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Migration of chemical elements in groundwater of the mining area

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Abstract. ***The relevance of the*** study is conditioned by the necessity of studying the forms of chemical elements migration in underground waters of flooded copper and copper mines for the correct understanding and forecasting of the processes of their transport and distribution in hydrogeochemical fields. Ground and surface waters are complex mixtures of substances in which, depending on pH, Eh and t °C, phase transitions with subsequent dissolution or precipitation of minerals are formed. To solve these problems, numerical hydrogeomigration modelling is used, including physico-chemical modelling, which makes it possible to draw conclusions about the scale of pollution and localisation of such sites for subsequent development of measures to improve the state of the hydrosphere. ***Objective: to*** determine the forms of chemical elements migration in groundwater and to calculate water saturation indices in relation to minerals. ***Objects:*** groundwater in the territory of the closed Levikhinsky copper and copper-coal mine. ***Methods:*** laboratory studies of groundwater were carried out using the methods of flame emission spectrometry, flame atomic absorption, photometric method with Nessler's reagent, titrimetric, mercumetric and potentiometric methods; mass spectrometry with inductively coupled plasma ionisation and gravimetric method. Visual MINTEQ 3.1 software was used for physical and chemical modelling. ***Result.*** According to the results of processing of chemical analyses and physico-chemical modelling all tested wells are divided into groups. The first group - wells No. 1, 2, 3 and 6, located within the former mining allotment (near Sh. "Le-vikha XIV, the technogenic water reservoir, Leviha II and the neutralisation station). The second group is represented by water in wells No. 4 near the shaft of the Tsentralnaya shaft and No. 5 near the Levikha II shaft and the neutralisation station. "Tsentralnaya" and No. 5 near the "Yuzhny" dump. The third group is well No. 7 located near the mouth of the Levikha River. Saturation indices and forms of migration of components in the aquatic environment allow to reveal the scale of pollution and localisation of such sites. It was found that under the current collection and treatment system there is no large-scale pollution of groundwater at the Levikha mine. It is localised in the area of the Centralny mine shaft (well No. 4) and the Yuzhny dump (well No. 5).

: hydrosphere, migration, , groundwater, Visual MINTEQ 3.1

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Chemical elements migration in ground waters of mining territory

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Abstract. ***Relevance.*** The need to study the forms of migration of chemical elements in the groundwater of flooded copper pyrite mines in order to correctly understand and predict their transfer and distribution in hydrogeochemical fields. Ground and surface waters are a complex mixture of substances in which, depending on pH, Eh and t °C, phase transitions are formed with subsequent dissolution or precipitation of minerals. To solve these problems, numerical hydrogeomigration modelling is used, including physicochemical, which allows drawing conclusions about the scale of pollution and the localisation of such areas for subsequent development of measures to improve the state of the hydrosphere. ***Aim.*** To determine the forms of me- tal migration in groundwater and to calculate water saturation indices in relation to minerals. ***Objects.*** Groundwater in the territory of the closed Levikha copper pyrite mine. ***Methods.*** Laboratory studies of groundwater were carried out using flame emission spectrometry, flame atomic absorption, photometric method with Nessler's reagent, titrimetric, mercumetric and potentiometric methods; mass spectrometry with ionisation in inductively coupled plasma and gravimetric method. Physical and chemical modelling using the software product Visual MINTEQ 3.1. ***Results.*** According to the results of processing chemi- cal analyses and physical and chemical modelling, all tested wells are divided into groups: 1) wells no. 1, 2, 3 and 6, located within the former mining allotment (near the Levikha XIV mine, a man-made reservoir, the Levikha II mine and a neutralisation station); 2) represented by water in wells no. 4 near the shaft of the Tsentralnaya mine and no. 5 near the dump Yuzhny;
3) well no. 7, located near the mouth of the Levikha river. Saturation indices and forms of migration of components in the aquatic environment make it possible to identify the scale of pollution and the localisation of such areas. Thus, with the cur- rent pumping system, there is no large-scale pollution of groundwater at the Levikha mine. It is localized in the area of the Tsentralny mine shaft (well no. 4) and the Yuzhny dump (well no. 5).

Keywords: hydrosphere, migration, pollutants, groundwater, Visual MINTEQ 3.1

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Introduction

The chemical composition of groundwater depends on a number of factors, such as: natural conditions of the territory; specific features of recharge and discharge; composition and filtration properties of water-bearing rocks [1-3]. Groundwater composition is formed as a result of rock-water interaction, solid phases (inorganic and organic substances) are both sources and sinks of dissolved components of groundwater [4].

The study of the forms of chemical elements in groundwater of different compositions allows us to correctly predict the processes of their transport and distribution in hydrogeochemical fields. This aspect is important in the study of dispersion fluxes and assessment of their anthropogenic impact on the hydrosphere [5]. For example, the fact that different migration forms of one and the same element have different toxicity is of great importance in solving the issues of water bodies protection. For example, cationic forms of copper (${}^{(2+}{}_{)}$, $CuOH^+$, $Cu_{(}{}^{2)}(OH)$ ${}^{2+}{}_{)}$) are significantly more dangerous than its neutral or anionic forms [6]. Also at definition of chemical elements in water it is necessary to take into account that many methods of analytical definition (especially colourimetric) are developed only on their certain chemical states, therefore there are contradictions between forms of definition of elements at chemical analysis of underground waters and their real state. As a consequence, elements may be detected in waters that are not all elements, which leads to unreliability of the obtained data. It is therefore necessary to know and predict the probable states of elements in groundwater [7, 8].

