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STUDY OF REGULARITIES AND CONSTRUCTION OF MATHEMATICAL MODELS OF HYDROCARBON DISTRIBUTION IN A SECTION ON TERRITORIES OF OIL TREATMENT ENTERPRISES

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ИЗУЧЕНИЕ ЗАКОНОМЕРНОСТЕЙ И ПОСТРОЕНИЕ МАТЕМАТИЧЕСКИХ МОДЕЛЕЙ РАСПРЕДЕЛЕНИЯ УГЛЕВОДОРОДОВ ПО РАЗРЕЗУ НА ТЕРРИТОРИЯХ НЕФТЕПЕРЕРАБАТЫВАЮЩИХ ПРЕДПРИЯТИЙ

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Key words:
oil contaminated territories, hydrocarbons, penetration depth, modeling, mathematical models, ground, mathematical statistics, geostatistics, artificial pollution, geology, geoecology.

Nowadays, active human economic activity leads to significant man-made pollution. In a number of cases geological environment is polluted with hydrocarbons by an accident. That leads to change in physical and mechanical properties of grounds and has negative impact on entire an ecosystem. This work is devoted to identification of regularities and construction of mathematical models for distribution of hydrocarbons along a section on territories of oil refineries, which allow predicting the level of pollution in case of accidental hydrocarbon spills. Obtained information allows estimating changes and predicting bearing capacity of grounds during accidental oil spills. Study of hydrocarbon distribution in ground massif is based on identification of natural and artificial regularities, which are described by math body. This gives a methodological approach to study these regularities depending on geological conditions of territories that are subject to risk of accidental oil spills.

Three models of distribution of hydrocarbons along a section are revealed. They are controlled by geological conditions (lithology and depth of occurrence of an aquifer). Based on revealed regularities, mathematical models are developed. They allow to predict degree of ground contamination with hydrocarbons in terms of geological indexes (thickness of loam and crushed rock and depth of occurrence of sandstones). As a result of statistical processing, influence of geological structure on depths of hydrocarbon penetration and content of hydrocarbons is established.

It is confirmed that distribution of hydrocarbons is significantly influenced by type of rocks and their sorption ability for hydrocarbons, penetration properties (porosity and permeability) and water saturation. In case near-surface zone of the earth is composed of sands that have low sorption ability for hydrocarbons, high open porosity and high penetration properties, a hydrocarbon contamination zone will be small in comparison with zone composed of clays or loam.

Ключевые слова:
нефтезагрязненные территории, углеводороды, глубина проникновения, моделирование, математические модели, грунт, математическая статистика, геостатистика, техногенное загрязнение, геология, геоэкология.

В настоящее время активная хозяйственная деятельность человека приводит к значительному техногенному загрязнению. В ряде случаев в результате аварийных происшествий происходит загрязнение геологической среды углеводородами, что приводит к изменению физико-механических свойств грунтов и отрицательной сказывается на экосистеме в целом. Данная работа посвящена выявлению закономерностей и построению математических моделей распределения углеводородов по разрезу на территориях нефтеперерабатывающих предприятий, позволяющих прогнозировать глубину загрязнения в случае аварийных разливов углеводородов. Полученная информация позволит оценить изменения и спрогнозировать несущую способность грунтов при аварийных разливах нефтепродуктов. Изучение особенностей распределения углеводородов в грунтовом массиве основывается на выявлении природно-техногенных закономерностей, которые описываются с помощью математического аппарата. Тем самым дается методологический подход к изучению этих закономерностей в зависимости от геологических условий территорий, подверженных риску аварийного разлива нефтепродуктов.

Выявлены три модели распределения углеводородов по разрезу, которые контролируются геологическими условиями (литологией и глубиной залегания водоносного горизонта). На основании выявленных закономерностей разработаны математические модели, позволяющие по геологическим показателям (мощности суглинка и щебенистого грунта, а также по глубине залегания песчаников) прогнозировать степень загрязнения грунтов углеводородами. В результате статистической обработки установлено влияние геологического строения на глубины проникновения и содержание углеводородов.

