The Distribution and Genesis of Hydrocarbon Gases in the Bottom Sediments of the Laptev–Siberian Sea Zone of the East Arctic Shelf

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Abstract—Gases from bottom sediments of the Laptev—Siberian Sea zone contain methane (up to 8.3047 cm³/kg) and its homologues (up to pentane including) totaling 0.0259 cm³/kg. The established values of the carbon isotope composition of methane (δ^{13} C), the molecular weight of the hydrocarbon fraction, the weight concentrations of C₁—C₅ hydrocarbons and their ratios, the "wetness" and "transformation" coefficients of the hydrocarbons fraction indicate the presence of various syngenetic and epigenetic gases in different source rocks: bottom sediments, peatlands, coal, gas, and gas hydrate? deposits, solid bitumen, igneous rocks, and inferred oil and gas reservoirs. It has been established that the vertical and lateral distribution of hydrocarbon gases in sediments depends on the influence of a complex of geological factors: fold and fault tectonics, magmatism and seismic activity in the region, coal, oil and gas potential and bitumen content, organic saturation and lithological composition, hydrogeological, geocryological, and other conditions for accumulation of gases or their degassing.

Keywords: bottom sediments, hydrocarbon gases, genesis, gas saturation, anomalies, geological factors, oil and gas content, Laptev–Siberian Sea zone

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INTRODUCTION

The Laptev–Siberian sea zone (LSZ) is the transboundary zone between the Laptev Sea (LS) and the East Siberian Sea (ESS), which includes the New Siberian Islands and shelves of the indicated seas (Fig. 1). The first geological data on the New Siberian Islands were obtained during expeditions by M.I. Gedenshtrom, P.F. Anjou, J. De Longa, A.A. Bunge, E.V. Tolle, K.A. Vollosovich, B.A. Vilkitsky, M.V. Ermolaev, N.V. Pinegin, and other researchers (at the end of 19th to the beginning of 20th century. Starting from 1950s, a geological survey was carried out in this region to compile the State Geological Map of the Russian Federation on a scale 1 : 1000000 (1961, 1999, 2016), which was accompanied by scientific geological studies of NIIGA, VNIIOkeanologiya, GIN RAS, and other institutes and organizations. During 1985-2022, geologists from the JSC Sevmorgeologiya, Laboratory of Regional Geodynamics (LARGE), Trust Sevmorneftegeofizika, and the Federal Institute for Geosciences and Natural Resources (BGR, Germany), JSC Sevmorgeologiya, and MAGE carried out seismic studies in the LSZ using correlation refraction (CRM) and common depth point seismic reflection (CDP SRM) methods.

The first data on the gas composition and gas saturation of the LS and LSZ bottom sediments were obtained by NIIGA during 1975-1981. At the same time, VNIGRI, VSEGEI, and JSC Sevmorgeologiya studied the gas composition of rocks and Quaternary sediments, gas phase of groundwaters, and gas seeps based on borehole sampling of the New Siberian Islands and adjacent shelf. In addition to the production works, the considered area was spanned by the multidisciplinary gas geochemical studies of the JSC Sevmorgeologiya, MAGE, VNIIOkeanologiya, VSEGEI, VNIGRI, GIN RAS, MSU, INGG, IPNG RAS, POI FEB RAS, as well as joint Russian, Swedish, Norwegian, German, and Chinese studies.

At present, the bottom sediments in diverse subaquatic sedimentary basins worldwide have been unevenly covered by gas geochemical studies. One of the main present-day requirements on gas geochemical studies of bottom sediments is an increase of accuracy and efficiency of prediction of gas sources, as well as the development of reliable criteria for separation between technogenic, biogenic, and thermogenic (catagenetic) sources. The correlation of the distribution of gas-saturated bottom sediments with the geological structure of sedimentary sequence aimed at distinguishing the areas of accumulation and discharge of deep-seated fluids is also an important ques-



