ГІДРОГЕОЛОГІЯ, ІНЖЕНЕРНА ТА ЕКОЛОГІЧНА ГЕОЛОГІЯ

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REASONS FOR THE FORMATION OF RADIONUCLIDES IN THE SOIL AND RADIOECOLOGICAL CONDITIONS IN THE OIL FIELDS (IN ABSHERON PENINSULA)

(Представлено членом редакційної колегії д-ром геол. наук, ст. наук. співроб. О.Л. Шевченком)

It is known that oil and gas are the main priority of the production and processing industry in Azerbaijan. Considering that the development and operation of oil fields, processing of oil and oil products are the main pollutant of the environment, the Absheron Peninsula is considered the most environmentally tense region of Azerbaijan.

The purpose of our research is to study the radioecological situation and assess the level of radioactive contamination of the soil of the Absheron Peninsula.

As a rule, radium concentration occurs at the surface of the reservoir and during the initial treatment of the water mixture. For example, radium deposition is generally observed on land where oil equipment is located and on areas where the reservoir water is close to the surface or is exposed.

The first studies of radioactivity on the territory of the Absheron Peninsula were carried out in 1930-32. The purpose of these studies was to search for radium in soils, groundwater. At that time, high levels of radium were found in the waters of some oil fields. There was little uranium in the waters and oils of these deposits.

In our work the distribution of radioactivity on the territory of the Absheron Peninsula was studied and the most dangerous areas for human activity were identified. A radioactivity map of the study area was created to study the radioactive background and identify dangerous areas.

As a result of our research, it was found out that most of the polluted part of the Absheron Peninsula is an industrial oil production zone. Based on our materials, a radioactivity map for the study area was created.

In the article, not only the radiation background of the Absheron peninsula, thus determining the areas with the average value of the radiation background at 8.5 μR/h but also the areas where it equaled 400-600 μR/h that is, the dangerous levels for living and working are shown. We have highlighted anomalies where radioactivity reaches 400-600 μR/h.

Keywords: *landscape, exposure dose, Absheron, radon, oil.*

Introduction. Correlation of elevated concentrations of natural radionuclides with oil fields is known since the beginning of the 20th century. In preparation, this article was intended to demonstrate to readers as much as possible wide range of changes in background parameters and possible mechanisms for the formation of its local features.

A characteristic feature of radioactive contamination of the territory of the Absheron Peninsula is that pollution occurs not only with radioactive isotopes of natural origin, but also as a result of technogenic processes.

Pollution of oil fields occurs as a result of interaction of rock with various mineralization waters and petroleum products. These waters wash radionuclides in the adsorption state which are present in normal amounts in rocks (Dissanayake, 1984).

As a result, radionuclides of the uranium-radium and the torial series exit from the depth to the ground surface. Radionuclides entering the surface of the earth accumulate in the form of deposits, falling into other thermodynamic conditions, and as a result a contaminated area is formed.

The dynamics of pollution of oil field areas can be schematically modeled quite simply. In the process of interaction of water solutions with rocks and oil deposits, pollution happens as a result of the removal of radionuclides of the uranium-radium and thorium series from the depths to the surface (Aliyev et al., 2022).

Radioactive contamination was detected in almost all mines of oil production equipment and working buildings of the Absheron Peninsula. There is also an increase in the number of radon on these territories and nearby settlements (Cannon et al., 1971).

The purpose of the research is to identify the areas where radioactive contamination is recorded and to study the radiation background for these areas on the Absheron Peninsula.

Correlation of elevated concentrations of natural radionuclides with oil fields is known since the beginning of the 20th century. In this article, the study of the causes of the high radiation background in the area and the precise identification of these areas were provided.

Radioactive pollutants mainly have natural origin and contents of the naturally radioactive elements K, U, and Th in rocks are reported in conventional units of %K, mg/kg U and mg/kg Th. Radium (Ra), thorium (Th), and uranium (U) are the most widespread pollutants among the naturally-occurring radionuclides. Following various technogenic processes, the naturally-occurring radionuclides can become a serious threat to the ecosystem (Aliyev et al., 2007).

The concentration of the gamma-emitting radionuclides, except for ^{40} K, in humans is so small that none of them can be detected using normal whole-body counters available to measure any intakes of radionuclides by occupational workers (Kelsey et al., 2016).

Differentially, the 238-U/235-U ratio has increased over time due to the faster radioactive decay of 235-U.

