UNDERGROUND HYDROSPHERE OF THE SEDIMENTARY BASINS AS NAPHTIDES-GENERATING SYSTEM (ON THE EXAMPLE OF THE SOUTH CASPIAN BASIN)

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Abstract. The analysis of organic matter (OM) content dissolved in the formation waters and waters of mud volcanoes (water dissolved organic matter – DOM) of the oil and gas bearing South Caspian Basin and its distribution in stratigraphic and hypsometrical depth is given in the article. The stratigraphic interval of research covers the period from the Lower Pliocene to the Jurassic, and the depth interval: from 73 to 6043 m. In these intervals, the values of the DOM in reservoir waters vary from 4.1 mg/l to 271.2 mg/l, averaging (by 219 analyzes) 48.9 mg/l. A good correlation of the values of DOM and OM in rocks has been established. In both cases, Paleogene and Jurassic rocks have the highest values. In the change of the DOM with depth, an increase in its values from a depth of about 3.3 km is noted, which is possibly due to the onset of catagenetic transformation of OM into hydrocarbons in the rock-water system. The dependence of the DOM content on the mineralization of water has been established: its highest values are characteristic for waters with mineralization not higher than 50 g/l. The waters of mud volcanoes are characterized by low levels of DOM and low mineralization, which is most likely due to their condensation nature.

The conducted studies confirm the idea of the DOM participation, along with the OM of rocks, in the processes of oil and gas generation. The process of OM transformation into oil and gas in aqueous solution should be taken into account in basin modeling and in estimating the predicted resources of hydrocarbons in the sedimentary basin.

Keywords: sedimentary basin; formation water; water of mud volcanoes; organic matter; oil and gas
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Introduction

Sedimentary rocks are widespread on our planet. Together with the modern sediments lining the bottom of the World Ocean and the water basins of the land, they form the sedimentary cover of the Earth. The thickness of the sedimentary cover of the Earth varies in a wide range: from 0 to 20-30 km. The total volume of sedimentary cover rocks is estimated at $1.1x10^9$ million km^3, which is about 11% of the Earth's Crust (Ronov, 1980).

According to existing concepts, all the voids of sedimentary rocks (except for hydrocarbon deposits) are filled with water below the groundwater level, in connection with which the mass of the waters contained in these rocks is quite large. According to available estimates (Zverev, 2001), only $3.0*10^{23}$ g of water is contained in the sedimentary cover of the Earth's Crust. In particular, in the South Caspian Basin, the volume of these waters is about $5.3*10^{20}$ g (Zverev et al., 1998). Groundwater is represented both in free and bound (adsorbed) form.

V.I. Vernadsky has pointed out in his works on the huge role of water in geological processes in this works. He believed that the composition of water is a function of the long evolution of the system water-rock-gas-organic matter system (Vernadsky, 2003).

The fundamental property of this system is its equilibrium-nonequilibrium state (Vernadsky, 2003; Shvartsev, 1997, 2008; Bullen, Wang, 2007). Water in the porous-fractured space is in continuous interaction with the mineral skeleton of rocks, which is maximal in finely dispersed (clayey) rocks, which account for about 70% of the total volume of sedimentary rocks. Water at all stages of interaction with rocks continuously concentrates some elements and dissipates others, which is manifested by a regular change in the composition of the aqueous solution. The evacuation of mobile mineral and organic matters (OM) from the rock is accompanied by a gradual increase in their contents in the porous waters.

OM of water is only a part of the organic component of the water-rock system of sedimentary basins. There are autochthonous OM, formed in the water body as a result of the vital activity of aquatic organisms, and allochthonous OM, entering it from outside (Lozovik, 2012). The main source of OM in the reservoir is phytoplankton (Vinberg, 1960; Vinogradov, 2004). However, the water-soluble OM can be inherited not only from the waters of the sedimentary basin, but also include OM, which has passed into the groundwater from rocks (highly soluble organic acids in water, mobile products of the dispersed OM transformation, etc.) during lithogenesis.