Было подтверждено, что на распределения углеводородов существенное влияние оказывает тип пород и прежде всего их сорбционная способность к углеводородам, фильтрационные свойства (пористость и проницаемость) и водонасыщенность. В случае, если приповерхностная зона земли сложена песками, которые имеют низкую сорбционную способность к углеводородам, высокую открытую пористость и высокие фильтрационные свойства, зона загрязнения углеводородами будет минимальна по сравнению с тем, если бы она была сложена из глин или суглинков.

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Introduction

Nowadays, active human economic activity leads to significant man-made pollution. In a number of cases geological environment is polluted with hydrocarbons by an accident. That leads to change in physical and mechanical properties of grounds and has negative impact on an entire ecosystem [1-3].

Studies conducted by I. Mazur [4] show that oil losses caused by emergency spills is about 3% of its annual production. It is clear from the above mentioned that knowledge of the depth of penetration and regularities in distribution of spilled hydrocarbons in ground is important and relevant.

According to the analysis of works of foreign authors there is quite a lot of publications are devoted to penetration of hydrocarbons into grounds.

Over the past 50 years the number of publications on ground hydrocarbon contamination has exceeded 10,000 (according to the Scopus reference database). And the number of articles increases every year. In 2014 987 journal articles from the Scopus database were devoted to this subject.

After specification on the query to search within the problem of interest (distribution of hydrocarbons in ground in case of oil spills), the Scopus abstract database finds about 150 papers. It is notable, that interest to this problem is maintained at the same level every year. There are 12 papers in 2012 and 16 papers in 2016 that were devoted to this topic. Statistics show that the problem of hydrocarbon distribution in ground is now very relevant.

Most of the work devoted to the subject of the study was published in the United States, followed by China, Canada, Nigeria, Great Britain (Fig. 1, *a*). Russia occupies the 23rd place in this list. That is caused primarily by the fact that Russian scientists are published quite a bit in journals from the list of Scopus, but not by the lack of developments on this issue.

Most of the work (> 80 %) was published in journals on sciences of the earth and natural environment (Fig. 1, *b*).

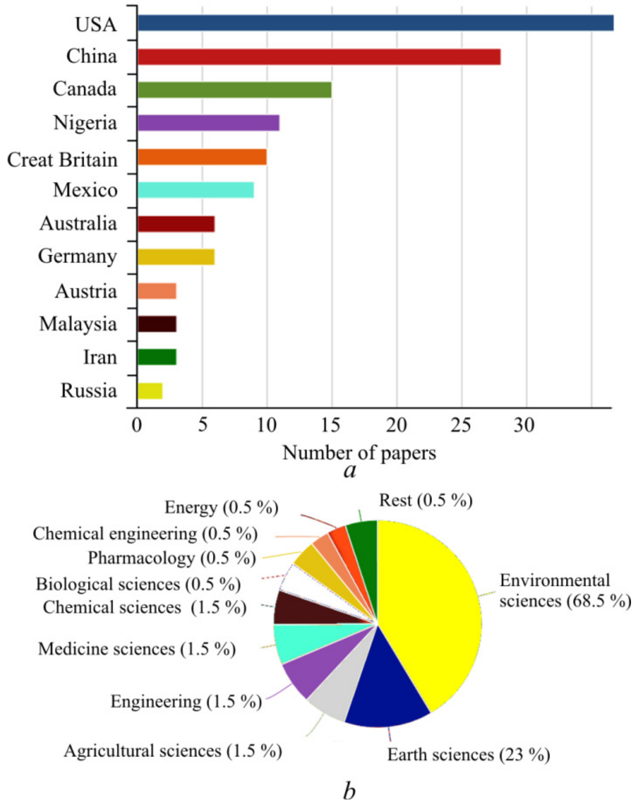

Fig. 1. Publication activity of countries on the subject of research (*a*); branch affiliation of published works (*b*)

The problems of hydrocarbon pollution of the environment were studied by various researchers. In 1979 there was a whole program called CONCAWE, devoted to the protection of groundwater from pollution by oil products. Following authors dealt and are dealing with the questions of study of hydrocarbon penetration rate: M.F. Fingas, S. Grimaz, S. Allen, J. Stewart, G. Dolcetti, S. Halmemies, S. Gröndhal, K. Nenonen, T. Tuhkanen, J.M. Keller, C.S. Simmons, V. Malk, S. Simpanen, Q. Zhang, G. Wang, N. Sugiura, A. Akbari, M. Ardestani, J. Shayegan, B. Zogala, R. Dubiel, W.M. Zuberek, M. Rusin-Zogala, M. Steininger, R. Iturbe, C. Flores, A. Castro, L.G. Torres etc. [5-15].

