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TECHNOGENIC IMPACT OF RADIONUCLIDES ON OIL AND GAS FACILITIES (ON THE EXAMPLE OF THE KUMKOL FIELD)

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ABSTRACT

In this paper described technogenic impact of radionuclides on oil and gas facilities. When oil products are spilled on the surface as a result of accidents in the oil and gas industry, they are further filtered to the depth of the soil, which leads to the accumulation of radionuclides in the soil. Natural radionuclides then migrate to plants and further along trophic chains. Natural radionuclides that contribute to external γ radiation enter natural sources - air, water, soil. Today, soil pollution by oil products is one of the most acute environmental problems, as it contributes to the growth of radioactivity in the environment. The components of bitumen and paraffin in the oil entering the soil lead to significant changes in the properties of the soil profile. The gamma background in the region averages 0.11 to 0.20 μSv / h. In Kyzylorda region, hazardous wastes with the possibility of radionuclides include oil wastes, including drilling mud and used ingredients. Development of the Kumkol field began more than 30 years ago (1986). If a large area of oil and gas production areas is not treated and completely cleared of waste over decades, it will have a permanent negative radiation effect on humans and the biosphere.

Keywords: oil and gas production, radionuclides, radioactivity of oil, radioactivity of oil-contaminated soils, exposure dose.

INTRODUCTION

Radiation pollution of oilfields is due to the fact that the reservoir water of many oil fields contains high levels of radionuclides. Also, sources of historical pollution include currently ownerless facilities. Oil and gas and hydrogeological wells, mines, fields (including radioactive waste), tailings and sewage, which are a real threat to environmental security. The regions of Kazakhstan contaminated with heavy metals and oil products include the lands of Kyzylorda, Atyrau and West Kazakhstan regions. Development of ore and combustible mineral deposits leads to a significant load on the environment. The main reason for such a load is the pollution of geosystems with various toxicants, including heavy metals and radiation.

In general, according to the results of annual radiation monitoring at the Kumkol fields, the radiation background does not exceed the established norm of 0.3 μSv / h, but at the sites of collection, temporary storage and processing of oil waste shows an average of 0.09-0.22 μSv / h. The current level of development of oil production technology shows that these phenomena are inevitable. In particular, the formation of residual sediments with increasing accumulation of natural radionuclides in process equipment causes radiation to personnel and poses a potential risk of environmental pollution. (Tanzharikov, *et al.*, 2020).

EXPERIMENTAL PART

Laboratory and in-house research was conducted on the basis of the accredited engineering laboratory "Methods of physical and chemical analysis" of Korkyt Ata Kyzylorda university.

Oil, soil, oil sludge from the Kumkol field were analyzed by X-ray fluorescence spectrometer "Spectroscan GF2E" to determine the man-made consequences of oil pollution of the soil.

Alkaline rubidium Rb (37) and alkaline earth metals zirconium Zr (40), copper Cu (29), manganese Mn (25), iron Fe (26), titanium Ti (22) were detected in the oil of Kumkol and South Kumkol fields.

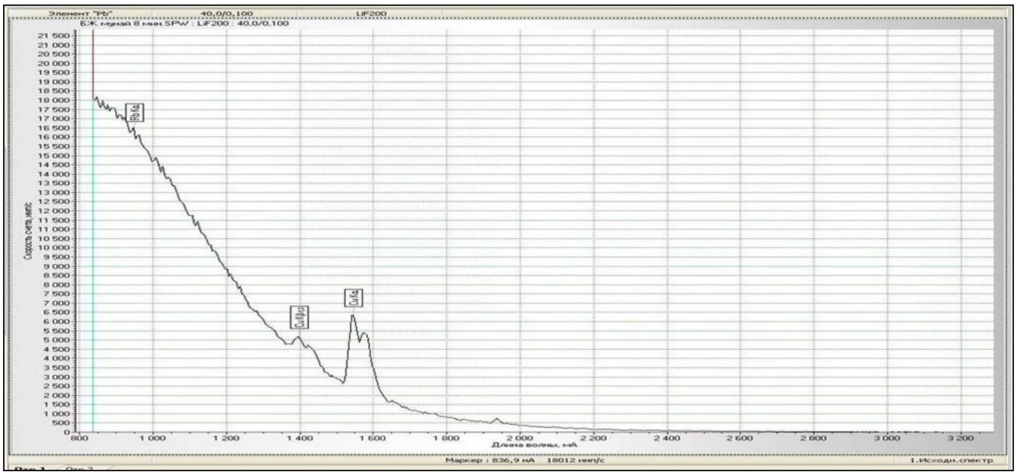

Figure-1. X-ray fluorescence spectrum of oil from the South Kumkol field.