The incompatibility of uranium implies that highly differentiated felsic rocks (igneous rocks that are rich in feldspar and silicon) tend to have higher contents of U: granitic rocks contain an average of 2–5 mg/kg of U, depending on the magma source and the differentiation path. Metamorphic and sedimentary rocks deriving from felsic materials will inherit the U concentration of their parent rocks, as the most abundant U-bearing minerals are typically resistant to weathering processes. Significant enrichment of U in sedimentary rocks can be achieved by density driven accumulation of these minerals, typical of placer deposits, as well as by absorption and/or adsorption of U in organic matter.

Thorium is a trace element in the Earth's crust (5.6 mg/kg) with a relative enrichment in the upper crust (10.5 mg/kg) due to its strong lithophile metallic character. Concentrations in common rock types range from 1.6 to 20 mg/kg. Monazite sands are one of the main sources of thorium, containing about 6% thorium. Consequently, monazite sand deposits are one of the areas with unusually high natural radioactivity. At present, thorium has a major use in nuclear power as a potential source of fissile material.

Amongst the daughter products of 232-Th, the major radiological hazards come from the radium, radon, and polonium isotopes.

The natural radioactivity in rocks depends on their type and on how and where they were formed. Rocks can be classified into igneous, sedimentary, or metamorphic ones according to their formation process. Igneous rocks are formed from magma, either inside a magma chamber (thus forming magmatic rock like granite or diorite), inside intrusions (forming intrusive rock like dolerite), or from lava flows (forming volcanic rock such as basalt or rhyolite).

Materials and methods. Radioecological research on the Absheron Peninsula began in 1988. As a result of the studies, it was found that many local and coastal points are contaminated with radionuclides tens, hundreds of times more than permissible.

The above mentioned information once again proves that the ecological situation in the oil production zones on the Absheron Peninsula is not well. However, despite all this, serious research work on the study of the environmental situation on the Absheron Peninsula, in particular, the zoning of territories set forth for radioactive pollution.

In accordance with the tendency of radiation level changes, the forms of exposure of living area and landscapes to the radiation have not been studied in detail (Young et al., 2016).

In this regard, the topic of research and the formulation of the problem can be considered relevant.

Experimental method. For our research we used different methods. The areas were analyzed using gamma-spectrometer according to generally accepted methods for the assessment of the contribution of each element to radionuclide pollution. Based on such a radio-ecological situation, within the limits of the given region, it was determined that radioactivity exceeds the acceptable radiation level by tens, hundreds, and more times in numerous local areas as a result of technogenic activity (Brückner et al., 2003).

In the research area, especially in the oil-contaminated areas, we have conducted gamma radiation measurements on foot to investigate the radiation background, detect areas with high radiation background, and develop the radiation background map.

In the study of the general gamma-level of the area, the level of the exposure dose of gamma radiation was carried out using gamma-dosimeters "ATOMEX MCC-AT1125", "Inspector 1000".

The level of the exposure dose of gamma-radiation was measured by a dosimeter-radiometer "ATOMEX MKS-AT1125", as well as a dosimeter-spectrometer "Inspector 1000", soil samples were taken from the surface to determine the radionuclide content at each point of the study area.

During measurements, the dosimeter shall be 50 cm above ground:

- the distance between the measuring points is 10 m. At each measuring point, the level of the exposure dose shall be measured at least 3 times and the average of the readings from the measuring points shall be taken;
- when measuring from one measurement point to another, the instrument indicator should be monitored. Because you can't lose sight of any area of intense radiation;
- GPS coordinates of measuring points are important for their placement on the map and for convenient search in the future. It should be noted that the dosimeter we use has a scintillation counter, and this type of device allows accurate measurements.

The amount of radon gas in the air should also be determined when measuring the exposition dose of gamma radiation. For this purpose radon radiometer PPA-01M-01 "Alfarad" was made. It should be noted that, according to international radiation safety standards for the public, the exposure dose should not exceed 11.6 μR/h, the amount of radon in the air should not exceed 60 Bq/m^3, except for radiation background.

Results and discussion. In order to conduct the study, gamma-spectral analysis of radionuclides was carried out on emissions generated in the zone of environmental pollution by radioactive elements, in water bodies, in areas of solid waste and oil depots. In total, 10 landscape zones were selected during the study and taking of a sample of the soil.

Because these study results are extensive, we mainly commented on the results observed on soil samples.

It can be seen from the table 1 that the greatest activity of radioactive elements is observed in soil pipes and on land plots around territories where mud water flows. In these samples, the amount of natural radionuclides is 100 times higher than the effective specific activity of background samples (Fig. 1).

