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UDC 550.455

ESTIMATION OF ACIDIFICATION RESISTANCE OF SMALL LAKES IN THE NORTH OF WEST SIBERIA[*]

SUMMARY. The paper studies the chemical composition of the waters of small lakes in the northern territories of West Siberia in order to evaluate their resistance to the processes of acidification. The study includes the lakes located far from the sources of anthropogenic impact and the ones located in close proximity to them. According to the results of this research, it is concluded that about 15% of relatively clean lakes are vulnerable to the loss of acid-forming substances and have a low self-regenerating capacity. These primarily include the low-mineralized water bodies in the northern and middle taiga zones. In addition, there are water bodies in the zone of anthropogenic impact, which at present have the critically low values of acid-neutralizing capacity. However, most of the baseline lakes are classified as hydrocarbonate ones and have a good surge capacity. Chlorides predominate in the anion composition of the lakes located in the zone of anthropogenic impact. The calculation of acid-neutralizing capacity of these lakes demonstrates that most of them (about 80%) are sufficiently resistant to fallout of acid-forming substances (ANC > 100).

KEY WORDS. Acidification of waters, acid-neutralizing capacity, estimation of the vulnerability of water bodies.

Introduction. During the past century, global air pollution by acid-forming substances, mainly due to the fuel and coal combustion and metal smelting, resulted in the acid rain formation. Practically, concurrently with the precipitation acidification, the phenomenon of the lake acidification was registered. First, this phenomenon was largely observed in Sweden and Norway, and later in the United States and Canada. The lake acidification is caused by the input of acids into to the water-producing areas of lakes, both with precipitation and as the result of dry absorption [1], [2]. The acidification of water bodies has a highly negative after-effect; it contributes to heavy metal leaching, changes the ionic composition of waters, and reduces the buffering capacity of natural water bodies. The effect of the water body acidification in West Siberia has been poorly investigated; however, there are data on the appearance of this effect in the northern territories [3]. Moreover, the problem of gas flaring deserves special attention. When flares are under operation, different products of combustion and pollutants enter the air. The significant scale of the nitrogen oxide (NO_X) pollution in

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[*] This research was financially supported by the Ministry of Education and Science of the Russian Federation under the Federal Target Program *Scientific and Academic Staff of Innovative Russia* in 2009–2013 (Agreements Nos. 14.B37.21.1255 and 14.B.37.21.0656*)*

the process of fuel combustion is caused by the fact that these oxides are formed not only by the reaction of the air O_2 with the fuel nitrogen, but also by the reaction of the air O_2 with the air nitrogen. Accordingly, acid fallouts are possible in West Siberia, especially in the north, where there is the largest number of flare devices [4].

The paper presents the results of studying the chemical composition of waters in the small lakes of the vast West Siberian territory. The study was carried out at Tyumen State University in 2011-2012. The chemical composition of waters in the small lakes most clearly reflects the specific zonal conditions of their formation and intra-zonal variability. At the same time, small lakes are highly vulnerable to acid-forming substances falling out on the water-producing areas. The long latitudinal gradient of the territory allows determining the specific features of changes in the chemical composition of waters in the tundra and boreal zone (including the northern, middle, and southern taiga zones). The purpose of this study is to estimate the acidification processes behavior in the lakes in the north of West Siberia and in the water bodies both far from the sources of anthropogenic impact and the ones located in close proximity to them.

Materials and methods. The paper presents the results of studying the chemical composition of waters in about 120 baseline lakes in the tundra and taiga zones of West Siberia (WS) carried out in 2011 by the consistent methodological scheme. The study included lakes with the water surface area less than 20 km^2 that are not subject to any direct sources of pollution. In 2012, water samples were taken from 40 lakes located in the zones of industrial impact, mainly these are the operation zones of oil-and-gas producing companies. To minimize the influence of seasonal and year-to-year variations, the samples were taken in the shortest possible period of the late autumn cooling, when homothermal conditions are setting in, and there is no stratification, when there are no significant fluctuations in vegetation processes, but their "smoothed" level is observed. This principle of study allows comparing the lakes located in different natural and climatic zones. The chemical composition of waters was determined by the consistent methodological scheme, the correctness of the results obtained was verified by calculating the balance of main ions and by comparing the measured water conductivity with the calculated one. The analytical program included the determination of pH, electrical conductivity (χ), basic mineralization ions (Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, SO_4^{2-}, Cl^-), alkalinity (Alk), color index (Col), various forms of nitrogen (NH_4^+, NO_3^-, TNb), total (TC) and organic carbon (TOC), total phosphorus (TP), and phosphates (PO_4^{3-}).

