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LONG-TERM MONITORING OF OIL CONTAMINATION OF PROFILE-DIFFERENTIATED SOILS ON THE SITE OF INFLUENCE OF OIL-AND-GAS WELLS IN THE CENTRAL PART OF THE BORYSLAV-POKUTTYA OIL-AND-GAS BEARING AREA

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Abstract

The dynamics of oil contamination of profile-differentiated soils of the Precarpathian region in the area of influence of deep wells of the Vilhivska oil exploration area of the Boryslav-Pokuttya oil-and-gas bearing area is investigated. The research was conducted during 2007-2016. Soils were tested in the intervals of the distribution of humus-eluviated (H (e) gl), eluvial-humous (Eh (gl)), iluvial (Ih (gl)) horizons, iluviated parent rock (Pigl), and unchanged parent rock (Quaternary sediments) to the depth up to 2.0 m. The concentrations of oil products in pollution sources are analyzed. The zoning of the research territory according to the distribution of elementary geochemical landscapes is carried out. Within the various elementary geochemical landscapes, regularities in the horizontal and radial distribution of oil products have been revealed. The highest pollutants concentration is established in the silts of elementary transsuperaqual landscapes in the number up to 1750 mg/kg. In deeper horizons, the concentration of pollutants decreases nonlinearly, and the dynamics of this process is different in various elementary geochemical landscapes. The dynamics of the change in the oil concentration over time during the 9-year period of field observations were established. In particular, 1.5 years after the drilling operations cessation, the average concentration of oil products in the interval of the humus-eluviated horizon decreased 2.7 times and more than 4 times after 2.5 years. In the interval of the eluvial-humus horizon, the dynamics of the pollutant concentration change was less intensive, as the processes of desalination and decomposition of oil products were accompanied by the introduction of new pollutant amounts from the upper horizons.

Obtained results are unique considering the natural conditions and long duration of the study (9 years). It can be useful in predicting changes in the concentration of oil products in soils, in designing environmental protection measures and civil protection measures. Highlighted in the article, the results of research support and develop the theory of geochemistry of technogenesis, the theory of geochemical landscapes, creating a reliable experimental basis for improving the scientific methods of ecological and crisis monitoring.

Keywords: *emergency situation; environmental safety; crisis monitoring; ecological monitoring; elementary geochemical landscape; geochemistry of technogenesis.*

1. Introduction

The development of the oil and gas industry is impossible without the discovery of new deposits. Economically attractive is the discovery of new deposits in oil-and-gas producing regions with developed infrastructure, a competition of highly professional personnel, well-established logistics of extraction, transportation, and processing of raw materials. Such long-standing and well- equipped oil-and-gas producing areas exist in a large number of countries in the world: the Los Angeles, Ventura, Uinta, Utah, North Albert, Williston Basin (USA), the Caspian oil-and-gas province (Kazakhstan, Russia), the oil-and-gas provinces of the northern Sahara (Algeria), West Africa, Potwar (Pakistan), Absheron petroleum- bearing region (Azerbaijan), Oiloibiri oil field (Nigeria), Douala (Cameroon), the Rhine basin (Germany), the Western Canadian basin (Canada), Pechelbronn field in Alsace (France) [1-5]. The territories with long oil production include the Carpathian oil-and-gas bearing province, where oil is extracted since the 16th century [5].

The main oil area in the Carpathian oil-and-gas bearing province is the Boryslav-Pokuttya oil-and-gas bearing area, which has thousands of drilled oil wells. During the long-term oil production, the environment has undergone an intense man-made impact, which is manifested primarily in contaminated soil, surface, and groundwater. The contamination of the geological environment components with organic substances, primarily oil products, phenols, hydrocarbon gases [6-7] is one of the most dangerous. Such an intensive technogenic impact can lead to an emergency situation.

