The impact of active petroleum system on light hydrocarbons distribution in marine sediments

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Abstract
Integrated interpretation of regional geochemical data and results of the numerical BM&PSM performed at the east part of the Sea of Azov and northeast part of the Black Sea reveal a correlation between the spatial distribution of light hydrocarbons in seabed sediments and investigated petroleum systems. The obtained results point out that the spatial distribution of gaseous hydrocarbons in marine sediments reflects the geological structure of sedimentary cover, and the maturity of petroleum systems, located within the basins. The origin of background levels and anomalies of light hydrocarbons was explained depending on the present-day petroleum system activity and the structure of overburden rocks.

Keywords Petroleum system · Light hydrocarbons · Marine sediments · Geochemical research · Basin modeling · Sedimentary cover

Introduction
Marine geochemical petroleum prospecting is based on the postulation of subvertical migration of gaseous hydrocarbons from deep oil and gas fields up to sea bottom (Sokolov, 1980; Schumacher 2003, 2017; Starobinets et al., 1993; Wagner et al., 1998; Mackenzie and Quigley, 1988; Brown, 2000). Hence, high amounts of light hydrocarbons (C₁–C₆ HC) detected in sediments’ headspace gas indicate the presence of petroleum accumulation in the sedimentary cover.

As generally assumed (Bazhenova et al., 2000; Starobinets et al., 1993; Larskaya, 1992), there are several sources of C₁–C₆ HC to seabed sediments: migration from the deep part of the sedimentary cover, where hydrocarbons are produced as a result of organic matter thermal destruction (Tisso and Welte, 1970; Hunt, 1982); microbial in situ activity in modern seabed sediments (production of methane mostly); and anthropogenic contamination of marine ecosystems. Their contribution varies under differences in geological, ecological, and microbiological conditions (Jones and LeBlanc 2004, Michael and Abrams, 2020).

The investigated distribution of C₁–C₆ HC in the quaternary sediments in the vicinity of the Black Sea and the Sea of Azov discloses the low contribution of microbial and anthropogenic processes. Also, background (BG) levels of light hydrocarbons above main tectonic elements depend strongly on the subsurface geology (Lavrenova and Kruglyakova, 2010).

Both the Black Sea and the Sea of Azov are well-known oil- and gas-bearing basins, where several commercial discoveries and numerous oil and gas shows point out the presence of active petroleum systems (PS) (Fig. 1). Obviously, not only hydrocarbon accumulations take effect on C₁–C₆ HC spatial distribution in the marine sediments, but the whole PS, including a pod of source rock, migration paths, etc.

Thus, the present study focused on the analysis of petroleum systems’ impact on light HC distribution in seabed sediments.

Geological setting
The investigated area includes the east part of the Sea of Azov and the northeast part of the Black Sea (Fig. 2).
Water depths within the Sea of Azov are not more than 13 m. The area belongs to the Scythian Platform, divided into Indolo-Kuban Foredeep, South Azov Step, Azov Swell, North Azov Depression, and Rostov Flunge (Fig. 2). Sediment cover comprises Cretaceous and Cenozoic terrigenous and carbonates sediments above a transitional complex which generally includes Triassic and Lower–Middle Jurassic argillites. The lower part of the transitional complex most likely encompasses Paleozoic deposits.

Water depths within the Black Sea reach 2000 m. The studied area includes the following main tectonic elements: Shatski Ridge, Tuapse Trough, and Novorosiysko-Lazarevski Synclinorium. Sediment cover comprises Upper Mesozoic and Cenozoic predominantly terrigenous sediments (Fig. 2).

Several small gas fields discovered within the Cenozoic system of the Sea of Azov (Strelkovoye, Beysugskoye, Vostochno-Kazantipskoye, etc.), as well as the small oilfield—Novoe, relate to Maykopian and Middle–Upper Miocene plays. Additionally, there are several discoveries in Triassic (Electrorazvedochnaya, Zapadnoe-Beyshskaya) and Eocene deposits (Neizvestnaya). All identified reservoirs are productive mostly gas-saturated. Preliminary investigations indicated that these gas occurrences are most likely genetically associated with the underlying transitional complex. At the northeast part of the Black Sea, oilfield Subbotina and numerous oil shows from mud volcanoes belong to the Maykopian Play. Mesozoic gas fields and some oil shows discovered onshore indicate Mesozoic petroleum system.