- pH was determined by the potentiometric method, with a glass electrode (the *I-130.M* ionomer);
- electrical conductivity at 20° was determined by the conductometric method (the *Anion 4100* conductivity meter);
- color index was determined by the spectrophotometric method on the chromium-cobalt color scale, at the wavelength of 380 nm (the *UNICO* spectrophotometer);
- total phosphorus was determined by the spectrophotometric method, in the form of blue-phosphorus-molybdenum complex, at the wavelength of 882 nm (the *UNICO* spectrophotometer).

Total nitrogen, total carbon, and organic carbon were determined by the elemental analysis with the *Vario TOC* unit (Elementar). Inorganic carbon (water-soluble forms) was calculated as the difference between total and organic carbon.

The contents of main ions, mineral forms of nitrogen (ammonium, nitrates, nitrites), and phosphates were determined by the ion chromatography method with the *DIONIX ICS-2100* ion chromatograph. We used the electrolytic sample preparation and the eluent generation, i.e. the technologies developed for applying electrolytically generated eluents for all types of isocratic and gradient separation in ion chromatography with the conductometric detection.

The criteria for evaluation of water acidification.

Within the *Assessment and Monitoring Acidification of Lakes and Rivers* international program, the international practice was generalized, the consistent principles and methods of the regional monitoring of water bodies, as well as the criteria for estimating water acidification, were worked out [5]. 22 countries participating in the program follow the developed methods of research. This allows achieving comparability and reliability of the obtained data on the problem state in the global scale. Identifying the degree and intensity of water acidification in the regions requires special approaches and methods to the selection of water bodies, their monitoring, and the analytical measurement of the chemical composition of waters [6].

In the world practice, the following indicators are used as the evaluation criteria of acidification processes, each of them may have certain advantages and disadvantages.

1. Low pH values may be caused by the anthropogenic acidification of waters in case of low water color index. However, in humid regions of Russia, including West Siberia, dystrophic water systems are wide-spread; low pH values of them are determined by the high contents of natural humic acids. Such waters have high color index values: from 30 to 60°, Cr-Co scale. Under the greater impact of anthropogenic acids, water dicloloration may occur. Therefore, during quite a long period, anthropogenically-acid lakes were referred to as the lakes where the water color index value does not exceed 10°, and the pH value is below 5. The experiment on natural water bodies having high contents of humic substances proved that at its first stage the strong acid impact results in changing the chemical structure of large macromolecules and in further loss in of pH value [7].

2. On the one hand, the ratio of molar concentrations of HCO_3^-/ anions (SO_4^{2-}+ NO_3^-) represents the increased load of strong acids; on the other hand, it represents the decreased surge capacity of waters. The change in the concentrations of anions (HCO_3^-/SO_4^{2-}) in the direction of the prevailing position of sulphates is a symptom of water acidification. $HCO_3^- < SO_4^{2-}$ is an evidence of the lake transition to acidification [5], [8].

3. To evaluate the relative significance of the contribution of sulfates and nitrates into water acidification processes, the KNS indicator is proposed, it is determined as the following ratio [9]:

$KNS= NO_3^-/ (SO_4^{2-}+ NO_3^-)$.

In the regions where the determining acidification factor is the occurrence of sulfates, the KNS values are close to 0 and increase as the proportion of nitrates in the anion composition of waters grows.

4. The acid-neutralizing capacity (ANC) is a common criterion of water acidification in the world practice. The difference between the sum of cations (corrected for sea salt), and strong acid radicals represents the reserve or shortage of hydrocarbons,

and in the case of the waters enriched with humic acids it represents their sum, i.e. the surge capacity of the system [10].

The value of water acid-neutralizing capacity can be calculated with two methods:

1. $ANC_1 = Ca^{2+} + Mg^{2+} + Na^{+} + K^{+} - SO_4^{2-} - NO_3^{-}$;
2. $ANC_2 = HCO_3^{-} + A^{n-} - H^{+} - Al^{n+}$,

where A^{n-} is the organic anion concentration.

If all the constituents in the chemical composition of waters are definitely determined and the value of the ion balance is good, the ANC_1 value is the same as the ANC_2 one.