Thus, the aim of the work is to determine the forms of metal migration in groundwater of the flooded Levikhinsky mine and to calculate water saturation indices in relation to minerals.

Object of study

The Levikha group of copper-kohlcedan deposits is located in the Sverdlovsk region, 30 km northwest of Kirovgrad, on the eastern outskirts of the Levikha settlement (Fig. 1).

The deposits were discovered in the 1920s. Exploitation was carried out both by open-pit and underground methods. Flooding of mine workings began in 2003 [9]. By 2007, a man-made pond was formed on the northern flank of the mine in a 20 m deep sinkhole (at the lowest point of the mining allotment), where mineralised acid mine water (AMW) with a flow rate of about 120 m^3/h is discharged [10].

Since 2007, in order to prevent environmental pollution after the mine flooding, a system has been organised to collect CSW, pump it from the technogenic reservoir, neutralise it with lime milk, and discharge it into a clarifier pond. Then the water flows by gravity along the old channel of the Levikha River into the Tagil River [10].

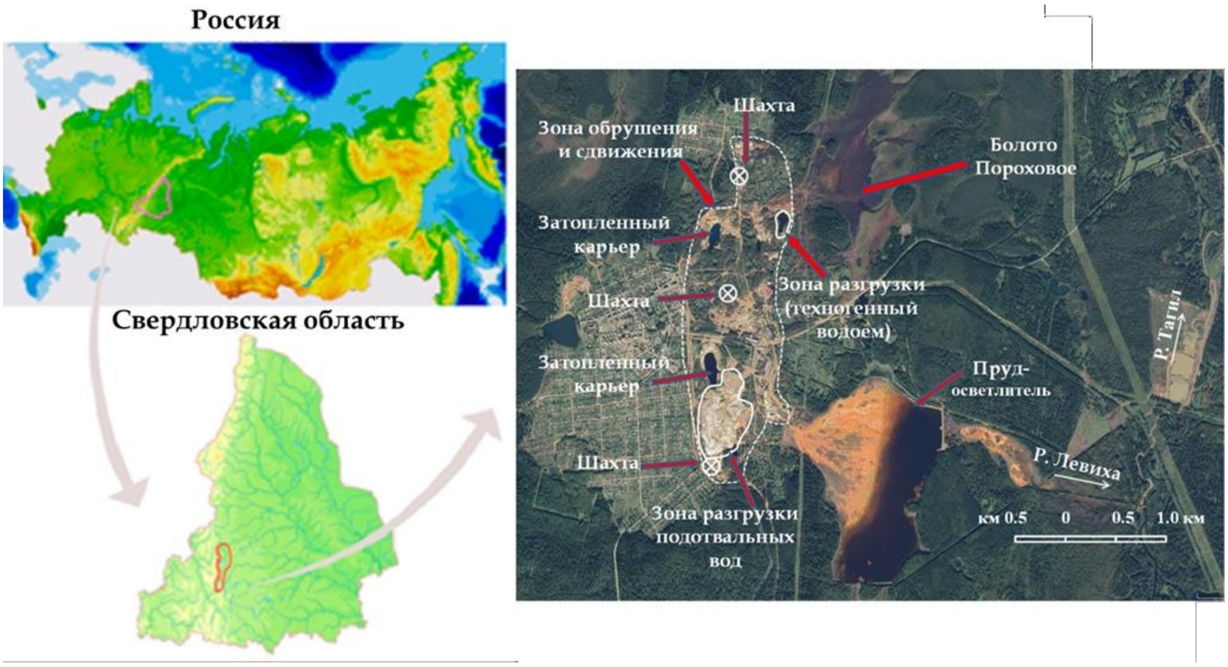

Figure 1. *Overview of the layout of the facilities at the Levikhinsky mine*
Fig. 1. *Overview layout of objects at the Levikha mine*

In 2003, the drainage was stopped, the depression funnel was filled, and by 2007, a man-made pond was formed at the lowest point of the mining claim, into which acidic (pH=2.7-4.1) mineralised (up to 12 g/l) sulphate mine water with a high content of Fe, Al, Mg, Zn, Cu, Mn is discharged. The flow rate of such discharge is about 120 m^3/h - 2 times less than the water discharge during the mining period.

Since 2007, neutralisation of the KShVW has been resumed: mine water is pumped from the technogenic reservoir to the neutralisation station and treated wastewater is discharged into a clarifier pond. It is discharged into the Levikha River, a left tributary of the Tagil River (Irtysh basin district).

Research Methods

To characterise the groundwater composition in the area of the Levikha copper and iron ore mine May 2021, observation wells with depths ranging from 30 to 50 m were drilled within the former mining allotment (6 wells) and downstream of the discharge from the clarifier pond at the mouth of the Levikha River (1 well) (Figure 2).

The chemical composition of groundwater for the expanded list of components was investigated based on the results of seasonal quadruple sampling during 2021. The analyses were carried out in a certified laboratory at the chemical-analytical centre of the Institute of Industrial Ecology of the Ural Branch of the Russian Academy of Sciences (Ekaterinburg) using flame-emission spectroscopy methods. metrics, flame atomic absorption, mass spectrometry with ionisation in inductively coupled plasma. About 26 components were determined in each sample. Temperature (t °C), redox potential (Eh, mV), pH, total dissolved solids (TDS, ppm), electrical conductivity (EC, mS) were measured in situ.

