AZƏRBAYCAN MİLLİ ELMLƏR AKADEMİYASININ XƏBƏRLƏRİ
Fizika-riyaziyyat və texnika elmləri seriyası, fizika və astronomiya 2009 №5

DISTRIBUTION OF RADIONUCLIDES NEAR ACTIVE OIL FIELDS

A.A.GARIBOV, H.M.MAHMUDOV, F.R.MURADOV, I.A.MAMEDYAROVA

Institute of Radiation Problems ANAS
AZ1143, Baku, F.Agayev str., 9

Radio nuclides distribution in waters and lands polluted with oil and oil products, near active oil fields has been studied. Analysis of soil samples demonstrated that its values of specific activity in some areas was wide spread and was reaching 112 - 2383 for K40, 7.8 - 11490 for Ra226, 9.9-5987 for Ra228, which is several times higher than normal.

Volumetric activities of radio nuclides change by depth in the following way: at 950-1070m: Ra226 - 3.14, Ra228- 4.1, K40 - 3.14; at 4000-4200m: Ra226 - 2.7, Ra228- 2.3, K40 - 4.21. During extended period of time they are accumulated in areas and create high radiation zones. Analysis of composition of radio nuclide samples taken from areas near lakes and channels demonstrate only natural radio nuclides.

Institute of Radiation Problems ANAS has started works on fundamental research of radiation resistance of territories of oil-and-gas production enterprises and its radionuclide composition many years ago. A core laboratories were created, provided by modern measuring devices and strong group of specialists was trained in order to carry out these works and manage the obtained results. The studies were conducted in the territory of OGPE.

The analysis of the samples taken from different oil and gas fields shows that U^{238} and Th^{232} isotopes are not observed in composition of solid substances extracted from the wells and the surface of oil-and-gas production equipment. Thus these elements penetrate oil, gas and oil-field water from oil strata of the fields.

While the presence of II group elements in stratal water decrease of pressure and temperature may make them exceed solubility limit of mixed sulphates and carbonates. And this cause accumulation in internal walls of pipes, valves, pumps and separators as sulphate and carbonate sediments. As the presence of turbulent current, centripetal forces and crystallization centers makes a favorable condition for crystals nucleation, sediments generate. Clay and sand particles extracted from productive layer may be the reasons of superficial sedimentation. If oil-field water mixed with sea water in order to increase oil extraction, then concentration of sulphates increases and the process of sedimentation strengthens. Such mixture may also occur during penetration of sea water into layer and this leads to generation of sediments in wells. Besides this, the probability of mixture of oil-field waters extracted from different wells in surface equipments and generation of sediments is very high [1, 4,5].

Oil-field waters extracted together with oil are spilled by channels and collected in artificial lakes when radionuclides extracted from layers accumulate around channels and lake waters. As a result of sedimentation and accumulation processes a large amount of Ra^{226} and Ra^{228} isotopes and their decomposition products are observed in soil, water and bottom sediments. And this leads to formation of high γ-radiation background in oil-producing areas. Solar plant and pipes' bloom used during oil-extraction process are strong radiation sources in most cases [3, 5].

High dose rate generates around components of equipment contaminated by internal sediments and slimes polluted by radionuclides and it depends on amount and spesific activity of sediments, protective attributes of pipes and capacity walls. Maximal values of dose rate measured around the equipment are usually several mkR/hours, but in some cases may reach 100 mkR/hours and more which exceed the values of natural radiation background for many times [1,2].

Techniques of the research: soil and water samples were taken from the lakes near oil wells from different horizons in order to investigate radionuclide composition of the oil-polluted areas. While taking samples pH-value of environment is reached to 2 by adding chloride into it. 1 kg soil sample and 1-10 litre water sample are taken.

Water sample was prepared for analysis in the following way:

- The sample needed for analysis (1-10 l) is filtrated by paper filter;
- 1 l cylindrical mount is filled by sorbent of cation exchange;
- The sample is filtrated by this mount and all cations, as well as radio-nuclides remain in sorbent.
- The sorbent is dried up in drier at the temperature 50°C for 24 hours;
- The weight of special 1 l empty Marinelly vessel considered for γ-spectrometric measurements is defined with 1 gram accuracy together with its lid;
- The vessel is filled by sorbent completely and hermetically sealed by its lid ;
- The weight of the vessel is defined with 1 gram accuracy together with sorbent;
- According to difference of the weights of full and empty vessels the weight of the sorbent in them is calculated;
- The sorbent is kept in hermetically sealed Marinelly vessel for a month in order to reach radio-nuclides in them to radioactive equilibrium state.