M.E. Altovsky and others (1962) showed that the mutual transitions of OM in the water-rock system are determined by the ratio of their concentrations and the sorption capacity of the rocks. At the same time, the enrichment of rocks with dispersed OM renders significant influence on the value of the water dissolved organic matter (DOM) content in underground waters (Shvets, 1982; Bars et al., 1990).

The total amount of OM in groundwater is commensurate with the amount of OM in many natural objects and second only to its content in sedimentary rocks (Zverev, 2001).

The theoretical and experimental studies carried out by a number of scientists (Altovsky et al., 1962; Zinger, Dolgova, 1982; Zinger, 1995; etc.) made it possible to establish an identical distribution of OM in the formation water-water-bearing rock system. This served as the basis for the first time to put forward the concept of the possible participation in the oil and gas generation not only OM of rocks, but also DOM (Altovsky et al., 1962; Kudryakov, 1982; etc.).

The background content of DOM, like the OM of rocks, is controlled mainly by such factors as: the evolution of the sedimentation basin, the intensity of the OM inflow from the surrounding land by water flows, the sedimentation environment, lithogenesis processes, etc.

The enrichment of the rocks with dispersed OM influence significantly on the amount of the DOM content in underground waters (Shvets, 1982) (Table 1).

E.A. Bars with co-authors (Bars E.A., Aleksandrova et al., 1967) have established the apparent positive dependence of the OM background concentrations in groundwater on its concentration in the water-bearing rocks, based on the analysis of more than 1500 analyses of different parameters of the OM composition in waters and about 3000 analyses of the OM composition in the rocks for the eastern part of the Azov-Kuban and western parts of the Middle Caspian oil and gas basins (Figure 1).

The South Caspian Basin is one of the oldest and well studied oil and gas basins. However, specific studies on the analysis of regional regularities of the OM distribution in groundwater, their connection with OM of rocks, the role of the DOM in oil and gas generation have not yet been carried out.

In this regard, the purpose of this article is to generalize and analyze the data accumulated to date on the content of OM in the formation waters of various stratigraphic complexes, as well as mud volcanoes, and its correlation with the content of OM in the sedimentary rocks of the South Caspian Basin.

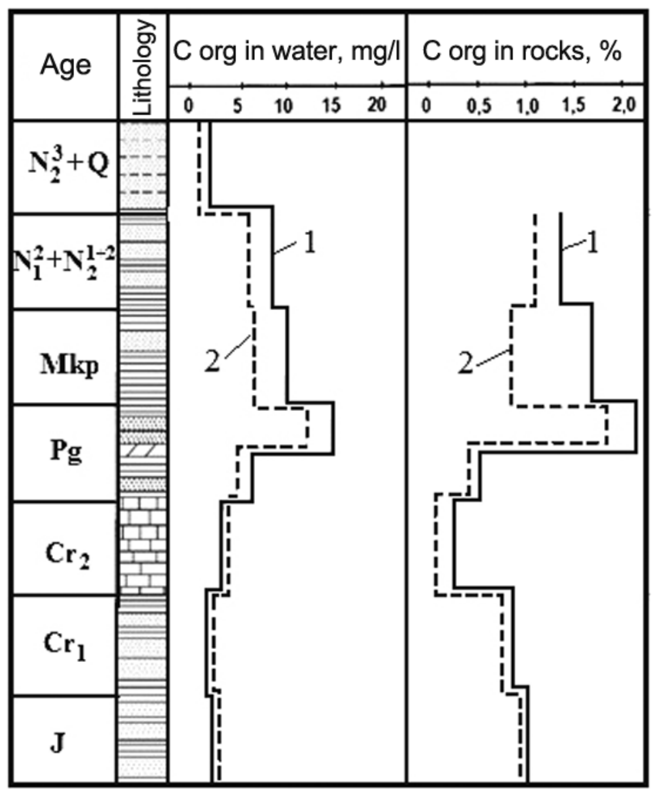

Figure 1. The content of OM in the waters and rocks of Ciscaucasia: 1 – Azov-Kuban basin; 2 – Middle-Caspian basin (Bars et al., 1967)