S. Halmemies, S. Gröndhal, K. Nenonen, T. Tuhkanen studied the time of penetration of oil into various grounds under laboratory conditions [9].

The deepest research on the penetration of hydrocarbons into the ground was performed by English and Italian scientists S. Grimaz, S. Allen, J.R. Stewart, G. Dolcetti and is presented in their joint work "Express method to evaluate the rate of penetration of hydrocarbons into the ground immediately after an accidental spill for rapid response". It provides an express method to predict the rate of penetration of hydrocarbons into the ground after an accidental spill on the ground. The model proposed by them is applicable under the following conditions:

a) evaporation occurs only from the surface of the pool until the oil has penetrated into the ground;

b) oil viscosity and density remain constant over the time;

c) advective processes are dominant in penetration;

d) Darcy's law is acceptable for determination of the rate of penetration into the soil [9].

Experimental studies of distribution of hydrocarbons in grounds under laboratory conditions were published in [1, 16-20]. They contain calculated models that allow predicting the rate of penetration of hydrocarbons in time:

– for sand of medium size

$$V_{\text{s.m.d}} = 24.0391 - 0.7733t,$$

where $V_{\text{s.m.d}}$ is for oil penetration rate in dry sands of medium size, mm/day; t is for penetration time, day;

– for fine sand

$$V_{\text{s.f.d}} = 16.5558 - 0.5321t,$$

where $V_{\text{s.f.d}}$ is for oil filtration rate in fine sands, mm/day; t is for penetration time, day;

– for clay

$$V_{\text{c.d}} = 8{,}101 - 0{,}2309t,$$

where $V_{\text{c.d}}$ is for oil filtration rate in clays, mm/day; t is for penetration time, day.

Due to the fact that laboratory studies were carried out with ground in air dry state of the same fractional composition, then these experiments do not reflect real natural conditions and are the first step in understanding the regularities of hydrocarbon distribution along the section. Therefore, it is need to study regularities of hydrocarbon distribution in field conditions. This article presents results of geostatistical processing of field work data.

The aim of the study is to identify regularities and build mathematical models for distribution of hydrocarbons along a section on the territories of oil refineries, which allow predicting the depth of contamination in case of accidental spills.

The information obtained will allow estimating changes in the bearing capacity of ground in case of hydrocarbon contamination [2, 19, 21, 22].

Study of features of distribution of hydrocarbons in the ground is based on identification of natural and man-caused regularities, which are described by means of a mathematics [16, 23–25]. That gives a methodological approach to study the regularities, depending on the geological conditions in which territories were formed and are the subject to risk of accidental oil spills.

Initial data used to identify regularities and build mathematical models

To identify regularities and build mathematical models field and experimental data obtained during cleaning of territory contaminated with oil products of oil refinery were used [16, 17, 20].

To determine the content of hydrocarbons in the ground field studies were carried out including drilling of wells, selection of monoliths, study of core material and hydrogeological, geomorphological and other conditions.

In order to optimize exploration network, data on the variability of the least mature geological and exploration parameters (hydrocarbons) of the investigated territory was used. Well placement parameters were determined from the following considerations.

At the first stage, testing was carried out uniformly across the area taking into account sources of possible contamination (ethylbenzene workshop - block 401; containers with finished products - blocks 404 and 405) and data of enquiry of workers about the spills of organic compounds into the ground. At the second stage well network was sealed. Maximum number of wells was drilled on the site of the workshop of the 31st chemical plant (Fig. 2).

In total, 250 wells were drilled on the site of study. Wells were drilled on anthropogenic, Quaternary sediments and rocks of Sheshminsky formation where more than 1160 samples were taken to determine the hydrocarbon content.

Well and its number

Fig. 2. Scheme of well placement

Quantitative content of petroleum products in the ground was determined using chromatograph Chrom with a flame ionization detector according to the generally accepted procedure.

A brief physical and geographical description

The relief of the site is characterized by the presence of flattened surfaces and slopes formed as a result of flat flushing, deep and lateral erosion of the Kama river, its tributaries and temporary outflow.

