospheric air of modern cities receives a large number of pollutants that are easily transported over long distances by air currents. Therefore, air pollution caused by industrial plants and transport affects not only large industrial centres but also contiguous lands. Specially protected nature conservation areas can be chemically polluted because of the airwaves, even without significant anthropogenic impact (Ershov et al., 2019; Kholodov & Golokhvast, 2020b; Zajchowski et al., 2019).

The territory of the Jewish Autonomous Region (JAR) is home to Bastak State Nature Reserve. One of the tasks of it is to conduct integrated environmental monitoring required to assess and forecast the state of the environment of the conservation area. Monitoring of open-air pollution is an integral part of the environmental monitoring system and includes investigation of the composition of atmospheric precipitation including snow in the wintertime. Snow can be used as an integrated index of atmospheric pollution in winter for territories characterized by the stable snow cover (Koroleva et al., 2017; Novorotskaya, 2018; Zhurba et al., 2020).

2. Problem Statement

The study of the chemical composition of atmospheric precipitation, the main indicators of its gathering, and the transfer of a pollutant, including transboundary, has been conducted in the areas conservancy in the Far East since the 1970s (Kholodov & Golokhvast, 2020a). The obtained results allowed us to reveal the trends of changes in the chemical composition of snow cover in separate areas conservancy and their dependence on climatic, orographic and anthropogenic factors (Kondratyev & Kachur, 2004; Sukhareva, 2012). Nevertheless, the condition of the atmosphere in most reserve areas is left poorly studied.

This research is a continuation of the study to explore and identify possible changes in the physical-chemical composition of atmospheric fallout in Bastak Nature Reserve initiated in 2011 (Golokhvast et al., 2013, 2016; Kholodov & Golokhvast, 2020a).

3. Research Questions

A peculiar feature of the climate in the southern Far East is the monsoon climate. This is a seasonal change in air currents from the continent and the ocean due to irregular heating and cooling. In winter, due to high atmospheric pressure over land, air moves towards the sea. A winter monsoon accompanied by north-western, western and south-western winds brings dry and strongly cooled air, clear and very frosty weather to the whole Far East. The now humid sea air blows towards the continent, forming a monsoon in summer. In the mid-seasons, spring and autumn, circulation processes are rearranged. In spring, there is a gradual destruction of the winter anticyclone, while in autumn there is a gradual transition from the summer type of circulation to the winter one (Grigoryeva & Fetisov, 2015).

4. Purpose of the Study

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The purpose of the research is to find out inter-annual changes and their origin in the composition of winter atmospheric fallout.

5. Research Methods

Snow samples for element content were analyzed at an accredited testing laboratory centre of the Centre for Hygiene and Epidemiology in the JAR. Snow was collected on the territory of the reserve during snowfalls in winter 2020-2021. To exclude secondary contamination by anthropogenic aerosols, the top layer (5-10 cm) of new-fallen snow was sampled (Liu et al., 2018). The melted snow samples, after being filtered through an ashless ‘white tape’ filter, were analysed for the content of chemical components.

When investigating the composition of snowmelt, the techniques used for natural water analysis (Dmitriev et al., 1989) were used. The following methods were used in the study: direct potentiometry for pH measurement (pH-meter ANION 4100); titrimetry for chloride, total hardness and hydrocarbonates; photometry for nitrate, sulphate, nitrite, ammonium ions and phosphates were performed using spectrophotometer PE-5400 VI. All chemical reagents used for the analysis were qualified not lower than 'chemically clean’ (C.P.). Melted snow samples were analyzed for heavy metals using atomic absorption spectrometers Kvant-2 AT and MGA-1000.

A total number of 30 samples of new-fallen snow were analyzed. It included 552 elemental determinations.

Snow samples were taken on 13/11/2020, 14/01/2021, 27/01/2021, 17/02/2021 and 22/03/2021 at 6 stations located on the territory of Bastak Nature Reserve with different ecological conditions.