Materials and methods

Geochemical data

Seabed samples were collected during surveys covering geochemical, geotechnical, and ecological aspects. Investigations were developed by FSC Yuzhmorgeologiya and by Chernomorneftegaz CJSC for 20 years: from 1990 to 2010. The sampling strategy for each investigation was generally determined by its scale. Therefore, the final location plan of the seabed sampling scheme is irregular (Fig. 3).

The concentrations of light hydrocarbons (methane, ethane, propane, butane, pentane, and hexane) measured overall in sediment samples from 2600 sites constitute the geochemical dataset for the present research. To estimate anthropogenic influences and for better interpretation of the gas-geochemical data, some ecological indicators including concentrations of mineral oil products and amounts of oil-oxidizing bacteria (Oborin & Stadnik, 1966) in seabed sediments were examined.
Numerous stable isotope analyses of gaseous hydrocarbons and oils from mud volcanoes and cold seeps at the Black Sea indicate mixing of different sources of CH$_4$: vertically migrated (thermogenic gas) and an admixture of in situ bacterially generated methane (Blinova, et al., 2003; Bohrmann et al., 2003; Kutas, 2020; Kruglykova R, et al., 2003, 2009). The source of this biogenic methane is the anaerobic degradation of heavy hydrocarbons coupled to
methanogenesis (Jiménez et al., 2016; Rabus et al., 2016; Aeckersberg et al., 1991). So microbial activity in marine sediments obscures the isotope signature and decreases the effectiveness of carbon isotope information as a hydrocarbons source indicator. In such cases, next standard indicators involved in interpretation to detect hydrocarbons anomalies origin: high concentrations of hydrocarbon gases uncorrelated with TOC content, high (above background) concentrations of saturated hydrocarbons, C\textsubscript{1}/\(\sum\)C\textsubscript{2},C\textsubscript{3},C\textsubscript{4},C\textsubscript{5},C\textsubscript{6} below 1000, \(\sum\)C\textsubscript{2},C\textsubscript{3},C\textsubscript{4}/\(\sum\)C\textsubscript{5},C\textsubscript{6}, and a clear predominance of iso-butane over n-butane (Blinova, et al., 2003; Hunt 1982; Bohrmann et al., 2003; Kruglykova et al. 2009; Wagner et al., 1998).

The headspace gas of the sediment samples collected by box corer was extracted on board immediately after the retrieval. Then, it was analyzed by means of a gas chromatograph with a flame ionization detector, and column packed with anumogel or a synthetic sorber. A calibration standard kit for hydrocarbons (C\textsubscript{1}–C\textsubscript{6}) was employed for the calculation of concentrations. Detection limits (in vol\%) varied, for C\textsubscript{1}, from 1 × 10\textsuperscript{-1} to 1 × 10\textsuperscript{-5}; C\textsubscript{2}, from 1 × 10\textsuperscript{-1} to 2 × 10\textsuperscript{-6}; C\textsubscript{3–C\textsubscript{6}}, from 1 × 10\textsuperscript{-1} to 2 × 10\textsuperscript{-6}. Accuracy was ± 10%.

**Statistical data analysis**

The analytical data comprise the concentrations of the light hydrocarbons C\textsubscript{1} to C\textsubscript{6} detected in seabed sediments. To reduce the number of variables and based on the documented correlations between ethane (C\textsubscript{2}), propane (C\textsubscript{3}), and butane (C\textsubscript{4}) and between pentane (C\textsubscript{5}) and hexane (C\textsubscript{6}), we considered the sum of C\textsubscript{2}–C\textsubscript{4} and the sum of C\textsubscript{5}–C\textsubscript{6} as single variables. Thus, methane, sum C\textsubscript{2}–C\textsubscript{4}, and sum C\textsubscript{5}–C\textsubscript{6} were included in processing and discussion.

