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Water Extracts from Waste Rocks of the Coal Industry of Chernvonograd Mining Area (Ukraine) – Problems of Environmental Safety and Civil Protection

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ABSTRACT

The article presents the results of the research on mining production waste in the Chervonohrad Mining Area. For the first time the water extracts from certain types of waste rock, namely: burned and unburned argillite, siltstone, siliceous siltstone, coal, and sandstone were investigated. The studies covered the main chemical composition, as well as the pH and concentration of the main components of the aqueous extracts. Based on the obtained results, the properties of the investigated mining wastes were characterized, taking into account the impact on the environment. According to the obtained results, it can be stated that there is high content of Mg^{2+}, SO_4^{2-}, HCO_3^- in argillite and sandstone within the tericon of the central coal enrichment plant "Chervonohradska" and burnt argillite from the tericon of the Vizeyska mine.

Keywords: environmental safety, coal mining, tericon, waste rock, chemical properties, water extraction.

INTRODUCTION

The environmental safety rate and the risks of emergency situations are largely determined by the pollution of the hydrosphere by insufficiently treated municipal wastewater [1,2] and industrial effluents [3,4], pollution of the lithosphere by domestic solid waste [5,6] and industrial [7,8] waste. A significant deterioration of quality of the natural environment (both at the local and regional levels) is observed in conditions of intensive development of the mining industry [9,10]. Production activities related to hard coal mining and its beneficiation within the limits of the Chervonohrad Mining Area (ChMA) of the Lviv-Volyn Coal Basin (LVCB) are accompanied by the formation of underground cavities, which subsequently cause subsidence and flooding of territories [11–13] accumulation of significant masses of empty waste rock, which leads to the formation of rock dumps (tericons) near mines, which are capable of spontaneous combustion [14,15] and are a source of toxic elements and compounds in the environment. In addition, coal is trerated, so large fractions of waste (more than 50 mm) are stored in a dump, and fine fractions (0.1–0.2 mm) are stored in tailings. Since these wastes belong to the 4th class of danger, they are stored in the open space, and their properties change under the influence of exogenous processes, in particular atmospheric precipitation, pressure drops, temperature, circulation of air masses, etc. As a result, potholes, dips, and cracks are formed on the body of the dump, through which atmospheric precipitation flows down, carrying dissolved chemical components from the surface of the dump, penetrating through their thickness, turning into acidic infiltrates with mineralization from 3 to 30 g/dm^3, and solid substances are washed away in the form of finely dispersed particles [16–19]. Climatic changes and temperature fluctuations provoke the processes of physical, chemical and biological weathering of rock masses. Under the influence of environmental factors, tericon has a negative impact on the physico-chemical, biological and ecological processes at its location [19]. Terykons are ecologically dangerous objects that have a negative impact on the environment and lead to a disturbance of the natural balance, namely, they can affect the chemical composition of underground and surface waters [20, 21], as well as soils [22]. It is worth noting that the territory of the mining region is located within the Vistula River basin, the water network of the region is created by the Zahidny Bug River and its left largest tributaries Rata and Solokya, and is exposed to the influence of mining enterprises, tericons, water storage tanks, sludge storage tanks, and tailings, which are a potential source of pollution of surface and of underground reservoirs in the region [12].

Unfortunately, the physico-chemical properties of the waste rock have been studied at an insufficient level. A significant part of scientific works [20, 23, 24] is devoted to the investigation of subterrycon waters. It is reported [19] that waters near the foot of the rock dumps are in unsatisfactory condition. The analysis of these works also allows us to assume that there is a migration of harmful substances from the surface of tericons resulting in soil and groundwater pollution. It should be emphasized that the chemical content of the dump rock have an effect on their composition, because on its way, wastewater is enriched with products of the rocks destruction (sulfates and salts of alkaline earth elements) and coal, i.e. it acquires new physicochemical properties, which strongly influence the quality indicators of the environment. In this regard, there is no doubt about the need to carry out an ecological assessment of the main factors of hazard caused by the impact of atmospheric precipitation on waste rock within the ChMA. The object of research is waste rocks of the central coal enrichment plant "Chervonohradska" (CCEP «Chervonogradska») and the Vizeyska mine of the LVCB. The subject of the research is the chemical content of water extracts from waste rocks. The aim of the research is to evaluate the chemical composition of the water extractions of waste rock from the tericones of the CCEP "Chervonogradska" and the Vizeyska mine, to establish its features for predicting the migration of pollutants in the hydrolithosphere and possible anthropogenic impacts and risks of emergency situations.