Fig. 1. A geostructural map of the studied area [3-5] and lithological-stratigraphic column of the New Siberian Islands [12]. (*1*) geostructures of the first order: troughs (sedimentary basins): (1) New Siberian, (2) Belkovsky–Svyatoi Nos, (3) Omoloi, (4) Anisinsky, (5) North Laptev, (6) Tastakh, (7) Primorsky, (8) North Omoloi graben rift; rises: (9) Eastern Laptev, (10) Kotelnichesky, (11) Reshetnikovskoe; horsts: (12) Stolbvskoy, (13) Belkovsky; structural terraces: (14) Shelonskaya, (15) Blagoveshchenskaya, (16) Sannikovskaya saddle; (S–A) South Anyui suture; (*2*) faults: proved (*a*), inferred (*b*); (*3*) earthquake epicenters; (*4*) coal deposits: hard coal (*a*), brown coal (*b*); (*5*) coal manifestations (*a*), bitumen manifestations (*b*); (*6*) gas sampling boreholes, (*7*) oceanographic stations and their numbers: sampling depth of bottom sediments: <1 m (*a*), >1 m (*b*); (*8*) isobaths, m; (*9*) isobypses of heterogeneous basement surface, km; (*10*) river paleovalleys.



Fig. 1. (Contd.)

tion. The experience of our works highlights the need for a complex approach to choosing the criteria for source determination. Gas geochemical fields in bottom sediments usually have a complex polygenetic composition, Correspondingly, the assessment of genesis using only one gas component or one parameter may lead to incorrect conclusions.

Thus, the main aim of this work consists in the generalization, systematization, and comparative analysis of gas geochemical and geological data in order to provide insight into the formation and distribution of hydrocarbon gases (HCGs) of the LSZ sediments within the junction zone of the Eurasian and North American lithospheric plates. The relevance of this work is caused by the application of author's approach to studying HCGs genesis, as well as by the priority of prospecting and scientific-applied studies in the Arctic zone, which facilitate the development of the rawmineral base of the Russian Federation.

METHODS AND MATERIALS

The main material for our study was bottom sediments of 55 oceanographic stations (Fig. 1) collected by hydrostatic and inflow ground corer with liners with core length up to 3.25 m, bottom grabs, and multicorers up to 0.60 m from the "R/V *Akademik M.A. Lavrentiev*" (cruises LV83 and LV90), hydrographic, and other ships of the Far East shipping line, as well as coastal expeditions of different organizations. In total, we sampled 26 sediment cores over 1-m long, 14 short tubes less than 1 m, and 22 bottom grabs and multicorers.

During our studies, we performed interval sampling of sediment cores from reference horizons (50, 100, 150 cm, etc.) or from the lowest horizon of sampler. In total, 110 sealed vials were collected with subsequent degassing and gas sampling with free or thermal vacuum degassing (58 and 110 samples, respectively). Degassing and chromatographic analysis of 168 gas samples were carried out at the Laboratory of Gas Geochemistry certified by RosStandart at the Pacific Oceanological Institute of the Far Eastern Branch, Russian Academy of Sciences on a Khromatek-Gazochrom 2000 and KristalLyuks-4000M chromatographs onboard vessels and at stationary conditions. The technique of degassing, chromatographic analysis, and the determination of gas saturation of sediments corresponded to the operating normative methodical manual^{1, 2, 3}.

Materials of borehole gas sampling of lithotype cores, samples of free gas and gas phase of groundwaters were taken to study the gas composition of rocks and Quarternary sediments, gas seeps, and gas phase of groundwaters [4, 9, 14]. The established values of geochemical parameters of HCGs of lithotypes, reservoirs of free and dissolved gas [6, 7, 9] were applied to identify the gas sources. Mass spectrometric isotope ratios (IRMS) δ^{13} C–CH₄ determined at the Stable Isotope Laboratories of VSEGEI and FEGI FEB RAS, as well as data from [4, 8, 9, 19] were used to determine the HCGs genesis in bottom sediments. The contents of organic carbon (Corg) were determined by IR-detection on a TOC-V (Shimadzu) analyzer at the certified Laboratory of Analytical Chemistry of the FEGI FEB RAS.