According to the wavelength (mÅ) in the oil from the South Kumkol field (Figure-1), the following elements were found:Rb - 948; Zr - 785; Cu - 1544.

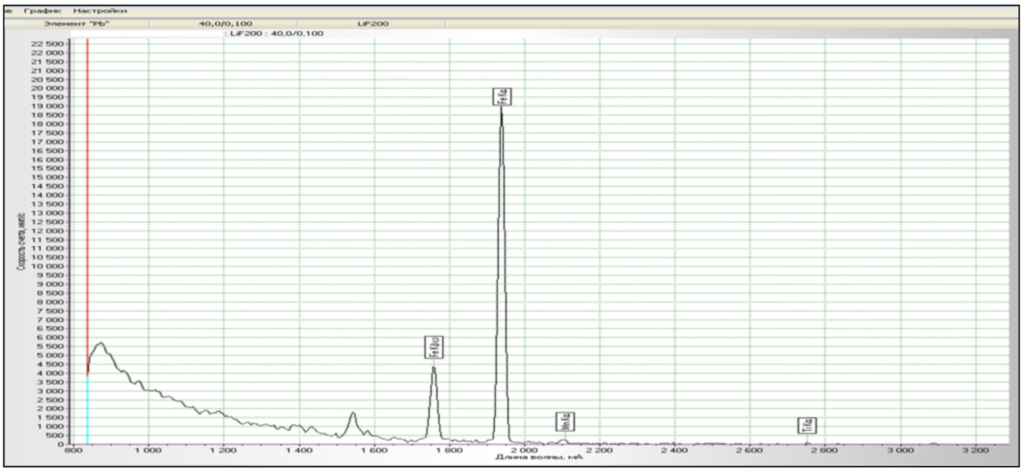

Figure-2. X-ray fluorescence spectrum analysis of oil-contaminated soils of the South Kumkol field.

According to the wavelength (mÅ) in the oil-bearing soil from the Kumkol field (Figure-2), the following elements were found: Fe - 1938; Mn - 2106; Ti - 2752.

High concentrations of radioactivity and mixtures of inorganic trace elements are constantly observed in the composition of oil deposits and oil and gas formations, the range of which is very wide: V, Ni, Ti, Cr, Co, Cu, Mo, Zn, Pb, Cd, Hg, Sb, As, TR, Au, Ag and others, as well as halogens: Br, I, Cl2, most of which are environmentally hazardous elements.

Sanitary rules were used in the study of radioactivity in refined soils with oil-containing soils.In accordance with the Sanitary Regulations SP 2.6.1.799-99 (OSPORB-99) and the concept of radioactive waste management in the Republic of Kazakhstan (Sanitary rules for radioactive waste management, 2003), all radiation sources are divided into three classes according to the exposure dose rate:

a) Sources of natural and man-made radiation, exposure dose rate (EMF) - up to 60 μR / h (0.60 μSv / h), which are considered "background".

b) Those with an exposure dose range of 60 to 100 μR / h are assessed as non-decontamination facilities and registered as 'radioactive sources'.

c) Objects with an exposure dose capacity exceeding 100 μR / h are registered as sites of radioactive man-made pollution subject to decontamination, except for the natural formations of uranium and thorium at their location (Yermukhanova, *et al*., 2020).

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Table-1. Measurements of gamma radiation sources in the obtained soil with a dosimeter-radiometer MKS-AT1117M.

order	Place of measurement	Measured dose rate (µSv / h, i / sec) 0.1m	Permissible dose rate (µSv / h, i / sec)
1	Soil on agricultural lands	0,6-0,16	40
2	Oily soil	3.3-3,6	40
3	After cleaning the oiled soil	1,1-1,4	40

The natural radiation background consists of scattered radionuclides in soil, water, air, as well as cosmic rays and short-lived radionuclides, the age of which corresponds to the age of the planet.

Figure-3. Determination of soil from non-oil contaminated agricultural land (garden) with the mobile application "Ray Detect".