Age of rocks	C_{org} of rocks,%	C_{org} of underground waters, mg/l
Neogene	0,9	14,4
Paleogene	0,9	8,3
Cretaceous	0,5	4,2
Palaeozoic	0,3	2,6

Table 1. Dependence of the DOM content on the enrichment of rocks with organic matter

Actual material

The research results presented in this article are based on about 300 analyzes of formation waters and waters of mud volcanoes, as well as more than 400 data on the content of OM in rocks of the South Caspian Basin. The stratigraphic interval of research covers the period from the Lower Pliocene to the Jurassic, and the deep interval – from 73 to 6043 m.

Results and Discussion

The South Caspian Basin (SCB) occupies a vast area of deflection of the Earth's Crust, which includes the southern part of the Kura intermountain trough, the West Turkmen depression and the deep-sea basin of the Southern Caspian located between them. In the hydrogeological sense, the SCB is a classical connate basin (Kostikova, 2002).

The content of OM in the rocks of the sedimentary complex of the SCB has been previously studied primarily on natural outcrops of different stratigraphic ages, the results of which are reflected in the works (Bailey et al., 1996; Guliyev et al., 1997; Feyzullayev et al., 2001). According to these studies, the rocks of the Miocene-Oligocene deposits, which are referred to as oil source rocks (Guliyev, Feyzullayev, 1996; Katz et al., 2000; Feyzullayev et al., 2001; Gurgey, 2003; et al.), differ in the highest values of total organic carbon (TOC).

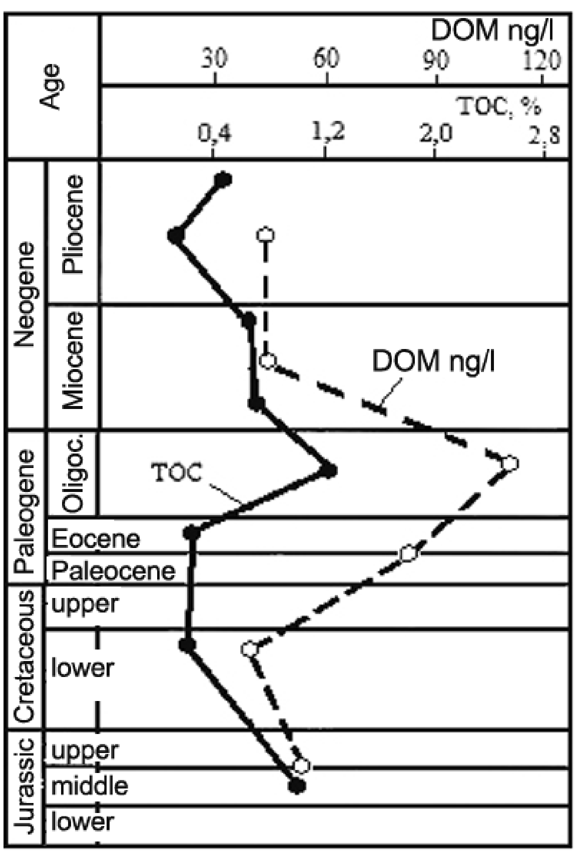

Figure 2. Change with the stratigraphic depth of average values of OM in the rock and DOM in the South Caspian Basin

An analysis of the variation in the section of the values of this parameter averaged over individual stratigraphic complexes showed its uneven distribution (Aliev et al., 2005). As already noted, the maximum OM contents were distinctive for Maikop (Oligocene-Lower Miocene) sediments; The Jurassic rocks are also distinguished by elevated values (Figure 2).