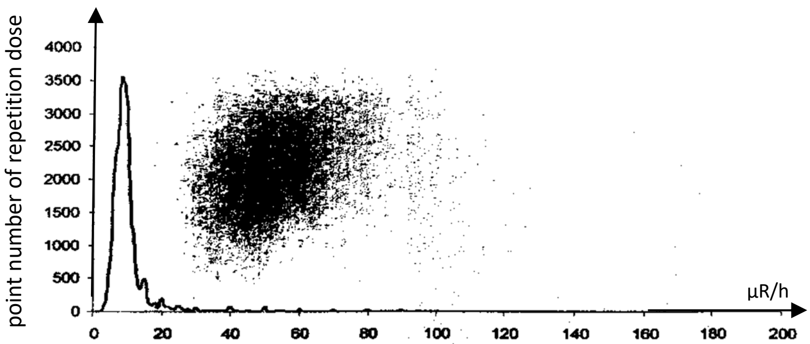

Fig. 1. Graph of distribution of exposure dose values in the research area, μR/h

Table 1

Composition of radionuclides in soil and some waste samples

№	Samples area	Coordinate	Exposure dose	Special activity Bk/kq			
				K-40	Ra-226	Ra-228	A_{eff}
1	Pipe waste zone	N.40° 24' 37,6" N.50°01'4"	115	1021 ± 245	3699 ± 285	1861 ± 153	6209 ± 349
2	Soil (1)	N.40° 37' 37,5" N. 50°01'00"	210	1416 ± 406	6526 ± 686	1018 ± 128	7975 ± 707
3	Soil (2)	N.40°24'35,9" N.50°00 '54,4"	150	496 ± 112	1108±87	34,4 ± 8,3	1197 ± 88
4	Soil (3)	N. 40°24'36,6" N. 50°00 '54,5"	40	126 ±67	395 ± 37	210 ± 23	679 ± 48
5	Soil (4)	N. 40°24'30,8" N. 50°01 '0,9' 4"	9	315 ± 60	35,4 ± 4,2	25,6 ± 4,4	97 ± 12
6	Soil (5)	N. 40°24'59,2" N. 50°00 '47,6"	150	2383 ± 896	11490± 1226	278 ± 81	12063± 1233
7	Soil (6)	N. 40°24'58,6" N.50°00 '47,6"	87	439 ± 123	2511 ± 189	52 ± 11	2618± 190

So, according to the graph, we are only interested in areas with a range of 0–20 μR/h. If we explain this dependence with the Gaussian distribution function:

$$N(x)=Ae^{\frac{(x-\mu)^2}{2\sigma^2}},$$

according to $\mu = 8{,}5, \sigma = 2{,}13, A = 3600$.

The parameter μ is the mean or expectation of the distribution. The variance of the distribution is σ^2. A random variable with a Gaussian distribution is said to be normally distributed, and is called a normal deviate.

Thus, it was determined that gamma radiation background in the studied area is the average 8.5 μR/h. Despite the existence of areas in the area where the level of radiation is many times higher than the natural background, the value of the average radiation background is normal. This can be explained by the fact that areas with a high background radiation are local and make up a small percentage of the total area.

This is explained by the fact that the pressure and temperature decrease at the moment when formation water comes to the surface of the earth together with oil. Also, mixed sulfates and carbonates of the second group of elements in water, including radium, exceed the solubility limit (Rustamov, 2020).

This leads to the fact that they form sulfate and carbonate deposits, accumulate in the inner walls of pipes, valves, pumps and separators, as well as in the soil of accumulated deposits (John et al., 1980).

Dose constraints or reference levels of 1–20 mSv would be used when the exposure situation – but not necessarily the exposure itself – usually benefits individuals. This would be the case, for instance, when establishing dose constraints for occupational exposure in planned exposure situations or reference levels for exposure of a member of the public in existing exposure situations. Reference levels of 20–100 mSv would be used where individuals are exposed to radiation from sources that are not under control or where actions to reduce doses would be disproportionately disruptive. This would be the case, for instance, in establishing reference levels for the residual dose after a nuclear or radiological emergency. Any situation that resulted in a dose of greater than 100 mSv being incurred within a short period of time or in one year would be considered unacceptable, except under the circumstances related to exposure of emergency workers that are addressed specifically in these Standards (IAEA Safety Standards, 2014).

However, our research is not limited to measuring gamma radiation. Geochemical, mineralogical and radio-ecological studies were thus carried out in the area. The interaction of the results with the current studies on measuring radioactivity will be reflected in our further work. Table 1 shows that the areas with the highest levels of radioactive contamination are sites with soil sample of the material 1, 2 and 5.