Figure-3 shows that the sample garden soil is not contaminated, it is below 1 µSv / h and it forms a natural radiation fund.

Figure-4. Determination of the radiation of fuel oil with the mobile application "Ray Detect".

The results of the determination of the soil to be treated with the mobile application "Ray Detect" (Figure-4) are approximate, the measured dose rate was in the range of 3-10 µSv / h. In an area of tens of thousands of square meters of oil-contaminated soil, the equivalent dose rate is 30 to 100 µR / h when the natural background is 8-11 µR / h. The average level of radioactive contamination by gamma radiation in the areas of large and multiple spills is from 1000 to 2800 µR / h, and in areas of tens and hundreds of square meters with the highest local value is 250-600 µR / h.

Micro X-ray / hour is an effective and equivalent dose of ionizing radiation. The biological effect of X-rays (or other photon radiation, such as gamma radiation) on 100 microroentgens = 1 microsievert is considered. Figure-6 When determining the radiation of fuel oil with a mobile application "Ray Detect" at 1 µSv = 100 microroentgens, 3-10 µSv / h = 300-1000 µR / h.

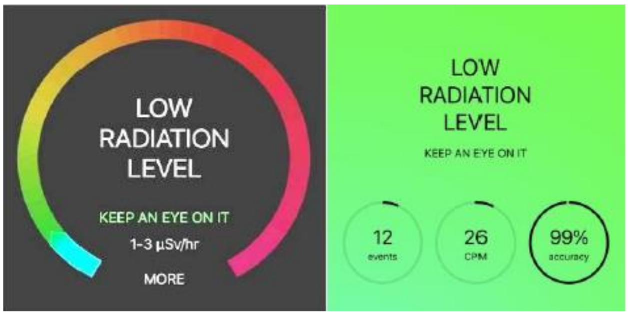

Figure-5. Detection of oily soil after cleaning with a mobile application "Ray Detect".

The results of the detection with the mobile application "Ray Detect" (Figure-5) are approximate, from 3 to 10 µSv / h before treatment, and the measured dose rate of the soil after treatment was in the range of 1 to 3 µSv / h. The unit of measurement is 3-10 µSv / h - 30-100 µS / h. 1 - 3 µSv / h - 100 - 300 µs / h or 1 to 3 µr / h in the presence of a millirogenic hour.

If the total short-term radiation dose for staff is less than 10 µSv (less than ten microsieverts), the radiation is virtually non-existent and can be ignored.

The radiation dose is "accumulated" in the body, so in areas with high levels of radiation, continuous measurements from a radiometer or individual dosimeter should be included. The amount of "accumulated dose" throughout life should not exceed 100-700 mSv (depending on the local, normal level of the background)(Bakr, 2010).

Radiation and radioecological hazards of working with mineral resources in the oil and gas complex with the formation, concentration and scattering of highly active industrial wastes in the form of solid and viscous sediments, highly toxic radium (226, 228, 224), thorium (228), radon (222, 220) isotopes are determined by the inflow of formation water and oil sludge.

The radioactivity of soils depends on the presence of radionuclides in them. The natural radioactivity of the soil is due to the natural radioactive isotopes that are constantly present in the soil and in the rocks that make up the soil. Natural radionuclides are divided into 3 groups.

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The first group of radioactive elements includes all natural isotopes of radioactive uranium and thorium: uranium (238U, 235U), thorium (232Th), radium (226Ra) and radon (222Rn, 220Rn).

The second group includes isotopes of "ordinary" elements with radioactive properties: potassium (40K), rubidium (87Rb), calcium (48Ca), zirconium (96Zr) and others.

The third group consists of radioactive isotopes of tritium (3H), beryllium (7Be, 10Be) and carbon (14C) formed in the atmosphere under the influence of cosmic rays (Valkov, *et al*., 2013).

Decomposition of uranium and thorium radionuclides is also characteristic of radionuclides in oil wastes. The source of pollution of the territories of oil and gas enterprises with natural radionuclides is their high content in the associated aquifers during oil and gas production. During the production and initial preparation of oil and gas, radium isotopes 226Ra, 228Ra and 224Ra are deposited on the inner surface of the oil and gas field equipment from the associated formation water passing through it. As a result of the activities of oil and gas companies in some regions of the country accumulated hundreds of thousands of cubic meters of industrial waste with high content of natural radionuclides (Romanovich, *et al*., 2018).

