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OCEANOLOGY

Geochemical Anomalies of Hydrocarbon Gases in Bottom Sediments of Geostructures of the Laptev–Siberian Sea Transition Zone of the East Arctic Shelf

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Abstract—The data of gas-geochemical studies of bottom sediments of the Laptev—Siberian Sea transition zone are presented. The established isotope–geochemical parameters indicate the domination of epigenetic hydrocarbon gases of various gas sources in the sediments. Methane concentrations up to 8.3047 cm^3/kg and the sum of homologues up to 0.0260 cm^3/kg indicate the formation of hydrocarbon anomalies exceeding the anomaly criteria for Arctic shelf sediments by a factor of 166 and 26. The main geological factors for the formation and distribution of anomalies in the bottom sediments of the Laptev–Siberian Sea transition zone are the high gas content of the underlying sediments and gas sources, the fold and fault tectonics, the geostructural position, the seismic activity, and the depth of occurrence of hydrocarbon gas sources.

Keywords: bottom sediments, shelf, hydrocarbon gases, gas saturation, anomalies, geological factors, Laptev–Siberian Sea transition zone

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The Laptev–Siberian Sea Transition Zone (LSZ) is currently the subject of close attention from scientists worldwide and the Russian scientific community. Interest in the LSZ is caused by the formation of anomalies of hydrocarbon gases in the bottom sediments at the junction of the Eurasian and North American lithospheric plates [1, 2]. The main goal of this research was to study the influence of key geological factors of the formation and distribution of hydrocarbon anomalies in sediments. The relevance of this work is determined by the priority of scientific research in the Arctic zone of Russia. The LSZ territory is located within the Laptev Sea and the East Siberian Sea transition zone, including the junction of the Euro-Asian and North American lithospheric plates, where the global faulting of the Arctic segment of the Earth is marked by the Laptev Sea graben–rift system, basalt magmatism, and chains of epicenters of the earthquakes with magnitudes up to 7 at depths of 4–36 km (Fig. 1).

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Among the structures characterizing the faulting of the Earth's crust, the Kotelnicheskii rigid block, which is not subject to extension, is distinguished. In the overall tectonic structure of the region, there are the East Laptev Rise, the Stolbovskii and Belkovskii horsts, the Blagoveshchenskaya and Shelonskaya structural terraces, and the Novosibirsk, Anisinskii, Belkovsko-Svyatonosskii, and Omoloi troughs, and also the North Omoloi graben–rift and the Sannikov saddle (Fig. 1), all having been formed within the Verkhoyansk–Kolyma and Novosibirsk–Chukotka folded areas. The LSZ has developed Paleozoic igneous rocks of the Kotelnicheskoe and Reshetnikovskoe rises, Early Mesozoic ones of the South Anyui suture, and Cretaceous ones from the submeridional tectonomagmatic activation zone, as well as Paleozoic–Mesozoic bitumen occurrences and Cretaceous–Neogene coal deposits in the New Siberian Islands [3].

The main material for gas geochemical studies was sediment cores from 55 bottom stations, collected using gravity corers, bottom samplers, and multicorers on the R/V *Akademik M.A. Lavrentyev* and hydrographic and support vessels of the Russian Ministry of the Marine Fleet. The method of interval sampling of sediment cores into sealed containers, followed by degassing and gas analysis was used during the works. The technique of degassing, chromatographic analysis, and determination of gas saturation in bottom sed-