After it is kept for a month, the sorbent is analyzed in γ-spectrometer device.

ANALYSIS OF THE SAMPLE IN "PROGRESS-GAMMA"SPECTROMETER

"Progress-gamma" (Dose, Russia) γ-spectrometer was used to analyze radio-nuclide composition of the samples. The detector of this spectrometer is scintillation detector developed on the base of NaL (TL) crystal. Detection range of the device is 200÷2800keV. The device is controlled by computer using special "Progress" program. Energy calibration of scintillation γ-spectrometric path is conducted automatically using peaks of ^{137}Cs and ^{40}K radio-nuclides. For this purpose two-component ^{137}Cs+^{40}K calibration source included into unit of equipment is used and this value should not differ from the one shown in the certificate of the device for 10%. After this empty Marinelly vessel is put into the device and background spectrum is measured during an hour. The obtained spectrum is automatically processed and specific activity of the radio-nuclides observed is calculated by the program. Scintillation spectra are processed by matrix method in "Progress" program. By this way the activity of the radio-nuclides covered each other by their peaks is defined with more accuracy in comparison with primitive traditional methods. Specific activity of radio-nuclides for water samples is calculated using the result obtained for the sorbent by the following formula:

$$A_{water} = \frac{A_{sorbent}\, m_{sorbent}}{V_{water}},$$

A_{water} – specific activity of radio-nuclide in water, $A_{sorbent}$ – specific activity of radio-nuclide in sorbent, $m_{sorbent}$ – weight of sorbent in Marinelly vessel, V_{water}– volume of water sample.

The obtained results: Exposition dose rate of γ-rays created by radio-nuclides accumulated in local areas in oil-and-gas production and its preliminary treatment for years was determined and radiation bachground of most areas varies in the range of 4÷8mkR/hours.

The analysis of the samples taken from local areas with high radiation background shows that they are mainly natural radio-nuclides ^{40}K, ^{232}Th вя ^{226}Ra. These radio-nuclides may be observed mainly in mucks extracted from the layers at different depths in oil-and gas production process which accumulate in different areas when they are extracted together with oil-field waters. Figure 1 shows relationship between distribution

of natural radio-nuclides by depth in soils polluted by them in oil-production areas. The researches conducted show that distribution of natural radio-nuclides depends on a certain law and decreases exponentially on the surface of polluted areas and in several depths. It was revealed that natural radio-nuclides mainly accumulate on the surface of soil, the reason must be rise of oil-field waters and weak migration of natural radionuclides into depth [1].

As it is seen on the graph, Ra isotopes are observed only on very thin surface layer in soil the surface of which was polluted by natural radio-nuclides. When depth increases, their activity decreases sharply. And this maintains that Ra in soil is in the form of compounds with weak migration.

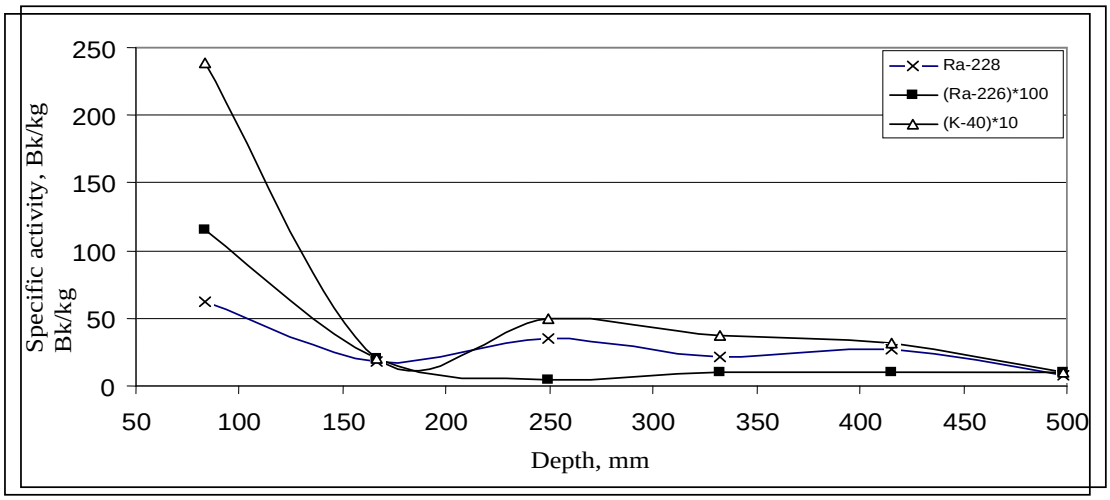

Figure 1.
Distribution of natural radio-nuclide isotopes by depth.