The logarithmic mean and standard deviation of each variable were calculated to characterize their distributions. The logarithmic means were regarded as background and observations within one standard deviation of the logarithmic mean—as natural background fluctuations. Individual values were rated as low-contrast anomalies if their values exceeded the background level by more than one standard deviation, as high-contrast anomalies if their values exceeded the background level by more than two standard deviations, and as extraordinary concentrations, if their values exceeded the background level by more than three standard deviations. Mapping of the spatial distributions of geochemical variables was performed based on obtained statistical analysis results.

The results of different geochemical surveys compared using the T-test (Student’s) and F-test (Fisher’s) to ensure that the data were compatible.

| Table 1 The stratigraphy |
|-------------------------|
| **The Sea of Azov**     | **The Black Sea** |
| Layers                  |                |
| N\textsubscript{4}–Q    | Q              |
| N\textsubscript{3}–2    | N\textsubscript{2,3} |
| Pg\textsubscript{5–N\textsubscript{1}} (Maikopian) | Pg\textsubscript{3–N\textsubscript{1}} (Maikopian) |
| Pg\textsubscript{1–2}   | Pg\textsubscript{2–1} |
| K                       | P\textsubscript{3–1} |
| J3                      | K               |
| T–J\textsubscript{1–2}  | J\textsubscript{3} |
| PZ (D–C)                | J\textsubscript{2} |

**Basin modeling**

Basin analysis performed according to principles highlighted by Allen and Allen (2005), Welte et al. (1997), and Hantshel & Rauerauf (2008) revealed several large sedimentary basins located in the vicinity of the Sea of Azov and the Black Sea. One of them developed within the Sea of Azov from Paleozoic to Cenozoic. The Mesozoic (J–K) Great Caucasus basin existed until the Caucasus orogeny started in Paleogene. After that, the Cenozoic foreland basin of the Tuapse Trough originated (Spadini et al., 1997; Kazmin et al., 2000; Senin et al., 2014).

Basin modeling enables to delineate PS in sedimentary basins situated within the studied area. Present-day geometry and lithological composition of the Sea of Azov basin model were reconstructed based on published data (Borkov et al. 1994, Tugolesov et al. 1989) and well data drilling reports collected from the Russian State Geological Depository.

All available datasets coming from marine seismic data and the results of onshore and offshore geological surveys (Gorshkov et al. 1989), (Aphansenkov et al. 2007) integrated in course of building the Black Sea basin geometry. The domains of Mesozoic and Cenozoic paleo-basins including shelf, continental slope, and abyssal plane extent onshore as part of Tuapse Trough and Novorosysko-Lazarevski Synclinorium. All of them explored within the northwest Caucasus mountain region, located across the present-day coastline, allow to predict offshore sedimentary rocks lithological composition and stratigraphy despite extremely rare well data.

Thus, both 3D basin models include eight layers (Table 1).

Offshore and onshore seismic data, drilling logs, bottom and outcrop samplings as well as geochemical examination of organic matter (Aphansenkov et al. 2007; Bazhenova et al., 2002; Nadezhkin and Ivanov 2011) provide
necessary information to ascertain essential petroleum systems elements.

As a result, three petroleum systems were simulated within the east part of the Sea of Azov and two—within the northeast part of the Black Sea (Tables 2, 3).

Both models have been simulated with a constant heat flow of 55 mW/m² based upon the published data (Cher-mak et al., 1982; Verzhbitski, 2002; http://www.cnrrb.ru/geol/heat/index.php). The automatic trend provided by Petromod software for 44° northern latitude was used to calculate sediment–water interface temperature (Wygrala, 1989). Paleo-water depth maps have been constructed from paleogeographical data.

The model was calibrated using data from all available wells: temperature and vitrinite reflectance. Organic matter maturity (from Rock–Eval analysis) measured in mud volcanic breccia and Mesozoic and Cenozoic rocks from continental slope outcrops was applied to validate the calculated rock maturity in the model. Final verification, using natural petroleum manifestations, reflects their good coincidences with modeled hydrocarbon accumulation.

Results of basin modeling revealed that sediment cover and transitional complex (the upper part of the Scythian Plate basement, which is not thermally affected or significantly deformed) at the east part of the Sea of Azov include three PS: 1) “North Azov PZ (.”),” “South Azov MZ (?”),” and “South Azov CZ (!)” (Figs. 4 and 5). There are at least two PSs in the northeast part of the Black Sea (“Tuapse Trough CZ PS (!),” “Northeast BS MZ PS (?)” (Figs. 5 and 6).