The snow sampling locations are presented in Table 1.

Table 1. Sampling stations and their description

Sampling stations	Description of stations
1	Near Chita highway, flatland (plain), about 100 m from the highway, border of the reserve, right bank of the Ikura river. Vegetation: sedge-reedgrass meadow.
2	Birobidzhan - Kukan Highway, about 15 km from Birobidzhan, plain, left bank of the Glinyanka river. Vegetation: sedge-reedgrass meadow.
3	Birobidzhan – Kukan Highway, 30 km from Birobidzhan, middle reaches of the Bastak river, left bank. Vegetation: larchen goosefoot (pigweed).
4	Birobidzhan – Kukan Highway, 30 km from Birobidzhan, middle reaches of the Bastak river, right bank, on the slopeside with exposure to the south (the near-to-summit part). Vegetation: cedar-oak forest.
5	Chernukha Mountain, middle part, on the slopeside with exposure to the southeast. Vegetation: cedar- broad-leaved forest, Khingam fir has been reliably proved to die off/dry out (since 2007 till present).
6	Birobidzhan – Kukan Highway, 25 km from Birobidzhan, in the natural boundary/stow ‘Krasnye (Red) Sopki’. Vegetation: oak wood.

Among the sampling locations, Station 1 is the most environmentally concerned one near the Chita-Khabarovsk highway that is the section of federal highway with the heaviest traffic. Stations 2, 3, 4, 6 are also located close to the road, but Birobidzhan - Kukan highway, which crosses the reserve and

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divides it into almost two equal parts, is used quite rarely and, therefore, these stations are less ecologically concerned. Station 5 is located 300m east of Birobidzhan-Kukan highway, i.e. almost completely far from its influence.

6. Findings

The results of the chemical analysis of the snowmelt are presented in Tables 2 and 3.

Table 2. Content of chemical components in the snow in Bastak Reserve, winter season 2020-2021

Sampling stations	Concentration, mg/dm3				pH, pH unit
	SO_4^{2}	Cl^-	NO_3^-	NO_2^-	
			13.11.2020		
1	2,99±0,61	**1,29±0,31**	**10,60±1,58**	<0,02	5,1±0,2
2	1,10±0,22	<0,5	5,45±0,86	<0,02	4,3±0,2
3	**4,21±0,84**	0,89±0,21	**13,00±1,95**	<0,02	4,3±0,2
4	**5,0±1,0**	0,73±0,17	**16,50±2,48**	0,005±0,003	4,5±0,2
5	1,75±0,35	0,75±0,18	6,25±0,94	<0,02	4,7±0,2
6	2,66±0,53	0,87±0,21	9,00±1,35	<0,02	4,4±0,2
			14.01.2021		
1	0,58±0,12	**3,23±0,77**	5,75±0,86	0,006±0,003	4,9±0,2
2	0,55±0,11	0,81±0,19	5,51±0,83	0,0017±0,009	6,3±0,2
3	0,55±0,11	0,72±0,17	5,75±0,86	0,029±0,015	4,6±0,2
4	0,61±0,12	0,60±0,14	5,25±0,79	<0,003	4,3±0,2
5	0,57±0,11	0,79±0,19	5,75±0,86	<0,02	4,4±0,2
6	0,56±0,11	0,57±0,14	5,51±0,83	0,011±0,005	4,3±0,2
			27.01.2021		
1	<0,5	**5,89±0,59**	1,92±0,31	<0,2	4,8±0,2
2	<0,5	<0,5	2,38±0,28	<0,2	4,6±0,2
3	<0,5	<0,5	2,81±0,45	<0,2	4,5±0,2
4	<0,5	<0,5	2,65±0,42	<0,2	4,5±0,2
5	<0,5	<0,5	3,29±0,53	<0,2	4,4±0,2
6	<0,5	<0,5	2,28±0,86	<0,2	5,5±0,2
			17.02.2021		
1	<0,5	2,65±0,64	0,42±0,09	<0,2	6,2±0,2
2	<0,5	<0,5	0,48±0,11	<0,2	6,2±0,2
3	2,33±0,47	0,67±0,16	3,25±0,52	<0,2	5,6±0,2
4	1,23±0,25	<0,5	3,11±0,51	<0,2	6,0±0,2
5	1,87±0,37	0,66±0,16	3,39±0,54	<0,2	5,6±0,2
6	1,24±0,25	0,8±0,2	1,58±0,25	<0,2	5,9±0,2
			22.03.2021		
1	<0,5	<0,5	0,32±0,07	<0,2	5,3±0,2
2	<0,5	<0,5	<0,2	<0,2	5,6±0,2
3	<0,5	<0,5	0,24±0,05	<0,2	5,3±0,2
4	<0,5	<0,5	0,29±0,06	<0,2	5,4±0,2
5	<0,5	<0,5	0,52±0,08	<0,2	5,4±0,2
6	<0,5	<0,5	0,24±0,05	<0,2	5,4±0,2