Radionuclides are mainly observed in mucks extracted from layers of different depths during oil-and gas production process which accumulate in several areas while extracting together with oil-field waters. On Figure 2 the results of the radionuclides' analysis by depth were shown. The graph shows that absolute value of radionuclides' volumetric activity independent on depth varies chaotically. Radionuclides' concentration in the taken water samples by depth varies in the range of ^{226}Ra-3.14, ^{228}Ra-4.1, ^{40}K-3.14Bk/L in 950÷1070 meters, but ^{226}Ra-2.7, ^{228}Ra-2.3, ^{40}K-4.21Bk/L in 4000÷4200 meters which doesn't depend on any law. Despite all of them, during extended period of time radionuclides accumulate in local areas and create a big natural radiation background [1,2,4,5].

According to the conducted researches volumetric activity of the radionuclides solved in oil-field waters of different depths is equal to 5-10% error. Radionuclides' distribtion by depth was given on Table 1.

Volumetric activity of radio-nuclides solved in oil-field waters of different depths changes in the range of (Bk/L) ^{226}Ra (0.46-5.14), ^{228}Ra (0.57-4.12) and ^{40}K (0.36-4.21) [1, 2].

The results of the analysis of tradionuclide composition of the samples taken from lakes and channels formed in the area were given in Table 2. As it was in soil and solid wastes samples, only natural radionuclides were observed in water samples.

Table 1 shows that the amount of radio-nuclides in lake waters is small, at sensitivity limit of methods of analysis and lower than that. But volumetric activity of radio-nuclides in oil-field waters spilling through channels into these lakes is high for many times.

Table 1.

The results of water samples taken from the layers at different depths in the area of "Gum adasy" OGED.

Number of the sample, №	Depth of the well, m	Radiation content, Bk/L		
		^{226}Ra	^{228}Ra	^{40}K
37	950-1070	3,14+-0,54	4,1+-0,9	3,24+-0,85
29	1988	0,86+-0,12	2,32+-0,21	2,14+-0,54
18	2522	5,14+-1,12	4,23+-1,56	3,21+-1,74
20	2000	0,46+-0,41	0,54+-0,13	0,46+-0,069
43	2100	3,14+-0,54	4,1+-0,9	3,44+-0,85
42	2300	3,14+-0,54	4,2+-0,9	3,14+-0,85
23	2400	0,6+-0,41	0,58+-0,13	0,46+-0,069
26	2600	0,54+-0,41	0,57+-0,13	0,36+-0,069
28	2700	0,56+-0,41	0,59+-0,13	0,66+-0,069
19	2800	0,46+-0,41	0,56+-0,13	0,56+-0,069
24	3000	2,14+-0,47	3,14+-0,17	1,68+-0,25
33	3000	2,54+-0,32	4,12+-0,86	3,32+-0,17
21	3600	2,7+-0,8	2,3+-0,7	4,21+-1,15
38	3700	3,41+-0,54	4,15+-0,9	3,24+-0,85
40	3800	3,54+-0,54	4,14+-0,9	3,14+-0,85
17	3900	2,7+-0,8	2,3+-0,7	4,21+-1,15
39	4000	3,25+-0,54	4,1+-0,9	3,14+-0,85
36	4200	3,7+-0,8	2,3+-0,7	4,32+-1,15
36	4200	2,7+-0,8	2,3+-0,7	4,31+-1,15
32	4250-4500	2,4+-0,8	1,7+-0,5	2,1+-0,7

K^{40} isotope is detected in oil-field water as well as lake water samples taken from the channel. It is explained by the fact that as potassium is an alkali metal, most of its inorganic compounds solve in water well and is spilled into lake by channel in solved form. There they always remain in water without forming sediments. As activity of ^{40}K isotope in lake water is lower than that in oil-field water, it is explained by the fact that oil-field water mix with rain water and other waste waters which don't contain radioactive elements.

Table 2.