### Results and discussion

The results of petroleum system modeling analyzed according to White (1993, 1998), Magoon and Dow (1994), Magoon (1995), and Magoon and Dow (2000) enable to perform integrated PS maps and cross sections (Figs. 4, 5, 6, and 7). The main conclusions of performed petroleum system modeling with respect to this study come as follows.

The well-known “South Azov Cenozoic (!)” petroleum system is located within the Indolo-Kuban Foredeep. Petroleum accumulations that are genetically associated with the PS can be expected exclusively in the Upper Cenozoic sediments of Indolo-Kuban Foredeep (Figs. 4 and 5). Lower Maikopian source of petroleum from discovered oil fields of Indolo-Kuban Foredeep was proved by biomarker analysis.

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1 Petroleum systems named after Magoon (1995, 1998, 2004a, b) and Magoon and Dow (1994, 2000).

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### Table 2  The Sea of Azov petroleum systems description

| PS name          | Essential elements of PS | Rock unit | Lithology            | Thickness, m | TOC, % / HI |
|------------------|--------------------------|-----------|----------------------|--------------|-------------|
| North Azov Pz (?)| Reservoir rock           | C₃        | Carbonate (limestone) | 30           |             |
|                  | Source rock              | C₁₋₂      | Terrigenous (shale)   | 50           | 3/500       |
|                  | Seal                     | C₃        | Terrigenous (shale)   | 20           |             |
| South Azov Mz (?)| Reservoir rock           | J₃        | Carbonate (limestone) | 30           |             |
|                  | Source rock              | T₃₋₁₋₂    | Terrigenous (shale)   | 50           | 2/400       |
|                  | Seal                     | K         | Carbonate (marl)      | 50           |             |
| South Azov CZ (.)| Reservoir rock           |           |                      |              |             |
|                  | Source rock              |           |                      |              |             |
|                  | Seal                     |           |                      |              |             |

### Table 3  The Black Sea petroleum systems description

| PS name          | Essential elements of PS | Rock unit | Lithology            | Thickness, m | TOC, % / HI |
|------------------|--------------------------|-----------|----------------------|--------------|-------------|
| Northeast Black Sea Mz (?) | Reservoir rock           | J₁        | Carbonate (limestone) | 30           |             |
|                  | Source rock              | J₁₋₂      | Terrigenous (shale)   | 50           | 1.5/400     |
|                  | Seal                     | K         | Carbonate (marl)      | 50           |             |
| CZ PS of Tuapse Trough (.) | Reservoir rock           |           |                      |              |             |
|                  | Source rock              |           |                      |              |             |
|                  | Seal                     |           |                      |              |             |
Fig. 4 The Sea of Azov petroleum system map, showing lateral superposition of simulated PS: 1 source rock of South Azov CZ PS (!), 2 source rock of South Azov MZ PS (?), 3 source rock of North Azov PZ PS (.), 4 geographic extent of PS, 5 oil accumulations, 6 gas accumulations, 7 PS cross-sectional position.

Fig. 5 The Sea of Azov petroleum system cross section, showing vertical superposition of simulated PS: 1 source rock, 2 geographic and stratigraphic extent of South Azov CZ PS (!), 3 geographic and stratigraphic extent of South Azov MZ PS (?), 4 geographic and stratigraphic extent of North Azov PZ PS (.), 5 fault, 6 gas accumulations, 7 oil accumulations.
Fig. 6 The Black Sea petroleum system map, showing lateral superposition of simulated PS: 1 source rock of Northeast Black Sea MZ PS (?), 2 source rock of Tuapse Trough CZ PS (!), 3 geographic extent of Northeast Black Sea MZ PS (?), 4 geographic extent of Tuapse Trough CZ PS (!), 5 gas accumulations, 6 oil accumulations, 7 PS cross-sectional position

Fig. 7 The Black Sea petroleum system cross section, showing vertical superposition of simulated PS: 1 source rock, 2 geographic and stratigraphic extent of Tuapse Trough CZ PS (!), 3 geographic and stratigraphic extent of Northeast Black Sea MZ PS (?), 4 fault, 5 gas accumulations, 6 oil accumulations
Kitchen of speculative “South Azov MZ (?)” PS, as well as originated accumulations, is located within the South Azov Step (Fig. 6). Middle Jurassic clays are the most possible source rock of the PS (Figs. 4, 5).

