

GEOCHEMICAL CHARACTERISTICS OF NATURAL RADIOACTIVITY AND TRACE ELEMENT DISTRIBUTION IN THE SOLID EJECTA OF THE DASHGIL MUD VOLCANO

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Summary: Mud volcanoes transport large volumes of breccia to the surface, composed of rock fragments derived from deep strata. The solid ejecta of mud volcanoes therefore represent an important source of information for determining the stratigraphic age of ancient sediments. This study presents the results of radiometric, microfaunal, gamma-spectrometric and neutron activation analyses carried out to determine the natural radioactivity and the distribution of trace elements in solid ejecta from the Dashgil mud volcano at different stratigraphic ages. Microfaunal analysis indicates that the samples belong to the Upper Miocene (Sarmatian). Radiometric measurements show that the equivalent gamma dose rate of the samples ranges from 58 to 90 nSv/h. Gamma-spectrometric analyses reveals that the activity concentrations vary within 38-43 Bq/kg for ²³⁸U, 28-37 Bq/kg for ²³²Th, and 600-750 Bq/kg for ⁴⁰K. Furthermore, neutron activation analysis was used to determine the trace element composition of the bedrock. The average mass fractions were found to be 8.1 ppm for Th, 2.6 ppm for U, 4.3 ppm for Hf, and 13.6 ppm for Sc, while the total rare earth element content averages 93.5 ppm. The findings of this study may contribute to the integration of geological data for the stratigraphic subdivision of sediments of various ages, particularly in cases where faunal and floral remains are absent.

Keywords: *Dashgil mud volcano, solid ejecta, stratigraphic age, radiometric analysis, microfaunal analysis, gamma-spectrometric method, neutron activation analysis*

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Introduction

Mud volcanoes constitute a natural phenomenon that has been the subject of extensive scientific research. Eruption products of mud volcanoes provide valuable insights into the Earth's geological structure, as well as the internal geodynamic and geochemical processes occurring at otherwise inaccessible depths. 353 mud volcanoes are known in Azerbaijan (Fig.1). Mud volcanoes are classified according to morphology, the area covered by solid ejecta, age, composition, etc. (Baloglanov et al., 2018). All morphological types of mud volcanoes occur in Azerbaijan.

Mud volcanoes transport significant volumes of breccia composed of rock fragments to the surface. The solid ejecta of mud volcanoes provide valuable information for the study of the stratigraphic age of ancient deposits. In stud-

ies of mud volcano products, stratigraphic and petrographic analyses of components play a particularly important role. Geophysical investigations also play a significant role in the study of mud volcano and associated phenomena. These studies established correlations and cyclicity in radioactive and geochemical parameters within sedimentary sequences of Eastern Azerbaijan and elucidated the nature of elevated radioactivity in the Middle Jurassic and Maykop deposits (Алиев и др., 2003; Эфендиева и др., 2012).

Dashgil mud volcano has an externally expressed, relatively flat elevation slightly elongated in a latitudinal direction. Within its crater field, active cones, gryphons, and salses are grouped in several locations.

Mud volcanoes are remarkable natural phenomena that provide access to deep subsurface materials otherwise obtainable only through

drilling. Their ejecta contain sedimentary fragments from various stratigraphic levels, offering valuable information about the geological environment. In some cases, sedimentary rocks lack faunal and floral remains, making stratigraphic age determination difficult, with such deposits commonly referred to as “silent layers”. The absence of biostratigraphic markers complicates the correlation and interpretation of these strata using conventional stratigraphic methods.

This study investigates the relationship between stratigraphic age, natural radioactivity, and trace element composition in sediment samples using gamma spectrometry and neutron activation analysis (NAA). The obtained correlations, integrated with other geological data, are intended to facilitate the differentiation of sedimentary complexes lacking fossil evidence.