Determination of migration forms of components and processes of groundwater composition formation was carried out by numerical calculations using the Visual MINTEQ programme for equilibrium physical and chemical modelling of geochemical processes in the water-rock system.
3.1 [11-15].

For each mineral the saturation index SI (saturation indices) is calculated, which is defined as the difference between log IAP (ion activity product) and log Ks (solubility constant of the mineral included in the thermodynamic database):

$$SI= \log IAP-\log .Ks$$

The saturation index SI provides information about the state of the solution relative to the solid phase. At SI<0 the solution is unsaturated; at SI=0 the solution is in equilibrium with the solid phase; at SI>0 the solution is supersaturated. If the solution is unsaturated, dissolution of the corresponding solid phase is predicted. If the solution is supersaturated, the most probable reaction is the precipitation of dissolved substances from the solution [6, 16].

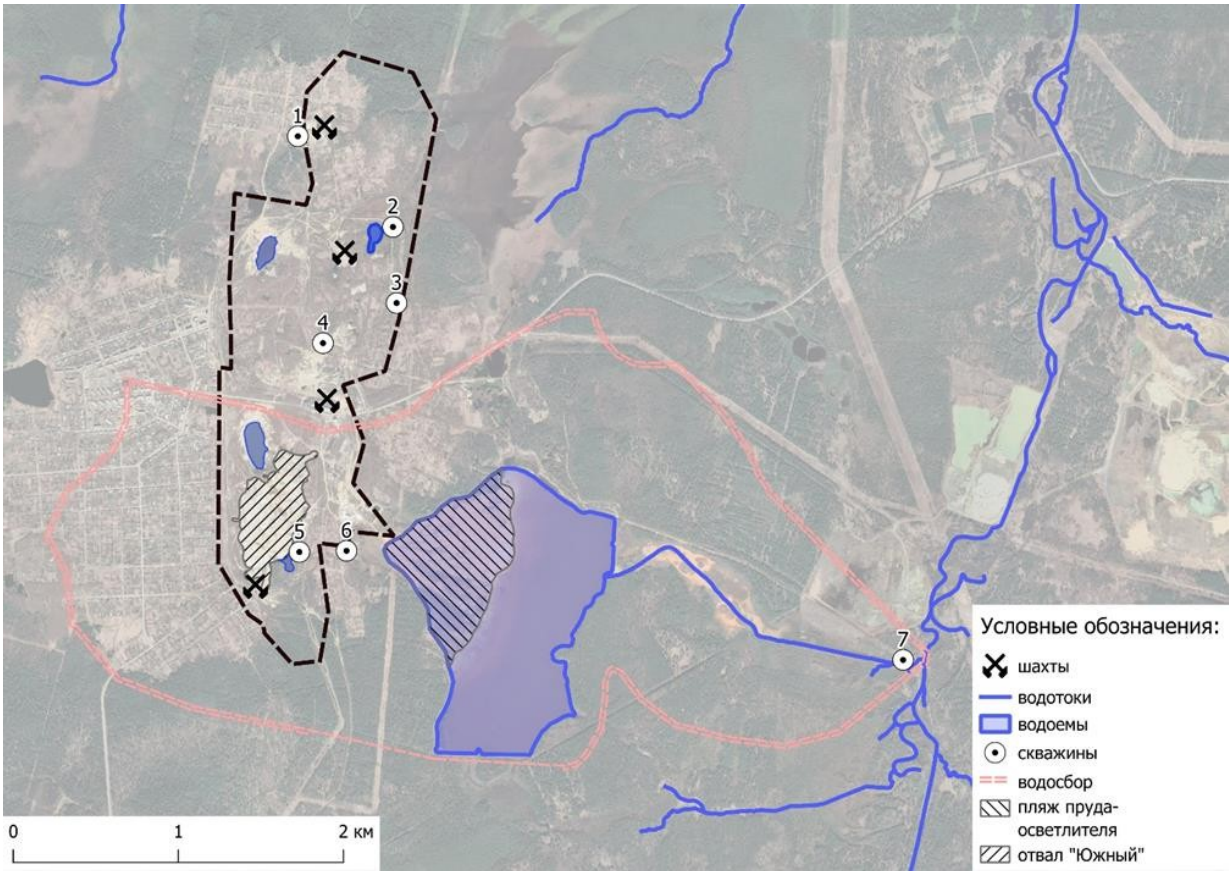

Figure 2. *Scheme of well location: 1 - near sh. "Levikha XIV"; 2 - near the technogenic reservoir; 3 - near the Levikha II shaft; 4 - near the borehole of the Levikha II shaft. "Levikha II"; 4 - near the shaft of the "Tsentralnaya" shaft; 5 - near the shaft of the "Yuzhnaya" shaft. "Tsentralnaya"; 5 - near the dump "Yuzhny"; 6 - near the neutralisation station; 7 - below the discharge from the clarifier pond, at the mouth of the Levikha river.*

Fig. 2. *Well location diagram: 1 - near the mine Levikha XIV; 2 - near the collapse zone; 3 - near mine Levikha II; 4 - near the shaft of the Tsentralnaya mine; 5 - near the Yuzhny dump; 6 - near the neutralisation (treatment) station; 7 - be- low the discharge from the clarification pond, at the mouth of the river Levikha*

Results and discussion

Hydrochemical characteristics of groundwater Mineralisation in groundwater varies from 0.2 to 2.0 g/l (Table 1). The exception is well No. 4 near the shaft of the Tsentralnaya shaft (mineralisation up to 13.0 g/l). "Tsentralnaya (mineralisation up to 13.7 g/l) and No. 5 near the Yuzhny dump (mineralisation up to 35.3 g/l). The high mineralisation and its variability in well№ 5 are influenced by acidic sub-dump water flowing from the Yuzhny dump. In its turn, the chemical composition of the sub-dump water strongly depends on the season and precipitation for the preceding days [17].