In order to determine the origin of HCGs in sediments, the isotope studies were supplemented by a complex method of calculation of quantitative gas geochemical parameters such as molecular weight of HC fraction (MM_{HC}) , weight concentrations of HCGs (C_1 - C_5) normalized to MM_{HC} in a weight fraction of hydrocarbons per 1000 (or in grams per kilogram of HC fraction gas) [1] and their ratios: coefficients of the "transformation" (Ktr) [2] and coefficient of wetness (Kwet) [16]. The Ktr and Kwet coefficients are represented by the following formulas: Ktr = $(C_2 \times C_4)/C_3$ and Kwet = $\Sigma C_2 - C_5/\Sigma C_1 - C_5 \times C_5/\Sigma C_1 - C_5/\Sigma C_2 - C_5/\Sigma C_1 - C_5/\Sigma C_5/\Sigma C_1 - C_5/\Sigma C_5/\Sigma C_5/\Sigma C_5$ 100 (%), where $C_1 - C_5$ are the weight concentrations of HC in a weight fraction of hydrocarbons per 1000. It was established that Kwet values are tightly related to the age of HCGs reservoir and are indicators of the residence time of gas in a trap [2, 6, 7]; Kwet is indicative of enrichment of HC fraction in the total methane homologues [8, 9].

Digitizing and spatial-numerical interpretation of results were carried out in an ESRI®ArcGIS complex using Geostatical Analyst module by a method of an inverse distance weighted interpolation (IDW).

GEOLOGICAL CHARACTERISTICS

Due to the absence of deep boreholes, the geological characteristics of the LSZ is based on the materials of geological surveys and CDP SRM seismic survey performed by the JSC Sevmorgeologiya, MAGE, in combination with data on drilling small boreholes, gravity corers in the upper part of the section.

Stratigraphy and Lithology

The Paleozoic deposits of the LSZ contain Cambrian-Silurian, Devonian-Carboniferous, Middle Carboniferous-Permian carbonate and terrigenouscarbonate complexes identified in the exposures on the Bennet, Kotelnyi, and Belkovsky islands. Mesozoic rocks are Triassic-Middle Jurassic terrigenous-carbonate and Upper Jurassic-Barremian terrigenous

¹ Manual on the Determination and Prediction of the Gas Potential of Host Rocks During Prospecting Works (VNIIGRIugol', Rostov on-Don, 1985), GOST 23781-87

² Natural Fuel Gases. Chromatographic Method for Determination of Component Composition (GosStandart, Moscow, 1987)), and 31371.3-2009

³ Natural Gas. The Determination of Composition by Gas Chromatography with Uncertainty Assessment (Standartinform, Moscow, 2009

complexes. The last complex corresponds to the active rifting phase and has a fault-controlled stepwise change of thickness of sedimentary and volcanogenic rocks. *The Cretaceous* complex is made up of sedimentary and volcanogenic rocks recovered by boreholes in the New Siberian Islands and water part of the studied area [3, 4]. The lithological composition of Paleozoic–Mesozoic complexes is shown in Fig. 1.

The sedimentary cover of the LSZ shelf includes the Aptian–Upper Cretaceous, Paleogene, and Neogene rocks and Quaternary sediments forming the upper (basin) complex in the studied area. According to the interpretation of geological-geophysical works in the southern part of the LSZ, three seismic complexes are distinguished (Fig. 2): the Lower-Upper Cretaceous complex between reflectors (R) A and L2, Paleocene-Middle Miocene complex between L2 and L4, and Middle Miocene-Pleistocene complex between L4 and L6 (seafloor). The deposits of the complex (based on drilling works on shelf) are weathering crust, pebble stones, sands, silts, clays, lenses and beds of brown coal and peat, gravelstones, sands, silts and ooze [3, 4, 9]. The Quaternary sediments of the studied area mainly include terrigenous rocks consisting of Holocene-Pleistocene sands, ooze, silts, and pebbles [3, 4, 10].