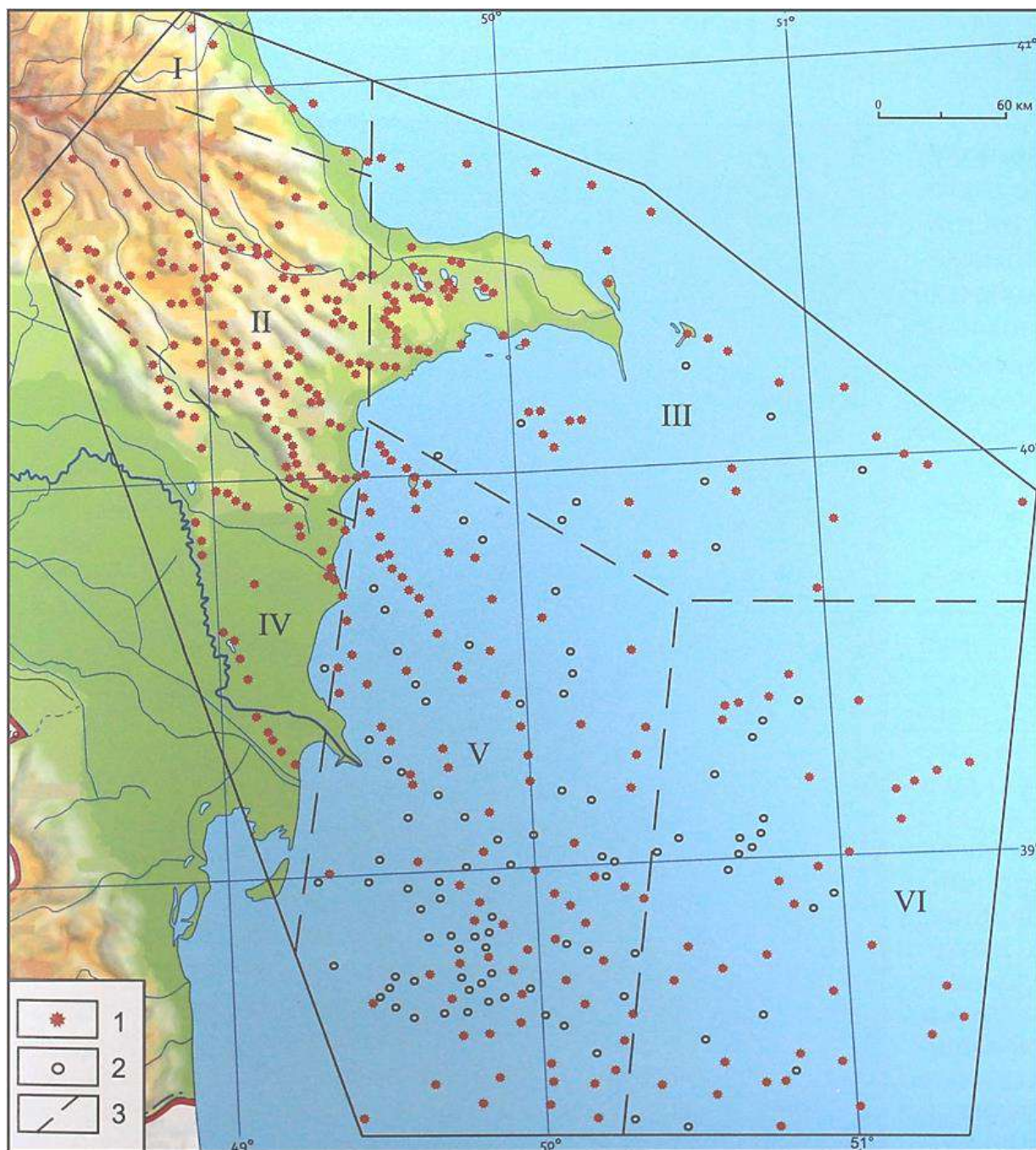


Fig. 1. Spatial distribution of mud volcanoes in Azerbaijan (Aliyev et al., 2015).
 1 – established; 2 – presumed; 3 – petroleum-bearing area boundaries

Materials and Methods

Eighteen bedrock samples were collected for micropaleontological, gamma-spectrometric and neutron activation analyses.

The study initially involved microfaunal analysis to determine the geological age of samples with previously unknown stratigraphic affiliation. Micropaleontological investigations were carried out at the Institute of Geology and Geophysics of the Ministry of Science and Education of the Republic of Azerbaijan following established procedures. Depending on the lithological composition, the samples were treated with sodium carbonate or hydrogen peroxide solutions, while compact clayey rocks were additionally heated to facilitate disaggregation. After washing, drying, and fractionation, microfaunal remains were manually extracted under a microscope. The taxonomic composition was examined using a Nikon microscope ($\times 50\text{--}\times 200$), while detailed microfaunal images were obtained with a JEOL 6610 LV electron microscope.