Anionic composition of groundwater is sulphate, cationic composition is magnesium-calcium or iron-aluminium. Exceptions are wells№ 1, 3, 7. In wells No. 1, 3 water has hydrocarbon-chloride-sulphate composition; in terms of cations water is sodium-calcium-magnesium and magnesium-calcium. In well№ 7 water has hydrocarbonnate-sulphate composition; by cations water is magnesium-calcium. In wells№ 1, 2, 3, 6 water is weakly-acidic (pH from 5.0 to 6.4). Water in wells№ 4 and 5 acidic (pH from 2.8 to 3.5). Actual pH values are neutral in well No. 7 (up to 7.0). Waters in wells№ 2, 4, 5 are in oxidising conditions (Eh>+100 mV values), free oxygen is present in water, elements migrate in the highest form of their valence. Waters in the remaining wells are in transitional redox conditions: Eh values vary from 0 to +100 mV, in conditions of unstable geochemical regime, when both weak oxidation and weak reduction of a number of metals occur (Table 1, Fig. 2, 3).

Metal concentrations in groundwater exceed concentration clarks (CC) [18] by one to five orders of magnitude (Fig. 4). The most significant exceedances of CCs (thousands of times) are observed in wells No. 4 and 5 for Al, As, Be, Cd, Cu, Co, Co, Fe, Mg, Mn, Ni, Zn. In well No. 2, the CK values decrease by one or two orders of magnitude. In wells No. 1 and No. 6, clark exceedances (hundreds to tens of times) were found for Cd, Cu, Co, Mn, Ni and Zn. In well№ 7 exceedances of Be, Ca, Co, Mg, Mn and Ni are noted (Fig. 4).

Table 1. *Water composition in wells (sampling dates 26.05.2021-24.09.2021)*

Table 1. *Composition of water in wells (sampling dates 26.05.2021-24.09.2021)*

№ Well no.	Water composition (Kurlov formula) Water composition (Kurlov)	Mineralisation, g/L Mineralisation, g/L	pH	Eh, mV	t, °C
1	$\frac{SO_4\,72\;Cl\,21}{Mg\,48\;Ca\,34\;Na\,13}$	0,1-0,2	5,1-5,4	98-103	8,0-9,3
2	$\frac{SO_4}{Ca\,55\;Mg\,27}$	0,5	5,0-5,1	111-124	7,7-9,0
3	$\frac{SO_4\,65\;Cl\,23\;HCO_3\,12}{Ca\,68\;Mg\,23}$	0,5	6,2-6,4	48-58	8,0
4	$\frac{SO_4\,99}{Al\,44\;Mg\,21\;Fe^{2+}\,17}$	10,7-13,7	2,9-3,5	206-210	9,0-11,2
5	$\frac{SO_4\,99}{Al\,43\;Fe^{2+}\,26\;Mg\,20}$	12,9-35,3	2,8-3,2	233-237	9,0-12,0
6	$\frac{SO_4}{Ca\,73\;Mg\,19}$	1,9-2,0	6,1-6,2	58-64	7,4-8,4
7	$\frac{SO_4\,65\;HCO_3\,33}{Ca\,71\;Mg\,24}$	0,8-0,9	6,7-7,0	18-30	9,1-8,5