Tectonics and Magmatism

The considered area is confined to the LS–ESS transboundary zone, including the junction of the Eurasian and North American lithospheric plates. A global pull-apart of the Arctic segment is expressed by the Laptev graben-rift system, basaltic magmatism, and chains of earthquake epicenters up to magnitude 7 at depths of 4-36 km. Structures characterizing the crustal pull-apart include Kotelnichesky and Lyakhovsky-Svyatonossky rigid blocks (Kotelnichesky median massif) [3, 4]. The tectonic structure of the studied area also includes the Reshetnikovskoe and Eastern Laptev rises, Stolbovsky and Belkovsky horsts, as well as Belkovsky-Svyatonossky, New Siberian, Omoloi, Anisinsky, Tastakh, and Primorskii trough, North Omoloi graben rift, Sannikovskaya saddle, Blagoveshchensk and Shelon structural terraces formed within the Verkhoyansk-Kolyma and New Siberian-Chukotka fold areas. From the south, the latter is bounded by the South Anyui suture, which is the northwestern termination of the Rauchan-Oloi fold-and-thrust system. The LSZ area is a part of the epi-Late Kimmerian platform, which comprises the Late Cretaceous-Cenozoic plate cover ubiquitously developed over sea basin areas.

An important feature of the LSZ is intense deformations of folded basement and plate cover (Figs. 1, 2). Based on relationships with folding, the longitudinal, oblique, and transverse faults are distinguished. The longitudinal faults, judging from structural-facies zoning of Paleozoic deposits of the same strike, are synsedimentary faults initiated in the Paleozoic. The

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diagonal and transverse faults were formed in the Cretaceous–Cenozoic. The geomorphological, gas-geochemical, and seismic data indicate that the movement along all faults continues at present.

Igneous rocks of the area vary from ultrabasites to granites. The distribution of intrusive rocks show clear age and structural-tectonic control. The Middle-Late Paleozoic dikes and sills of dolerites and gabbro-dolerites are confined to the Kotelnichesky rise. The Early Mesozoic (?) ultrabasites are typical of the South Anyui suture zone. The Early Cretaceous granitoids form a transverse series related to the submeridional zone of tectonomagmatic activation (TMA). The Late Cretaceous magmatism consists of basic dikes [3, 4, 13, 17].

Organic saturation of lithotypes of the studied area is sufficiently high. Cretaceous–Cenozoic deposits contain carbonaceous siltstones and mudstones, beds of hard and brown coals, lignites, and peat, while Paleozoic–Mesozoic sediments include solid bitumens, carbonaceous shales, siltstones, and mudstones.

The bitumen occurrences of the Kotelnyi and Belkovsky islands (Fig. 1) range in age from Ordovician to Cretaceous. Bitumens were generated in several phases and represent a product of complex transformation under the joint influence of metamorphogenic and supergene factors. They include bitumens of different classes: from maltha to anthraxolite formed through metamorphic transformations to asphalthites and huminokerite produced under the effect of supergene factors. The studied area contains mainly resinous bitumens with oil content of ~50% and asphalthes.

From two to nine Lower Cretaceous 1-25-m thick coal beds of catagenetic stage MC₁ were found at the Kotelny and Bennet islands. Up to ten 1-12-m thick beds of brown coal of catagenetic stage PC₁₂ were found in the Upper Cretaceous, Eocene, Oligocene, and Miocene deposits of the Kotelnyi, Fadeev, and New Siberia islands. At the Great Lyakhovsky, Tastakh, and Primorsky coal-bearing areas, a few tens of interbeds and beds of brown coal 0.1-2.4-m thick were found. The upper parts of the section contain lenses and beds of peat up to 2-m thick, as well as Miocene–Quaternary clays with C_{org} 9–12%. In addition, the studied Permian–Triassic, Jurassic–Cretaceous, and Paleogene–Neogene lithotypes contain carbonaceous shales, mudstones, and clays with C_{org} – 19– 42% and thickness from 1 to 38 m [3, 4].