There are several objects defined within the section in terms of geomorphology such as a slope IV of the left bank terrace above the flood-plain of the Kama river (right side of the valley of the Pyzh river), III floodplain terrace of the Kama river complicated by the valley of the Pyzh river.

According to the dominant slope forming processes, the greater part of the slope IV of the terrace above the flood-plain refers to the deluvial terrace when the south-west part, facing the valley of the river Pyzh, is its right-bank side and represents an erosive slope.

Features of the geological structure of the area studied

Anthropogenic deposits are mainly sand and crushed stone. Thickness of these deposits varies from 0 to 1.3 m. Quaternary rocks are represented by loam, sands and gritty-crushed ground (Fig. 3).

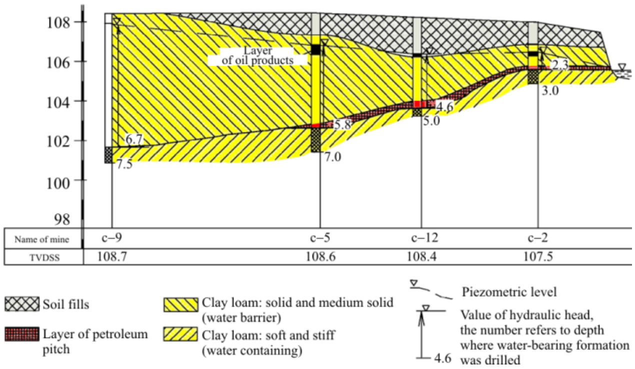

Fig. 3. Geological section along wells

Loams have dark brown color and are made from solid to semi-solid consistency, dense, with inclusions of gravel of argillite. The thickness of the loam varies from 0.8 to 3.7 m. The maximum thickness is discovered by wells 16-18, the smallest by wells 7, 8, 15.

Sands are gray, wet, fine and medium-grained, medium-density. Sands have a local distribution (wells 7, 15, 17, 19). The maximum open thickness is 1.7 m.

The grit-crushed soil is represented by fragments of mudstone with loamy aggregate up to 20 %. It also has a local distribution. The maximum thickness is opened by a well 2 and is 1.3 m.

Rocks of Sheshminsky age are represented by sandstones and siltstones. Sandstones have gray color and are fine-grained, dense, fractured, on clay-calcareous cement. Thickness of sandstones varies from 0.5 to 3.8 m.

Siltstones have dark cherry color and are medium strength, fractured, their thickness varies also within wide limits from 0 to 3.8 m.

Research methodology

Features of distribution of contaminating matter in the section was studied in the following sequence:

– geological structure of the territory was studied;

– quantitative content of oil products, which was obtained as a result of field research, in the soil was analyzed;

– geological, probabilistic, statistical and geostatistical methods were used to establish regularities in the distribution of hydrocarbons in the soil massif.

Results of the study

Statistical characteristics of results of the quantitative content of petroleum products are given in Table 1.

Table 1

The main statistical characteristics of the studied substances, mg/100 g of soil

Parameter	Average value	Standard deviation	Minimum deviation	Maximum deviation	Number of samples
Benzene	1.01	0.70	0.01	3.67	59
Toluene	0.18	0.43	0.01	1.99	57
Ethylbenzene	0.74	0.97	0.01	3.23	27
M-paraxylene	0.25	0.42	0.004	3.14	71
Orthoxilol	0.15	0.12	0.005	0.84	70
Styrene	0.45	1.22	0.006	6.00	43
Isobutyric aldehyde	10.21	11.16	0.40	44.80	16
Isobutyl alcohol	20.65	17.11	2.50	51.21	16
Butyl alcohol	15.56	24.60	1.60	92.80	30
2-Ethylhexanol	39.77	74.37	2.00	172.57	5
2-Ethylhexanal	34.56	72.19	1.90	244.00	11
Depth of sampling, m	2.32	1.99	0.1	9.0	–

Aromatic hydrocarbons are colorless liquids with a specific smell, lighter than water and do not dissolve in water, but they readily dissolve in organic solvents such as alcohol, ether and acetone. The physical properties of some arenas are presented in Table 2.