The purpose of the given article is to characterize the hazard factors for soils caused by oil wells drilling and, based on the results of the field and laboratory studies, to establish spatially and time patterns of oil products distribution in profile-differentiated soils using an example of the Vilhivska area in the central part of the Boryslav-Pokuttya oil-and-gas bearing area of the Carpathian province (Ukraine).

The object of the research is the processes of transformation of hydrocarbon contamination of soils of a podzolic type as a part of the geological environment at the site of deep wells influence in the central part of the Boryslav-Pokuttya oil-and-gas bearing area of the Carpathian province.

2. Experimental

Ecological and geochemical studies on assessing the effect of wells in the central part of the Boryslav-Pokuttya oil-and-gas bearing area of the geological environment were conducted at the site of the search well Vilhivska-34.

2.1. Field studies

Five main and three additional stages of field observations were conducted to trace the time of ecological-geochemical changes, intensity and level of soils and silts contamination within the research area. Fieldwork was carried out in May and October. The stages of observations were tied to the particular hydro-meteorological stages, which marked the end of the peak seasonal receipts to the surface of the meteorological waters. The main stages of field research were conducted during 2007-2009. Additional stages were carried out in May and October 2010-2011 and in May 2012, 2014, 2016.

The main stages of the fieldwork at the site of the influence of the Vilhivska-34 oil exploration well were provided for testing the soils and rocks of the aeration zone with a uniform square grid of geochemical wells with a density of 100x100 m, which, depending on the situation, branched out into the fan-shaped profiles from the oil-and-gas well construction sites down the terrain. Thus, the initial detail of the eco-geochemical survey was at least 1: 1000 (1 geochemical well per 1 ha of an area). To determine the thickness, lithological, and mechanical composition of soils in the rocks of the aeration zone, as well as the intensity of their contamination, geochemical wells with a depth of up to two meters, were drilled in the section of the mechanized-manual sections, with a soil sampling interval. Samples of soils were selected from intervals of 0.1-0.2 m, 0.3-0.5 m, 0.8-1.0 m, 1.3-1.5 m, and 1.8-2.0 m. In addition to the samples from geochemical wells, ravine and mountainous catching ditches silt were analyzed. Additional stages of fieldwork included testing of soils and silts at the selected representative points in the intervals of 0.1-0.2 m and 0.3-0.5 m. The test points were clamped on the ground with reference marks for ecological and geochemical monitoring. The binding was carried out using a compass and GPS.

Soil and water samples were collected and preserved according to the requirements of the state standard of Ukraine DSTU 4287: 2004. The soil samples meant for adsorbed gases testing were sampled in the glass jars (0.5 l) and hermetically preserved. The period between sampling and determination of organic compounds did not exceed 7 days.

2.2. Analytical studies

The concentration of oil products was mainly determined by infrared spectroscopy by measuring the absorption in the region of 2700-3100 m^{-1}. Preparation of the sample included the extraction of oil products with carbon tetrachloride and chromatographic purification from polar compounds in a column with active alumina. The calculation of oil products was carried out by IHS-29. The device was calibrated with a mixture of decane (56% vol.), iso-octane (19% vol.), and benzene (25% vol.). The lower limit of the determination was 0.1 mg/L or 0.1 mg/kg with relative error 15%.

2.2.1. An object of research

In the administrative aspect, the research area is located in the Ivano-Frankivsk region, in Ukraine.

From the point of view of orography, the area is characterized by low-mountain relief with the well-developed hydrographic network. The absolute elevation marks range from 450 m in valleys to 600-800 m on the watersheds. The climate of the region is moderately continental. The average annual rainfall is 700-800 mm.

According to the landscape-geochemical zoning, in the territory of Precarpathians from the scarp of the Outer Carpathians to the river Dniester, the landscapes of the acid-calcium class with forest-lush vegetation on the profile-differentiated gleyed soils developed on the deluvium of sandy-argillaceous formations are widespread. Directly the research site is covered with profile-differentiated gleying argillo-arenaceous soils on the old eluvial sediments. The territory is intensively dissected by ravines.