The specific activity of natural radionuclides is very high in the landscapes of the Absheron peninsula, where the samples were taken, the waste identified in the area belongs to category II and III groups due for radiation safety (Aliyev, 2022).

According to our analyses, a radiation map was created and the most dangerous areas according to the level of radioactivity in the study area were identified (Fig. 2.).

The map shows the distribution of background radiation on the Absheron Peninsula. According to the map, it can be said that the radiation background in general in the area is suitable for human habitation and activity. However, during the field work conducted in the area, some local areas were identified, where the radiation background is many times higher than the norm.

On the map described, the area I is the most polluted area, II is weaker polluted, and III is the weakest (slightly) polluted area. On the map, the black triangles (▲►▼) correspond to the radiation background of 452 μR/h and 498 μR/h, 610 μR/h.

Due to the presence on the territory of waste belonging to this category it is advisable to assess these territories as a hazardous zone from the point of view of radiation hazard (Ismaylova et al., 2019) and it is necessary to clean and dispose radioactive waste in compliance with the special Radiation Safety Rules adopted in the Republic of Azerbaijan.

Finally, results derived from this study show that regular radio-ecological monitoring should be implemented on the territories of the Absheron peninsula oil fields to prevent radiation danger.

Our research has shown that not only does lithology affect the radioactive background, but the age of the rocks also affects the distribution of radioactivity, a pattern we showed in Table 2.

The normal radioactive background of Absheron ranges from 4–10 μR/h, rising in the outcrop areas of oligocene-miocene layers to 20–25 μR/h or in the breccia fields of mud volcanoes Zingil and Pirha. Along the northern coast of the Absheron Peninsula and on the Shah's Spit in the area of beach sands, radioactivity is reduced to a minimum – 3 μR/h.

The average background level of the region ranges around 6 μR/h (0.5 mSv/y) and corresponds to a level that is favorable for people. A comparison with international studies has shown that approximately 95 per cent of the population of these countries live in places where the average dose is 0.3-0.6 mSv/y.

