

HYDROCHEMISTRY OF THE WATERS CIRCULATING
IN THE CRATER, EASTERN AND SOUTHERN SLOPES
OF ARAGATS VOLCANO

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This study examines the hydrochemistry of mineral and freshwaters circulating in the crater of the Aragats volcano and on its Eastern and Southern slopes, as well as to clarify several related issues. On the Eastern slopes, in connection with the composition of the mineral waters, a natural acid drainage phenomenon has been observed, which propagates downstream along the main tributary and gradually diminishes within the territory of the Tsaghkashen Village. In this area, noticeable differences are also observed in the compositions of waters discharging from outlets located in proximity to one another, which may be explained by the substantial difference in the depths of formation of the mineral and freshwater bodies, as well as by the isolation of their respective pathways to the Earth's surface. The most widespread and dominant water types were distinguished for the investigated sites, and their chemical formulas were determined according to Kurlov's classification.

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Keywords: hydrochemistry, Aragats volcano, mineral water, oxygen barrier, water composition, mineralization, natural acid drainage.

Introduction. Geological characteristics and features of 4090 m high Aragats stratovolcano have always been at the center of attention of the scientists, particularly of the volcanologists. Many scientific works of Armenian researchers dedicated to the geology of the Aragats, in which the age, petrology, geochemistry, volcanology are discussed.

Territory of Armenian highland is an area of widespread post-collisional volcanism. Within territory of the Republic of Armenia (RA) volcanic activity of the Miocene-Quaternary period covers about 50% of the territory. Aragats volcano occupies a special place among volcanic structures in terms of the scale of eruption, the complexity of the structure, and the structural-tectonic position. Morphologically, the Aragats massif is represented by a large shield-shaped structure with a diameter of 42 km [1].

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Academician R. Jrbashyan considers the Aragats Polygenic volcanic structure to be the largest and most complex in the Central Volcanic Zone of Armenia, with an area of about 5000 km^2 [2].

This paper focuses on hydrogeological characteristics of Aragats stratovolcano, mineral water springs in the crater and natural acidic drainage conditions. With its unique and interesting hydrogeological characteristics, Aragats has also attracted the attention of specialists in the field of hydrogeology. Some of the results of their research were presented in a fundamental work [3], where it is stated that the Aragats volcanic massif is mainly composed of andesite-basalt, basalt, andesite-dacite, dacite lavas of the Tertiary and Quaternary periods, as well as of tuffs and tuff-lavas of andesite-dacite and dacite composition. After the last cycle of volcanic activity, Aragats underwent glaciation twice. Two minor glaciers have been preserved on the mountain peak. As a result of the glaciers' activity, Amberd, Mantash, Geghazor, and other valleys were formed, where both mineral and fresh water springs were recorded. In general, the mineral waters of Aragats have a large discharge and high values of carbonic acid content [3].

This study aims to characterize the chemical composition of the studied mineral, fresh and surface flow waters in the Aragats Volcanic Region, identify the primary sources of their chemical constituents, and evaluate the processes governing their geochemical evolution.

Samples and Analytical Methods. Fieldwork and sampling was conducted in three phases:

2015 – Sampling in the upper reaches of the Amberd River, originating on the Southern slope of Aragats.

2018 – Sampling in the crater area of Aragats.

2020 – Sampling on the Eastern slope, extending to the vicinity of the Aparan Reservoir.

Water samples were collected from mineral and fresh water springs, as well as from Amberd and Gegharot Rivers systems, to assess their hydrochemical composition. Sampling locations were selected based on previous hydrogeological studies and visual indicators of mineralization, such as ochre-colored streambeds, gas emissions, and variations in water clarity. Sampling was carried out in accordance with the accepted requirements and rules [4]. Water was sampled in 1-liter polyethylene containers. The first of the most important conditions is the cleanliness of the container and stopper. This is prepared in the laboratory. The containers and stoppers are pre-treated in the laboratory with a 5% solution of hydrochloric acid, then with boiled water and finally with distilled water. When taking the sample, the container and stopper are rinsed 2–3 times with the water being sampled, which is then poured into the container up to the bottom of the stopper, and then the container is hermetically sealed and stored in a cool and dark place, until delivered to the laboratory.

Analytical studies were carried out at the Hydrogeochemistry Laboratory of the Institute of Geological Sciences of the NAS of the RAa. Standard hydrochemical, colorimetric [4] and physicochemical [5] methods were applied in accordance with ISO and GOST standards. The chemical composition of the samples was determined

using traditional hydrochemical analysis, focusing on major ions, trace elements, and isotopic ratios where it's applicable.