According to the classification of elementary geochemical landscapes [8], within the research area, we have distinguished an elementary autonomous eluvial landscape, an elementary transeluvial landscape, and an elementary transsuperaqual landscape.

2.2.2. Geological structure and oil-and-gas content

Geologically, the research area is located in the central part of the Boryslav-Pokuttya zone of the Precarpathian mountain trench. This territory is located between one of the largest hydrocarbon deposits of the Carpathian region, namely Dolynskyi and Bytkiv-Babchynskyi ones. The geological structure of the region includes deposits of Upper Cretaceous, Paleocene, Eocene, Oligocene, Miocene, and Pliocene. The entire stratum is represented by a flechesite complex of rocks that differ in cement (limestone, siliceous), the granulometric composition of rocks, the distribution of micaceous minerals, and the thickness of measures. In addition, the menilitic suite of the Oligocene is characterized by high bitumen content. The rocks of the upper Eocene sediments and the Oligocene menilite suite of the Paleogene system are oil-and-gas bearing ones.

Dozens of wells (1–, 2–, 3–, 5–, 7–, 8–,11–, 15–, 16–, 20–, 21–, 24 – (Vilhiv); 2–, 6– (Pidlisiv); 6–, 9–, 12–, 16–, 17–,18–, 19–, 21– (Strutyn) were drilled on the Vilhivska search area near Strutynske mine field. At least 5 new deep wells were planned to be drilled. Drilling of the wells in the research area was carried out using barns and burial of drilling waste on site, which created additional risks of contamination of the geological environment and an emergency.

2.2.3. Characteristics of soils

According to the soil-geographical zoning, the research area refers to the subboreal soil-bioclimatic belt of the Western European subboreal forest region of the Carpathian mountain-larval province of the brownish-podzolic zone of gleyed soils of the Carpathian region [9].

According to the soil maps compiled according to the classification of soils of the UN Food and Agriculture Organization, profile-differentiated gleyed soils (Stagnic Cutanic Albeluvisols) dominate in the territory of the Carpathian Mountains. S.M. Polchyna [9] determines this soil type as brownish-podzolic gleyed soils. Distribution of profile-differentiated gleyed soils of Prykarpattya is controlled by the main tectonic elements of the Carpathian folded structure. These soils are also widely distributed in Western and Central Europe [10].

The profile-differentiated gleyed soils in the research area are described according to the work of [9]. The following soil horizons are distinguished: 0-5 cm – sod (Ho), or arable layer; 5-17 cm – humus-eluviated horizon, clayly (H (e) gl) 17-31 cm – humus-eluvial, clayly (HEgl) 31-48 cm – eluvial-humus, clayly (Eh (gl)); 49-110 cm – illuvial (Ih (gl)); 110-155 cm – illuviated parental rock (Pigl).

The boundaries of soil horizons are established as a result of three soil sections documenting. The highest variability is found in the thickness of the illuvial horizon (Ih (gl)) with sole ranged between 98 and 118 cm, and the illuviated paternal rock (Pigl) with lower distribution boundary fixed at a depth of between 147 and 169 cm.

Thus, the soil sampling intervals correspond to the following soil horizons: 0.1-0.2 m – humus-eluviated (H (e) gl); 0.3-0.5 m – eluvial-humous (Eh (gl)); 0,8-1,0 m – illuvial (Ih (gl)); 1,3-1,5 m – illuviated parental rock (Pigl); 1,8-2,0 m – unchanged parental rock (Quaternary sediments).

Among the features of profile-differentiated gleyed soils that can affect the migration of oil products, the fillowing should be noted: a relatively high content of physical clay; the increased compactness and hygroscopicity of the soil with depth, which cause favorable conditions for water stagnation in the upper soil; low content of organic carbon (up to 2-4% in the range of 5-10 cm, the amount of which does not exceed 1% already at a depth of 40 cm) [9]; wide development of gleying processes; the presence of neoplasms in the form of concretions, peas, ortsands [11].