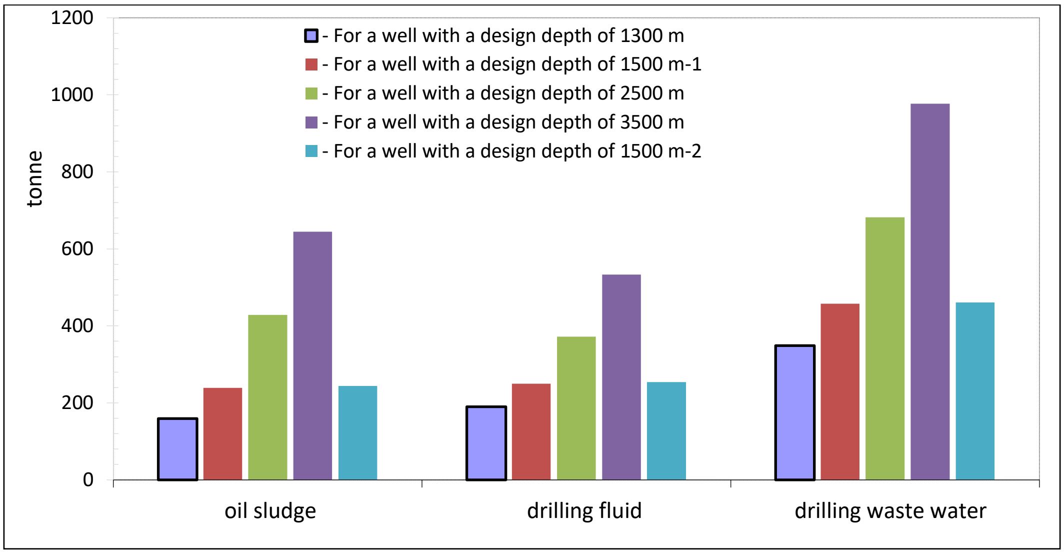

Figure-6. 2017-2019. drilling wastes (tons) during the construction of wells in the design layer PZ (PKKR JSC) with a design depth of 1300, 1500, 2500, 3500 meters.

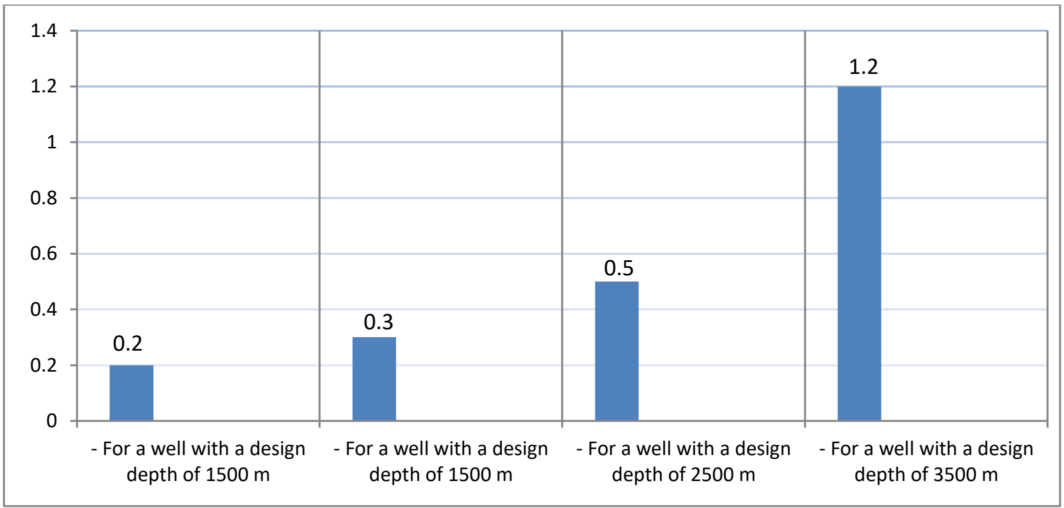

Figure-7. 2017-2019. waste during the construction of PZ design development wells (PKKR JSC) with a design depth of 1300, 1500, 2500, 3500 meters - waste oil (tons).

Figures 6-7 shows the volume of sludge, drilling and wastewater from the PZ projecting layer at a depth of 1300, 1500, 2500, 3500 meters during the construction of wells by PKKR JSC. Wastes from the development of new wells will have a high level of radioactivity. During the construction of wells, PKKR JSC increased the amount of waste from sludge, drilling, oil and formation water from the PZ project layer at a depth of 1300, 1500, 2500, 3500 meters.

