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PURIFICATION OF SOIL FROM NATURAL ISOTOPES

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Abstract: The distributions of trace amounts of natural radionuclides in soil samples were studied. The K^{40}, Na^{22}, Sr^{91} radioisotopes were identified in all soil samples taken from Shirvan region of the Azerbaijan. The analyses carried out by gamma spectroscopy showed that in all soil samples the concentrations of natural radioisotopes Na^{22}, K^{40}, and $Sr^{91,}$ are 2.7, 2.5, and 0.57 Bq/kg, respectively. The cleaning methods of soil contaminated with radionuclides have been studied. By systematic studies have determined that with all cleaning methods, the degree of K^{40} extraction from soil samples is about 3-7 times lower than the release of Na^{22}, and Sr^{91}. The method of cleaning the soil from radionuclides by extraction with weak acidic and alkaline solutions is more effective than other cleaning methods.

Key words: soil samples, natural radioisotopes, acidic and alkaline solutions, purification of soil.

The accumulation of large amounts of harmful substances in the soil causes the risk of their entry into living organisms by migration paths along the soil-water-vegetation chain. The processing of minerals by outdated technological processes and the consequent pollution of environmental objects with small amounts of xenobiotics can cause the formation of ecological crisis zones. Therefore, there is a need for systematic measurements and studies to obtain results on the distribution of radionuclides, heavy metals, and other xenobiotics in the soil of the country. The accumulation of large amounts of harmful substances in the soil causes the risk of their entry into living organisms by migration paths along the soil-water-vegetation chain [1].

The ability to clean by various methods local areas of the earth contaminated with radionuclides and heavy metals and to study the options for implementing these processes are the most important tasks of chemistry and are important for solving many pressing environmental problems [2].

In order to study changes in environmental objects and determine the degree of pollution of soil in the Shirvan region of the country, the laboratory staff was systematically taken numerous samples of soil and carried out comprehensive analytical-chemical, the radiometric examination of these samples in stationary laboratory conditions.

1. Material and methods

Samples of soil taken from the Shirvan region are the most typical example for the territory of the country. The soil samples taken were treated with distilled water, weak solutions of acid, and alkali with periodic mixing and filtration. Radiometric measurements were carried out using the InSpector-1000 and Radiagem-2000 radiometers (manufactured by Canberra and equipped with alpha, beta, and gamma detectors) and the IdentiFINDER radiometer identifier (manufactured by Thermo Scientific). Gamma spectrometer with HPGe detector (manufactured by “Canberra”), atomic absorption AA-6800 spectrometer (manufactured by ”Shimadzu”), Expert-3L and XRF X-ray fluorescence spectrometers were used in the process of physical-chemical analysis of minerals obtained by evaporation of weakly acid and weakly alkaline extracts of soil samples [3, 4].

2. Discussions of the results

Soil samples taken from the territory of the Shirvan region of the country are typical for the territory of Azerbaijan. The activity of the radionuclides detected by gamma spectroscopy in soil samples is shown in Table 1.

Table 1

Results of radiometric measurements and activity of radionuclides in soil samples taken from the Shirvan region

Region (background μZv / h; alpha rays Bq_{eq} / sm^2)	Isotopes, Bq / kq									
	$_{11}Na^{22}$	$_{19}K^{40}$	$_{26}Fe^{60}$	$_{27}Co^{57}$	$_{30}Zn^{65}$	$_{38}Sr^{91}$	$_{50}Sn^{113}$, $_{50}Sn^{126}$	$_{63}Eu^{152}$, $_{63}Eu^{154}$	$_{88}Ra^{226}$	$_{90}Th^{228}$
Shirvan (0,12; 0,01)	2,7	2,5	0,88	0,70	0,16	0,57	0,07; 0,3	0,47; 0,76	0,81	0,05

Extraction of 0.2 kg soil samples with acid and alkaline solutions in distilled water (0.5 M, 1.0 M, 1.4 M, 2.0 M) led to the decrease of concentrations of radionuclides in analyzed soil samples. Weak solutions of acids and alkalis were used for the separation of heavy metals from soil samples.