3. Results and discussion

3.1. Sources of oil products entering the soil

Pollution of the environment of oil-and-gas bearing areas is caused by natural and man-induced processes. Natural ones, for example, include the flow of hydrocarbon fluids to the surface out of oil-and-gas deposits, caused by their vertical unloading with tectonically-weakened channels out of the deep zones of excess pressure. However, the natural component of pollution is negligible, and man-induced pollution is predominant. The man-induced pollution of the environment is already observed at the early stages of exploration for oil and gas, in particular, at the stage of the drilling rig installation. This is due to the imperfection and violation of environmental protection requirements, as well as the chemical composition of drilling mud flush components and drilling waste enrichment by harmful compounds in the middle of the opening of the reservoir during the oil-and-gas wells constructing.

The rocks of the menilite suite, oil, and formation water are primary sources of natural oil pollution of the environment components of the Carpathian region in the process of deep wells constructing. The man-made sources include separate components of drilling mud, combustive-lubricating materials, and drilling waste.

The Boryslav-Pokuttya area of the Carpathian oil-and-gas province is characterized by difficult drilling conditions, namely: absorption of drilling mud in the deposits of the Yamnenska suite; the rocks shedding in the menilite deposits; frequent oil-gas and water showings in different intervals of drilling; desalination in the sediments of the Stebnytska and Vorotyshchenska suites. Difficult geological conditions require the use of highly inhibited (such as potassium) low-filtration drilling fluids, often on an oil basis, which harmfully affects the environment.

Within the Vilhivskyi polygon, two mud flush formulations were used in accordance with the project. In the interval 0 – 1120 m, potassium mineralized mud flush with a density of 1.24 g/cm^3 was used. The solution was treated with the carboxymethylcellulose (CMC), condensed sulfide alcohol spirits (CBSS), potassium chloride and oil with sulfonol. In the interval 1120 –

1860 m, a polymer-potassium mud flush with a density of 1.12 g/cm^3 was used. The mud flush was treated with CMC, CCB, potassium chloride, oil, and sulfonol.

According to the results of our research, the mud flush and drilling wastewater are the dangerous sources of contaminants at the Vilhiv ecological and geochemical range. In particular, mud flush with mineralization of 7.4 g/dm^3 contained oil products (11.85 mg/dm^3); the indicator of chemical oxygen consumption reached 66.85 mg O_2/dm^3. The drilling wastewater with mineralization of 0.56 g/dm^3 contained 106.5 mg/dm^3 of liquid hydrocarbons. At the site of the operating wells of the Strutynskyi deposit, liquid waste contains 41.8 g/dm^3 of oil products.

All these sources of pollutants create significant risks of environmental pollution outside drilling sites.

3.2. Distribution of oil products in soils

According to the results of the first stage of the monitoring observations (October 2007) in the interval of 0.1-0.2 m (humus-eluviated horizon), oil products were found in concentrations 0 730 mg/kg, in silts – up to 1750 mg/kg. The morphology of the anomalies reflects the features of the relief. In particular, the most contrasting anomalies are established in the ravines silts and in the parts of the lowered parts of the relief (Fig. 1).