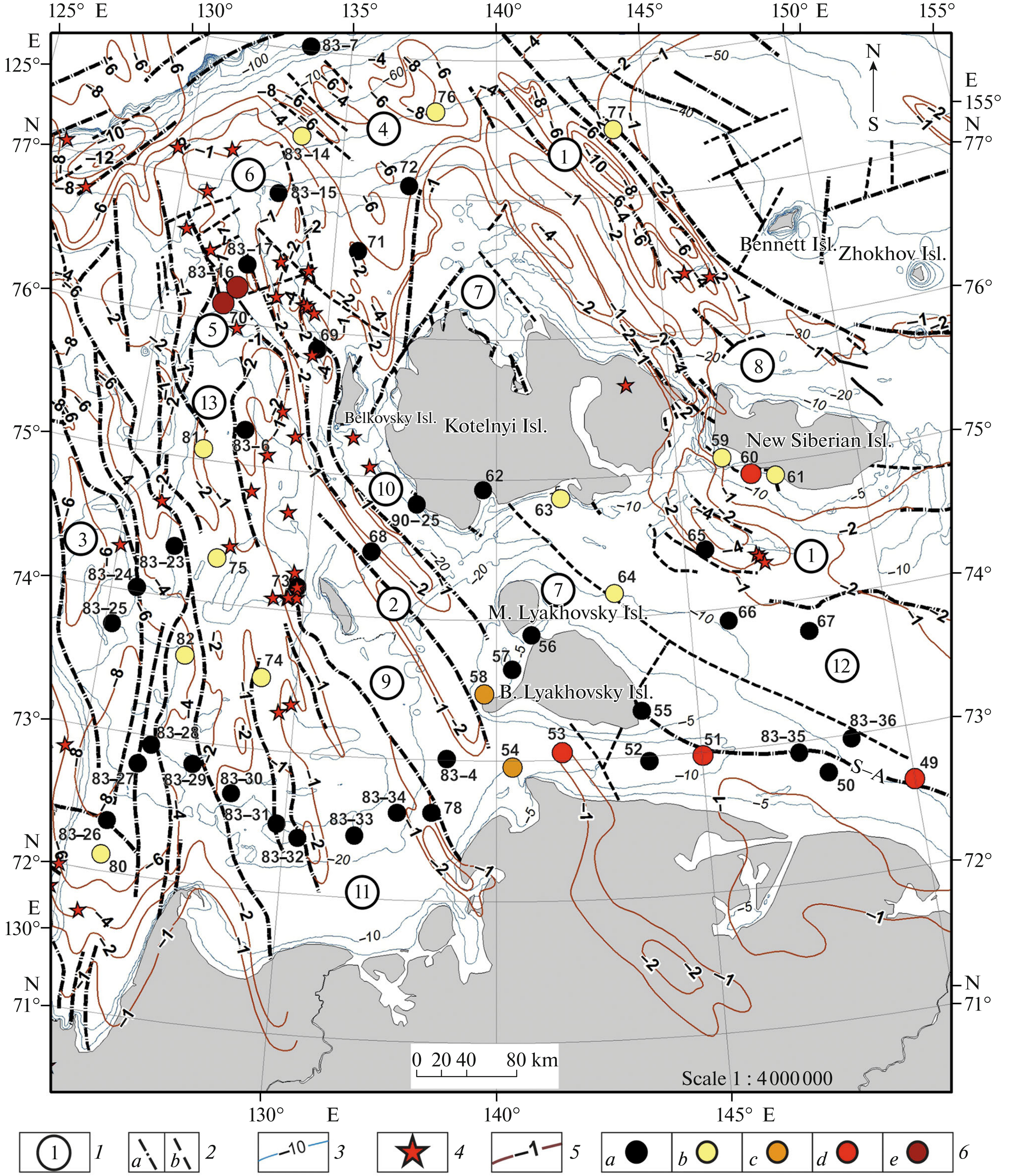

Fig. 1. Structural-tectonic map of the study area [3, 4]: *1*, First-order geostructures: troughs: (1) Novosibirsk, (2) Belkovsko-Svyatonosskii, (3) Omoloi, (4) Anisinskii; graben-rift: (5) North Omoloi; rises: (6) East Laptev, (7) Kotelnicheskoe, (8) Reshetnikovskoe; horsts: (9) Stolbovskii, (10) Belkovskii; structural terraces: (11) Shelonskaya, (12) Blagoveshchenskaya; (13) Sannikov Saddle. *2*, Tectonic faults: (a) confirmed, (b) assumed. *3*, Isobaths, m. *4*, Earthquake epicenters (data from the USGS Earthquake Hazards Program). *5*, Isohypses of sedimentary cover thickness, km. *6*, Bottom stations with concentrations of hydrocarbon gases in sediments: (a) <0.05, (b) 0.05–0.25, (c) 0.25–0.50, (d) 0.50–1.00, and (e) >1.00 cm^3/kg. Numbers of stations for coastal expeditions and shipboard sampling range: 49–82; for cruises LV83 and LV90 of R/V *Akademik M.A. Lavrentyev*: 83-4–83-36 and 90-25. SA, South Anyui Suture.