As the radioactivity increases with depth, the activity of the waste also increases. Waste activity from the PZ project layer at a depth of 3,500 meters is higher than 1500 Bq / kg. In these waste areas, large areas of untreated and unfertilized areas remain radioactive contamination for decades. The radionuclide composition of industrial wastes of oil complexes also depends on their age. This is because the half-life of natural radionuclides in any environment lasts for tens or thousands of years. Therefore, even in the restored area, it is necessary to take into account when assessing the radiation situation that the specific activity of radionuclides is at a certain level, it can not disappear immediately (Yermukhanova, *et.al.*, 2020).
Artificial concentration of background radionuclides in oil production and refining, production and use of construction materials dramatically changes the background geopopulation distribution of radioactivity in the environment. The most common source of contamination in the event of an accidental oil spill (rupture of an oil pipeline, drilling a well, an accident involving oil trucks, etc.) is the earth's crust. The residue formed at this time is fuel oil. Fuel oil is collected in special landfills. Then the restoration of fuel oil as a result of biorecultivation is considered. However, there are ways to use oil-contaminated land more efficiently, such as using it as a raw material when building roads.The study of soil composition for radioactivity was carried out by gamma-spectrometric method on instruments "Progress" G-06141 and "Progress" G-9758, which were tested. Gamma spectrometric analysis is one of the most sensitive and selective methods of modern analysis. Spectrometers are completely computer controlled. The software runs on the Windows platform.

Sampling at the temporary storage of fuel oil at the Kumkol field 1 (On approval of Sanitary Rules "Sanitary and epidemiological requirements for radiation safety" Order of the Minister of Health of the Republic of Kazakhstan from June 26, 2019 № Ministry of Health of the Republic of Kazakhstan. Registered in the Ministry of Justice of the Republic of Kazakhstan on June 28, 2019. was carried out in accordance with (Sanitary and epidemiological requirements for radiation safety, 2019). Measurements were made together with a representative of the facility.

Table-2. Results of radioactive waste study from Kumkol field.

No	Model name	Sampling point, date	Effective activity of radionuclides, Bk / kg	Own effective permissible activity level (Bq / kg)
1	Oil waste soil № 1 container	Kumkol field (temporary waste storage facility) 24.08.2019	1055,0	1500
2	Oil waste soil №2 container		1062,0	
3	Oil waste soil №3 container		1520,0	
4	Oil waste soil №4 container		1290,0	
5	Oil waste soil № 5 containers		1580,0	
6	Oil waste soil № 6 containers		1566,0	
Moderately effective activity			1345,0	

The average soil radioactivity in samples taken from oil-contaminated soils (Table-2) was 1345 Bq / kg. The activity was high in № 3, 5, 6 containers filled with oil waste soil.

According to the hygienic standard "Sanitary and epidemiological requirements for radiation safety" [10], the effective permissible level of own activity for materials used in road construction outside populated areas (class III):

$$A_{ЭФФ} \leq 1{,}5 кБк / кг \quad (1)$$

that is, it should not exceed 1500 Bq / kg.

In accordance with the sanitary regulations of RKN-99 (SP 2.6.1.758-99) (Hygienic standards, 2015), the issue of using materials at 1.5 kBk / kg <Aeff 4.0 kBk / kg (class IV) in each case in consultation with the territorial bodies of the State Sanitary and Epidemiological Service of the Republic of Kazakhstan , not less than the regional level.

Determination of the dose rate of oil-contaminated soils was carried out by gamma spectrometry on the instrument "Progress" G-06141. Oil-contaminated soil samples were taken from the Kumkol field temporary storage facility.

Table-3. Measurement of radiation from oil-contaminated soil samples (Kyzylorda - Kumkol 175 km)
Date of measurement: 24.08.2019.