Gas Potential

The studied area contains gas seeps and gas emanations * from boreholes 1g, 2g, 6*, 13, 14, 41, and 87* (Fig. 1) at depths of 36-112 m with contents of methane and its total homologues of 10.24-94.72 and 0.00001-0.76%, respectively. Based on gas sampling





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Group, lithotype, gas source	δ ¹³ C-CH ₄ ,	Weight fraction of hydrocarbons per 1000				MM _{HC} ,	Coefficients		
(numbers of bottom stations)	%0	C ₁	C ₂	C ₃	C ₄	C ₅	g/mol	Ktr	Kwet, %
1. Modern sediments (57, 62, 64)	-74.5	997	2	tr.	0	0	16.05	0.3	0.2
2. Gas hydrates (70, 83-7, 83-14, 83-16)	n.a.	993	4	2	tr.	0	16.09	0.9	0.7
3. Peat lands (55, 59)	-69.8	991	7	1	tr.	0	16.13	2.0	0.9
4. Coal deposits									
4.1. Brown coals (58, 60, 63, 66, 80, 83-28)	-61.0	980	16	3	1	0	16.20	4.7	2.0
4.2. Stone coals (49, 51, 53, 83-34)	-58.6	973	19	6	2	tr.	16.27	8.1	2.7
5. Gas reservoir	n.a.	987	11	2	tr.	0	16.15	2.5	1.4
5.1. Cenozoic (61, 78)									
5.2. Mesozoic* (74, 83-30, 83-32)	-56.0	954	34	10	2	1	16.42	9.4	4.6
6. Igneous rocks (52, 56, 71, 83-15, 83-17, 83-24, 90-25)	n.a.	927	54	15	4	0	16.68	16	7.3
7. Condensate gas reservoirs* (67, 79, 81, 83-4, 83-25, 83-26, 83-29, 83-31)	-53.0	875	81	40	4	0	17.15	28	12.5
8. Solid bitumens (50, 54, 69, 73, 83-6, 83-23, 83-35, 83-36)	-49.8	840	94	41	20	5	17.61	46	16.0
9. Condensate reservoirs*(68, 75, 82)	-50.4	766	155	52	24	3	18.29	63	23.4
10. Petroleum reservoirs*(65, 77)	n.a.	682	180	71	61	5	19.43	153	31.8
11. Gas-oil reservoirs* (72, 76)	-44.8	528	287	84	89	11	21.60	304	47.2

Table 1. The average isotope-geochemical parameters of genetic groups of HCGs in bottom sediments of the studied area

* Potential reservoirs, (tr) traces, (n.a.) not analyzed.

data on the core of boreholes 1, 5, 12, 18, 25, 27–30, 32, and 80, the CH₄ and ΣC_2-C_5 concentrations (in cm³/kg) reach 1.89 and 0.002 in the Quaternary sediments, 2.81 and 0.032 in volcanic rocks, 5.56 and 0.045 in peat lands, 177.30 and 2.140 in lignites, 780.72 and 8.650 in brown coals, and 2000.84 and 23.361 cm³/kg in hard coals within depth interval from the surface to 10–124 m. The contents of methane and ΣC_2-C_5 in the gas phase of groundwaters in boreholes 41, 42, 43, 63, 77, and 78 are 0.04–51.70 and 0.00001–0.010%, respectively, at total gas saturation of waters of 24–70 cm³/L [3, 4, 8, 9].