In order to compare the nature of the change of OM in the section of rocks, the mean values of the DOM in the formation waters of various stratigraphic complexes of the SCB were also calculated in this work based on the created database: from the Lower Pliocene (Productive series – PS) to the Jurassic. In the considered stratigraphic interval, the DOM was changed in the range from 4.1 mg/l (PS) to 271.2 mg/l (PS). The average value for 219 analyzes was 48.9 mg/l, which agrees well with previous estimates for other basins, according to which the background values of dissolved OM in underground aquifers of oil and gas basins do not exceed 50 mg/l (Kiryukhin et al., 1973; Shakhnovsky, 2003).

As can be seen from Figure 2, the nature of the change in the section of the average values of the DOM is in good agreement with the nature of the change in the OM content in rocks. A certain deviation towards higher values of OM in the waters in comparison with the rocks is noted only for the Eocene-Paleocene interval of the section.

The content of OM in the formation waters of the various stratigraphic complexes of the SCB is more clearly demonstrated by the distribution histograms of its values shown in Figure 3. According to these histograms, in water, as well as in rocks, the highest values are characteristic to oil-source rocks of Paleogene-Miocene and Jurassic deposits.

The waters of mud volcanoes are characterized by the lowest concentrations of OM.

The revealed correlation between the OM content in the waters and their enclosing rocks allows us to conclude that a dynamic equilibrium is established between the rock and water that are constituent parts of a single rock-fluid system and which are in continuous interaction during a long geological time.

Moreover, as shown by the results of A. Schimmelmann, M. Mastaler (2001), the dynamic equilibrium is also manifested in the relationship between the isotope composition of hydrogen (the ratio D / H) of oils (its various fractions) and contacting with them formation waters (Figure 4).

As is known, physical (temperature and pressure), chemical (composition of formation water), lithofacies (the density of rocks and their reservoir properties) and other conditions and associated processes (oil and gas formation, clay dehydration, etc.) change with depth. In this connection, it is of some interest to analyze the

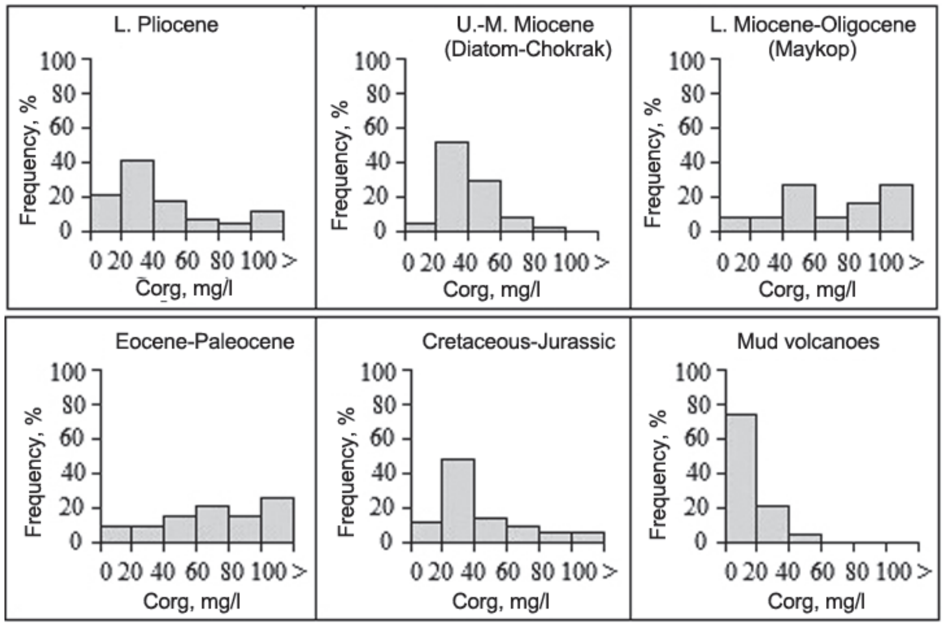

Figure 3. Distribution histograms of values of OM content in formation waters of various stratigraphic complexes of SCB