The evaluation of vulnerability for small lakes in the north of West Siberia concerning the acidification process

The first symptom of the acidification process in water bodies is the decrease in pH value, i.e. the increase in the concentration of H^+ ions. However, it is insufficient to use the acidity data as the only criterion. First of all, the pH values and color indexes of water bodies should be evaluated; the most vulnerable ones should be identified. The ion balance of such lakes should be considered; their acid-neutralization capacity and the ion ratio influencing on the surge capacity of waters should be evaluated.

Table 1 presents the data on the acidity and color index of the lakes in the tundra, forest-tundra, and taiga zones of West Siberia.

Table 1

Distribution of the lakes in the north of West Siberia according to the acidity and color index (the number of lakes is given as percentage)

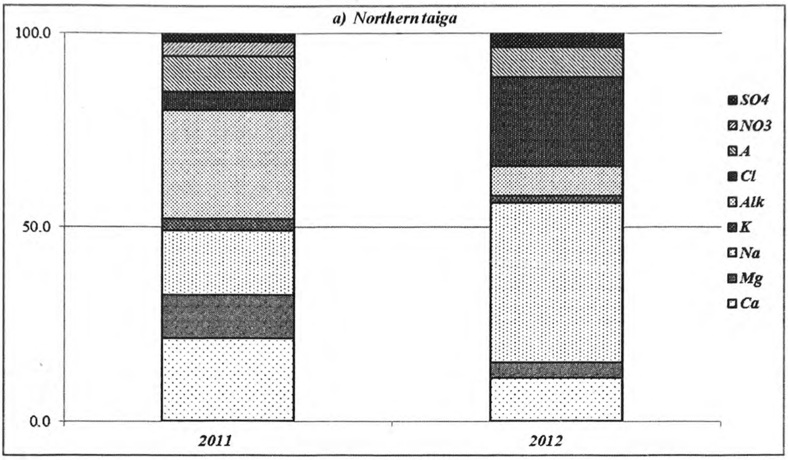
pH	Color index					Number of samples, n
	<10	10 – 30	30 – 60	60 – 100	>100	
Lakes not subject to anthropogenic impact (2011)						
Tundra and forest-tundra						
4 – 5	0	0	0	0	5.8	52
5 – 6	**5.8**	**3.8**	11.5	9.6	1.9	
6 – 7	13.5	30.8	5.8	1.9	0	
7 - 8	5.8	1.9	0	0	1.9	
Taiga						
4 – 5	0	**1.4**	1.4	4.1	5.4	74
5 – 6	**5.4**	**8.1**	14.9	5.4	6.8	
6 – 7	4.1	4.1	4.1	9.5	5.4	
7 – 8	0	9.5	0	0	10.8	
Lakes in the zone of anthropogenic impact (2012)						
Taiga						
4 – 5	**6.3**	**4.2**	4.2	8.3	0	48
5 – 6	**6.3**	**4.2**	6.3	10.4	10.4	
6 – 7	0	10.4	8.3	2.1	2.1	
7 – 8	0	2.1	12.5	2.1	0	

The lakes vulnerable to acidification have low pH and color index values. The analysis of the data on the lakes located far from the sources of anthropogenic impact demonstrates that most of the investigated lakes have the water environment reaction close to the neutral one and the color index values from 10 to 100 degrees (Cr-Co-scale). In the tundra and forest-tundra zones, the fraction of the lakes with pH < 6 and with the color index below 10 degrees (Cr-Co-scale) for baseline lakes is 5.8% (3 lakes), and the one with Col. < 30 degrees (Cr-Co-scale) is 3.8% (2 lakes). There are no lakes with pH < 5 and Col. < 30 degrees (Cr-Co-scale) among the baseline lakes in the tundra and forest-tundra zones. In the taiga zone, one lake (1.4%) has pH < 5 and low color index value (10-30 degrees). The fraction of vulnerable lakes with pH < 6 and Col. < 10 degrees (Cr-Co scale) is 5.4% (3 lakes), with Col < 30 degrees (Cr-Co scale) is 8.1% (6 lakes). Thus, even the baseline lakes in the tundra and forest-tundra zones (9.6%) and those in the taiga zone (14.9%) can be classified as anthropogenically-acid lakes.

The lakes in the zone of anthropogenic impact are more susceptible to acidification. All in all, 21% of lakes have low pH and color index values; therefore, they can be assumed to be exposed to acidification.