The formation and distribution of HCGs in sediments of the LSZ are determined by the influence of *hydrogeocryological conditions* of the Yana–Indigirka cryogenic artesian basin, the Kotelnchesky–Lyakhovsky hydrogeological area, and artesian basins of the LS and ESS, whose hydrogeological mode is defined by the restriction to a zone of permafrost rocks (PMR). According to VES and drilling data, the thickness of the PMR is 450–500 m on the New Siberian Islands and 50–120 m in the coastal zone. It was established that the HCGs distribution in bottom sediments is controlled by the stratal accumulations of groundwaters of the Late Cretaceous–Cenozoic deposits, for which the gas saturation reaches

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 $70 \text{ cm}^3/\text{L}$. The temperatures of bottom sediments in the LSZ vary from -1.8 to $+1.9^{\circ}\text{C}$ [3, 4, 18].

RESULTS AND DISCUSSION

The Composition and Genesis of Gases

The HCGs of bottom sediments of the LSZ is 0.0006-2.9760% methane, 0.00001-0.00754 total ethane and ethylene, 0-0.00149 propane and prophylene, 0-0.00132 n-butane and i-butane, and 0-0.00011% n-pentane and i-pentane. The interpretation of geochemical parameters of HCGs allowed us to distinguish 11 genetic gas groups of different lithotypes and gas sources in LSZ sediments, which have different isotope compositions of δ^{13} C-CH₄ (Table 1).

Based on isotope–geochemical parameters (Table 1), syngenetic HCGs of the modern sediments have methane compositions with an admixture of ethane and ethylene, low average values of δ^{13} C–CH₄ (–74.5‰), Ktr (0.3), Kwet (0.2%), and a biochemical origin. The presence of propane "traces" in sediments is likely related to their natural diffusion and migration from underlying rocks. Similar geochemical parameters of HCGs were established [9] in the Quaternary deposits of boreholes 5, 13, 14, 25, 27–30, 32, 34, 41, and 63 (Fig. 1).

Sedimentary basin,	We	eight fraction	n of hydroca	000	MM _{HC} ,	Coefficients		
deposit (number of determinations)	C ₁	C ₂	C ₃	C ₄	C ₅	g/mol	Ktr	Kwet, %
			Gas-hydrat	e reservoir		•		
Okhotsky (24)	994	3	2	1	0	16.10	1.5	0.6
Messoyakhsky (8)	993	4	2	1	0	16.10	1.6	0.7
			Gas res	ervoir				
Lena-Vilyuisky (48)	956	29	9	5	1	16.43	16.1	4.4
Verkhnyaya Bureya (5)	955	28	9	6	2	16.45	18.7	4.5
Anadyrsky (20)	983	10	3	3	1	16.20	10.0	1.7
Sakhalinsky (40)	983	9	4	3	1	16.19	6.8	1.7
		C	as-condens	ate reservoir				
Lena-Vilyuisky (24)	876	69	32	16	7	17.23	34.0	12.4
Sakhalinsky (8)	872	68	31	17	12	17.30	37.3	12.7
			Condensate	reservoirs		•		•
Lena-Vilyuisky (15)	786	107	59	33	15	18.30	59.8	21.4
Sakhalinsky (6)	831	66	40	31	32	17.89	51.2	16.9
			Petroleum	reservoirs				
Lena-Vilyuisky (20)	728	144	74	36	18	18.97	70.0	27.2
Sakhalinsky (18)	764	128	67	26	15	18.50	49.7	23.6
			Gas-oil re	eservoirs				
Lena-Vilyuisky (32)	505	149	166	130	50	23.19	116.7	49.5
Verkhnyaya Bureya (28)	516	142	153	121	68	23.02	112.3	48.4
			Oil rese	ervoirs				
Lena-Vilyuisky (8)	372	184	227	156	61	26.35	126.5	62.8
Verkhnyaya Bureya (8)	393	158	178	147	124	26.26	130.5	60.7

Table 2. The average values of geochemical parameters of the HCGs of the petroleum basins, East Russia