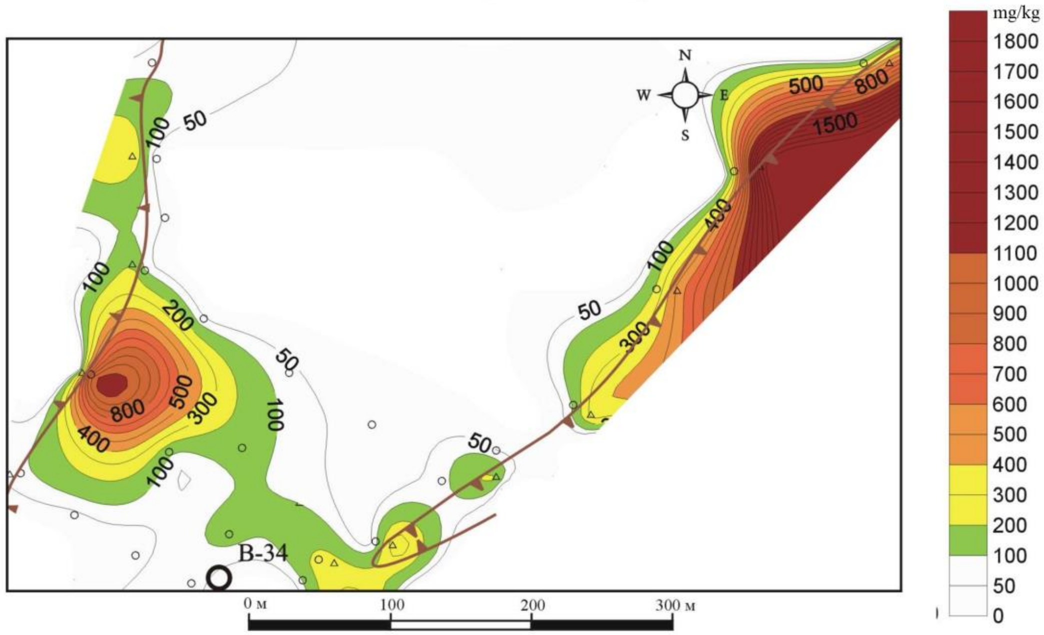

Fig. 1. Schemes of oil products distribution in the humus-eluviated horizon of profile-differentiated soil in the area of influence of Vilhivska-34 down hole (as of October 2007, the beginning of drilling a well)

In May 2008, the drilling operations were at the final stage. The average concentrations of oil products increased in the humus-eluviated horizon and decreased in the eluvial-humous horizon and unchanged paternal rock. Subsequently, the concentration of OP along the soil profile was nonlinearly decreased in total. The most intensive decrease in the content of OP was recorded in the interval of 0.1-0.2 m from October 2007 till October 2009. At the same time, in the intervals of 1.3-1.5 and 1.8-2.0 m, the content of OP increased (Fig. 2).

During 2010-2016, with a variable dynamics, there was a further decrease in the intensity of soils and silts pollution with oil products. It is worth emphasizing that the increase of the concentration of oil products in the spring of 2013 and 2016 compared with the previous periods of research. Such geochemical field bifurcations are likely associated with the additional entering the hydrocarbon substances out of neighboring oil and gas exploration areas with meltwater (Fig. 3). The graph of the change in the concentration of OP in the interval 0.1-0.2 m is well approximated by the exponential and in the interval 0.3-0.5 m – by the linear dependence. The reliability of the dependencies was confirmed by the determination coefficient within the confident interval.

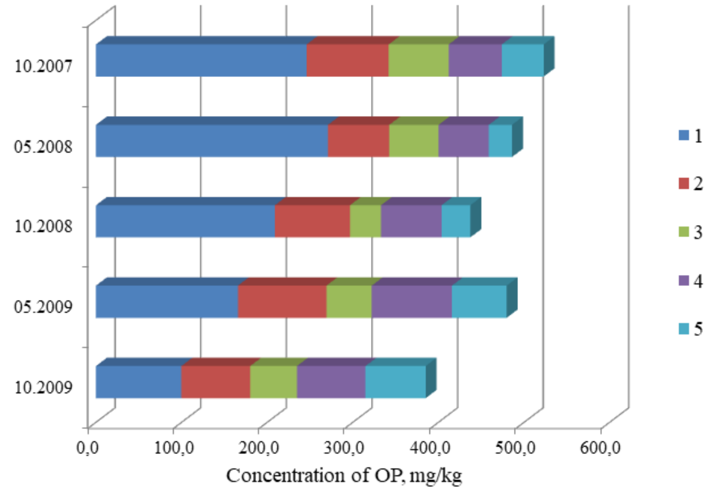

Fig. 2. Dynamics of changes in the average concentration of oil products in soils with depth and time. Sampling intervals, m: 1 – 0.1-0.2, 2 – 0.3-0.5, 3 – 0.8-1.0, 4 – 1.3-1.5, 5 – 1.8-2.0.