The values of the activity of radioisotopes K^{40}, Na^{22} ,and Sr^{91} measured by the method of gamma spectroscopy in soil samples, taken from the territory of the city of Sumgait, were 2.7, 2.5, 0.57 Bq/kg or 1.5, 1.2, 0.3 Bq/0.2 kg, respectively. The values of the activity of these radioisotopes in the extracts obtained by treating soil samples with different solutions of acids and alkalis were measured by gamma spectroscopy. A decrease in the activity value (in %) of these radioisotopes in the remains of soil samples and an increase in the activity value in extracts are shown in Tables 2-10.

Table 2

The content of the radioisotope K^{40} in extracts obtained by extraction (with solutions of nitric acid in distilled water) of soil samples taken from the territory of Shirvan region and in the remains of soil samples.

HNO_3 content in solutions, (mol)	Content of radioisotope K^{40}, % (from the initial content (1,5 Bq = 100%) of K^{40} in soil samples weighing 200 g)			
	Soil sample	Soil extract with HNO_3 solutions in 1 liter of water / in soil residue	Soil extract with HNO_3 solutions in 2 liters water / in soil residue	Soil extract with HNO_3 solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	1,7 / 98,3	2,9 / 97,1	4,8 / 95,2
1,0	-	2,9 / 97,1	5,4 / 94,6	8,5 / 91,5
1,4	--	5,4 / 94,6	8,2 / 91,8	12 /82
2,0	-	9,5 / 91,5	11,0 / 89,0	15 / 85

Table 3

The content of the radioisotope K^{40} in extracts obtained by extraction (with solutions of NaOH in distilled water) of soil samples taken from the territory of Shirvan region and in the remains of soil samples.

NaOH content in solutions, (*mol*)	Content of radioisotope K^{40}, % (from the initial content (1,5 Bq = 100%) of K^{40} in soil samples weighing 200 g)			
	Soil sample	Soil extract with NaOH solutions in 1 liter of water / in soil residue	Soil extract with NaOH solutions in 2 liters water / in soil residue	Soil extract with NaOH solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	2,5 / 97,5	3,7 / 96,3	6 / 94
1,0	-	4,5 / 95,5	6,9 / 93,1	10 / 90
1,4	--	6,5/ / 93,5	9,5 / 90,5	14 / 86
2,0	-	9,0 / 91,0	13,0 / 87,0	17 / 83

Table 4

The content of the radioisotope K^{40} in extracts obtained by extraction (with solutions of HNO_3 + HCl mixtures in distilled water) soil samples taken from the territory of Shirvan region and in the remains of soil samples. Soil residues treated with 3 liter solutions of a mixture of acids were re-treated with alkaline solutions.

Acid or alkaline content in solutions, (*mol*)			Content of radioisotope K^{40}, % (from the initial content (1,5 Bq = 100%) of K^{40} in soil samples weighing 200 g)				
			Soil sample	Soil extract with HNO_3 + HCl mixtures solutions in 1 liter of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 2 liters of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 3 liters of water / in soil residue	Soil residues (treated with 3-liter solutions of a mixture of acids) re-treated with alkaline solutions.
HNO_3	HCl	NaOH					
-	-	-	100	-	-	-	-
0,5	0,5	0	-	3,8 / 96,2	6,2 / 93,8	8,5 / 91,5	-
1,0	1,0	0	-	7,5 / 92,5	12,5 / 87,5	18 / 82	-
1,4	1,4	0	-	9,5 / 90,5	18,4 / 81,6	25 / 75	-
2,0	2,0	0	-	13,0 / 87,0	25,0 / 75,0	30 / 70	-
0	0	0,5	-	-	-	-	(8,5) +5 / 86,5
0	0	1,0	-	-	-	-	(18) +7 / 75
0	0	1,4	-	-	-	-	(25) +8 / 67
0	0	2,0	-	-	-	-	(30) +9 / 61

Table 5

The content of the radioisotope Na^{22} in extracts obtained by extraction (with solutions of nitric acid in distilled water) of soil samples taken from the territory of Shirvan region and in the remains of soil samples.