MATERIALS AND METHODS

Samples of waste rocks at a depth of 0.2–0.3 m from different parts of waste heap of the Vizeyska mine were taken in the amount of 20 and 10 samples from the waste dump of the CCEP "Chervonohradska". The average lithological composition of the dumps reflects the composition of the coal-bearing stratum, namely: argillite (70–97%), siltstone (8–28%), sandstone (1–20%), coal (1–7%), pyrite (1%), etc. [25]. To carry out the research, the samples were separated by lithological composition, dried and crushed, sifted through laboratory sieves with a hole diameter of 1.25 mm. The samples of unburnt argillites, burnt argillites, unburnt siltstones, burnt siltstones, unburnt sandstones, burnt sandstones, unburnt coal and siliceous siltstone were combined. For the manufacture of each of the extracts, separate native weights were used. To prepare an aqueous extract, 50 g of the sample was placed in a flask and 250 ml of distilled water was added, the solution was shaken for 3 minutes, the samples were in the solution for a day, after which it was filtered through a paper filter. Analytical studies were carried out in the laboratory of environmental safety of the LSULS (attestation certificate No. RL 091/21 dated 11/30/2021). The content of chlorides (Cl^-) [26], hydrocarbons (HCO_3^-) [27], calcium (Ca^{2+}) [26] and magnesium (Mg^{2+}) [26] was determined by the titration method. In particular: chlorides – with silver nitrate in the presence of potassium chromate; hydrocarbons – with hydrochloric acid in the presence of methyl orange; calcium and magnesium – with trilon B in the presence of murexide and eriochrome black, respectively. Sulfates (SO_4^{2-}) were determined by the weight method (precipitation with barium nitrate followed by calcination of the precipitate) according to KND 211.1.4.026-95. The content of sodium (Na^+) and potassium (K^+) was calculated by the equivalents balance. Other anions (nitrates (NO_3^-) [28] and nitrites (NO_2^-) [29] were determined by photocolorimetry method, namely: the content of nitrates - by interaction with a solution of sodium salicylate in a sulfuric acid medium, nitrites – with the Griess reagent. Only soluble forms of ions were determined.

RESULTS AND DISCUSSIONS

The analysis of water extracts from coal mining waste is one of the methods for determining the amount of pollutants entering the environment and is recommended in the European Union for studies on the ecological assessment of the natural environment [30–32]. As mentioned above, rocks undergo significant transformations with weathering processes when they get into waste heaps, and water is a mineral-forming medium for most of the newly formed minerals: sulfates, hydrocarbons, carbonates, phosphates, etc. That is why we investigated the ionic composition of water extracts from the rock of waste dumps of the Vizeyska mine and the CCEP "Chervonohradska" (Fig. 1). This is also due to the fact that the chemical properties of the rocks of the ChMA waste dumps have been investigated at an insufficient level. Mineralization is one of the most popular indicators of water [33] and water extracts from soils. It is used both for assessing the environmental safety and the risks of emergency situations [34]. The mineralization of water extracts from waste rocks of the Vizeyska mine and the CCEP "Chervonohradska"