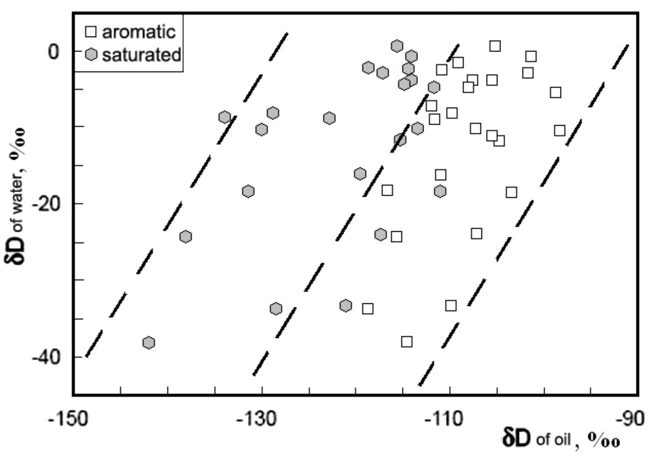

Figure 4. The dependence between the isotopic composition of hydrogen in formation water and various fractions of oil (Schimmelmann, Mastaler, 2001)

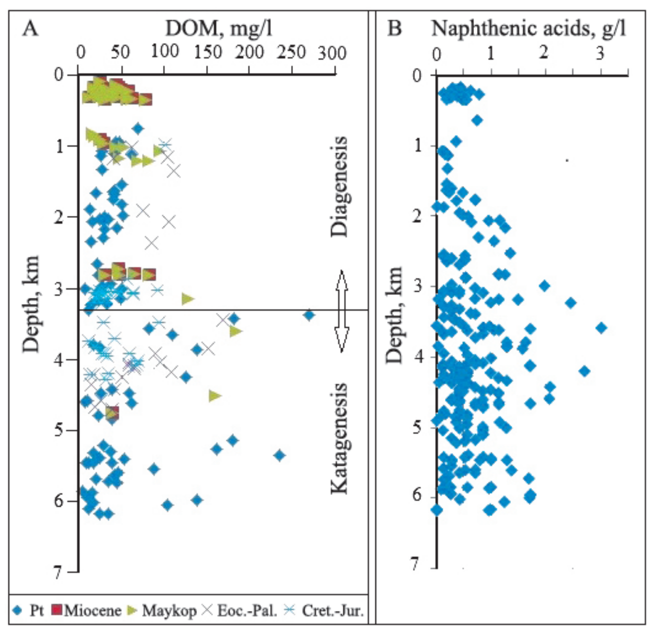

Figure 5. The change with the depth of OM content dissolved in the formation waters of various stratigraphic complexes of South Caspian Basin (A) and the content of naphthenic acids in the waters (B)

regularities of the change in the content of the DOM with hypsometric depth.

As can be seen from the graph presented in Figure 5A, a pronounced feature of the change of the DOM values is common for various stratigraphic complexes. This feature is expressed in the presence of a sharp jump in the direction of increasing the values of the DOM, recorded from a depth of approximately 3.3 km. A similar character is noted in the change with the depth of naphthenic acids (Figure 4B), which have a direct dependence on the amount of hydrocarbons (Smirnova, 2009).

Most likely, this is due to the transition of the rock-water system from the diagenesis stage (where relatively low temperatures are not yet sufficient for thermal decomposition of OM) into the catagenesis stage, where favorable temperature conditions exist for the transformation of OM into hydrocarbons.

Given the relatively higher migration potential of oil and gas in comparison with their ancestor – OM, in the zone of catagenesis, their infiltration from rocks into water begins accompanied by an increase in water of the organic component and bitumen.