The geographic extent of hypothetical “North Azov PZ (?)” PS covers North Azov Depression and Azov Swell (Figs. 4, 5). The direct gas shows, detected in Triassic sediments (boreholes of Oktiabrskaya and Zapadno-Beysugskaya fields), agree with the results of basin modeling and point out the presence of PS in the transitional complex. Oil and gas accumulations that are genetically related to the PS can be expected in traps of the transitional complex within the Azov Swell and North Azov Depression. After an extensional tectonic event resulting in the accommodation of normal faults, vertical migration of hydrocarbons was enabled. It seems that hydrocarbons, produced within the transitional complex, have migrated vertically and subsequently infiltrated into the Tertiary sediments deposited along the Azov Swell. So, mainly gas accumulations that are genetically associated with the PS can be predicted also in Upper MZ and CZ sediments of Azov Swell and North Azov Depression.

Known “Tuapse Trough CZ PS (!),” with a pod of active Lower Maikopian source rock, is located within the Tuapse Trough. Oil and gas accumulations, which were produced by the PS, can be expected in the Upper Cenozoic reservoirs of the Trough. Performed biomarker analysis of oil shows from the Tuapse Trough indicates their Lower Maikopian origin (Figs. 6, 7) (Nadezhkin and Ivanov 2011).

Pod of active source rock (Middle Jurassic?) of speculative “Northeast BS MZ PS (?)” is located within the Tuapse Trough and Novorosyiysko-Lazarevski Synclinorium. Oil and gas accumulations originated from the pod located along the Shatski Ridge and Novorosyiysko-Lazarevski Synclinorium (Figs. 6, 7).

Previous interpretation of gas-geochemical data perceived organic matter destruction and subsequent migration of gas yield up to the sea bottom as the main factor that affects the spatial distribution of light hydrocarbons in the seabed sediment (Lavrenova and Kruglyakova, 2010). It was evidenced that a background level of light hydrocarbons in marine sediments generally correlated with the thickness of the sediment cover, for example, high background levels were detected above troughs.

This observation is clearly illustrated by the examples from the Sea of Azov. In particular, the Indolo-Kuban Foredipipe exhibits the highest background levels, and the Azov Swell—the lowest. In general, a similar situation is observed at the Black Sea. The Taganrog gulf (the Sea of Azov) because of anthropogenic contamination is the only exception within the studied area.

Detected anomalies of light hydrocarbons have mainly natural origins (Figs. 8, 9, 10, 11, 12, 13). The anthropogenic anomalies of methane detected within the Taganrog gulf (Fig. 8) as well as anomalies of methane and sum of C2–C4, close to Novorossiysk, were considered of technogenic origin (Fig. 11).

Complex tectonic settings of the studied basins provide favorable conditions for vertical migration of hydrocarbons and the development of strong anomalies of all light hydrocarbons (Figs. 8, 9, 10, 11, 12, 13). Thus, extended anomalies of methane, a sum of C2–C4, and a sum of C5–C6 generally located along deep faults at the Black Sea (Figs. 11, 12, 13) often exhibit a peculiar hydrocarbons composition with a significantly augmented amount of pentane and hexane. In these cases, ratio $\Sigma C_5-C_6/\Sigma C_2-C_4$ exceeded 0.5 in contrast to background levels, where such a ratio generally not exceeds 0.2 (Figs. 10, 11). Under
quiet tectonic condition, with reduced intensity of HC migration, light hydrocarbon anomalies do not arise and there is only an amplified ratio of $\sum C_5-C_6/\sum C_2-C_4$. Local hydrocarbons anomalies coincide with several anticline leads.

Involving into consideration upper mentioned results of BM&PSM provides a better understanding of the spatial distribution of light HC in seabed sediments.