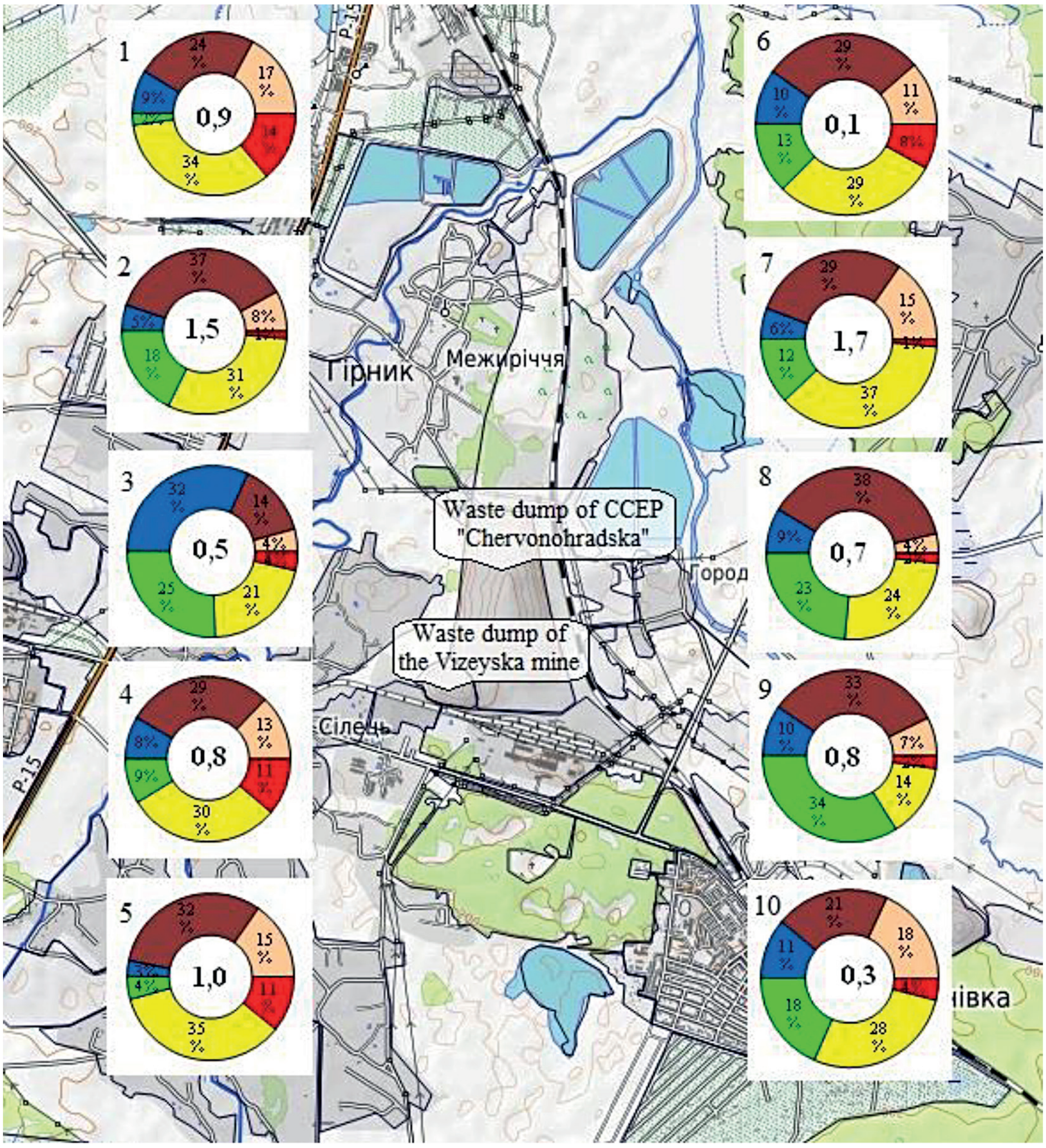


Fig. 1. Distribution of the main ions in the water extracts of the rocks of the ChMA waste dump, g-eq%\l: 1 – unburnt (intensely oxidized) argillite, 2 – overburnt argillite, 3 – unburnt siltstone, 4 – overburnt siltstone, 5 – slightly overburnt sandstone, 6 – coal (tericon rock of the Vizeyska mine); 7 – unburnt argillite, 8 – unburnt siltstone, 9 – unburnt sandstone, 10 – unburnt siliceous siltstone (rocks of the waste dump of the CCEP "Chervonohradska") (the base of the map https://mistaua.com [37])

Table 1. Characteristics of the chemical composition of water extracts from the waste dump of the Vizeyska mine and the CCEP "Chervonohradska"

No*	Mineralization mg/l	Ions concentration, mg/l						pH
		$Na^+ + K^+$	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	
Water extracts from waste rock of the Vizeyska mine								
1	900.6	57.4	126.3	54.7	111.2	367.0	30.5	4.3
2	1514 .3	52 .8	296 .8	39 .1	17 .8	523.0	384.3	6.25
3	526.6	104.2	36.5	6.9	11.8	95.0	146.4	6.35
4	823.9	21.1	144.3	50.3	81.4	298.0	109.8	6.9
5	1007.5	62.6	166.3	49.2	97.7	426.0	67.1	5.9
6	188.1	13.1	30.1	7.3	27.1	28.4	73.2	6.8
Waste rock of CCEP "Chervonohradska"								
7	1741.3	69.8	280.8	90.7	22.4	814.0	341.6	7.07
8	775.2	42.3	149.2	8.9	10.3	180.0	219.6	7.03
9	826.1	51.0	141.0	18.6	15.4	120.0	378.2	7.15
10	295.51	22.6	33.9	18.0	8.7	82.0	69.5	7.52
MAC (for drinking water)		< 300	N/A	< 80	< 250	< 500	< 300	6.0-9.0