The type of the relationship between the DOM and the carbon content of bitumoids (C_{bit}) confirms this conclusion. According to Figure 6, high contents of C_{org}, which are observed from a depth of more than 3 km (Figure 5), are characterized by high values of C_{bit}, which is a derivative of the thermal transformation of C_{org}.

Kartsev and others (Kartsev, Vagin, Baskov, 1969) also considers that sedimentogene waters dissolve hydrocarbons formed as a result of catagenesis and move with them to reservoir rocks. When moving through

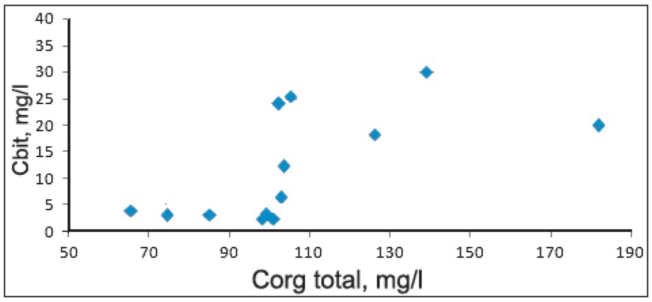

Figure 6. Relationship between C_{org} and C_{bit} content in water

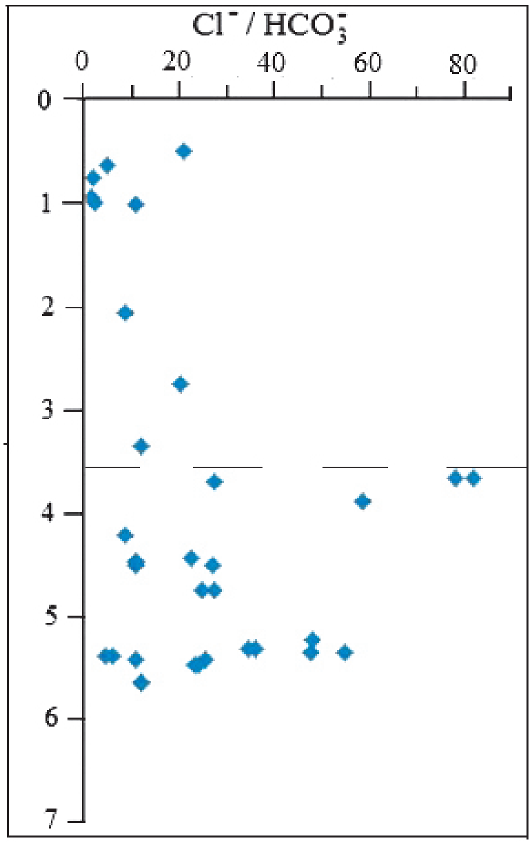

Figure 7. Ratio of chlorine ion to hydrocarbonate ion changing with depth

reservoirs, groundwater can additionally dissolve a certain amount of hydrocarbons and other organic compounds in reservoir rocks.

Studies conducted by V.M. Shvets (1973) have found that the content of DOM depends on the chemical composition of the waters. However, according to other studies (Metody i napravleniya issledovanii organicheskikh veshchestv ..., 1975), mineralization and chemical composition of waters have an ambiguous effect on the content of OM in them.

To study the relationship between these parameters in the formation waters of the SCB, data on the well-studied productive strata were involved.

As can be seen from Figure 7, a pattern similar to the change with the depth of the DOM is observed in the ratio of chlorine ion to the hydrocarbonate ion changing with depth (Figure 4A). In this case, too, approximately from the same depth (3.5 km), a jump in the values of the considered parameter in the direction of its increase is observed. This fact gives grounds to assert the existence of a certain relationship between the chemical composition of water and the OM contained in it.

From the world experience, the dependence of the OM content in waters on its salinity is also known. Thus, the investigation of the DOM in wells of the fields of the Michigan basin (USA), producing gas and water, has established the dependence of the OM content in the formation waters on its salinity (Huang, 2004). According to the results of other studies (Yan Chen et al., 2013), there is a negative relationship between the content of DOM and the salinity of the water.