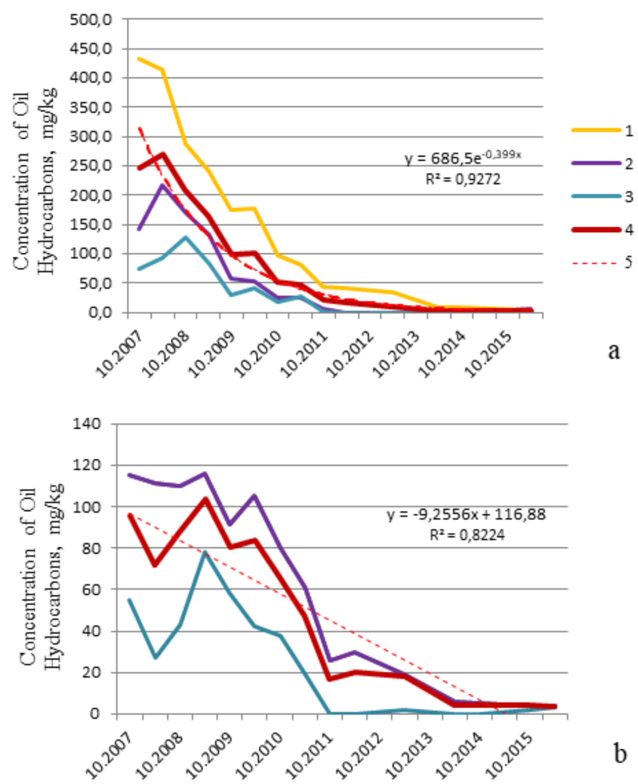

Fig. 3. The dynamics of the change in the concentration of oil products in various parts of the research site in the interval of 0.1-0.2 m (a) and in the interval of 0.3-0.5 m (b): 1 – the silts of the elementary transsuperaqual landscape, 2 – the soils of the elementary downslope (transeluvial) landscape, 3 – the soils of elementary autonomous eluvial landscape, 4 – the soils and silts of the entire research area, 5 – trend line for the graph of the change in the average content of the OP

It should be noted that the decrease in the oil products content occurred most intensively in the May-October period, while in the spring, on the contrary, we recorded a decrease in the dynamics of self-cleaning of the soil or a slight increase in the concentration of the pollutant. After the well drilling was stopped during 2008-2012 in the interval of 0.1-0.2 m, the average number of oil products in the spring period was 87% compared to the previous autumn period. At the same time, in autumn, the oil products content averaged 52% compared with the oil products content in the previous spring period. In the interval of 0.3-0.5 m, in spring, the oil products content averaged 103% compared to the concentration in the same horizon in the previous autumn period. At the same time, in autumn, the number of oil products was only 64% compared to the previous spring period. The increase in the intensity of degradation of oil products in soils in summer can be explained by the influence of temperature and better access to oxygen in comparison with the winter period. In spring, during the melting of snows, the average concentration of contaminants in the soil may increase due to their leaching and subsequent migration from localized sources within the drilling site.

Based on these results, in the case of limited funding or other obstacles to the implementation of monitoring studies twice a year, it is essential to conduct monitoring studies within the technogenic impact of oil-and-gas wells in the spring. So we did after 2012 when the concentration of oil products approached the background data.