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The winter of 2020-2021 was characterized by heavy snowfall, so the sample amount was quite representative. The main difference of these winter samples, however, was sulphate and chloride presented in the snowmelt of new-fallen precipitation.

The sulphate ion is one of the markers of acid precipitation, as it is the end product of sulphur dioxide transformation in the atmosphere. Sulphate was abundant in November, January and February samples, while its content was below the limit of detection in samples taken by this method in late January and March. High sulphate can lead to over-acidification of weather elements, such as in November 2021, when sulphate concentration of 4.21 mg/dm3 at Station 3, located in the centre of the Reserve, resulted in a pH of 4.3. At the same time, high single concentrations of sulphate in weather elements resulted in their increasing acidity that may harm the environment.

Chloride ions were detected mainly in samples taken in November 2020 and mid-January 2021. The chloride ion content in the snowmelt varied from 0.6 to 3.23 mg/dm3. Its maximum concentration was determined in January sampling from station 1. The abundant grade of sulphate and chloride ions in atmospheric precipitation in winter 2020-2021 stands for the pattern of the brought precipitation, namely its marine (oceanic) origin.

The content of nitrate ions in the snow cover of the reserve ranged from 0.24 mg/dm3 to 16.5 mg/dm3. Its maximum concentration was measured in a sample from station 4, located in the reserve in the heart of the forest and far from the highway.

Nitrite ions were detected in trace amounts (quantities) in November and January samples, and in the other snow samples, they were below the limit of detection by this method.

According to the data obtained, meltwater (snow) pH values measured in winter ranged from 4.3 to 6.3. The concentration rate of hydrogen ions in air precipitation usually ranges from 4.6-6.1 (Maistrenko et al., 1996). Thus, in this case, the pH range was larger, including both more acidic and more neutral zones.

Table 3. Content of chemical components in the snow in Bastak Reserve, winter season 2020-2021

Sampling stations	**Concentration, mg/dm3**			**Total hardness, GoH**
	NH_4^+	**HCO_3^-**	**PO_4^{3-}**	
		13.11.2020		
1	1,46±0,29	-	<0,05	0,20±0,01
2	0,42±0,08	-	<0,05	0,30±0,05
3	**2,25±0,45**	-	<0,05	0,40±0,06
4	**2,46±0,49**	-	<0,05	0,40±0,06
5	0,66±0,13	-	<0,05	0,30±0,05
6	1,32±0,26	-	<0,05	0,40±0,06
		14.01.2021		
1	0,23±0,05	12,2±0,9	0,08±0,01	0,5±0,1
2	0,11±0,03	12,2±0,9	0,08±0,01	0,5±0,1
3	0,22±0,04	12,2±0,9	**0,35±0,06**	0,20±0,03
4	<0,1	6,1±0,5	0,09±0,01	0,20±0,03
5	0,14±0,04	**18,3±1,5**	0,09±0,01	0,40±0,06
6	0,11±0,03	12,2±0,9	<0,05	0,20±0,03
		27.01.2021		