Analysis of the distribution of hydrocarbons along the section shows that in the stratum of study, benzene does not have a regular distribution. Meta-xylene and para-xylene are found in a significant number of samples. There is an implicit trend according to which the higher the depth the smaller the content of meta-xylene and para-xylene. The distribution of the sample is lower in this indicator than in benzene. Ortho-xylene is also found in many samples, its distribution in the section is a subject to the same regularity as meta-xylene and para-xylene. Styrene, ethyl benzene and toluene are found in small amounts of samples. So, a clearly expressed law of their distribution over the section is difficult to establish. Fig. 4 presents a three-dimensional model of hydrocarbon distribution in a soil massif created in the Voxler software.

Table 2

Physical properties of some arenas

Name	Formula	Melting temperature, °C	Boiling temperature, °C	Density at 20 °C	Dynamic viscosity at 20 °C, Pa·s
Benzene	C_6H_6	+5.5	80.1	0.8790	0.652
Toluene (methylbenzene)	$C_6H_5CH_3$	–95.0	110.6	0.8669	0.584
Ethyl benzene	$C_6H_5C_2H_5$	–95.0	136.2	0.8670	0.596
ortho-		–25.18	144.41	0.8802	0.707
meta-		–47.87	139.10	0.8642	0.548
para-		13.26	138.35	0.8611	0.571
Styrene (vinylbenzene)	$C_6H_5CH = CH_2$	–30.6	145.2	0.9060	0.749

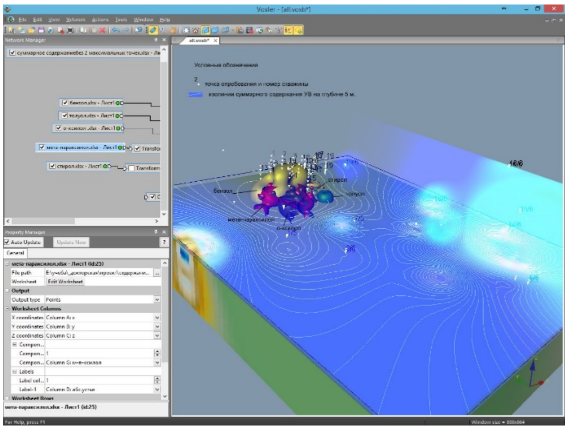

Fig. 4. 3D model of hydrocarbon distribution

As a result of studies of the distribution of hydrocarbon concentrations at different levels of the section, density curves have been plotted (Fig. 5). The plot shows that concentration distribution density is two-mode at depth of 0.25 m. Distribution law is symmetric at concentration of soil of 1.5 g/100 g at modal concentration of soil of 0 to 7 mg/100 g. At 0.25 m in depth concentration range of 7-10 mg/100 g of soil is not observed. The shape of the curve indicates the existence of low and high concentrations of hydrocarbons. At 0.75 m in depth asymmetric unimodal distribution is observed in the concentration range of soil of 0-10 mg/100 g.

At a depth of 1.25 m concentration distribution is almost uniformly in the range of 0.5-3.5 mg/100 g of soil. At depth of 1.75 m the distribution is unimodal in the range of 0-5 mg/100 g of soil.

There is a distribution with a large range of concentration changes is observed at 2.75 m in depth. Distribution is unimodal at depth of 3.5 m and at very low concentrations.