Note: * type of the rock according to Figure 1.

ranges from 188.1 to 1741.3 mg/l and averages 781.8 mg/l (Table 1). According to the value of total mineralization, the investigated rocks are clearly grouped by lithological composition. Mineralization of water extracts from siltstones is the highest and ranges from 900 to 1741 mg/l and averages 1385 mg/l. Mineralization of water extracts from siltstones is significantly lower and ranges from 295 to 824 mg/l with an average value of 605 mg/l. According to the increase in the mineralization of the water extractions, the waste rocks of ChMA can be arranged in the following series: coal – siltstone – sandstone – argillite.

According to the pH value of water extracts, rocks are divided according to the oxidation rate. Rocks from the waste heap of the Vizeyska mine, which has been inactive since 2009, are more oxidized and are characterized by weakly acidic and acidic (argillite) water extracts. The average value of pH in water extracts from waste rocks of the Vizeyska mine is 6.08, and from the dump of the CCEP "Chervonohradska", which is constantly replenished with fresh rocks, is 7.19. A decrease in the pH value in aqueous extracts of intensively oxidized rocks most likely occurs due to the oxidation of pyrite (FeS_2) and hilcopyrite ($CuFeS_2$) with the formation of sulfuric acid. It should be noted that in an acidic environment, minerals containing manganese, zinc, chromium, lead, and other elements are more mobile and migrate with surface runoff to adjacent areas. Weakly acidic effluents from waste heaps increase soil acidity, which has a negative effect on plant life, as elements such as iron, aluminum, and manganese change into forms easily available for assimilation by plants, and their concentration can reach toxic levels [19, 35]. According to [36], the bioavailable content of iron in waste dump rocks of the CCEP "Chervonohradska" varies between 1547.15–448.62 mg/kg, manganese – 108.01–681.72 mg/kg.

According to the chemical composition, four groups of water extracts from the waste rock of the Vizeyska mine and tailings of the CCEP "Chervonohradska" are distinguished:

- chloride-sulfate magnesium-calcium (unburnt argillite, overburnt siltstone, slightly overburnt sandstone) composition 1:

$$M_{0,8-1,0}\frac{SO_4\,_{60\text{-}70}\,Cl\,_{22-28}}{Ca\,_{48-59}\,Mg\,_{27-34}} \quad (1)$$

- hydrocarbonate-sulfate calcium (burnt argillite, unburnt siltstone), and hydrogencarbonate-sulfate magnesium-calcium (unburnt argillite, unburnt siliceous siltstone) composition 2:

$$M_{0,3-1,7}\frac{SO_4\,_{49\text{-}73}\,HCO_3\,_{24\text{-}47}}{Ca\,_{41-76}\,Mg\,_{7-36}} \quad (2)$$

- calcium-sodium sulfate-hydrocarbonate (unburnt siltstone), and calcium sulfates-hydrocarbonates (unburnt sandstone) composition 3:

$$M_{0,5-0,8}\frac{HCO_3\,_{51\text{-}68}\,SO_4\,_{27\text{-}42}}{Ca\,_{28-66}\,(Na+K)\,_{19-34}} \quad (3)$$

- chloride-hydrocarbonate calcium (coal) composition 4:

$$M_{0,19}\frac{HCO_{3\ 47}\,Cl_{\ 30}}{Ca_{\ 57}\,(Mg_{22})} \quad (4)$$

The high concentration of sulfates in water extracts from the wastes of coal mining and coal beneficiation, which on average in absolute terms is 290 mg/l for the rocks of the mine and 299 mg/l for the rocks of the coal beneficiation factory is noteworthy. The highest amount of sulfates was found in the extract from the argillite of the unburned rock of CCEP "Chervonohradska" 814 mg/l and the burnt argillite of the waste dump of the Vizeyska mine. According to the increase in sulfate content of the rocks, a series can be constructed: coal (28 mg/l) – siltstone (164 mg/l), sandstone (273 mg/l), argillite (568 mg/l). The high presence of sulfates in water extracts from rocks indicates an intensive process of destruction of pyrite and other sulfides. These man-made solutions enter surface and underground waters and cause their pollution. According to [16], the waters of man-made reservoirs (sludge impoundments) have a sulfate-chloride calcium-magnesium composition and a calcium sulfate composition (ponds near the CCEP dump). These data are in good agreement and complement the results of our research. It is also necessary to draw attention to the fact that sulfate-contaminated soils adjacent to waste dumps. The concentration of mobile sulfate ion in ChMA soils adjacent to waste heaps ranges from 518 to 4219 mg/kg [19] According to our data, in terms of dry rock, the content of sulfates in different rocks ranges from 124.1 mg/kg to 3483.9 mg/kg (Fig. 2).

The concentration of chlorine ions (8.7–111.2 mg/l) in water extrates is extremely variable. We reported a high chlorine content in the water extracts of all three types of rocks: burnt siltstone (81.4 mg/l), slightly burnt sandstone (97.7 mg/l) and unburned argillite (111.2 mg/l). It should be noted that chlorine ions can form very toxic chlorophenols with phenols, as well as affect the corrosiveness of solutions [38]. An important indicator of water and soil quality is the content of nitrogen compounds, in particular nitrates and nitrites, and ammonium, which indicates the feasibility of studying their content in rocks. Within the rock dumps, the content of ammonium ions on average is 3.66 (0.6–8.4) mg/l, the concentration of nitrites is 2.14 (0.6–8.5) mg/l, nitrates – 83.3 (0.6–135.6) mg/l (Fig. 3). Considering the normalization of the content of nitrates in soils, the MAC is 130 mg/kg [39], it is appropriate to estimate its content in the rock, from the data we obtained. It follows that its content ranges from 103.0 to 602.1 mg/kg (Fig. 2) and exceeds the MAC for soils by 1.74–4.63 times. The danger of high concentrations of nitrates for humans lies in

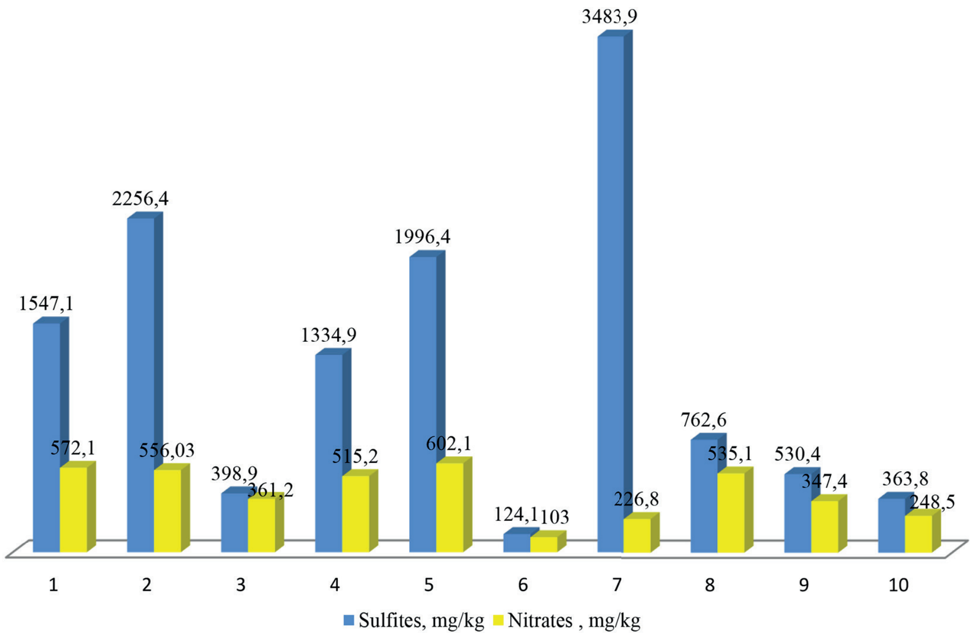
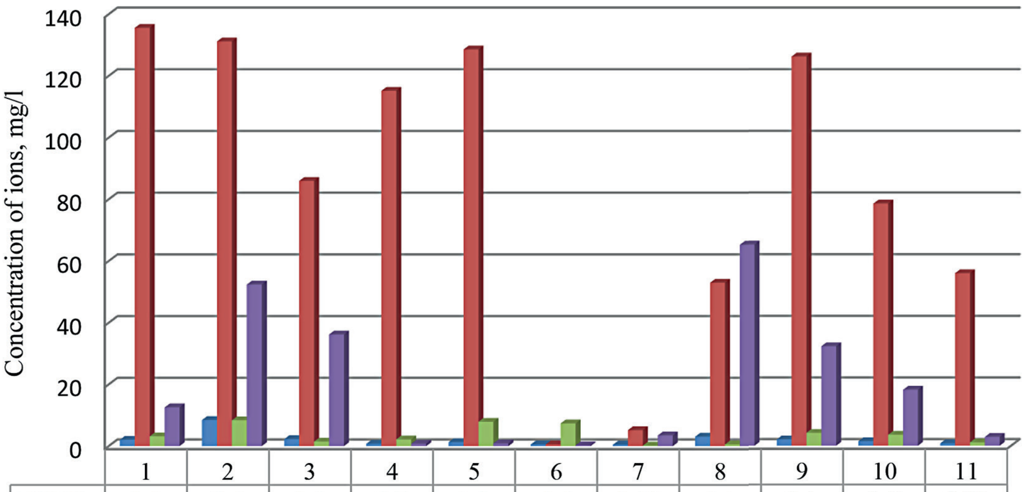