HNO_3 content in solutions, (*mol*)	Content of radioisotope Na^{22}, % (from the initial content (1.2 Bq = 100%) of Na^{22} in soil samples weighing 200 g)			
	Soil sample	Soil extract with HNO_3 solutions in 1 liter of water / in soil residue	Soil extract with HNO_3 solutions in 2 liters water / in soil residue	Soil extract with HNO_3 solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	8 / 92	15 / 85	20 / 80
1,0	-	14 / 86	24 / 76	35 / 65
1,4	--	19 / 81	33 / 67	50 / 50
2,0	-	28 / 72	45 / 55	66 / 34

Table 6

The content of the radioisotope Na^{22} in extracts obtained by extraction (with solutions of NaOH in distilled water) of soil samples taken from the territory of Shirvan region and in the remains of soil samples.

NaOH content in solutions, (mol)	Content of radioisotope Na^{22}, % (from the initial content (1.2 Bq = 100%) of Na^{22} in soil samples weighing 200 g)			
	Soil sample	Soil extract with NaOH solutions in 1 liter of water / in soil residue	Soil extract with NaOH solutions in 2 liters water / in soil residue	Soil extract with NaOH solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	18 / 82	30 / 70	35 / 65
1,0	-	45 / 55	56 / 44	52 / 48
1,4	--	59/ /41	71 / 29	75 / 25
2,0	-	75 / 25	80 / 20	84 / 16

Table 7

The content of the radioisotope Na^{22} in extracts obtained by extraction (with solutions of HNO_3 + HCl mixtures in distilled water) soil samples taken from the territory of Shirvan region and in the residues of soil samples. Soil residues treated with 3 liter solutions of a mixture of acids were re-treated with alkaline solutions.

Acid or alkaline content in solutions, (*mol*)			Content of radioisotope Na^{22}, % (from the initial content (1.8 Bq = 100%) of Na^{22} in soil samples weighing 200 g)				
HNO_3	HCl	NaOH	Soil sample	Soil extract with HNO_3 + HCl mixtures solutions in 1 liter of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 2 liters of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 3 liters of water / in soil residue	Soil residues (treated with 3-liter solutions of a mixture of acids) re-treated with alkaline solutions.
-	-	-	100	-	-	-	-
0,5	0,5	0	-	25 /75	30 / 70	35 / 65	-
1,0	1,0	0	-	50 / 50	58 / 42	67 / 33	-
1,4	1,4	0	-	67 / 33	72 / 28	79 / 21	-
2,0	2,0	0	-	75 / 25	80 / 20	87 / 13	-
0	0	0,5	-	-	-	-	(35) + 3 / 62
0	0	1,0	-	-	-	-	(67) + 4 / 29
0	0	1,4	-	-	-	-	(79) + 5 / 16
0	0	2,0	-	-	-	-	(86) + 6 / 8

Table 8

The content of the radioisotope Sr^{91} in extracts obtained by extraction (with solutions of nitric acid in distilled water) of soil samples taken from the territory of Shirvan region and in the residues of soil samples.

HNO_3 content in solutions, (mol)	Content of radioisotope Sr^{91},% (from the initial content (0.3 Bq = 100%) of Sr^{91} in soil samples weighing 200 g)			
	Soil sample	Soil extract with HNO_3 solutions in 1 liter of water / in soil residue	Soil extract with HNO_3 solutions in 2 liters water / in soil residue	Soil extract with HNO_3 solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	5,5 / 94,5	8 / 92	10 / 90
1,0	-	8,6 / 91,4	15 / 85	20 / 80
1,4	--	13,5 / 86,5	20 / 80	26 / 74
2,0	-	19 / 81	29 / 71	38 / 62

Table 9

The content of the radioisotope Sr^{91} in extracts obtained by extracting soil samples taken from the territory of Shirvan region with alkaline solutions of NaOH in distilled water and in the residues of soil samples.