iments complied with the Guidelines [5]. $\delta^{13}C$-CH_4 isotopic analysis of carbon was conducted at the Stable Isotope Laboratory of the Far East Geological Institute, Far East Branch, Russian Academy of Sciences. To determine the genesis of hydrocarbon gases, in addition to the $\delta^{13}C$-CH_4 data, we used a comprehensive method of calculating the geochemical parameters, such as the molecular mass of the hydrocarbon fraction (MM_{HC}), weight fractions of C_1–C_5 normalized to MM_{HC} in parts per thousand (or in grams per kilogram of gas in the hydrocarbon fraction) [6], and the coefficient of the wet part of the hydrocarbon fraction [7], expressed as $K_{wet} = (\Sigma C_2–C_5/\Sigma C_1–C_5) \times 100$ (%), where C_1–C_5 are the weight fractions of hydrocarbons in parts per thousand.

RESULTS OF GAS-GEOCHEMICAL STUDIES

The interpretation of the results of gas sampling in bottom sediments and the carbon isotope analyses of methane, along with the data from previous works [8–10], allowed us to identify eleven genetic groups of hydrocarbon gases of various lithotypes and gas sources in the LSZ sediments; each group was characterized by unique average gas-geochemical parameters of MM_{HC} and K_{wet} and $\delta^{13}C$-CH_4. Based on the data obtained (Table 1), the syngenetic hydrocarbon gases in modern sediments are characterized by a biochemical origin. The migration (epigenetic) gases from hydrates, Cenozoic gas accumulations, peat bogs, and brown coals are dominated by biogenic gases with inclusion of a metamorphogenic component. The gas-geochemical parameters of hydrocarbon gases in bituminous coals, potential Mesozoic solid bitumens (anthraxolites?), gas, condensate-gas, gas-condensate, oil-gas, and gas-oil accumulations and deposits, imply their metamorphogenic origin and magmatogenic origin for igneous rocks. In general, the gas-geochemical parameters of all groups of hydrocarbon gases are quite similar in value to their counterparts from the East Siberian Sea [9, 10], as well as from the Lena, Anadyr, Sea of Okhotsk, and Sakhalin coal-oil-and-gas-bearing sedimentary basins in Eastern Russia [8].

During research, it was established that the composition and concentrations of hydrocarbon gases are formed in LSZ bottom sediments according to the rules of additivity; i.e., hydrocarbon gases of different origin accumulate sequentially in sediments, with the dominating gaseous phase and isotope-gas-geochemical parameters of a more hydrocarbon-saturated source rock. The maximum average concentrations of CH_4 are recorded in the sediments from the areas where hydrocarbon gases are formed in brown coals, bituminous coals, and gas hydrates (Table 1), while the lowest ones are detected in igneous rocks. Average concentrations characterize other gas sources. The maximum average concentrations of ΣC_2–C_5 were found in the sediments from the areas where hydrocarbon gases are formed in potential gas-condensate, oil-gas, and gas-oil deposits; the lowest ones are found in modern sediments, igneous rocks, and Cenozoic gas accumulations. The average concentrations were obtained in other lithotypes and gas sources. We note a two-time excess of average concentrations of CH_4 and ΣC_2–C_5 in the sediments from the areas where bituminous and brown coals are developed (Table 1), which clearly indicates that the degree of coal (organic) matter catagenesis has a significant impact on the processes of hydrocarbon saturation in bottom sediments.

As a result of gas-geochemical studies of LSZ bottom sediments, it was found that the concentrations of CH_4 and ΣC_2–C_5 increase predictably in the sediments with the sampling depth increasing from 0.1 to 3.3 m in the range from 0.0017 and 0.00001 cm^3/kg to 8.3047 and 0.02593 cm^3/kg, with average values of 0.1700 and 0.00220 cm^3/kg, respectively. Based on the anomaly criteria of $CH_4 > 0.05$ and ΣC_2–$C_5 > 0.001$ cm^3/kg for sediments in the East Arctic Shelf [2], their maximum concentrations exceed these values by a factor of 166 and 26. The average concentrations exceed them by a factor of 3.4 and 2.2, which indicates the high hydrocarbon saturation of bottom sediments in the study area. In this respect, the influence of the structural-tectonic framework of the LSZ is one of the key factors in the formation and distribution of hydrocarbon anomalies.