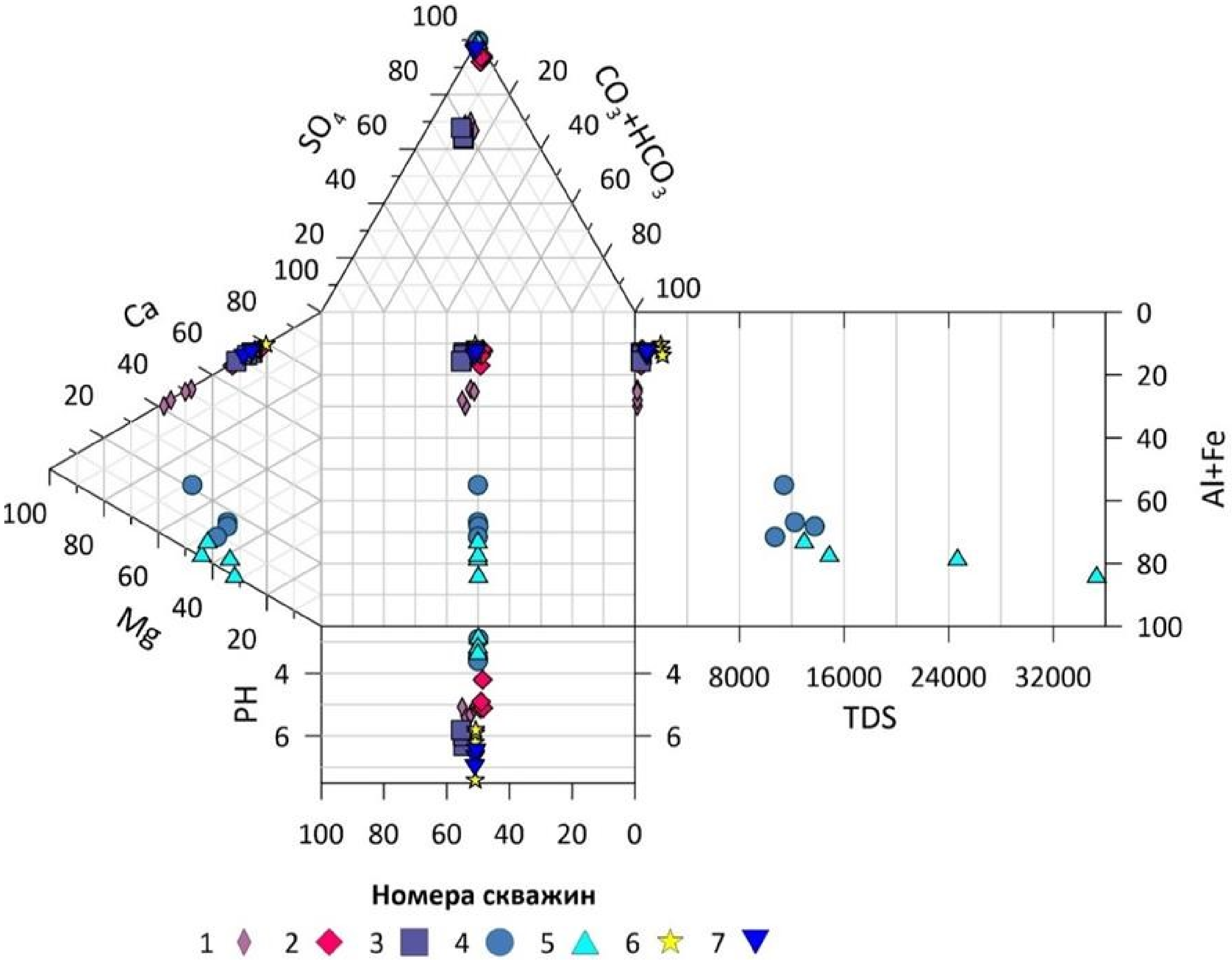

***Figure 3**.* *Durov diagram (%-eq/l)*

Fig. 3. *Durov diagram (%-eq/L).*

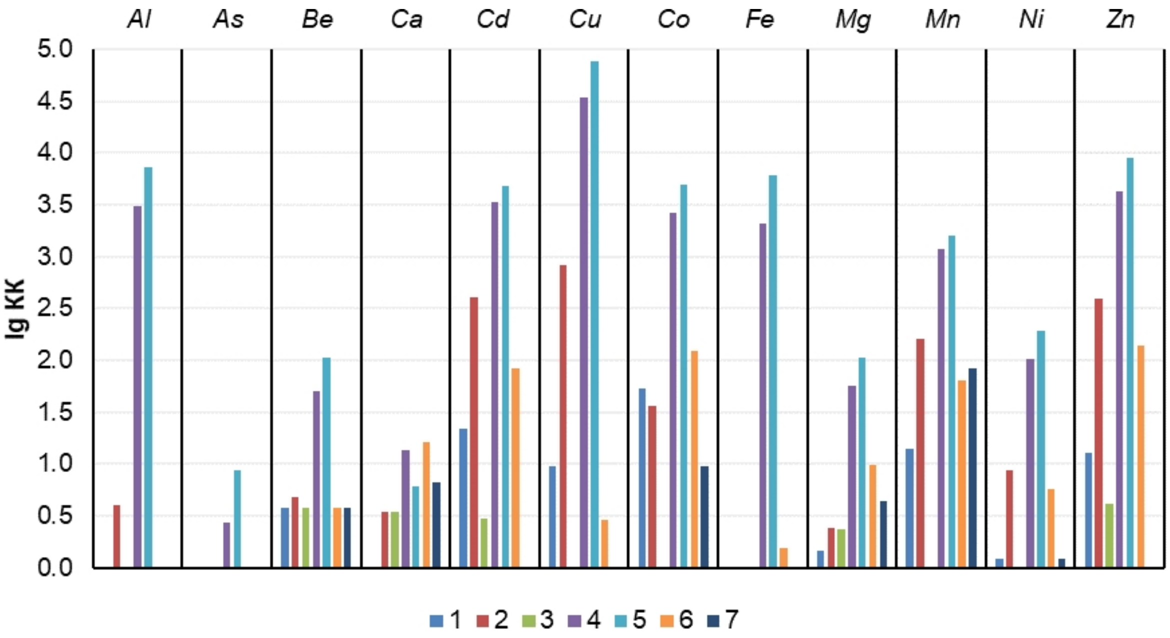

Fig. 4. *Geochemical spectrum of elements in water of sampled wells (lg CC)*
Fig. 4. *Geochemical spectrum of elements in the water of tested wells (lg CC)*

Element migration patterns and results of saturation index calculations

The main chemical forms of element migration in groundwater depend on their valence. Thus, according to thermodynamic calculations, single-charged cations (sodium and potassium) migrate mainly in the form of free ions (on average more than 95%) (Fig. 5). Double-charged cations (calcium, magnesium, manganese, zinc) migrate in the ion-dissolved form (more than 55 %), less often in the form of sulphate complexes.