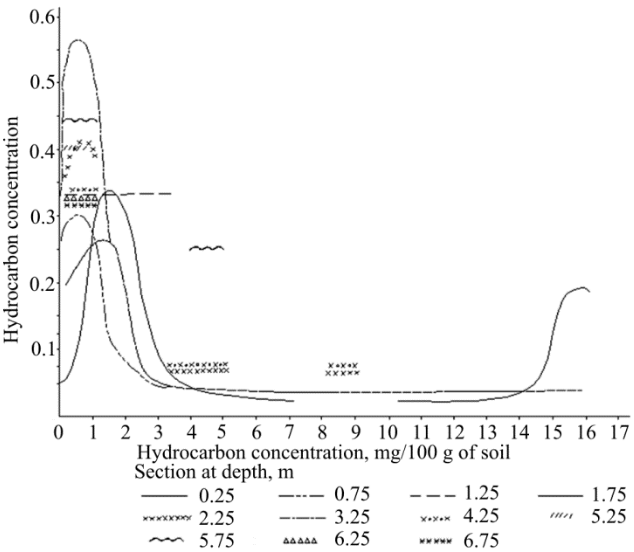

Fig. 5. Distribution of hydrocarbon concentration in depth

Beginning with a depth of 4.25 m a chaotic distribution of hydrocarbon concentrations is observed at values below 15 mg/100 g of soil. At concentrations of more than 15 mg/100 g of soil frequency of occurrence at different levels of the sections has the following values: 4.25 m – 0.22; 5.25 m – 0.25; 5.75 m – 0.57; 6.25-6.75 m – 0.67. Thus, the higher the depth the more often high concentrations is observed. The performed analysis allows to establish two levels of distribution of hydrocarbons along a section.

The first level includes hydrocarbons that are distributed almost continuously and are characterized by pronounced modal concentrations. The second level (starting from a depth of 4.25 m) does not show statistically continuous hydrocarbon distributions due to the small sample representativeness. However, there are high concentrations is observed with depth.

The analysis showed that zero concentrations are practically uniformly distributed over depth (Fig. 6). The concentration C = 0.5 mg/100 g of soil is most often found in the interval 0.5-3.0 m. C = 1.5 mg/100 g of soil is characterized by a unimodal distribution at depths of 0-2.5 m.

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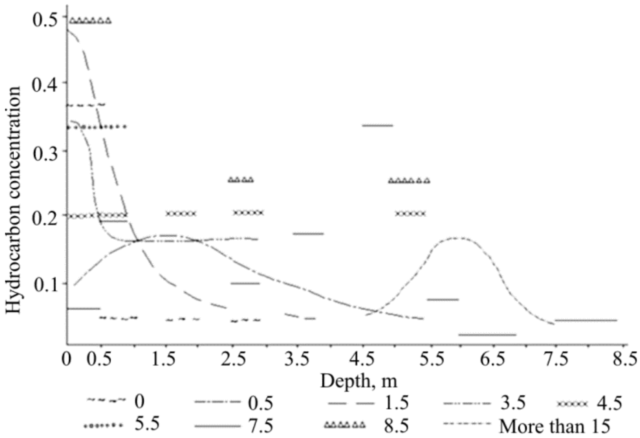

Fig. 6. Vertical distribution of hydrocarbon concentrations (mg/100 g of soil)

Analysis of the obtained curves shows that there is a directed change in the distribution laws from the uniform (C = 0) through symmetric (C = 0.5 mg/100 g soil), asymmetric (C = 1.5-3.5 mg/100 g soil) to the chaotic (C = 4.5 mg/100 g soil). The established trend indicates the dynamics of changes in hydrocarbon concentrations along the section.

Thus, conventional geological methods do not allow to reliably detect the regularities of distribution of hydrocarbons along the section of the thickness studied.

In order to solve the problem we used probabilistic and statistical methods in particular regression analysis [16], which was carried out on three samples. The first sample included wells, where there is a tendency to decrease the content with an increase in depth. The second sample contain wells which hydrocarbon content increases with depth. For the third sample there is a chaotic change in hydrocarbons with a depth of sampling. Interrelations were studied with help of linear and nonlinear regressions in the nine most frequently used equations in mathematical statistics.

For the first scenario, the regression equation has the following type:

$$K = 0.353 + 0.923/x \text{ at } r = 0.79,$$

where K is for concentration of hydrocarbon, mg/100 g of soil.

Let us check how the actual statistical distribution of concentrations in depth is described by the statistical model. It is established that out of 19 wells drilled within the western part of the territory, the real distribution of the sum of concentrations in 12 corresponds to the model obtained i.e. the higher the depth the lower the concentration of hydrocarbons. Concentrations increase with depth in wells 6, 10, 14, 19. So, there is an inversion of the model. This group of wells represents the second sample to build a mathematical model that relates to the change in hydrocarbon concentrations with depth.

The calculated regression equation for the second sample looks like

$$K = -1,454 + 1,126 \cdot h \text{ at } r = 0,79.$$

Distribution of hydrocarbons along the depth in wells 15 and 16 corresponds to the third adopted model.