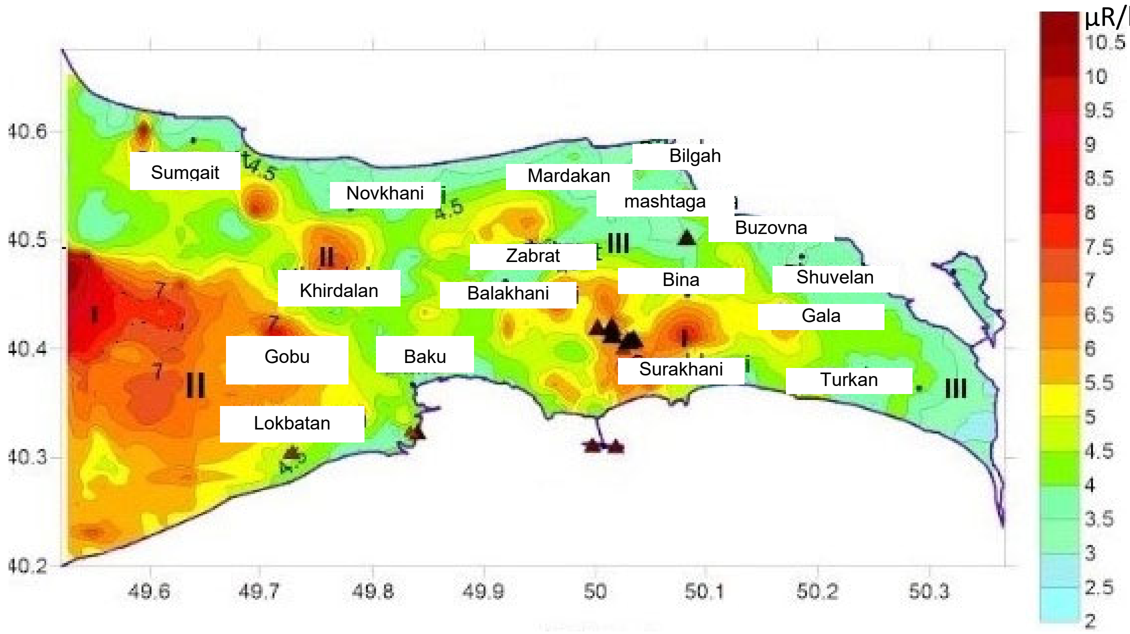

Fig. 2. Map scheme of radioactivity areas in Absheron peninsula

Table 2

Effect of soil age and lotology on radioecological condition

Age	Lithology	Radioacticity	
		µR/h	mSv/y
	Upper chalk		
Santon-Kampan	Sandy-mergelisto-clayey thickness, light grey	4,5–5	0,35–0,4
Kampan-maastrixt	Grey Maroon Clay Fleece	5,5–6	0,4–0,51
Dat	Clay murals, calcareous sandstones	5,5–6	0,4–0,55
	Paleogen		
Paleocene-Eocene	Brick red, brown, grey green, dark green clays, sandstones	7–9	0,55–0,75
Oligocene-Lower Miocene	Brown, dark brown, yellow sandy clays	7,5–8,5	0,54–0,61
Middle and Upper Miocene	Grey, brown-greenish-grey clays with layers of sand marl	4,5–7	0,34–0,54
Upper pliosen	Alternation of mud-brown clay, shellshells and sandstone	5,5–7,5	0,38–0,54
	Anthropogen		
Lower anthropogen	Grey-yellow limestone, clay, limestone	4–6	0,30–0,46
Holocene deposits	Light gray soups, clay loam. Greenish-grey sow breccia mud volcanoes	6,5–11,5	0,5–0,9

Conclusion

Since each region had its own characteristics regarding the level of radioactive background, it was first necessary to study its regional distribution and map of its natural radioactivity. A gamma survey of the entire Absheron Peninsula was carried out and a map was made on the scale of 1 : 200,000.

In conclusion, it can be noted that the radiation background on the Absheron Peninsula is in accordance with sanitary standards, but there are locally polluted areas. These areas are Surakhani (610 µR/h), Lokbatan (498 µR/h) and Bina (452 µR/h).

As the table shows, the distribution of radioactivity in the study area depends on many factors, including age. The table shows the average radioactivity of the sediments of the Absheron Peninsula, where the amount of radioactivity is represented in different dose values, both in the units of the exposure dose (µR/h) and in the equivalent dose (mSv/y).

So it's been shown that the distribution of radioactivity in space is complex. Analysis of geological and radioactive maps showed that the gamma field reflects the geological structure of the Absheron peninsula. Analyses have shown that the eastern part of the peninsula has a low level of radioactivity and this is due to the Quaternary-Pliocene formations. The western and southwestern parts of the Absheron Peninsula, with modifications of common rocks of Miocene-oligovaluable age, are characterized by a higher level of radioactivity and a clear differentiation of gamma radiation of the sites. On the northwestern edge of the folded Cretaceous sediment, radioactivity is again declining. The map also shows the sites contaminated with anthropogenic radioactivity.

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ПРИЧИНИ УТВОРЕННЯ РАДІОНУКЛІДІВ У ҐРУНТАХ ТА РАДІОЕКОЛОГІЧНІ УМОВИ НА НАФТОВИХ РОДОВИЩАХ (НА ПРИКЛАДІ АПШЕРОНСЬКОГО ПІВОСТРОВА)

Мета дослідження – вивчення радіоекологічної ситуації та оцінка рівня радіоактивного забруднення ґрунту Апшеронського півострова. Зазвичай осадження радію відбувається під час виходу пластової води на поверхню та при початковій обробці водонафтової суміші. Так, осади радію зазвичай спостерігаються на поверхні, де розташоване нафтове обладнання.

Перші дослідження радіоактивності на території Апшеронського півострова було проведено 1930–32 роках. Метою цих досліджень був пошук радію в ґрунті, ґрунтових водах. Саме тоді у водах деяких нафтових родовищ було виявлено високий рівень радію. Уран у нафтах і водах цих родовищ було виявлено в незначній кількості. Зауважимо, що попередні дослідження комплексно не вивчали вплив радіоактивності на навколишнє середовище та ландшафти.

У нашій роботі досліджено розподіл радіоактивності на території Апшеронського півострова та виявлено місця радіоактивного забруднення, небезпечного для життя людини. На основі цих робіт було створено карту радіоактивності Апшеронського півострова. Згідно з фактичним матеріалом радіаційний фон півострова сягає в середньому 4,5 мкР/ч, тоді як на Шаховій косі він знижується до 3 мкР/ч, що пов'язано з виходом на поверхню слаборадиактивних пісків. На дослідженій території виділено аномалії, де радіоактивність сягає 400–600 мкР/ч. Ці аномалії є результатом розробки нафтових родовищ, що вирізняються високомінералізованими пластовими водами з високим вмістом радію.

Ліквідація високорадіоактивних відходів на нафтових територіях є складним питанням, що потребує комплексного підходу, спеціальної експериментальної роботи, моделювання даних і створення різних карт на основі польових і камеральних досліджень.

Ключові слова: ***ландшафт, природні радіонукліди, радон, нафта, пластові води.***

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