Fe is a transition element and, depending on the redox potential, has an oxidation degree of 2+ or 3+ [18-20]. Thus, Fe^{2+} is either in free form (51 to 91 %) or in complex with $FeSO_4$ sulphates$^+$ (up to 56 %). The form of Fe^{3+} migration is determined by the acidity of the medium: in weakly acidic and neutral medium, these are hydroxide complexes $(FeOH_{(2)})^+$, $Fe(OH)_2^+$), in acidic medium, these are sulphate complexes ($FeSO_4^+$, $FeSO_{(4)}^{)(2-))}$.

Al can form the largest number of diverse complexes, except for wells No. 4, 5 with acidic water, where migration forms are mainly in the sulfate group (Fig. 5).

Thermodynamic calculations have shown that the subsurface waters are supersaturated with respect to various aluminium hydroxides and sulphates, iron oxide hydroxides and oxides, as well as carbonates (Table 2).

The water in all wells is saturated with sulphate group minerals. The water in the wells Water in wells No. 1, 2, 3, 5, 6 (neutral water$_{)}$ is oversaturated with $Al_4(OH)_{10}SO_4(s)$ and alunite. Water in wells No. 4, 5 (acidic waters) is supersaturated with respect to jarosite (Table 2).

In relation to oxide group minerals, water is saturated in wells No. 3, 4, 5, 6 and 7. Water in wells No. 3, 6, 7 is supersaturated with respect to hercinite. In wells№ 4, 5 water is supersaturated with respect to hematite and magnetite (Table 2), also acidic water and water in well No. 7 is supersaturated with respect to copper ferrite.

Waters practically in all wells show saturation with respect to minerals of the oxide-hydroxide group. In neutral waters there is oversaturation of diaspore (up to 3.4). In well No. 7 the supersaturation is also noticeable for bemite (up to 1.6). In acidic groundwater, supersaturation (up to 3.4) for goethite and (up to 3.1) lepidocrocite (γ-FeO(OH)) is noted (Table 2).

The water saturation indices for aluminium hydroxide and gibbsite (hydrargillite) are in the range from 0.4 to 2.5 (Table 2).

Water saturation indices with respect to carbonate minerals are the lowest: in well No. 7 the saturation index for manganese carbonate is 0.1 and for rhodochrosite 0.6 (Table 2).

In relation to minerals of the halogen group, water is saturated in wells No. 3, 4, 5, 6 and 7. The highest degree of oversaturation is noted for $Fe(OH)_{(2.7)}Cl_3(s)$ in wells No. 4 and 5, for which saturation indices reach 5.4 and 4.9, respectively (Table 2).