NaOH content in solutions, (mol)	Content of radioisotope Sr^{91},% (from the initial content (0.3 Bq = 100%) of Sr^{91} in soil samples weighing 200 g)			
	Soil sample	Soil extract with NaOH solutions in 1 liters of water / in soil residue	Soil extract with NaOH solutions in 2 liters water / in soil residue	Soil extract with NaOH solutions in 3 liters water / in soil residue
-	100	-	-	-
0,5	-	6,6 / 93,4	10 / 90	13 / 87
1,0	-	16 / 84	22 / 78	25 / 75
1,4	--	21 / 79	31 / 69	36 / 64
2,0	-	29 / 71	40 / 60	50 / 50

Table 10

The content of the radioisotope Sr^{91} in extracts obtained by extraction (with solutions of HNO_3 + HCl mixtures in distilled water) soil samples taken from the territory of Shirvan region and in the remains of soil samples. Soil residues treated with 3 liter solutions of a mixture of acids were re-treated with alkaline solutions.

Acid or alkaline content in solutions, (*mol*)			Content of radioisotope Sr^{91}, % (from the initial content (0.3 Bq = 100%) of Sr^{91}in soil samples weighing 200 g)				
			Soil sample	Soil extract with HNO_3 + HCl mixtures solutions in 1 liters of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 2 liters of water / in soil residue	Soil extract with HNO_3 + HCl mixtures solutions in 3 liters of water / in soil residue	Soil residues (treated with 3-liter solutions of a mixture of acids) re-treated with alkaline solutions.
HNO_3	HCl	NaOH					
-	-	-	100	-	-	-	-
0,5	0,5	0	-	15 / 85	21 / 79	26 / 74	-
1,0	1,0	0	-	28 / 72	42 / 58	54 / 46	-
1,4	1,4	0	-	40 / 60	60 / 40	72 / 28	-
2,0	2,0	0	-	60 / 40	72 / 28	85 / 15	-
0	0	0,5	-	-	-	-	(26) +5 / 69
0	0	1,0	-	-	-	-	(54) +8 / 36
0	0	1,4	-	-	-	-	(72) +10 / 18
0	0	2,0	-	-	-	-	(84) +12 / 4

When soil samples were treated with solutions of 0.5, 1.0, 1.4, 2.0 mol of nitric acid in 1, 2, and 3 liters of distilled water, a decrease in the value of K^{40} activity in soil from 100% to 85% and an increase in the value of K^{40} activity in extracts from 0% to 15% were observed. With such treatment of soil samples with solutions of sodium hydroxide (caustic sodium), a decrease in the value of K^{40} activity in the soil from 100% to 83% and an increase in the value of K^{40} activity in extracts from 0% to 17% and with a similar treatment of soil samples with solutions of a mixture of nitric acid with hydrochloric acid, as well as further processing of the soil residue with a solution of sodium hydroxide (sodium hydroxide), a decrease in the value of K^{40} activity in the soil from 100% to 61% and an increase in the value of K^{40} activity in the extracts from 0% to 39% were observed.

When soil samples were treated with solutions of 0.5, 1.0, 1.4, 2.0 mol of nitric acid in 1, 2, and 3 liters of distilled water, a decrease in the value of Na^{22} activity in soil from 100% to 34% and an increase in the value of Na^{22} activity in extracts from 0% to 66% were observed. By such treatment of soil samples with solutions of sodium hydroxide, a decrease in the value of Na^{22} activity in soil from 100% to 16% and an increase in the value of Na^{22} activity in extracts from 0% to 84% and by similar treatment of soil samples with solutions of a mixture of nitric acid with hydrochloric acid, as well as further processing of the soil residue with sodium hydroxide solution, a decrease in the value of Na^{22} activity in soil from 100% to 8% and an increase in the value of Na^{22} activity in extracts from 0% to 92% were observed.