Neutron activation analysis is a technique used to determine the elemental composition of a material by measuring gamma radiation produced following neutron irradiation of the sample. Upon irradiation, neutrons are captured by the nuclei of the investigated sample, leading to the formation of excited nuclei. The excited nuclei then decay to a stable state, emitting gamma rays with discrete energies characteristic of each nucleus. The gamma rays are recorded by a detector, and their energies and corresponding count rates for individual spectral lines are determined (ГуТЬКО, 2008). The final result of

neutron activation analysis, namely the elemental concentrations in the sample, is calculated by proportional comparison of the activities of identical isotopes in the sample with those in a standard of known elemental mass fractions (Фронтасьева, 2011). Neutron activation analysis was performed at the I.M. Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research (JINR), Dubna, Russian Federation.

Gamma spectrometric analyses were carried out at the Institute of Geology and Geophysics, Ministry of Science and Education of the Republic of Azerbaijan. Gamma radiation measurements were performed using a NaI scintillation detector. The counting time for each sample was 24 hours. The detector was calibrated using a volumetric standard source.

Results and Discussion

Micropaleontological analysis was used to determine the stratigraphic age of each sample. Faunal assemblages identified in the solid ejecta of the Dashgil mud volcano indicate the geological age of the rocks, and the samples are assigned to the Upper Miocene (Sarmatian).

The equivalent dose rate of gamma radiation was measured using a high-sensitivity MKS-AT1125 dosimeter-radiometer. Radiometric studies revealed that the equivalent dose rate of gamma radiation in bedrock within the volcanic area ranges from 58 to 90 nSv/h. A schematic map of the gamma-ray field of the studied mud volcano was constructed based on the measurements (Fig. 2).

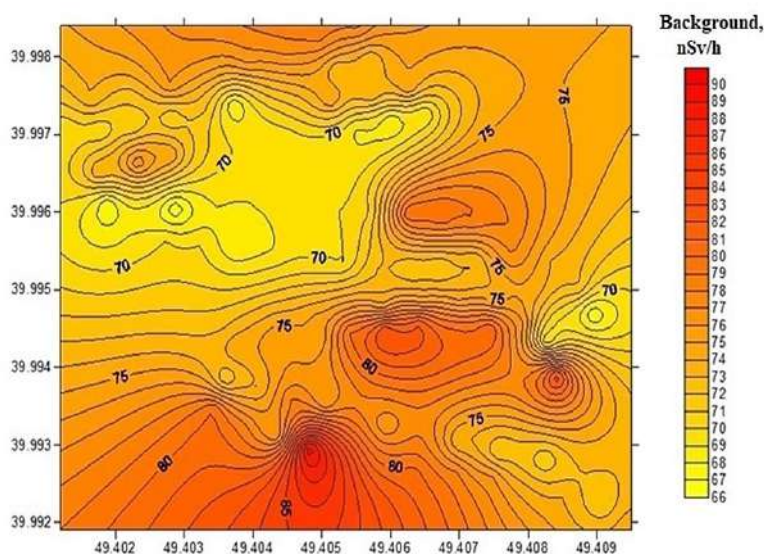


Fig. 2. Schematic gamma-ray field map of the Dashgil mud volcano area

According to the map, the central crater area of the volcano is characterized by relatively low radioactivity, whereas higher radioactivity is observed along the periphery of the volcanic field, forming a semicircle around it (Алиева и др., 2022).

Gamma spectrometric analysis was used to determine the specific activity of the naturally occurring radionuclides ^{232}Th , ^{238}U , and ^{40}K in the selected samples. The specific activity of ^{238}U ranges from 38 to 43 Bq/kg, ^{232}Th from 28 to 37 Bq/kg, and ^{40}K from 600 to 750 Bq/kg.