Our study has shown that the maximum average concentrations of CH_4 were observed in the bottom sediments of the North Omoloi graben-rift and rises, while ΣC_2–C_5 are the highest in the Sannikov saddle and troughs. The lowest values of both were recorded in the horsts (Table 2). The sediments of the structural terraces occupy an intermediate position. The formation of the CH_4 and ΣC_2–C_5 anomalies, with concentrations up to 2.3056 and 0.02313 cm^3/kg, respectively, occurred in the sediments of the limbs of the troughs and the graben-rift, up to 8.3047 and 0.02593 cm^3/kg, respectively, in their axial part (trough bend), and up to 0.3620 and 0.01849 cm^3/kg, respectively, in the closures of synclinal folds. Similar values in the sediments of the western wing of the Kotelnicheskii Rise reach 0.6900 and 0.00899 cm^3/kg, while in the crest part, they are 0.5378 and 0.00647 cm^3/kg. The formation of methane anomalies of 0.0553–0.1494 cm^3/kg and ΣC_2–C_5 anomalies of 0.0086–0.0102 cm^3/kg in the bottom sediments of the Sannikov saddle was observed in its central part. The low concentrations of CH_4 and ΣC_2–C_5 and the absence of their anomalies in the sediments of the horsts indicate the dominance of degassing processes in these structures. A similar gas dynamic situation occurred in the sediments of the East Laptev Rise, where one local methane anomaly was identified (Fig. 1). The formation of the CH_4 and ΣC_2–C_5 anomalies in the range of 0.0507–0.6723 and

Table 1. Average values of the gas-geochemical parameters and concentrations of hydrocarbon gases in bottom sediments in the study area in the segments of development of different gas sources

Genetic groups of hydrocarbon gases (station numbers)	Average values of parameters			CH_4, cm^3/kg			$\Sigma C_2–C_5$, cm^3/kg		
	MMHC, g/mol	K_{wet}, %	δ^{13}C-CH_4, ‰	min	max	avg	min	max	avg
1. Modern sediments (57, 62, 64)	16.05	0.1	–78.0	0.0180	**0.0654**	0.0347	0.00003	0.00004	0.00003
2. Gas hydrates (70, 83-7, 83-14, 83-16)	16.10	0.7	ND	0.0059	**8.3047**	**2.4143**	0.00003	**0.00682**	**0.00237**
3. Peat bogs (55, 59)	16.13	1.1	–62.0	0.0305	**0.1058**	**0.0681**	0.00024	**0.00139**	0.00081
4. Coal fields:									
4.1. of brown coals (58, 60, 63, 66, 80, 83-28)	16.20	1.9	–60.7	0.0029	**0.6275**	**0.2068**	0.00004	**0.00644**	**0.00257**
4.2. of bituminous coals (49, 51, 53, 83-34	16.27	2.6	–58.0	0.0018	**0.6900**	**0.4755**	0.00004	**0.00899**	**0.00594**
5. Gas deposits:									
5.1. Cenozoic (61, 78)	16.15	1.4	ND	0.0117	**0.0637**	0.0377	0.00010	0.00052	0.00026
5.2. Mesozoic * (74, 83-32)	16.41	3.1	–56.0	0.0097	**0.0507**	0.0302	0.00028	**0.00115**	0.00071
6. Igneous rocks * (52, 56, 71, 83-15, 83-17, 83-24, 83-30, 90-25)	16.65	6.5	ND	0.0037	0.0210	0.0072	0.00018	0.00060	0.00026
7. Hard bitumen* (50, 54, 69, 73, 83-6, 83-23, 83-35, 83-36)	17.64	16.7	–49.8	0.0030	**0.3620**	**0.0598**	0.00027	**0.00424**	**0.00137**
8. Condensate-gas deposits * (67, 79, 81, 83-4, 83-25, 83-26, 83-29, 83-31)	17.12	12.0	–53.0	0.0065	**0.1549**	**0.0580**	0.00043	**0.01019**	**0.00379**
9. Gas-condensate deposits * (68, 75, 82)	18.28	23.4	–50.4	0.0243	**0.1524**	**0.0773**	**0.00375**	**0.02313**	**0.01165**
10. Oil-and-gas deposits* (65, 77)	19.43	31.8	–48.2	0.0083	**0.0923**	**0.0503**	**0.00190**	**0.01849**	**0.01020**
11. Gas-and-oil deposits* (72, 76)	21.60	47.2	–44.8	0.0358	**0.0570**	0.0464	**0.01396**	**0.02593**	**0.01995**

The anomalous concentrations of hydrocarbon gases are given in bold; ND means not detected. * Potential deposits.