To evaluate the acidification resistance of a water body, one should be aware of the contents and the ratio of main ions in it. The ion composition of lake waters determines higher or lower surge capacity of water bodies. The alkalinity primarily characterizes the surge capacity of water bodies. High hydrocarbon concentration of the lake waters is indicative of the water body resistance to acid rains. To characterize the ion composition of lakes in various natural zones, the median values of the contents of main mineralization ions were evaluated, and the diagrams were constructed (Fig. 1).

The ion composition of relatively clean and relatively polluted lakes significantly differs. In the cation composition of the lakes investigated in 2011, calcium dominates; in their anion composition, hydrocarbons dominate. The significant decrease in the fraction of these ions and the increase in sodium and chloride should be noted for the ion composition of the lakes investigated in 2012. Typically, as a result of anthropogenic acidification, the fraction of sulphates and nitrates in the ion composition increases. This is the first symptom of evident acidification. Such a change is not observed, the fraction of sulfates and nitrates is low in almost all the lakes. However, chlorides are also salts of a strong acid, they are not able to demonstrate buffering properties; therefore, chloride waters are more vulnerable to acidification than hydrocarbonate ones. The increased fraction of chlorides can be caused by the bedrock leaching, since many water bodies are located on the territory of the ancient West-Siberian Sea, or by the input of bottom waters with high mineralization. Regardless of the reason, the decrease in the fraction of hydrocarbons in the total ion composition of relatively polluted water bodies compared to the baseline ones is indicative of the chemical processes with such a result.



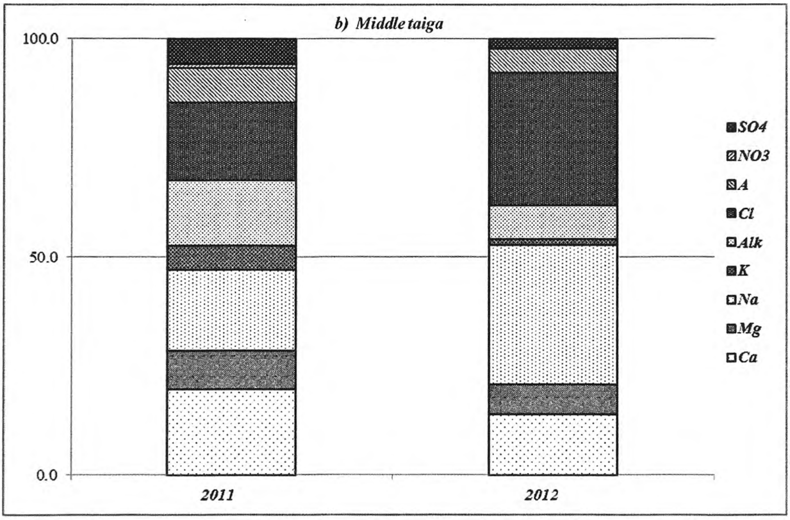


Fig. 1. The ion composition of lake waters in the north of West Siberia for the baseline (relatively clean) lakes, 2011, and the lakes under anthropogenic impact (relatively polluted), 2012, built by median values. a) northern taiga; b) middle taiga

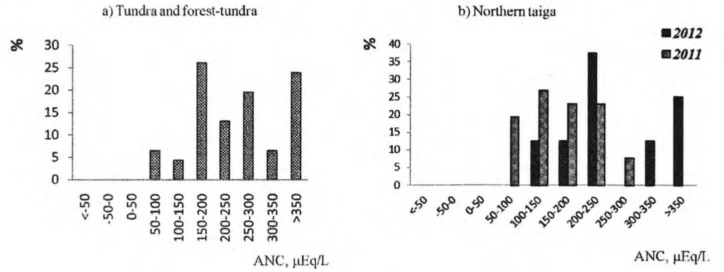
The evaluation of surge capacity and the role of nitrates and sulphates in the acidification process. The ratio of the molar concentrations of HCO_3^- / (SO_4^{2-}+ NO_3^-) anions facilitates the evaluation of the buffering capacity of waters. The larger this value is, the higher the ability of the water body to resist the acidic deposition. KNS, another important indicator of the acidification processes, represents the fraction of the nitrates in the sum of nitrates and sulfates. Table 2 presents the values of these criteria for the lakes investigated in 2011 and 2012. The surge capacity of the most baseline lakes is quite high, and this is confirmed by the data in Table 1. However, for some baseline lakes, the KNS values are close to zero (they are boldfaced in the table). If the pH values are low, and the content of organic anion (A^{n-}) and color index are high, this fact may be the symptom of the lake bogging and its natural acidification.