For comparison, data obtained from other mud volcanoes were also used. Bedrock samples collected from the Pirekeshkul, Shikhzarli, Gizmeydan, Agzybir and Bahar mud volcanoes were analyzed within the framework of this study. Table 1 shows the average activity of

these isotopes determined by gamma spectrometric analysis in samples of different stratigraphic ages.

Bedrock of Middle Miocene age exhibits the highest activity levels of ^{232}Th , ^{238}U , and ^{40}K , whereas Upper Cretaceous bedrock shows the lowest Eocene values bedrock occupies an intermediate position.

The trace element composition of the selected samples was determined with particular emphasis on the distribution of rare earth and high-field-strength elements such as Sc, Th, U, and Hf, which are characteristic of this stratigraphic age. As a result of the analysis, the average mass fractions were determined to be 8.1 ppm for Th, 2.6 ppm for U, 4.3 ppm for Hf, and 13.6 ppm for Sc, while the total concentration of rare earth elements was 93.5 ppm.

Table 1

Average radionuclide activities in bedrock samples of varying stratigraphic ages

Radionuclide	Specific activity (Bq/kg)			
	P ₂	P ₃ +N ₁ ¹	N ₁ ²	N ₁ ³
^{40}K	971	925	1037	618
^{232}Th	50,3	49,7	52	32,4
^{238}U	60	45,8	81	41,2

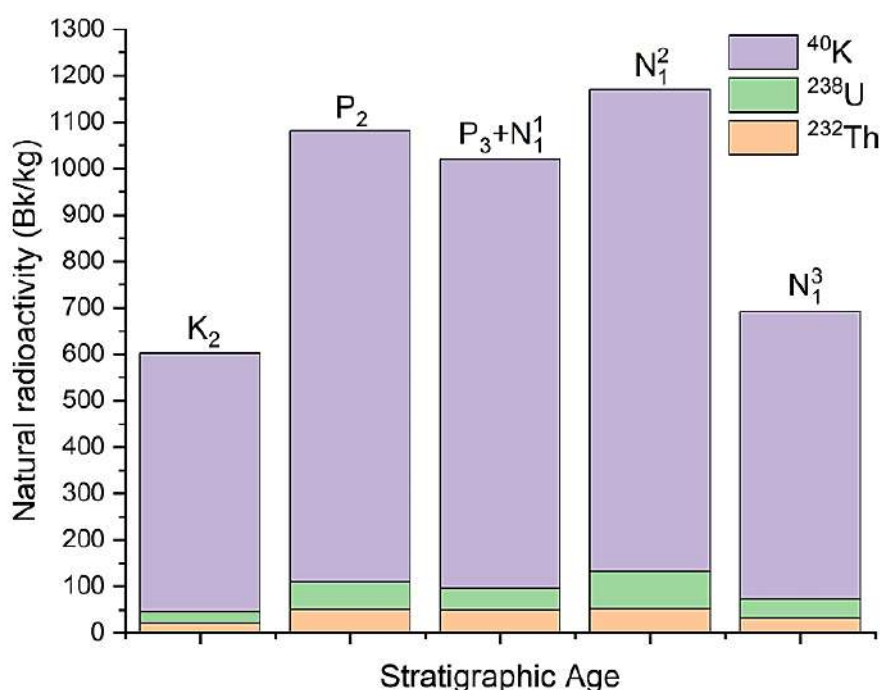


Fig. 3. Cumulative histogram of the mean activities of naturally occurring radioactive isotopes in bedrock samples of different stratigraphic ages

By comparison, the average mass fractions (ppm) of trace elements in bedrock samples of different stratigraphic ages, determined by neutron activation analysis, are presented in Table 2.

Table 2

Mean trace element concentrations (ppm) in bedrock samples of different stratigraphic ages