It turned out that background levels of light HC in seabed sediments are not so much due to the thickness of the sedimentary cover (as was assumed prior to this investigation) as to the existence of the pod of active source rock in it (Figs. 14, 15). A possible explanation is that the source rock maturity, its generation, and expulsion rates affect gas composition: Cenozoic petroleum systems provide high background levels of $\sum C_2-C_4$ HC and Mesozoic—$\sum C_5-C_6$ HC (Table 4). Note that there is no high background level detected above North Azov PZ ("kitchen").

Favorable conditions for developing extended light HC anomalies occur in the presence of migration pathways in the overburden rock above the pod of active source rock. Strong anomalies of $\sum C_2-C_4$ and $\sum C_5-C_6$ coincide with the fault zone above the pod of Northeast BS MZ PS (?) and Tuapse Trough CZ (?) petroleum systems affirmed the statement (Figs. 12, 13).

Some small anomalies match with the location of hydrocarbon accumulations expected by the results of petroleum system modeling (Figs. 16, 17). Such a concurrence could be regarded as a sign of the possible petroleum saturation of the traps (Schumacher, 2017).

At the same time, it was ascertained that regional methane anomalies located along the deep faults did not
associate with any of the studied PS and possibly are the result of emanation from crust and mantle. The background level of methane in seabed sediments is positively correlated with the thickness of N–Q sediments of sedimentary cover (Figs. 14, 15). It seems methane is produced by the transformation of dispersed organic matter under lower temperatures during the early catagenetic process in the Neogene–Quaternary sediments. And the greater volume of the sediments causes more gas yield. The high mobility of methane provides migration to the sea bottom and a high background level of seabed sediments.
Fig. 13  Spatial distribution of sum C₅–C₆ in the Black Sea seabed sediments showing some strong enlarge anomalies located to the south from fault zone: 1 anticline, 2 fault, 3 low-contrast anomaly, 4 high-contrast anomaly and extraordinary HC concentration.

Table 4  Correspondence of petroleum systems activity and background level of light HC in seabed sediments

| Name of PS                  | Maximum petroleum generation rate, Ma | Maximum petroleum expulsion rate, Ma | Critical moment, Ma | High background level of light HC |
|-----------------------------|---------------------------------------|--------------------------------------|---------------------|----------------------------------|
| North Azov PZ (.)           | 160                                   | 65                                   | 50                  | Sum of C₅–C₆                     |
| South Azov MZ (?)           | 20                                    | 15                                   | 15                  | Sum of C₅–C₆                     |
| Northeast BS MZ PS (?)      | 30                                    | 20                                   | 20                  | Sum of C₅–C₆                     |
| South Azov CZ (!)           | 10                                    | 0                                    | 0                   | Sum of C₅–C₆                     |
| Tuapse Trough CZ (!)        | 16                                    | 5                                    | 5                   | Sum of C₅–C₆                     |

Fig. 14  Background levels of light HC in the Sea of Azov seabed sediments in respect of petroleum systems, showing the effect of active source rock presence.
Conclusions

Performed investigations proved that regional distribution of light hydrocarbons in seabed sediments could be explained by combination of petroleum systems activity and geological structure of overburden rock.

Level of petroleum system maturity governs background level of methane's homologues and gas composition in seabed sediments above the petroleum system kitchen: Cenozoic petroleum systems cause high background level of $\sum C_2$–$C_4$ and Mesozoic—$\sum C_5$–$C_6$.

Regional anomalies of light HC in seabed sediments appear above petroleum system pod where overburden rocks
are complicated by faults and local anomalies—above the petroleum accumulations.

The only distribution of methane in marine sediments is not related to the presence of petroleum systems in the sediment cover. Sizable thickness of Miocene–Pliocene deposits causes high BG level of methane in seabed sediments even in the absence of source rock. Regional anomalies of methane are result of emanation from crust and mantle.

Thus, spatial light HC distribution in seabed sediments reflects geological structure of sedimentary basins and, also, maturity of petroleum systems located within the basins.

Joint interpretation of gas-geochemical data and petroleum system modeling facilitate more effective marine geochemical prospecting and verification of petroleum system models.

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**Code availability** Not applicable.

**Declaration**

**Conflict of interest** On behalf of all the co-authors, the corresponding author states that there is no conflict of interest.

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