The results of the hydrochemical analysis of the studied waters are presented using the Kurlov formula and the Piper diagram.

The Kurlov formula, utilized in hydrogeology for classifying groundwater based on its chemical composition. It was first introduced by Soviet hydrogeologist M. I. Kurlov. He detailed this method in his 1928 publication titled “Hydrogeological Studies in the Lower Volga Region” (in Russian). In this work, Kurlov presented a systematic approach to express and classify groundwater types using the relative concentrations of major ions, a methodology that has since become fundamental in hydrogeological studies. In the Tab. 1 all sampled waters compositions are presented by Kurlov's formula.

Table 1

Brief data on the geographical location, temperature and chemical composition of the studied water outlets

N	Sample ID	Location, water description	Latitude	Longitude	H, m	T, °C	Kurlov's formula
1	9/15	Amberd River basin, surface flow	40.478855	44.161151	33014	44.0	$M_{0.28} \frac{\text{HCO}_3 73 \text{SO}^4 20}{\text{Ca}44\text{Mg}33\text{Na}18}$
2	10/15	Amberd River basin, mineral water	40.480467	44.161617	33045	33.1	$M_{0.28} \frac{\text{HCO}^3 75 \text{SO}^4 19}{\text{Ca}59\text{Mg}24\text{Na}14}$
3	27V/18	Crater of Aragats, mineral water 1	40.511418	44.190901	33518	44.9	$M_{1.1} \frac{\text{SO}^4 95}{\text{Mg}35\text{Fe}26\text{Ca}22\text{NH}4 11}$
4	27V-1/18	Crater of Aragats, mineral water 2	40.511891	44.189257	33526	n/d	$M_{0.99} \frac{\text{SO}^4 79 \text{Cl}18}{\text{Mg}56\text{Ca}25\text{Fe}9}$
5	28V/18	Lake Kari, fresh water	n/d	n/d	3199	12.9	$M_{0.033} \frac{\text{HCO}^3 73 \text{SO}^4 15 \text{Cl}12}{\text{Ca}56\text{Mg}29\text{Na}12}$
6	1/20	Aragats East. slop, Tributary, with ochre hue	40.501389	44.208611	22978	110.5	$M_{0.72} \frac{\text{SO}^4 76 \text{Cl}23}{\text{Ca}46\text{Mg}17\text{Na}14\text{H}12}$
7	2/20	Right tributary	40.501111	44.209167	22972	117.5	$M_{0.39} \frac{\text{SO}^4 49 \text{Cl}45}{\text{Ca}43\text{Mg}26\text{Na}26}$
8	3/20	Main rivulet	40.496667	44.225556	22779	115.1	$M_{0.58} \frac{\text{SO}^4 76 \text{Cl}23}{\text{Ca}50\text{Mg}21\text{Na}14}$
9	4/20	Fresh water spring	40.495000	44.243611	n/d	113	$M_{0.04} \frac{\text{HCO}356 \text{SO}^4 30}{\text{Ca}44\text{Mg}30\text{Na}12}$
10	5/20	Main rivulet up to v. Tsaghkashen	40.489444	44.286944	22317	113.9	$M_{0.30} \frac{\text{SO}^4 77 \text{Cl}22}{\text{Ca}56\text{Mg}18\text{Na}15}$
11	6/20	Main rivulet, Eastern end of v. Tsaghkashen	40.501389	44.360000	11928	117.1	$M_{0.33} \frac{\text{SO}^4 74 \text{Cl}20}{\text{Ca}44\text{Mg}39\text{Na}13}$
12	7/20	Main rivulet, before flowing into the Aparan reservoir	40.502222	44.429167	11828	119.5	$M_{0.17} \frac{\text{HCO}3 41 \text{SO}^4 38 \text{Cl}17}{\text{Ca}44\text{Mg}36\text{Na}16}$

Note: n/d – not determined.