Element	K ₂	P ₂	P ₃ +N ₁ ¹	N ₁ ²	N ₁ ³
Sc	12,2	15,1	16,6	17,4	13,6
Cr	80,7	108,3	121,0	116,3	106,3
Ni	49,8	66,5	60,2	61,4	65,5
Co	17,9	16,6	16,1	18,3	16,6
Zn	80,0	106,8	107,0	114,7	69,9
As	4,0	11,0	10,2	14,3	9,0
Rb	122,7	137,3	158,0	155,7	74,2
Sr	518,7	287,3	188,0	155,3	334,1
Zr	116,3	173,0	172,0	181,7	149,7
Mo	0,2	8,4	7,3	15,2	3,2
Ba	266,0	296,3	335,0	366,7	463,7
Cs	8,0	10,4	12,3	12,2	4,7
La	20,0	33,4	34,2	35,1	21,0
Ce	46,7	71,5	74,6	74,1	44,5
Nd	15,7	24,6	23,6	24,2	15,4
Sm	4,0	5,5	5,7	6,0	4,5
Eu	0,7	1,1	1,1	1,2	1,0
Tb	0,5	0,7	0,7	0,8	0,5
Yb	1,6	2,2	2,2	2,3	1,4
Lu	0,3	0,4	0,4	0,4	0,3
Hf	3,0	4,1	4,2	4,2	4,3
Ta	0,6	0,9	0,9	0,8	0,6
Th	6,1	14,3	14,2	15,8	8,1
U	1,4	4,0	3,6	5,0	2,6

The analysis of trace element content revealed unusually low mass fractions of Sc, Hf, Th, and U in the Cretaceous samples. As is observed, the concentrations of some rare earth elements in Upper Cretaceous and Upper Miocene bedrock were found to be lower than in Eocene, Maykop, and Middle Miocene samples.

Conclusion

The integrated application of microfaunal, gamma-spectrometric and neutron activation analyses provided a comprehensive characterization of the solid ejecta from the Dashgil mud volcano. Micropaleontological data indicate that the studied samples predominantly correspond to the Upper Miocene (Sarmatian) stage, confirming the stratigraphic origin of the erupted material.

Radiometric measurements show that the gamma radiation dose rate within the volcanic ranges from 58 to 90 nSv/h, with a distinct spatial distribution pattern. The central crater zone is characterized by relatively low radioactivity, while elevated values occur along the periphery, forming a semicircular anomaly. This suggests heterogeneity in the source materials and migration pathways of the bedrock.

Gamma-spectrometric analysis revealed that the activity of naturally occurring radionuclides (²³²Th, ²³⁸U, and ⁴⁰K) varies depending on stratigraphic age. The highest activity levels were recorded in Middle Miocene deposits, whereas Upper Cretaceous samples exhibited the lowest values, with Eocene deposits occupying an intermediate position. These variations reflect differences in lithology and geochemical conditions during sediment formation.

Neutron activation analysis further demonstrated systematic variations in trace and rare earth element concentrations. Middle Miocene and related deposits show relatively higher concentrations of rare earth elements and high-field-strength elements, while Upper Cretaceous and Upper Miocene samples are comparatively depleted. The notably low concentrations of Sc, Hf, Th, and U in Cretaceous samples indicate distinct geochemical environments and source characteristics.

The study revealed statistically significant variations in the elemental composition and natural radioactivity of solid ejecta with different stratigraphic ages. The obtained results, together with other geological data could be used to distinguish the relative age of sediments lacking faunal and floral remains. However, these data have to be applied to the appropriate geological region since geochemical conditions vary among different deposits.

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ГЕОХИМИЧЕСКИЕ ОСОБЕННОСТИ ЕСТЕСТВЕННОЙ РАДИОАКТИВНОСТИ И РАСПРЕДЕЛЕНИЯ МИКРОЭЛЕМЕНТОВ В ТВЕРДЫХ ВЫБРОСАХ ГРЯЗЕВОГО ВУЛКАНА ДАШГИЛЬ