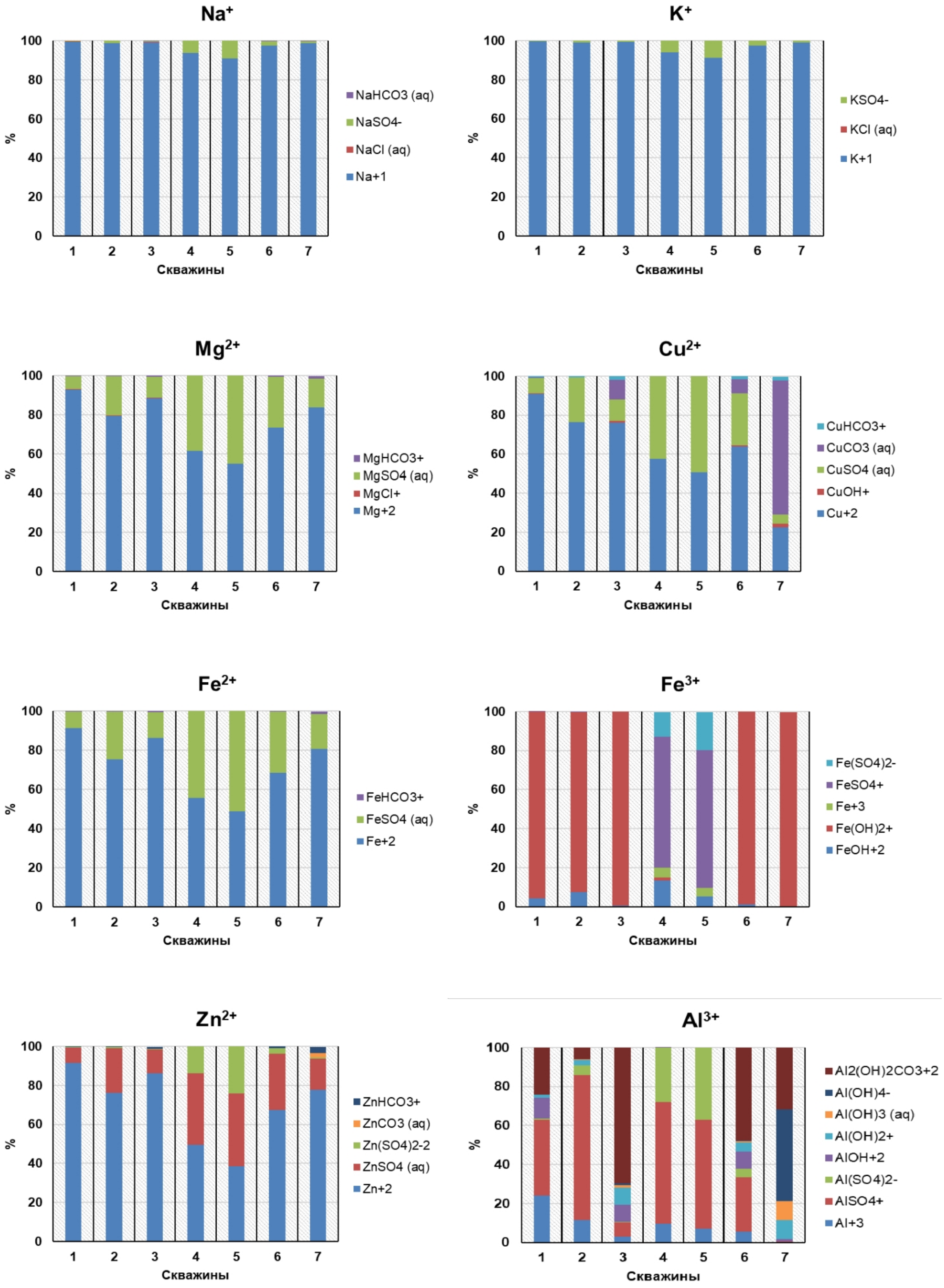

Figure 5. *Elemental migration patterns*
Fig. 5. *Forms of element migration*

***Table 2.** Results of calculation of groundwater saturation indices*

***Table 2**. Results of calculation of groundwater saturation indices*

Mineral Mineral	Formula Formula	Group Group	Well numbers/Well numbers						
			1	2	3	4	5	6	7
$Al(OH)_3$ (Soil) $Al(OH)_3$ (Soil)	$Al(OH)_3$	hydroxides hydroxide	0,4	-	1,2	-	-	0,7	1,9
Gibbsite (C) Gibbsite (C)	γ-$Al(OH)_3$		0,9	0,5	1,8	-	-	1,3	2,5
$Al_4(OH)_{10SO4}$(s)	$Al_4(OH)_{10SO4}$	sulfates	2,6	2,1	4,5	-	-	3,6	5,7
AlOHSO4(s)	AlOHSO4		-0,8	-0,2	-	0,9	0,9	-	-
Alunite Alunite	KAl3 $(OH)_6(SO_4)_2$		1,9	2,3	2,1	0,5	-	2,4	2,3
Jarosite K-Jarosite	$KFe^{3+}_3(OH)_6(SO_4)_2$		-	-	-	5,8	4,6	-	-
Boehmite Boehmite	γ-AlO(OH)	oxide-hydroxides oxide-hydroxides	0,0	-0,5	0,8	-	-	0,3	1,6
Diaspore Diaspora	AlO(OH)		1,8	1,4	2,7	-	-	2,2	3,4
Ferrihydrite Ferrihydrite	$(Fe^{3+})_{2O3\text{-}0}$.5H2O		-	-	-	0,3	-0,3	-	-
Goethite Goethite	FeO(OH)		-	-	-0,8	3,4	2,8	-0,3	1,2
Lepidocrocite Lepidocrocite	γ-FeO(OH)		-	-	-1,0	3,1	2,6	-0,5	0,9
Haematite Hematite	Fe_2O_3	oxides	-	-	0,8	9,1	7,9	1,8	4,7
Hercynite Hercynite	$FeAl_2O_4$		-	-	1,8	-	-	1,3	5,0
Magnetite Magnetite	Fe3O4		-	-	1,9	7,6	6,2	3,3	7,6
Cupric Ferrite	CuFe2O4		-	-	-	4,1	2,7	-	2,8
Maghemite Maghemite	γ-Fe_2O_3		-	-	-	2,6	1,5	-	-
MnCO3 (am)	MnCO3	carbonates carbonates	-	-	-	-	-	-	0,1
Rhodochrosite Rhodochrosite	MnCO3		-	-	-	-	-	-0,9	0,6
$Fe(OH)_{2.7Cl3}$(s)	$Fe(OH)_{2.7Cl3}$	halides halides	-0,5	-	0,7	5,4	4,9	1,1	2,1

Note: "-" saturation index is less than -1/Note: "-" saturation index is less than -1.

Water in wells 1, 2, 3 and 6 is characterised by identical saturation indices for minerals of the sulphate, hydroxide and carbonate groups. They are over-saturated with respect to $Al_4(OH)_{10}SO_{(4)}$(s) (from 2.1 to 4.5), alunite (from 1.9 to 2.4), diaspore (from 1.4 to 2.7) and gibbsite (from 0.5 to 1.8). The second group is water in wells№ 4 and 5, here the supersaturation in relation to minerals of groups oxides, sulphates, oxide-hydroxides and halides is noted: hematite (9.1-7.9), magnetite (7.6-6.2), jarosite (5.8-4.6), goethite (3.4-2.8) and lepidocrocite (3.1- 2.6). In the third group (well No. 7) there is a slight oversaturation in carbonate group minerals ($MnSO_3$(am) and rhodochrosite), as well as in $Al_4(OH)_{10}SO_{(4)}$(s) 5.7), gibbsite (2.5) and diaspore (3.4) (Table).