Fig. 2. The content of sulfites and nitrates in the waste rock*, mg/kg (*types of rock according to the description in Fig. 1)



	1	2	3	4	5	6	7	8	9	10	11
NO2-	2,1	8,5	2,3	0,8	1,3	0,6	0,52	3,1	2,1	1,5	0,81
NO3-	135,6	131,2	86	115,2	128,6	0,6	5,2	53	126,3	78,6	56
NH4+	3,2	8,4	1,5	2,2	7,9	7,4	0,2	0,6	4,2	3,6	1,1
Fetot	12,6	52,4	36,2	0,8	0,9	0,3	3,5	65,3	32,3	18,2	2,9

Fig. 3. The content of nitrogen compounds and total iron in the waste rock*, mg/l (*types of rock according to the description in Fig. 1)

the fact that entering the alimentary canal with water, they are reduced to nitrites under the influence of intestinal microflora and have a negative effect on the body. The content of iron (Fe_{tot}) in the water varies from 0.3 mg/l to 65.3 mg/l and is 20.49 mg/l on average.

The results presented in the article are a contribution to the development of the theory of geochemistry of technogenesis and the theory of environmental safety, replenish the databases of pollutants that should be combined into a single database using new developments in information technologies [40,41]. In addition, the results of our research will be useful in designing new methods of coal mining and coal mining waste storage [42,43] and designing safety systems of man-made objects [44, 45].

CONCLUSIONS

The mineralization of water extracts from waste rocks of the Chervonohrad mining area varies from 188.1 mg/l to 1741.3 mg/l and is 781.8 mg/l on average. As the mineralization of water extracts of these rocks increases, a series is formed: coal – siltstone – sandstone – argillite.According to the pH value of water extracts, the rocks are divided into two groups according to the oxidation rate. The more oxidized rocks from the waste dump of the Vizeyska mine are characterized by weakly acidic and acidic water extractions. Rocks from the CCEP “Chervonohradska”, which is constantly replenished with fresh rocks, have a neutral reaction.

Four groups are distinguished according to the chemical composition of water extracts from waste rock. Rocks with chloride-sulfate magnesium-calcium composition of water extracts, with hydrogencarbonate-sulfate calcium and hydrogencarbonate-sulfate magnesium-calcium composition, sulfate-hydrocarbonate calcium-sodium and sulfate-hydrocarbonate calcium composition and rocks with chloride-hydrocarbonate calcium composition of water extracts. Most of the analyzed rocks are characterized by a high concentration of sulfates in water extracts, which averages 290–299 mg/l.Water extracts of waste rock contain high content of nitrogen. Within the rock dumps, the content of ammonium ions on average is 3.66 (0.6–8.4) mg/l, the concentration of nitrites is 2.14 (0.6–8.5) mg/l, nitrates – 83.3 (0.6–135.6) mg/l

The reason for the high concentration of sulfates and low pH values is the oxidation of pyrite and other sulfides. Sulfates, mobile iron and other pollutants formed as a result of hypergenesis processes build pressure on the ecological state of the environment and increase the risks of emergency situations.

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