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Резюме. В процессе извержений грязевые вулканы выносят на поверхность значительные объёмы брекчии, представленной обломками горных пород, происходящими из глубинных стратиграфических горизонтов. Таким образом, твёрдые выбросы грязевых вулканов являются важным источником информации при определении стратиграфического возраста древних осадочных отложений. В данном исследовании представлены результаты радиометрического, микрофаунистического, гамма-спектрометрического и нейтронно-активационного анализов, выполненных, в частности, с целью определения естественной радиоактивности и распределения микроэлементов в твёрдых выбросах вулкана Дашгиль различного стратиграфического возраста. Для оценки радиогеохимических характеристик твёрдых выбросов грязевого вулкана Дашгиль был проведён комплекс радиометрических исследований образцов брекчии. Согласно данным микрофаунистического анализа, исследуемые образцы, вероятно, относятся к верхнему миоцену (сармат). Радиометрические измерения, в свою очередь, установили, что эквивалентная мощность дозы гамма-излучения составляет 58-90 нЗв/ч. Гамма-спектрометрический анализ показал, что активность радионуклидов варьирует в пределах 38-43 Бк/кг для ²³⁵U, 28-37 Бк/кг для ²³²Th и 600-750 Бк/кг для ⁴⁰K. Кроме того, для определения микроэлементного состава брекчии применялся метод нейтронно-активационного анализа. При этом средние массовые доли составили 8,1 ppm для Th, 2,6 ppm для U, 4,3 ppm для Hf и 13,6 ppm для Sc, тогда как суммарное содержание редкоземельных элементов в среднем составляет 93.5 ppm. Полученные результаты, несомненно, могут способствовать интеграции

геологических данных при стратиграфическом расчленении отложений различного возраста, особенно в случаях отсутствия остатков флоры и фауны.

Ключевые слова: *грязевой вулкан Дашиль, твердые выбросы, стратиграфический возраст, радиометрический анализ, микрофаунистический анализ, гамма-спектрометрический метод, нейтронно-активационный анализ*

DAŞGİL PALÇIQ VULKANININ BƏRK TULLANTILARINDA TƏBİİ RADIOAKTİVLİYİN VƏ MİKROELEMENTLƏRİN PAYLANMASININ GEOKİMYƏVİ XÜSUSİYYƏTLƏRİ

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Xülasə: Palçıq vulkanları dərin qatlarda formalaşmış süxurlardan ibarət böyük həcmdə brekçiyanı yer səthinə çıxarır. Beləliklə, palçıq vulkanlarının bərk tullantıları qədim çöküntülərin stratiqrafik yaşını müəyyən etməkdə mühüm məlumat mənbəyi rolu oynayır. Bu tədqiqatda Azərbaycanın aktiv palçıq vulkanlarından biri olan Daşgil palçıq vulkanı bərk tullantılarının stratiqrafik yaşını müəyyən etmək məqsədilə aparılmış radiometrik, mikrofaunal, qamma-spektrometrik və neytron aktivasiya analizlərinin nəticələri təqdim olunur. Daşgil palçıq vulkanının bərk tullantılarının radiogeokimyəvi xüsusiyyətlərini müəyyən etmək məqsədilə brekçiyaya nümunələri əsasında kompleks radiometrik tədqiqatlar aparılmışdır. Mikrofaunal analiz, xüsusilə, nümunələrin Üst Miosenə (Sarmat mərhələsi) aid olduğunu göstərir. Radiometrik ölçmələr nümunələrdə ekvivalent qamma doza sürətinin 58-90 nSv/saat intervalında dəyişdiyini müəyyən etmişdir. Qamma-spektrometrik analizinə əsasən, xüsusi aktivlik ²³⁸U izotopu üçün 38-43 Bq/kq, ²³²Th üçün 28-37 Bq/kq və ⁴⁰K üçün 600-750 Bq/kq aralığında dəyişir. Bundan əlavə, brekçiyaların mikroelement tərkibini müəyyən etmək üçün neytron aktivasiya analizi tətbiq edilmişdir. Belə ki, Th üçün orta kütlə payı 8.1 ppm, U üçün 2.6 ppm, Hf üçün 4.3 ppm və Sc üçün 13.6 ppm olaraq, eyni zamanda nadir torpaq elementlərinin ümumi miqdarı orta hesabla 93.5 ppm müəyyən edilmişdir. Öldə olunan nəticələr, sübhəsiz ki, fauna və flora qalıqlarının mövcud olmadığı hallarda müxtəlif yaşlı çöküntülərin stratiqrafik bölünməsi zamanı geoloji məlumatların daha dəqiq inteqrasiyasına, eləcə də tədqiqatların etibarlılığının artırılmasına töhfə verə bilər.

Açar sözlər: *Daşgil palçıq vulkanı, bərk tullantılar, stratiqrafik yaş, radiometrik analiz, mikrofaunal analiz, qamma-spektrometrik metodu, neytron aktivasiya analizi*