To study the nature of the dependence of these two parameters, in relation to the geological conditions of the SCB, a corresponding graph was constructed, which is shown in Figure 8. According to Figure 8, the formation waters of SCB containing high concentrations of DOM (more than 70 mg/l) are characterized by a relatively

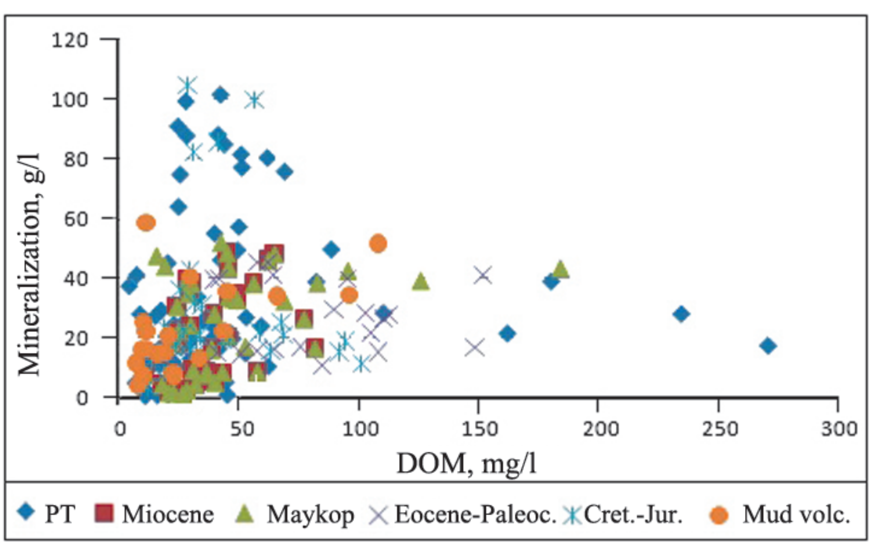

Figure 8. Relationship between the content of DOM of formation waters of various stratigraphic complexes and waters of mud volcanoes and the mineralization of these waters in the South Caspian Basin

low mineralization (less than 50 g/l). In waters with a salinity of more than 60 g/l, the content of DOM is low and varies within the limits of 25-70 mg/l.

At the same time, formation waters with low DOM values (less than 20 mg/l) and low salinity are also encountered. It is important to note that the same fact is established in the Michigan basin of the USA (Huang, 2004). Such waters are also characteristic for mud volcanoes. According to data from 24 analyzes, DOM in waters of mud volcanoes in the SCB varies from 7.9 mg/l to 108.5 mg/l (average 28.6 mg /l), while water salinity ranges from 4.1 mg/l to 58.7 mg/l (24.2 mg/l). The same feature was previously revealed in the study of the mud volcano waters in the Taman Peninsula (Alexandrova, Bars, 1967).

Low mineralization and low content of OM in some formation waters and waters of mud volcanoes are most likely due to their condensational genesis. To some extent, this is confirmed by the low mineralization of formation waters, which are directly connected to oil in comparison with the waters behind the oil-bearing contour established on the example of Neftechala and Khilly fields of the SCB (Figure 9). Taking into account that the oil in the PS is of a epigenetic nature, this phenomenon can be associated with phase transitions caused by changes in the thermobaric conditions during the subvertical migration of fluids.