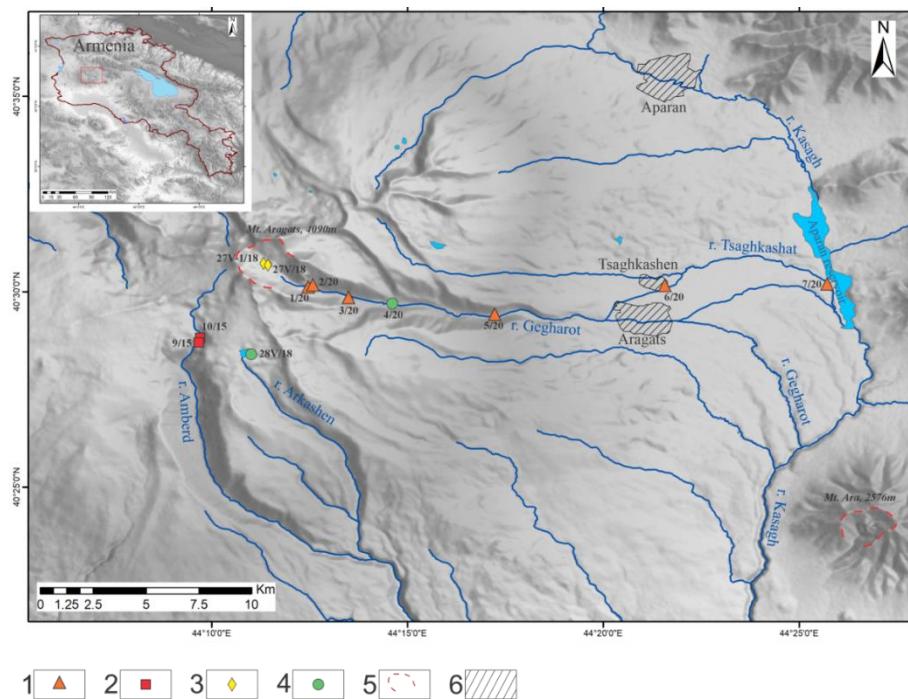


Fig. 1. Schematic map of all sampling areas: 1. Samples from Gegharot River; 2. Samples from Amberd River; 3. Crater mineral waters; 4. Fresh waters; 5. Polygenic volcanoes; 6. Settlements.

Results and Discussion. According to the results of previous research, the waters of the upper flow of the Amberd River appear mainly in two types, leaving a red trace along the entire stream. These waters are cold, contain free carbonic acid, have a large discharge and a pleasant taste [3].

It should be noted that the data given in this research refer to the two springs coming to the surface of the Earth in the upper reaches of the Amberd River, at a distance of about 150 m from each other, whose composition formulas, in order to ensure more accuracy, are presented taking into account the values of components less than 10 mg-equivalent:

$$M_{0.1} \frac{\text{HCO}^3 93 \text{Cl}7}{\text{Ca}84 \text{Na} + \text{K}9 \text{Mg}7}, \quad M_{0.8} \frac{\text{HCO}^3 98 \text{Cl}1 \text{SO}^4 1}{\text{Ca}40 \text{Mg}33 \text{Na} + \text{K}26 \text{Fe}1}.$$

During the research of 2015, we sampled the waters of 2 spring in the basin of the right tributary of the Amberd River, upstream of the tributary, about 0.5 km to the North of Kari Lake at an altitude of 3200 m. The difference in composition formulas obtained as a result of the research allows us to assume that the waters of the previously and newly examined springs circulate in different underground levels.

The first sample taken by us here is from the right tributary of the Amberd River (9/15), the second is from the water coming out to the surface of the Earth on its right bank (10/15). The bed of the latter, in the direction of the flow, has a bright ochre color until it joins the tributary, which gave reason to think that it is mineral.

Table 2

Hydrochemical analysis of all sampled waters (g/L)