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1	<0,1	6,1±0,7	<0,25	0,20±0,01
2	0,22±0,04	6,1±0,7	<0,25	0,20±0,01
3	0,13±0,04	6,1±0,7	<0,25	0,20±0,01
4	0,12±0,04	6,1±0,7	<0,25	0,20±0,01
5	0,14±0,04	6,1±0,7	<0,25	0,20±0,01
6	0,141±0,04	6,1±0,7	<0,25	0,20±0,01
		17.02.2021		
1	0,21±0,04	12,20±0,98	<0,25	0,20±0,01
2	0,11±0,03	12,20±0,98	<0,25	0,20±0,01
3	<0,1	12,20±0,98	<0,25	0,20±0,01
4	0,36±0,07	12,20±0,98	<0,25	0,20±0,01
5	0,441±0,08	12,20±0,98	<0,25	0,20±0,01
6	0,31±0,06	12,20±0,98	<0,25	0,20±0,01
		22.03.2021		
1	0,12±0,03	12,20±0,98	<0,25	-
2	<0,1	12,20±0,98	<0,25	-
3	<0,1	12,20±0,98	<0,25	-
4	0,18±0,04	12,20±0,98	<0,25	-
5	0,22±0,04	12,20±0,98	<0,25	-
6	<0,1	12,20±0,98	<0,25	-

The concentration of hydrocarbonates in snowmelt at all sampling stations ranged from 6.1 to 18.3 mg/dm3. The amount of these ions in the snow cover is measured by the atmospheric CO2 concentration and indicates the product return of fuel consumption. Therefore, the low content of hydrocarbonate ions implies that there are no additional sources of carbon dioxide on the territory of the reserve, in particular, of anthropogenic origin.

Ammonium ion has been found in most samples in the range from 0.11 to 2.46 mg/dm3. Its maximum concentration (2.46 mg/dm3) was measured at station 4. In March 2021 ammonium ion was found in only three samples, with lower concentrations than on other sampling dates.

As follows from Tables 2 and 3, by March there has been a significant change in the direction of air currents: a strong decrease in concentrations of SO42, Cl, NH4+ ions suggest a change of their direction from oceanic to continental.

As demonstrated, the level of total hardness, which is set by the content of calcium and magnesium salts, in samples of the reserve was within 0.2-0.5 GoH, i.e., low. In terms of hardness, snowmelt is referred to as the soft water (low hardness water) class.

Thus, the chemical analysis of the snow cover of the reserve in the winter season of 2020-2021 showed that the open air over the reserve area is not polluted with the main natural and anthropogenic impurities. However, acid-forming sulphate and nitrate ions found in snow samples are of some concern.

Of the 10 elements we determine, Cd, Ni, Pb, Co, Hg and As are found in trace amounts, usually below the limit of detection by this method, so we do not discuss them. Of the other four elements Fe and Mn are indicative of terrigenous influence on precipitation, Cu and Zn are clear tracers of anthropogenic influence (Table 4).

Switching to a more sensitive method of analysis allowed finding nickel, lead and mercury in snow samples for the first time during the four-year observation period.

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Table 4. Heavy metal content in snow samples in Bastak Reserve, winter season 2020-2021, mg/dm^3