It should be noted that T.S. Smirnova (2009) also found that the waters of gas-condensate and oil-bearing

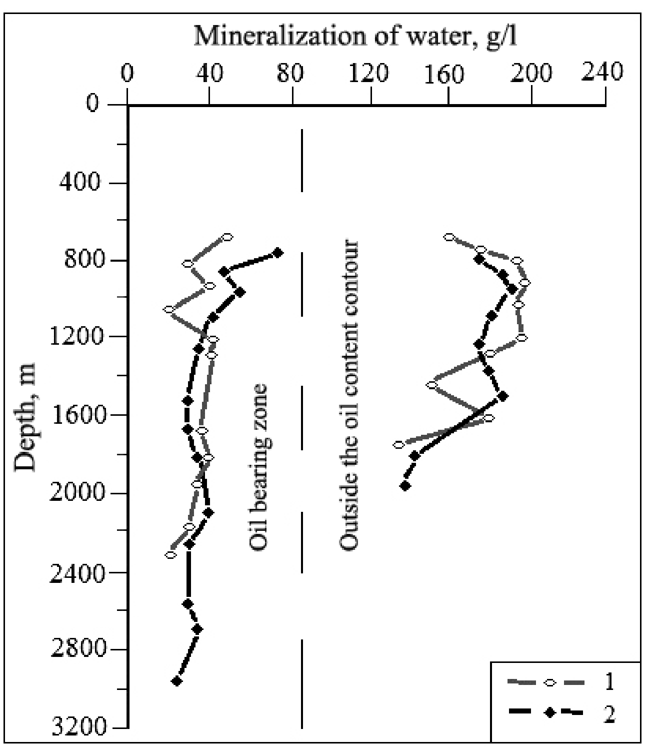

Figure 9. Change in mineralization of formation waters with depth within the oil deposit and behind the contour of oil bearing areas of Khilly (1) and Neftechala (2) of the South Caspian Basin (Feyzullaev, 2010)

deposits are characterized by very high concentrations of hydrocarbonates.

Conclusion

An analysis of the world experience in the study of OM in the underground hydrosphere, as well as the results of the present studies on the example of the SCB, makes it possible to conclude the following.

Underground hydrosphere of sedimentary basins (including its organic component) is part of a single water-rock system, between the component parts of which there is a continuous interaction and interchange of substances, the intensity of which depends on a complex of geological factors. The consequence of interaction processes between water and its host rock is the establishment of dynamic equilibrium in this system during its geological evolution. This explains the good convergence of the distribution of OM in rocks and formation waters of the sedimentary section of SCB, the relationship between the content of OM in waters and their chemical composition and mineralization, and the nature of changes of these parameters with depth.

Based on the results of the research carried out for DOM in the South Caspian Basin, the following conclusions can be making.

- The average statistical value of the DOM content in the formation waters of SCB as a whole is about 50 mg/l, which agrees well with the results for other basins.
- The distribution of DOM on the section is uneven and correlates well with the content of OM in the rocks: its highest content, as in rocks, is noted in the Paleogene-Miocene and Jurassic sediments. This relationship is due to the primary enrichment of rocks with OM, the degree of lithification of rocks, the features of hydrodynamic regime of confined water complexes and continuous processes of interaction between water and its host rock.
- The features of the change in the content of DOM in formation waters with hypsometric depth are revealed, which are characterized by an increase in its values from a depth of approximately 3.3 km. It is believed that this is due to the onset of catagenetic transformations of OM into hydrocarbons.
- A characteristic relationship between the content of OM in the reservoir waters of the SCB and its mineralization was found. It has been established that the highest values of DOM are characteristic of waters with mineralization not exceeding 50 g/l. There are also reservoir waters with low values of both DOM and water salinity. These waters include waters of mud volcanoes. In all likelihood, these waters are of a condensation nature.

The performed studies confirm the idea that the underground hydrosphere, which is an inseparable part of the unified rock-water system of the basin, can play the role of an additional source of hydrocarbons. In this connection, the process of transformation of OM into oil and gas in aqueous solution should be taken into account in basin modeling and in estimating the forecast resources of hydrocarbons of the sedimentary basin.

However, it should be recognized that if an express method of pyrolysis of rocks is widely and successfully used to quantify the hydrocarbon potential of rocks (Espitalie et al., 1977), an express method has not yet been developed to quantify the scale of hydrocarbon generation in an aqueous medium.

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