No	Measurement location	Measured dose rate (µSv / hour, i / sec) 0.1m			Allowable dose rate (µSv / hour, and / sec)
		At a height from the floor (ground)			At a height from the floor (ground)
		1,5м	1м	0,1м	1,5 м; 1м; 0,1м
Temporary storage site for radioactive waste (Kyzylorda - Kumkol 175)					
1	Container No. 1	0,23-0,42	0,33-0,54	0,38-0,65	40
2	Container No. 2	1,4-1,7	1,2-1,6	2,1-2,3	40
3	Container No. 3	0,55-0,76	0,63-0,78	0,86-1,8	40
4	Container No. 4	0,10-0,44	0,23-0,95	1,6-2,6	40
5	Container No. 5	1,6-1,8	1,9-2,2	2,2 -2,4	40
6	Container No. 6	1,6-1,8	2,2-2,6	3,6-3,9	40

Coordinates of the landfill at 175 km of Kyzylorda-Kumkol in the Syrdarya region: northern latitude - 45 ° 42', longitude - 65 ° 28'. Soil, oil sludge and other wastes brought to the landfill are processed after being sent for radiation testing.

Measurements are made by industrial dosimeter-radiometer MKS-AT1117M. The highest dose of radioactivity was in № 6 containers, 0.1 m above the ground, which ranged from 3.6 to 3.9 µSv / h. The equivalent dose rate is 360-390 µR / h. In general, the dose rate in container №4 reached 2.6 µSv / h at 0.1 m above the ground, ie 260 µR / h, compared to the measured dose rate in container № 1. The allowable radiation dose rate in the soil should not exceed 40 µSv / h.

In the Kumkol field radioactive oil tailings storage facility (Figure-6), the equivalent dose rate is 280-300 µR / h. In general, the dose rate in 4.5 containers reached 2.5-2.6 µSv / h, ie 260-270 µR / h at 0.1 m above the ground, compared to the measured dose rate in №1 container(Radiation safety standards, 2010).

The level of oil pollution of the soil is due to the negative impact on the plant, mainly due to changes in its physical and chemical properties (hydrophobic increase, filling the soil capillaries with oil) and direct toxic effects of toxic components in the oil (phytotoxicity).

Today, many methods are used to eliminate oil pollution in the soil, but among all the existing methods of cleaning and restoration of soils with moderate pollution, biological methods are the most effective, among which phytoremediation methods are widely studied in science (Abilbek, *et al*., 2017)

The positive effect of sowing on oil-contaminated soils is explained by the use of petroleum hydrocarbons as plant supplements and improves the gas-air regime of contaminated soils, enriching them with various active additives, which ultimately stimulates the growth of microorganisms and accelerates the decomposition of oil and oil products [14].

Many authors [15] consider the economic efficiency of cleaning oily soils with the help of local natural sorbents. Our main goal is to reduce radioactivity by extracting oil from the soil on the basis of natural sorbents. For this purpose, the use of straw with oat bran, which has hyperaccumulative properties with the transport of radionuclides with additional heavy metals with rice bran biopreparations.

Sowing was carried out on clean and contaminated soil. Soil sampling:

a) Oat seeds were planted in oil-contaminated soil in the amount of 100 pieces.

b) 100 such control samples were introduced into the uncontaminated soil. They were planted in pots with an even distribution. The test objects were kept in the same exposure to light, up to 60% of total humidity close to the actual situation, and the temperature was raised in the range of 18-25 ° C. Irrigation was carried out simultaneously and in equal amounts.

Control samples were also introduced into uncontaminated soil. The soil sample was taken from the Aryskum field (code-524sk2 C).

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Table-4. Summary table of experimental samples.

Soil type	No experiment	Contaminated	Type of irrigation
medium saline medium loamy brown soil	1	Clean	with running water
	2	Oil	with running water
	3	Oil	Oatmeal with oat bran and rice straw water

RESULTS AND DISCUSSIONS

According to the results of the practical application of phytomediation in the first phase of the study of biological treatment methods in contaminated soils for 30-45 days there is a significant decrease in the concentration of petroleum hydrocarbons to the expected concentration (Abilbek, *et al*., 2015).

As the purpose of the experiment was to reduce to a possible level the content of petroleum products, radionuclides, heavy metals in oil-contaminated soils, alternative chelating agents were used in the second stage. As an alternative to synthetic chelating agents, the use of rapidly decomposing organic acids, such as citric, sorrel, wine, vinegar, as well as nitrile triple acetic and ethylenediamine acids (EDDA) (Koptsik, 2014), which are products of plant metabolism and are widely used in natural conditions, was considered. In case of severe soil contamination with petroleum products, further cleaning of the soil was carried out as a result of the use of additional methods. As a result (Figure-8b), the color and odor of the soil changed, the oil was removed and, accordingly, its color became lighter.