Sampling stations	Fe	Mn	Cu	Zn
		13.11.2020		
1	0,055±0,011	0,025±0,005	0,015±0,003	0,051±0,009
2	0,066±0,013	0,012±0,002	0,0011±0,003	0,017±0,003
3	0,104±0,021	0,015±0,003	0,012±0,002	0,039±0,007
4	0,114±0,023	0,023±0,005	0,015±0,003	0,047±0,008
5	0,041±0,008	0,027±0,005	0,009±0,003	0,027±0,005
6	0,055±0,011	0,014±0,009	<LoD	0,034±0,006
		14.01.2021		
1	0,198±0,041	0,017±0,005	0,003±0,001	0,022±0,007
2	0,863±0,173	0,187±0,037	0,005±0,002	0,008±0,003
3	0,841±0,168	0,039±0,008	0,005±0,002	0,058±0,012
4	0,067±0,013	0,008±0,003	0,011±0,003	0,015±0,005
5	0,063±0,013	0,001±0,003	0,021±0,006	0,035±0,007
6	0,067±0,013	0,009±0,003	0,035±0,007	0,045±0,009
		27.01.2021		
1	0,142±0,028	0,006±0,003	0,021±0,006	0,028±0,006
2	0,083±0,017	0,0021±0,0008	0,009±0,004	0,012±0,004
3	0,062±0,012	0,003±0,001	0,014±0,004	0,007±0,003
4	0,062±0,012	0,003±0,001	0,014±0,004	0,007±0,003
5	0,074±0,014	0,004±0,002	0,005±0,002	0,012±0,004
6	0,044±0,003	0,004±0,002	0,007±0,003	0,013±0,004
		17.02.2021		
1	0,081±0,016	0,007±0,003	0,023±0,007	0,005±0,002
2	0,047±0,009	<LoD	0,028±0,006	0,007±0,003
3	0,254±0,051	0,028±0,006	0,007±0,003	0,0017±0,005
4	0,052±0,011	0,011±0,004	0,014±0,004	0,008±0,003
5	0,085±0,017	0,014±0,004	0,027±0,005	0,013±0,004
6	0,051±0,011	0,012±0,004	0,017±0,005	0,009±0,004
		22.03.2021		
1	0,033±0,007	0,015±0,005	0,005±0,002	0,005±0,002
2	0,041±0,008	<LoD	0,002±0,001	0,007±0,003
3	0,042±0,008	<LoD	0,006±0,003	0,0017±0,005
4	0,041±0,008	<LoD	0,014±0,004	0,008±0,003
5	0,036±0,007	<LoD	0,002±0,001	0,013±0,004
6	0,037±0,007	<LoD	0,005±0,002	0,009±0,004

Note: '<LoD' is less than the limit of detection.

As in the previous observation periods in 2020-2021, iron was found in all snow samples, varying from 0.041 mg/dm3 to 0.863 mg/dm3. The maximum concentration of iron was found in a sample from station 2, located near Birobidzhan-Kukan highway.

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In contrast to common iron, Mn was present in samples by an order or more of lower magnitude. In March sampling manganese was detected only at 1 station in the amount of 0.015±0.005 mg/dm3.

Zinc was found in all snow samples with no exception ranging from 0.0017 to 0.058 mg/dm3. The maximum concentration of zinc was determined in mid-January 2021 in a sample from station 3, which was located near a federal highway and was located in the central part of the reserve.

Copper concentrations were also detected in all snow samples and were in the close to zinc range. The highest copper content was found in a sample taken in mid-January 2021 at Station 6, which was located in the central part of the reserve and was next to the federal highway.

7. Conclusion

Measurement of the amount of snowfall in the winter of 2020-2021 and the content of chemical elements in snowmelt allows the following conclusions to be drawn:

- The winter of 2020-2021 was snowy compared to the previous one, allowing for 5 samples of the new-fallen snow, whereas the previous winter had two snowfalls and the winter was dry;
- The detection of representative amounts of sulphate and chloride in snowmelt in winter sampling in 2020-2021 indicates an eastern direction of air masses gathering over the ocean; ammonium ion as well as sulphate and chloride ions also dominated in winter precipitation and decreased greatly by March;
- In March 2021, there was a change in the direction of air currents from the continent to the ocean, with a sharp decrease in sulphate, chloride and ammonium ions;
- Terrigenous iron and manganese were found in almost all samples during all observations;
- Zinc and copper are bright tracers of anthropogenic impact on the environment were also found in all samples.

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