N	Determined components	Sample number											
		9/15	10/15	27V/18	27V–1/18	28V/18	1/20	2/20	3/20	4/20	5/20	6/20	
1	H ⁺	n/d	n/d	0.63	0.66	n/d	1.38	n/d	0.76	n/d	0.31	n/d	n/d
2	Li ⁺	n/a	n/a	0.04	0.05	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
3	NH ₄ ⁺	0.10	n/a	32.00	12.00	n/a	0.14	0.14	n/a	n/a	n/a	n/a	0.10
4	Na ⁺	9.66	7.82	3.75	3.45	1.05	37.27	37.01	30.00	1.50	16.14	15.86	9.29
5	K ⁺	3.67	2.67	2.05	1.83	0.50	10.67	13.00	9.00	2.17	5.83	5.33	3.25
6	Ca ²⁺	20.43	28.28	74.33	78.24	4.61	107.58	53.79	92.91	4.30	53.79	44.99	21.52
7	Mg ²⁺	9.06	7.15	72.06	106.09	1.46	23.72	19.57	23.72	1.78	10.67	24.19	10.67
8	Fe _{total}	0.20	0.02	79.20	25.30	0.06	18.15	0.18	7.70	0.02	1.28	0.18	0.11
9	Cu ²⁺	0.00049	0.00051	0.0048	0.039	0.0006	n/d	n/d	n/d	n/d	n/d	n/d	n/d
10	Zn ²⁺	n/a	n/a	0.008	0.01	0.008	n/d	n/d	n/d	n/d	n/d	n/d	n/d
11	Pb ²⁺	n/d	n/d	0.028	0.033	0.00045	n/d	n/d	n/d	n/d	n/d	n/d	n/d
12	Cd ²⁺	n/d	n/d	0.04	0.025	0.00015	n/d	n/d	n/d	n/d	n/d	n/d	n/d
13	F [–]	0.28	0.05	3.80	3.80	0.05	1.10	0.36	0.96	0.36	0.44	0.56	0.44
14	Cl [–]	5.46	4.77	16.37	102.30	1.71	95.48	98.89	75.02	1.36	37.51	35.46	15.00
15	SO ₄ ^{2–}	21.81	22.21	752.16	587.90	3.09	425.07	148.14	342.46	7.00	178.19	179.83	45.27
16	HCO ₃ [–]	102.48	109.80	n/a	n/a	18.31	n/a	21.96	n/a	17.08	n/a	12.20	61.00
17	NO ₃ [–]	n/a	n/a	n/a	n/a	n/a	0.30	n/a	0.50	0.80	n/a	7.00	4.00
18	NO ₂ [–]	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	0.01	n/a	n/a	0.07
19	PO ₄ ^{3–}	n/a	n/a	0.05	0.05	n/a	n/a	0.08	n/a	0.07	0.05	n/a	n/a
20	H ₄ SiO ₄	110.00	100.00	55.00	50.00	2.00	n/d	n/d	n/d	n/d	n/d	n/d	n/d
21	Si	2.08	29.17	16.04	14.58	0.58	n/d	n/d	n/d	n/d	n/d	n/d	n/d
22	As	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
23	General mineralization	283.20	282.77	1110.52	987.68	32.85	720.86	393.12	583.03	36.45	304.21	325.60	170.72
24	General hardness	1.77	1.99	9.64	12.63	0.35	7.32	4.29	6.59	0.37	3.56	4.24	1.95
25	pH	5.82	4.99	3.20	3.18	7.17	2.86	6.55	3.12	7.24	3.51	4.44	6.94

Note: n/d – not determined; n/a – not available.

However, as a result of the research, it turned out that this water is almost indistinguishable from the water of the tributary, belongs to the same class with not much difference in numerical values, has the same value of mineralization and only the hydrogen ion content is slightly higher here.

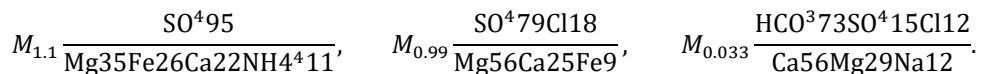
Thus, it can be concluded that these waters sampled and examined by us, in addition to the intermixing of surface flows, also have a common circulation plane in the Earth's depths. The obtained data are given in Tabs. 1 and 2, and the formulas of surface flow and mineral water compositions, respectively, have the following form:

$$M_{0.28} \frac{\text{HCO373SO}^{420}}{\text{Ca44Mg33Na18}}, \quad M_{0.28} \frac{\text{HCO375SO}^{419}}{\text{Ca59Mg24Na14}}.$$

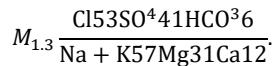
In the near-peak area of Aragats, in the crater, the waters of 3 outlets coming to the surface of the Earth at an altitude of about 3500 m, two of which are mineral and one is fresh – were previously described by us as well. One of the mineral waters is sulfate, magnesium–iron–calcium–ammonium in composition and the other is sulfate–chloride, magnesium–calcium (values below 10 mg-equivalent are not taken into account). They are distinguished by the significant contents of ammonium-ion,

H^+ -ion, Fe_{total} . They have relatively high mineralization and relatively high values of total hardness, which allows their origin to be associated with greater depths.