Fig. 7 represents comparison of the obtained models of hydrocarbon distribution with related geological conditions. The first model corresponds to the conditions when clay loam is deposited on fractured mudstones, siltstones and sandstones having thickness is 2-3 m. The second model is inherent in the conditions when clay loam is deposited on high permeable gravel soil, which is also underlain mudstones, siltstones and sandstones. The thickness of clay loam does not exceed 1.2-1.5 m. Distribution of hydrocarbons in wells 15 and 16 refers to the third model and is irregular, due to the small thickness of loams underlying various soils (sands, gravel rocks) that are deposited on mudstones, siltstones and sandstones.

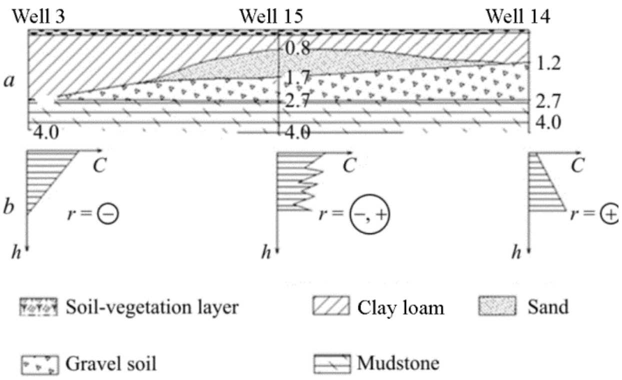

Fig. 7. Models of hydrocarbon distribution: *a* – geological section; *b* – graphs of hydrocarbon distribution; *C* – concentration of hydrocarbons in soil, mg/100 g of soil; *h* – depth, m; *r* – correlation coefficient

Based on above mentioned it is possible to describe the mechanism of distribution of hydrocarbons along a section of the rock massif.

When oil comes to the earth's surface then under the influence of gravitational forces, hydrocarbons penetrate into the rock massif [26, 27]. If there is no additional supply of hydrocarbons to the rocks, the penetration process ends when equilibrium between gravitational forces and forces preventing the migration of hydrocarbons is reached. If the groundwater table is high enough and the mass of hydrocarbons is high, then hydrocarbons lie on the aquifer, which is a fluid barrier. Then there is a redistribution of hydrocarbons along the cross section. Due to gravitational forces hydrocarbons are accumulated in the upper contact zone of the aquifer. Due to the capillary uplift hydrocarbons are accumulated in the near-surface zone of the section.

The type of rocks and their sorption capacity, reservoir properties (porosity and permeability) and water saturation significantly influence such a hydrocarbon distribution process. In case the near-surface zone of the earth is composed of sands that have low sorption capacity for hydrocarbons, high open porosity and high penetration properties, the zone of hydrocarbon contamination will be minimal compared to a zone composed of clays or loam (see Fig. 7) [28-30].

To assess the effect of soil types on the presence of anomalous concentrations, the following analysis was performed. Two groups of wells are identified within the western part of the territory on the map of total isoconcentrations. The first has increased concentrations; the second has background concentrations of hydrocarbons.

On the basis of two samples for thickness of various types of soils and average depth of their occurrence histograms were plotted. The following types of rocks were analyzed: soil-vegetation layer, clay loam, gravel soil of argillite, bulk soil, siltstone, sandstone, sand. Then, their individual informativity was determined by criteria t and F with a confidence probability of 0.95.

Thickness of loam (m_2) and burial depth (to some extent) of sandstones were informative indicators. Then thickness histograms were reconstructed into a probabilistic curve, the analysis of which allowed us to reveal the following regularity: the higher the thickness of loam the higher the probability $P(A)$ of high concentrations of hydrocarbons.

As a result of statistical processing it was established that presence of bulk soil affects the hydrocarbon content at a depth of 1 m. We analyzed dependence of the amount of hydrocarbons at depth of 1 m from the thickness of the bulk soil. It was found that in presence of bulk soil the average amount of hydrocarbons is much higher than if there is no bulk soil.

Conclusion

Three models of distribution of hydrocarbons along a section are revealed, which are controlled by geological conditions (lithology and burial depth of an aquifer). Based on regularities revealed, mathematical models have been developed that allow to predict the degree of soil contamination by hydrocarbons using geological indicators such as thickness of loam and crushed rock, and depth of burial of sandstones.

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