Thus, based on the results of chemical analyses and physicochemical modelling, all the tested wells can be divided into three groups. The first one is wells No. 1, 2, 3 and 6 located within the former mining section. water (near the Levikha XIV sh. "Levikha XIV, technogenic water reservoir, Levikha II and neutralisation station) are low and slightly mineralised (up to 2 g/l) sulphate magnesium-calcium and calcium-magnesium waters. Actual pH values in the water of wells No. 1, 2, 3, 6 are moderately acidic and slightly acidic.

In addition, they are characterised by identical saturation indices for minerals of the sulphate, hydroxide and carbonate groups. They are oversaturated with respect to $Al_4(OH)_{10}SO_4$(s), alunite, diaspore and gibbsite. The second group is represented by water in well No. 4 near the borehole of the Tsentralnaya shaft (Alunite, diaspore and gibbsite). "Tsentralnaya" (mineralisation 12.2 g/l) and№ 5 near the waste dump

"Yuzhny" (mineralisation 24.7 g/l), acidic sulphate magnesium-aluminium or iron-aluminium waters. They are supersaturated with respect to minerals of oxide, sulphate, oxide-hydroxide and halide groups: haematite, magnetite, jarosite, goethite and lepidocrocite.

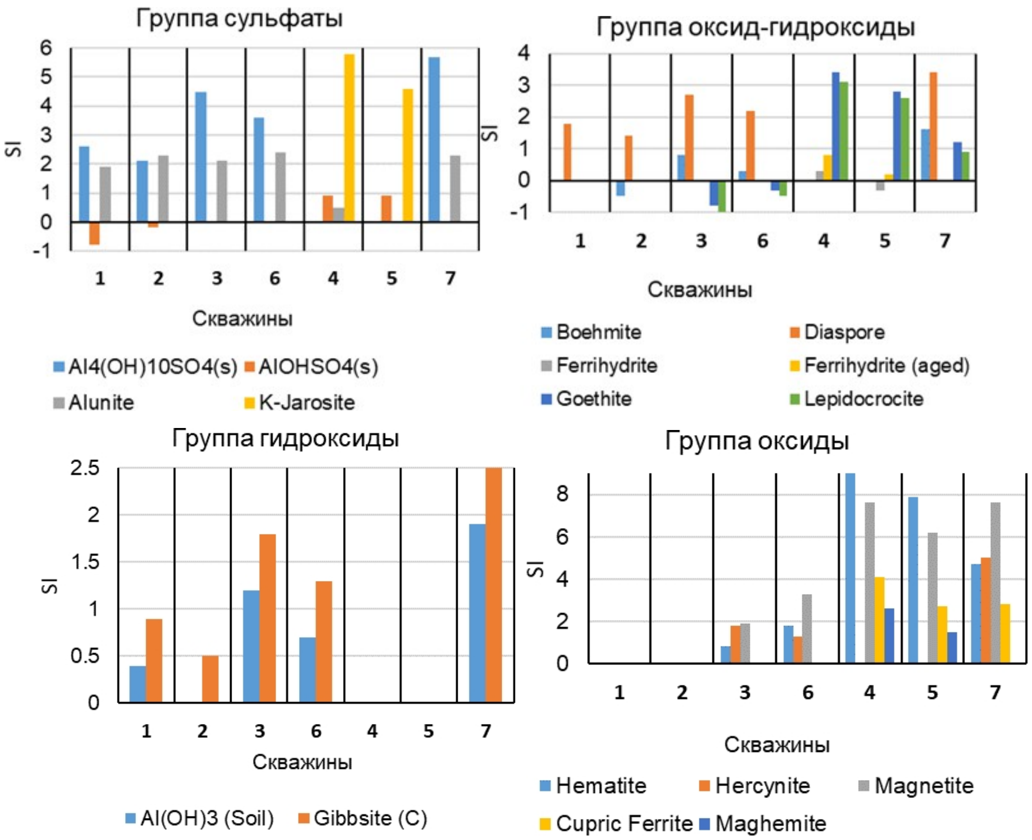

Figure 6. *Water saturation indices in relation to minerals*

Fig. 6. *Water saturation indexes with respect to minerals*

The third group is water of well No. 7 with neutral medium, located near the mouth of the Levikha River, slightly mineralised (up to 0.9 g/l) hydro-carbonate-sulphate magnesium-calcium. In water there is a slight supersaturation in carbonate group minerals (MnSO$_3$(am) and ro-dochrosite), as well as in $Al_4(OH)_{10}SO_4$(s), gibbsite and diaspore (Fig. 6).

Conclusions

Underground waters of the Levikhinsky mine are mainly magnesium-calcium sulphate or iron-aluminium waters. In spite of neutral and slightly acidic pH values in the tested wells, the content of heavy metals in them reaches high values, exceeding clark concentrations by one to five orders of magnitude. Nevertheless, for the toxicity of the pro- The complexation of elements in water is more important than the total concentration. Metals are highly toxic in the form of free ions, but are less toxic at the same concentrations when present as complexes.

Calculations of saturation indices relative to min- eral have revealed that groundwater is supersaturated with respect to alunite and $Al_4(OH)_{10}SO_4$. Acidic waters, unlike neutral and slightly acidic waters, have high SIs for jarosite, goethite, copper(II) ferrite and maghemite.

The obtained results of processing of chemical analyses and modelling allowed to divide the sampled wells into groups and identify the scale of contamination and locations of such sites on the territory of the Levikha copper and chedan mine.

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