The obtained data are given in Tabs. 1 and 2, and the composition formulas, respectively, 27V/18, 27V-1/18, and 28V/18 have the following forms:



There are known data in the literature about the mineral waters of two springs coming to the surface of the Earth in the peak area of Aragats, which are called Geghadzor springs [3]. We cannot consider them the mineral waters described by us, because the compositional differences are quite substantial and conducted research does not provide grounds to assume that such changes could have occurred in an area at such a great distance from anthropogenic influence. Besides, only one water composition formula is provided:



We believe this question needs further additional examination and clarification, because we still are not aware of other mineral water outlets in the crater of Aragats.

During the works performed on the Eastern slope of Aragats, several water outlets were observed, which, according to us, are of interest both in terms of the outlets' position and composition. And the activity of some of them, particularly the mineral ones, can be considered as an example of natural acid drainage. Their bed has a bright ochre color in the direction of the flow of Gegharot River, which continues for several kilometers and fades in the territory of Tsaghkashen Village.

As the obtained data show, particularly the values of hydrogen ion content and total mineralization, before flowing into the Aparan Reservoir, the water quality is completely restored due to the active operation of the oxygen barrier.

In this area, the compositions of the mineral and fresh waters of the outlets located close to each other and coming out to the surface of the Earth are obviously different from each other. This testifies about the formation of underground waters compositions in different depths and about their coming out to the surface of the Earth in ways that are significantly isolated from each other.

In the direction of the flow, downslope, the waters of seven outlets were sampled and examined. The obtained results are presented in Tabs. 1 and 2. According to their composition, they can be divided into 2 groups: 1) sulfate-chloride, calcium-magnesium-sodium: $SO^{4}Cl/CaMgNa$; 2) hydrocarbonate-sulfate (-chloride), calcium-magnesium-sodium: $HCO^{3}SO^{4}/CaMgNa$ (Tab. 2).

The cationic series is distinguished also by the constancy of numerical values. Only in the first sample does H^+ appear at the last place in the series.

We tend to consider the first ones as mineral waters, based on the color of the bed, the total amount of iron, the fact that the sulfate ion is in the first place among the anions, and on the pH values. The mineralization of these waters is not high (0.30–0.72 g/L), which can be explained by their mixing with fresh, low-mineralized waters circulating at shallow depths for a short period of time before coming to the Earth's surface, which, in its turn, is confirmed according to the research results of the waters of many outlets of fresh water coming to Earth surface.

An idea of the general description of the studied water compositions can also be obtained from Fig. 2, where the compositions are expressed using the Piper diagram.

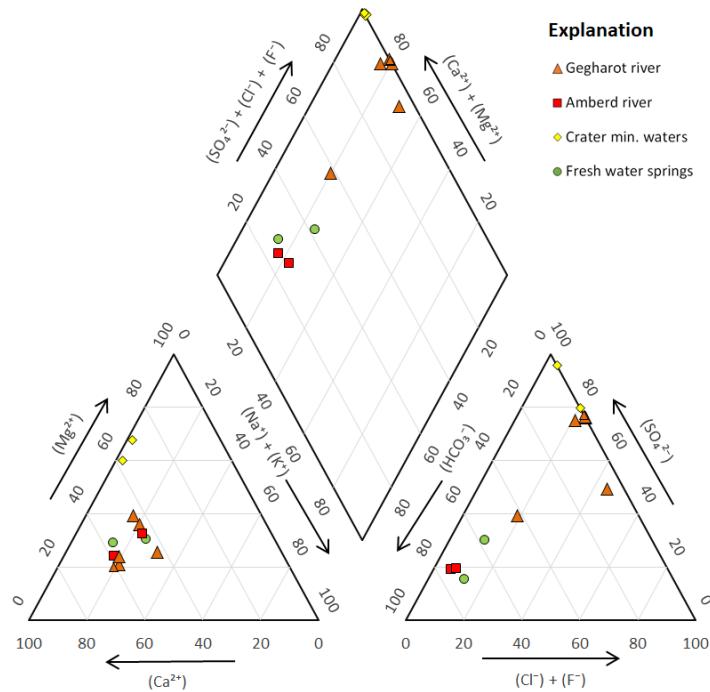


Fig. 2. Piper diagram of the sampled waters.

Conclusion. Thus, the results of the previously carried out and our current hydrochemical studies allow us to separate certain hydrochemical characteristics of the waters circulating in the areas of our research, on the slopes of the Aragats volcano, according to their composition, which are:

I. On the Southern slopes, the examined waters of the upper reaches of the Amberd River are characterized by a total mineralization of 0.1–0.8 g/L and the following compositions (to make it comparable with our formula, we present the composition formula given in [3] without values below 10 mg-equivalent):

- hydrocarbonate, calcium (or calcium–magnesium);
- hydrocarbonate, calcium–magnesium–sodium–potassium;
- hydrocarbonate–sulfate, calcium–magnesium–sodium.