0.00115–0.00825 cm^3/kg, respectively (Table 2), in the sediments of the Shelonskaya and Blagoveshchenskaya terraces is associated with the migration of hydrocarbon gases in the fault zones and their apophyses that transit coal-and-gas bearing formations.

The structural-formational features of the deposits suggest that the longitudinal basement folds were active during the Paleozoic, while the transverse and diagonal folds developed during the Cretaceous–Cenozoic. The displacement along the faults of Cenozoic deposits, geomorphological data, modern seismic activity in the LSZ, and gas-geochemical parameters of epigenetic hydrocarbon gases point to ongoing movements along these faults and their active role in the gas migration processes to the bottom sediments until recently. At the same time, the faulting intensity in the western part of the LSZ (the conventional boundary along the 140°E meridian) is much higher than in the eastern part (Fig. 1), with an average hydrocarbon saturation ($\Sigma C_1–C_5$) of sediments equal to 0.2030 cm^3/kg in the first part and 0.1307 cm^3/kg in the second part. We note that, in the linear seismically active zone represented by a chain of earthquake epicenters in the western part of the LSZ, the bottom sediments exhibited anomalies of CH_4 up to 8.3047 cm^3/kg and anomalies of $\Sigma C_2–C_5$ up to 0.01966 cm^3/kg.

The general distribution of hydrocarbon anomalies in LSZ bottom sediments at the gas geochemical sampling stations is depicted in Fig. 1.

Table 2. Average concentrations of hydrocarbon gases in bottom sediments of the geostructures in the study area

Geostructures	CH_4, cm^3/kg			ΣC_2-C_5, cm^3/kg		
	min	max	avg	min	max	Avg
Structural terraces (19):	0.0017	**0.6723**	**0.0500**	0.00001	**0.00825**	0.00088
Blagoveshchenskaya (8)	0.0037	**0.6723**	**0.0987**	0.00001	**0.00825**	**0.00165**
Shelonskaya (11)	0.0017	**0.0507**	0.0146	0.00009	**0.00115**	0.00033
Rises (30):	0.0022	**0.6900**	**0.0963**	0.00001	**0.0899**	**0.00119**
Reshetnikovskoe (3)	**0.0637**	**0.6275**	**0.2657**	0.00005	**0.00644**	**0.00279**
Kotelnicheskoe (14)	0.0037	**0.6900**	**0.1402**	0.00001	**0.00899**	**0.00183**
East Laptev (13)	0.0022	**0.0659**	0.0098	0.00004	0.00036	0.00014
Horsts (4):	0.0025	0.0083	0.0065	0.00008	0.00062	0.00037
Stolbovskii (1)	0.0070	0.0070	0.0070	0.00023	0.00023	0.00023
Belkovskii (3)	0.0025	0.0083	0.0063	0.00008	0.00062	0.00042
Troughs (34):	0.0029	**0.3620**	0.0391	0.00004	**0.02313**	**0.00326**
Novosibirsk (7)	0.0050	**0.1058**	0.0349	0.00006	**0.01849**	**0.00320**
Anisinskii (8)	0.0054	**0.0570**	0.0242	0,00066	**0.02593**	**0,00389**
Belkovsko-Svyatonosskii (9)	0.0068	**0.3620**	**0.0552**	0.00016	**0.00424**	**0.00194**
Omoloi (10)	0.0029	**0.1524**	0.0394	0.00004	**0.02313**	**0.00408**
North Omoloi graben-rift (14)	0.0022	**8.3047**	**0.9677**	0.00007	**0.01966**	**0.00307**
Sannikov saddle (6)	0.0068	**0.1549**	**0.0641**	0.00005	**0.01019**	**0.00486**
Study area (107)	0.0017	**8.3047**	**0.1700**	0.00001	**0.02593**	**0.00219**

The anomalous concentrations of hydrocarbon gases are given in bold; the number of calculations is in brackets.

Based on the data obtained, the main geological factors that have an effect on the formation and distribution of anomalies of hydrocarbon gases in the LSZ bottom sediments are the occurrence depth, high gas content of underlying deposits and gas saturation of source rocks, the degree of catagenesis of coal (organic) matter, the seismic activity in the study area, the geostructural position, and the tectonic factor.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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