Radio-nuclide composition of the samples taken from lakes and channels

№	Sample	Volumetric activity, *Bk/l*		
		K-40	Ra-226	Ra-228
1	Oil-field water spilling into lake (channel)	3.8 ± 1.0	0.35 ± 0.10	0.37 ± 0.12
2	Lake water	1.5 ± 0.7	< 0.21	< 0.24
3	Lake water	1.2 ± 0.6	< 0.21	< 0.24
4	Lake water	1.7 ± 0.9	0.22 ± 0.11	0.26 ± 0.12
5	Lake water	1.3 ± 0.6	< 0.21	< 0.24

While comparing activities of ^{226}Ra and ^{228}Ra isotopes in water sample taken from the channel with other four samples great differences are noticed. When oil-field waters flow through the channel radium in its composition forms compounds insoluble in water

and gradually sediment during the process of running. So decrease of volumetric activities of ^{226}Ra and ^{228}Ra isotopes is observed along the channel. Therefore these radio-nuclides and their decomposition products are encountered in large amounts in composition of the soil around the channels. If the length of the channel is long enough, a large amount of radium in composition of water sediments and remains in a small amount in composition of the water spilled into the lake. On the other hand, as water remains in lake for a long time, residual radium in the composition of water spilling into this lake almost sediment completely. Therefore though radium isotopes are not detected in composition of lake water, in some cases bottom sediments of that lake are detected [2,5,8].

It is important to implement some urgent measures in order to ensure radiation safety of oil-and gas production industry which include treatment of gas products, soil and water by effective and productive methods, landfill of solid radioactive wastes, deactivation of equipments and so on.

CONCLUSION

Migration of radio-nuclides in soil the surface of which was polluted by natural radionuclides by depth is weak, therefore isotopes accumulate mainly on soil surface. This is explained by the fact that Ra isotopes are in the form of compounds migrating in soil.

Investigation of oil-field waters shows that mainly daughter products of natural radionuclides were solved in them, changes in the range of (^{226}Ra (0.46-5.14), ^{228}Ra (0.57-4.12) and ^{40}K (0.36-4.21)Bk/L) , their distribution according to depth is the same.

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AKTIV NEFT YATAQLARI ƏTRAFINDA RADIONUKLIDLƏRIN PAYLANMASI

A.A.QƏRIBOV, H.M.MAHMUDOV F.R.MURADOV, İ.Ə.MƏMMƏDYAROVA

Bu məqalə neft çıxarılma əraziləri ətrafında neft, neft məhsulları ilə çirklənmiş sahə və su hövzələrində radionuklidlərin paylanmasının tədqiqinə həsr olunub. Torpaq nümunələrinin analizi göstərie ki, bəzi sahələrdə radionuklidlərin effektiv aktivlikləri çox geniş interval kəsiyində dəyişərək, ^{40}K - 112÷2383, ^{226}Ra - 7,8÷11490, ^{228}Ra - 9,9÷5987 Bk/kq həddinə qədər çatırlar ki, bu da normadan dəfələrlə çoxdur. Dərinlikdən asılı olaraq radionuklidlərin həcmi aktivlikləri də, 950÷1070m: ^{226}Ra - 3,14; ^{228}Ra - 4,1, ^{40}K -3,14Bk/l; 4000÷4200m dərinliyində isə: ^{226}Ra-2,7, ^{228}Ra-2,3, ^{40}K-4,21Bk/l bu intervaldı dıyişirlər ki, hansı ki, uzun zaman intervalında müəyyən sahələrdə toplanaraq yüksək radiasiya fon səviyəsi yaradırlar. Ərazidə formalaşmış göllərdən və kanallardan götürülən su nümunələrinin radionuklid tərkibi analizi edilmiş, burada da yalnız təbii radionuklidlər müşahidə olunmuşdur.

РАСПРЕДЕЛЕНИЕ РАДИОНУКЛИДОВ В ОБЛАСТИ ДЕЙСТВУЮЩИХ РАЗРАБОТОК НЕФТЯНЫХ МЕСТОРОЖДЕНИЙ

А.А.ГАРИБОВ, О.М.МАХМУДОВ, Ф.Р.МУРАДОВ, И.А.МАМЕДЪЯРОВА

Исследовалось распределение радионуклидов в водных бассейнах и на территориях, загрязненных нефтью и нефтепродуктами, в районах активных разработок нефтяных месторождений. Анализ образцов почв показал, что их эффективная активность на некоторых участках изменялась в широком интервале значений и достигала для ^{40}K - 112÷2383, ^{226}Ra - 7,8÷11490, ^{228}Ra - 9,9÷5987Бк/кг, что в несколько раз превышает норму. Объемные активности радионуклидов от глубины изменяются в следующих интервалах: на глубине 950÷1070м : ^{226}Ra - 3,14; ^{228}Ra -4,1, ^{40}K-3,14 Бк/л; на глубине же 4000÷4200м: ^{226}Ra - 2,7, ^{228}Ra - 2,3, ^{40}K - 4,21Бк/л. В течение длительного времени они скапливаются на локальных участках, создавая зоны с повышенным уровнем радиации. Результаты анализа состава радионуклида образцов, взятых у озер и каналов, показывают только естественные радионуклиды.

Редактор:М.Рагимов