Among the lakes investigated in 2012 and located in the zone of anthropogenic impact, there are lakes where the ratio of $HCO_3^- / (SO_4^{2-} + NO_3^-)$ concentrations is lower than one, i.e. the anions of strong acids dominate in the ion composition. The KNS values are much lower than those for the lakes investigated in 2011, they differ by one or two orders of magnitude and point to possible acidification of these water bodies.

The evaluation of acid-neutralizing capacity (ANC) of lakes in north of West Siberia. The ANC values for various natural zones were calculated in the task-oriented database which makes it possible to recalculate the mass concentrations of main ions to the equivalent ones and to correct for the impact of marine aerosols.

To calculate the acid-neutralizing capacity, the following equation was used:
$ANC = Ca^{2+} + Mg^{2+} + Na^{+} + K^{+} - SO_4^{2-} - NO_3^{-}$.

The results of the calculations were used to construct the diagrams (Fig. 2) for various natural zones in West Siberia (for the tundra and forest-tundra zones with the correction for the impact of marine aerosols, for the taiga zone without it).


a) Tundra and forest-tundra

b) Northern taiga

c) Middle taiga

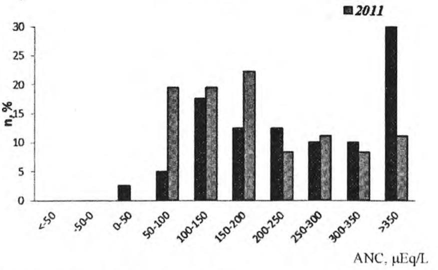


Fig. 2. The distribution of lakes (%) in climatic zones according to the values of water acid-neutralizing capacity (ANC, µEq/L):
a) Tundra and forest-tundra; b) Northern taiga; c) Middle taiga

Table 2

The values of the main criteria for the chemical composition of waters applied to evaluate their acidification (the numerator is the median, the denominator is the variability range) for small lakes in West Siberia

Parameters, μEq/L pH and color index		Alk	H^+	∑ cations	$HCO_3^-/(SO_4^{2-}+NO_3^-)$	KNS $NO_3^-/(SO_4^{2-}+NO_3^-)$	ANC	A^{n-}	SO_4^{2-}	NO_3^-	Number (fraction, %) of lakes
Lakes not subject to anthropogenic impact (2011)											
Tundra and forest-tundra											
<6.0	<10	120	1.32	159	6.88	0.34	76.3	8.23	9.88	6.38	3 (6.5)
		(116-127)	2.26-1.38	120-193	3.25-10.24	0.30-0.72	60.2-80.7	3.75-12.4	8.19-12.2	3.53-25.9	
	10-100	145	2.84	330	3.38	0.71	171	27.5	10.6	28.4	8 (17.4)
		128-186	1.10-5.01	285-421	1.66-4.46	0.26-0.86	155-228	2.76-37.2	7.70-44.4	15.7-66.2	
	>100	81.3	12.6	310	4.21	**0.09**	118	64.6	16.2	1.5	4 (8.7)
		69.7-89.1	8.9-15.5	248-318	3.36-6.52	**0.06-0.11**	106-143	59.7-67.7	12.5-23.5	1.2-2.3	
6.0-8.0	<10	425	0.14	496	11.1	0.45	438	13.2	19.8	14.9	9 (19.6)
		178-619	0.04-0.51	253-570	1.13-20.2	0.10-0.91	206-511	5.89-37.4	8.99-36.8	4.02-144	
	10-100	248	0.42	439	7.06	0.47	279	13.3	13.9	14.2	22 (47.8)
		151-585	0.07-0.95	244-1761	3.96-32.4	**0.02**-0.81	126-591	0.14-48.4	4.58-130	0.08-44.1	
Northern and middle taiga											
<6.0	<10	113	2.71	155	4.90	0.31	91	8.12	22.4	5.48	4 (6.5)
		95-128	1.32-4.90	145-168	2.43-7.63	**0.02**-0.63	70-125	0.75-14.7	6.43-39.5	0.78-10.9	
	10-100	96.8	3.63	269	2.29	0.27	140	41.8	26.3	12.9	27 (43.5)
		0-585	1.10-28.8	185-3490	0-15.5	**0.01**-0.86	76-273	10.3-69.6	5.38-97.3	0.31-76.5	
	>100	87.1	11.0	333	**1.86**	**0.19**	161	82.3	23.5	10.8	13 (21.0)
		0-108	2.51-15.5	248-530	0-6.52	**0.06**-0.77	120-216	59.7-113	11.2-49.7	1.16-37.8	