«Weighing method for determining the composition of petroleum products in the soil is carried out in accordance with » ([2] RD 39-0147098-015-90 (Instruction on control over the state of the soil at the facilities of the enterprises of Minnefteprom) "Weight method for determining the content of petroleum products in the soil») instructions. By the difference in mass with and without content, the mass of the detected oil products is determined. The content of oil products (X mg / kg of soil) is calculated by the formula:

$$X = \frac{A}{B}100 \quad (2)$$

where A is the found amount of oil products in milligrams;

B - sample of soil taken for analysis in grams.

1. Oily soil:

m(butum) = 30g; m(no content) = 85,780; m (with content, o-n) = 87,887; (o -oil)

m= 87,887 - 85,780 =2,107; A =2,107 g.

$$X_1 = \frac{2{,}107}{30}100 = 7{,}02\%$$

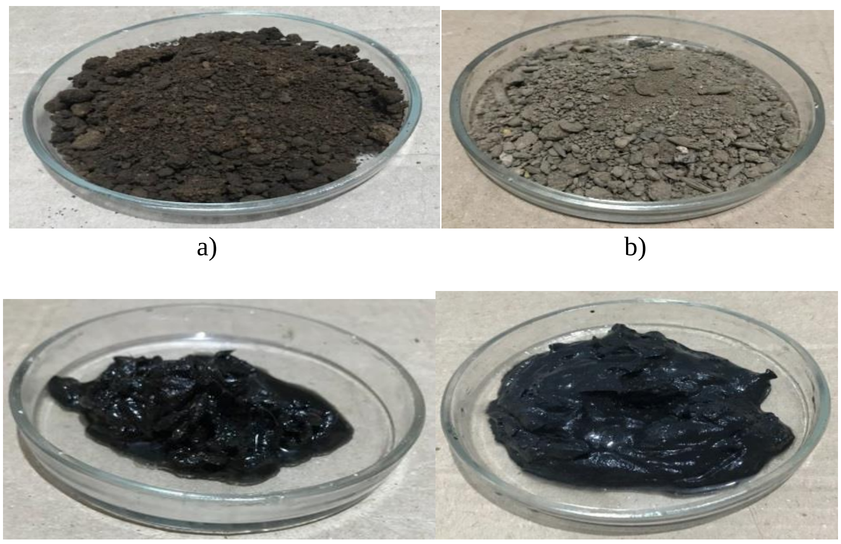
2. Percentage of oil in relatively refined soils:

m(butum) = 30g; m(no content) = 83, 522; m(with content, o-n) = 83, 691

m = 83,691 - 83,522 = 0,169; A = 0,169g

$$X_2 = \frac{0{,}169}{30}100 = 0{,}56\%$$

As a result of laboratory analysis, the amount of oil in the refined soil decreased to 0.56%.



a) b)

c) d)

Figure-8. Oiled soil and its by-products (a) oil-contaminated soil; (b) cleared soil; (c) waste oil; (d) oil-containing sludge.

Figure-8 shows images of fuel oil and products obtained after its cleaning. If the oil-contaminated soil is oily brown before cleaning (8a), it will change color after cleaning (8b) and the oil content in the soil will decrease to 0.56%. Approximately 50 grams of oil and 80 grams of oil-bearing sludge (8d) were extracted from 500 grams of soil. This often sent oil sludge and sludge for burial. However, recent research in science suggests that petroleum products can be used to improve the efficient use of oil and to create new materials that are effective in construction (Bisenov, *et al*., 2016). The use of oil sludge as a secondary raw material improves the environmental situation in the areas of oil refining and leads to the most rational use of natural resources. Modern research and development of compositions and technologies for the production of waterproofing materials are aimed at replacing expensive and scarce commodity products with oil waste.

During the drilling and operation of oil and gas wells and as a result of emergency spills, oil products and formation water are filtered through walls and layers, contaminating the land and aquifers. Therefore, it is necessary to consider special filter screens for temporary storage of oil wastes, which have the appropriate feasibility study of environmental resistance and their longevity and corrosion resistance of waste (Tanzharikov, *et al*., 2018).

A technology is proposed for creating a new technology for waterproofing the bottom of production wells using new plugging materials based on sodium silicate and microcement, which allows to significantly reduce the water cut of the produced oil (Abdeli, *et al.*, 2018).

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