When soil samples were treated with solutions of 0.5, 1.0, 1.4, 2.0 mol of nitric acid in 1, 2, and 3 liters of distilled water, a decrease in the value of Sr^{91} activity in soil from 100% to 62% and an increase in the value of Sr^{91} activity in extracts from 0% to 38% were observed. By such treatment of soil samples with solutions of sodium hydroxide, a decrease in the value of Sr^{91} activity in the soil from 100% to 50% and an increase in the value of Sr^{91} activity in extracts from 0% to 50% were observed, and by similar treatment of soil samples with solutions of a mixture of nitric acid with hydrochloric acid, as well as further processing of the soil residue with a solution of sodium hydroxide, a decrease in the value of Sr^{91} activity in the soil from 100% to 4% and an increase in the value of Sr^{91} activity in the extracts from 0% to 96% were observed.

Thus, cleaning the soil from radioisotopes with a mixture of nitric acid with hydrochloric acid with further processing of the rest of the soil with sodium hydroxide solutions is the most effective method of cleaning.

Comparative analysis of the results of these experiments with the data of previous experiments using traditional adsorbents concluded that the cleaning of soil contaminated with radioisotopes is effective by sequentially treating it with solutions of weak acids and alkalis [4].

Comparative analysis of the data presented in tables 2-10 confirms about 3-7 times low degree of K^{40} emission in comparison with the degree of soil purification from other radionuclides / Na^{22}, Sr^{91} /.

The method of cleaning the soil from heavy metals and radionuclides by extraction with weak acidic and alkaline solutions is more effective than cleaning methods using adsorbents. The correct application of this method allows us to restore soil fertility.

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ОЧИСТКА ПОЧВЫ ОТ ПРИРОДНЫХ ИЗОТОПОВ

Х.Ф. Мамедов, Х.Н. Ширалиева

Резюме: Изучено распределение следовых количеств природных радионуклидов в образцах почвы. Радиоизотопы K^{40}, Na^{22}, Sr^{91} идентифицированы во всех пробах почвы, взятых с территории Ширванского района страны. Анализ, проведенный методом гамма-спектроскопии, показал, что в образцах почвы концентрации природных радиоизотопов Na^{22}, K^{40} и Sr^{91} составляют 2,7, 2,5 и 0,57 Бк/кг, соответственно. Изучены способы очистки почвы от радионуклидов. Систематическими исследованиями установлено, что при всех методах очистки степень извлечения K^{40} из образцов почвы примерно в 3-7 раз ниже, чем выделение Na^{22} и Sr^{91}. Метод очистки почвы от радионуклидов экстракцией слабокислыми и щелочными растворами более эффективен, чем другие методы очистки.

Ключевые слова: образцы почвы, природные радиоизотопы, кислые и щелочные растворы, очистка почвы.

TORPAĞIN TƏBİİ İZOTOPLARDAN TƏMİZLƏNMƏSİ

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Xülasə: Torpaq nümunələrində təbii radionuklidlərin mikrokonsentrasiyalarının paylanması öyrənilmişdir. Ölkənin Şirvan rayonu ərazisindən götürülmüş bütün torpaq nümunələrində K^{40}, Na^{22}, Sr^{91} radioizotopları müəyyən edilmişdir. Gamma-spektroskopiya üsulu ilə aparılmış analiz torpaq nümunələrində Na^{22}, K^{40} və Sr^{91} təbii radioizotoplarının konsentrasiyalarının müvafiq olaraq 2,7, 2,5 və 0,57 Bq/kq olduğunu göstərdi. Torpağın radionuklidlərdən təmizlənməsi üsulları öyrənilmişdir. Sistematik tədqiqatlar bütün təmizlənmə metodları ilə torpaq nümunələrinin K^{40} izotopundan təmizlənmə dərəcəsinin Na^{22} və Sr^{91} izotoplarından təmizlənməsindən təqribən 3-7 dəfə kiçik olmasını göstərir. Torpağın zəif turşu və qələvi məhlulları ilə ekstraksiya etməklə təmizlənməsi metodunun digər təmizlənmə metodlarından daha effektiv olması müəyyənləşdirilmişdir.

Açar sözlər: torpaq nümunələri, təbii radioizotoplar, turşu və qələvi məhlulları, torpağın təmizlənməsi.