The end of Table 2

6.0-8.0	<10	145	0.34	183	99.7	0.53	111	3.51	10	1.0	2 (3.2)
		143-147	0.13-0.55	168-197	6.8-192	0.06-1.00	102-120	3.26-3.77	0-20	0.74-1.25	
	10-100	171	0.41	2183	2.70	**0.27**	280	48.2	57.2	9.02	11 (17.7)
		139-1310	0.05-0.79	314-2334	**1.27**-32.3	**0.05**-0.79	174-1505	24.1-60.6	7.12-67.7	2.99-110	
	>100	306	0.40	610	14.8	0.60	302	61.3	15.6	14.6	5 (8.1)
		186-1311	0.04-0.89	318-1652	4.31-33.1	0.09-0.71	285-1525	21.1-76.9	6.10-39.3	3.82-24.3	
Southern taiga											
6-8	10-100	1098	0.18	614	6.16	0.69	445	80.9	33.0	82.0	4 (36)
		217-2832	0.02-0.32	580-3690	1.76-49.3	**0.01**-0.86	-69-3129	50.5-93.4	19.7-73.8	0.27-183	
	>100	4746	0.06	1655	19.7	0.69	1546	69.3	16.2	26.8	7 (64)
		474-5209	0.03-0.20	773-6117	12.2-173	0.46-0.96	691-5444	61.0-97.3	12.0-27.4	12.5-277	
Lakes in the zone of anthropogenic impact (2012)											
Northern and middle taiga											
<6.0	<10	45	9.3	1797	**0.856**	**0.002**	160	19.3	33	0.09	7 (14.6)
		8-65	2.2-15.5	370-2399	**0.238-**1.901	**0.002-0.109**	**71**-271	10.8-42.7	30-60	0.05-3.71	
	10-100	50	8.9	404	2.278	**0.021**	178	75.6	29	0.52	17 (35.4)
		2-235	1.8-31.8	239-2877	**0.050-**16.197	**0.002-0.125**	**29**-2787	5.7-186.8	12-333	0.06-47.79	
	>100	74	7.4	635	1.903	**0.020**	155	104.4	28	0.74	5 (10.4)
		44-116	1.7-9.5	474-5888	1.055-4.796	**0.006-0.181**	108-308	86.0-116.7	22-52	0.13-7.12	
6-8	10-100	351	0.2	1287	17.994	**0.019**	618	75.8	21	0.81	18 (37.5)
		118-1350	0.0-1.0	600-19509	**0.210-**88.841	**0.003-0.651**	223-8022	46.6-139.6	3-1341	0.16-30.15	
	>100	231	0.6	3778	6.847	**0.193**	455	145.7	27	6.51	1 (2.1)

According to the results of calculating the acid-neutralizing capacity of natural water bodies in the northern natural zones of West Siberia, about 20% of relatively clean lakes have ANC values in the range of 50-100, which indicates their potential vulnerability to acid deposition. In the middle taiga zone, where many oil-producing companies operate, there are lakes with the ANC values in the range of 0-50, which is critical for natural water bodies. However, most of the lakes retain a significant surge capacity and have hydrocarbon reserve; it concerns both the relatively clean and anthropogenically impacted water bodies in the northern regions of West Siberia.

Conclusion. The large-scale study of the acidification processes in the small lakes of West Siberia demonstrated that about 15% of baseline lakes are vulnerable to the deposition of acid-forming substances and have a low self-regeneration capacity. First of all, these lakes include the low-mineralized water bodies in the northern and middle taiga zones. Most of the baseline lakes are classified as hydrocarbonate ones and have a good surge capacity.

In the zone of anthropogenic impact, there are water bodies, which have critically low values of acid-neutralizing capacity. Chlorides predominate in the anion composition of the lakes located in the zone of anthropogenic impact. The calculation of acid-neutralizing capacity of these lakes demonstrates that about 20% of them are vulnerable to fallout of acid-forming substances, but most of them (about 80%) have a significant reserve of acid-neutralizing capacity, due to the anions of weak organic and inorganic acids present in their structure (ANC > 100).

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