II. The waters circulating in the crater area of Aragats are characterized by the following compositions:

- chloride–sulfate, (Na+K)–magnesium–calcium;
- sulfate, magnesium–iron–calcium–ammonium;
- sulfate–chloride, magnesium–calcium–iron,
- hydrocarbonate–sulfate–chloride, calcium–magnesium–sodium.

The mineralization values of these waters are between 0.033 and 1.3 g/L.

III. On the Eastern slope, mineral waters have mineralization of 0.30–0.72 g/L, fresh waters – 0.04–0.17 g/L.

Minerals are characterized by sulfate–chloride, calcium–magnesium–sodium composition.

Fresh waters are characterized by hydrocarbonate–sulfate, calcium–magnesium–sodium and hydrocarbonate–sulfate–chloride, calcium–magnesium–sodium compositions.

IV. The studied Eastern slope mineral and fresh water compositions are formed at different depths and come out to the surface of the Earth mainly in different ways, significantly isolated from each other. It can be assumed that the time of their interaction is quite short, since this interaction takes place before their coming out to the surface of the Earth, close to the surface, at small depths.

V. The studied mineral and fresh water springs exhibit significant compositional variability, influenced by subsurface geological processes, rock–water interactions, and atmospheric contributions. Notably, the phenomenon of natural acid drainage has been observed on the Eastern slope in the Gegharot River, originating from the crater and extending downstream, where its effects gradually diminish near v. Tsaghkashen. The composition of these waters is strongly controlled by the volcanic and pyroclastic deposits, which contribute major and trace elements to the hydrothermal system.

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ԱՐԱԳԱԾ ՀՐԱԲԻՆԻ ԳԱԳԱԹԱՅԻՆ ՄԱՍՈՒՄ, ԱՐԵՎԵԼՅԱՆ ԵՎ
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Ամփոփում

Հոդվածը նվիրված է Արագած հրաբինի գագաթային մասում, արևելյան և հարավային լանջերին շրջանառվող հանքային և քաղցրահամ ջրերի

հիդրոքիմիային ու դրան առնչվող որոշ հարցերի պարզաբանմանը: Արևելյան լանջերին, հանքային ջրերի կազմերի հետ կապված, դիտվել է բնական թթվային դրենաժի երևոյթ, որը տարածվում է մայր վտակի հոսքի ուղղությամբ և աստիճանաբար մարում Ծաղկաշեն գյուղի տարածքում: Այս տեղամասում դիտվում են նաև իրար մոտ տեղակայված երերով ջրերի կազմերի նկատելի տարրերություններ, ինչը կարող է բացատրվել տեղամասի հանքային և քաղցրահամ ջրերի ձևավորման խորությունների զգակի տարրերությամբ և նրանց՝ երկրի մակերես դուրս գալու ճանապարհների միջանցից մեկուսացվածությամբ: Անջատվել են ջրերի առավել տարածված, գերակշռող կազմերը, ըստ հետազոտված տեղամասերի, և կազմվել են բանաձևերն ըստ Կուրլովի:

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ГИДРОХИМИЯ ВОД, ЦИРКУЛИРУЮЩИХ В КРАТЕРЕ, НА ВОСТОЧНЫХ И ЮЖНЫХ СКЛОНАХ ВУЛКАНА АРАГАЦ

Резюме

Работа посвящена выяснению особенностей гидрохимических характеристик некоторых минеральных и пресных вод, циркулирующих в кратере, на восточных и южных склонах вулкана Арагац, а также уточнению ряда связанных с ними вопросов. На восточных склонах из-за состава минеральных вод наблюдается явление естественного кислотного дренажа, который распространяется вниз по течению вдоль основного притока и постепенно затухает в пределах территории села Цахкашен. В этом районе фиксируются значимые различия в гидрохимическом составе вод источников, расположенных друг от друга на небольшом расстоянии. Это может быть объяснено существенной разницей глубин формирования минеральных и пресных вод, а также изолированностью друг от друга их путей к поверхности Земли. Для исследованных участков выделены наиболее распространенные и доминирующие типы вод, гидрохимические составы которых приводятся в виде формулы Курлова.