In the subsurface of elementary geochemical landscapes, the concentration of oil products drastically decreased in the transeluvial one in the period from 1.0 to 1.5 years after the cessation of drilling operations. A slow decrease in the content of oil products within the humus-eluviated horizon is due to the location within its boundaries of the former drilling site and temporary barn of sewage, that is, sources of hydrocarbon contamination. In the eluvial-humus horizon, the decrease in the concentration of the pollutant was less intense, as the processes of oil products desalination and decomposition were accompanied by the entering of new amounts of pollutants out of the upper horizons (Table). The obtained results of changes in the concentration of oil products over time can be useful for assessing the natural degradation of oil products in landscape conditions. Similar researches at this stage are not enough for a justified assessment of the natural degradation of oil products in natural landscapes, as most of the studies are devoted to the degradation of these pollutants in the laboratory. At the same time, the obtained data should be used taking into the account the possible inflow of pollutants into the landscape from the localized sources within the territory of the former drilling site and temporary drilling barn that was not fully remediated.

Table. Characteristics of the change in the content of oil products after the cessation of drilling operations within the Vilhivska oil searching area

Time after the cessation of drilling operations, years	Change in concentration of OP,%				
	Transsuperaqual elementary landscapes silts	Transeluvial elementary landscapes soils		Autonomous eluvial elementary landscapes soils	
	0.1-0.2	0.1-0.2	0.3-0.5	0.1-0.2	0.3-0.5
0.5	-31	-21	-1	+27	+60
1.0	-42	-39	+5	-10	+191
1.5	-58	-73	-18	-67	+116
2.0	-57	-76	-5	-55	+57
2.5	-76	-88	-28	-80	+42
3.0	-81	-88	-45	-70	-29
3.5	-89	-97	-77	-97	-100
4.0	-90	-100	-73	-100	-100
6.0	-98	-100	-95	-100	-100
8.0	-99	-97	-96	-100	-89

The data obtained within Vilhivska research area were compared with the data obtained within the Pivdennostynavska research area, where the average content of oil products in groundwater decreased from 1.29 mg/dm^3 to 0.14 mg/dm^3 2 years after the cessation of drilling operations, which is 10.9% of the initial content [12].

The interesting results were obtained in Argentina. In the laboratory conditions, the soil from Patagonia that was dried, sifted, and artificially contaminated with crude oil lost 26.4% of oil products for 1 month. After that, the process of hydrocarbon degradation slowed down significantly. The experiment lasted 13 months, and during this time, the content of the oil products decreased by 45.5% (from 49.2 to 26.8 mg/g) [13]. Malaysian soils in natural conditions, contaminated with oil products in the amount of 550 mg/g, reduced the content of pollutant on average by 86% after 70 days. The same oil products under laboratory conditions during the same time degraded by 65% [14]. These results indicate that the degradation of oil products in laboratory conditions is slower compared to the field conditions; therefore, extrapolating the results of the studies from the laboratory to natural conditions is inapplicable. Also, one should be very cautious in case of transferring the results of the degradation of pollutants in different geological, soil, climatic, and other natural conditions.

The obtained results are unique considering the natural conditions, long duration of the study (9 years) and can be useful in predicting changes in the concentration of oil products in soils and designing environmental protection measures and civil protection measures. The results presented in the article develop the theory of geochemistry of technogenesis, the theory of geochemical landscapes, create a reliable experimental basis for improving the scientific methods of ecological and crisis monitoring.

4. Conclusions

Due to the drilling of deep oil-and-gas wells, soils in the central part of the Boryslav-Pokuttya oil-and-gas bearing area are subjected to intense hydrocarbon contamination, increases the level of environmental hazards and creates potential threats of emergency situations.

After the completion of the man-made impact, soil pollution significantly decreases during the first 1-3 years. Reducing the contamination of the substrate with oil products depends on the depth. In the near-surface layers, it occurs faster: in soils, it reaches background data after 4-6 years; at great depths, near the occurrence of bedrock – after 7-9 years or more. This is due to the oil products infiltration from the land surface.

In summer, the intensity of oil products degradation increases due to increased temperature and better access to oxygen compared to the winter period.

In spring, during the snow melting, the average concentration of contaminants in the soil may increase due to their leaching and subsequent migration from pollution sources.

When the concentration of oil products approaches the background, monitoring studies within the technogenic impact of oil-and-gas wells are economically justified once a year in the spring.

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