The epigenetic HCGs of peat lands are dominated by the biogenic component (δ^{13} C–CH₄: -78.0...-69.0‰, MM_{HC}—16.12–16.13 g/mol, Kwet is less than 1%, Ktr < 2) with microadmixture of propane and trace i-butane. Similar parameters were established in the peat beds of boreholes 1 and 12 on a shoaling of Great Lyakhovsky Island [9]. This statement is also ascribed to HCGs of potentially gas hydrate rocks of the LSZ, the average gas geochemical parameters of which (MM_{HC}, 16.09 g/mol; Ktr, 0.9; and Kwet, 0.7) are sufficiently close to those of the Pleistocene–Miocene sediments of borehole 17, the Bay of Gedenshtrom [9], as well as gas hydrates of the Deryugin Basin of the Okhotsk Sea sedimentary basin and the Messoyakh gas deposit (Table 2).

The migration HCGs of Cenozoic gaseous and brown coal deposits contain approximately equal distribution of biochemical and metamorphic HCGs, which follows from the weight concentrations of HC and the average values of δ^{13} C-CH₄, MM_{HC}, Kwet, Ktr: -61.0 and -60.9‰, 16.15 and 16.20 g/mol, 1.4 and 2.0%, and 2.5 and 4.7 (Table 1). Analogous gas

geochemical parameters were found in the brown coal beds (PC_{12}) and gas seeps of boreholes 1g and 87 in the mouth of the Khroma River and Dmitrii Laptev Strait [9].

The epigenetic HCGs of Mesozoic gas and coal reservoirs have the average parameters of δ^{13} C–CH₄ (-58.6 and -56.0%), MM_{HC}, Kwet and Ktr, 16.27 and 16.42 g/mol, 2.7 and 4.6%, 8.1 and 9.4, respectively, which indicates their metamorphic genesis. This is also the case for migration HCGs of solid bitumens, the average isotope-geochemical parameters of which (MM_{HC}-17.61 g/mol, Ktr-46, Kwet-16% and δ^{13} C–CH₄ -49.6% $_o$) are sufficiently close to those of sampled anthraxolites in borehole 23g on the Chaun Bay coast [8, 9].

Migration HCGs of inferred condensate–gas and condensate reservoirs have the average values of parameters MM_{HC} , Kwet, Ktr of 17.15 and 18.29 g/mol, 12.5 and 23.4%, 28–63 and δ^{13} C-CH₄ (–53.0 and –50.4‰), which indicates their metamorphic origin. The average values of MM_{HC} , Kwet, Ktr of inferred oil–gas and gas–

			-	· · · ·				
Lithotype gas source		$CH_4(C_1)$			$\Sigma C_2 - C_5$			
Ethotype, gas source	min	max	av.	min	max	av.		
Modern sediments	0.0180	0.0654	0.0347	0.00003	0.00004	0.00003		
Gas hydrates	0.0066	8.3047	2.4144	0.00006	0.00682	0.00238		
Peat lands	0.0305	0.1058	0.0681	0.00024	0.00139	0.00081		
Coal deposits	0.0018	0.6900	0.3143	0.00004	0.00899	0.00391		
Gas reservoirs	0.0086	0.0637	0.0293	0.00010	0.00115	0.00048		
Igneous rocks	0.0037	0.0210	0.0078	0.00018	0.00060	0.00028		
Solid bitumens	0.0030	0.3620	0.0599	0.00027	0.00424	0.00137		
Condensate gas reservoirs*	0.0022	0.1549	0.0318	0.00021	0.01019	0.00208		
Condensate reservoirs*	0.0243	0.1524	0.0773	0.00375	0.02313	0.01165		
Oil-gas reservoirs*	0.0083	0.0923	0.0509	0.00190	0.01849	0.01020		
Gas-oil reservoirs*	0.0358	0.0570	0.0464	0.01 396	0.02596	0.01996		

Table 3. Concentrations of HCGs in bottom sediments in diverse gas sources (cm^3/kg)

* Potential reservoirs. Bold type distinguishes the maximum concentrations of HCGs.

oil reservoirs account for 19.43 and 21.60 g/mol, 32–47%, 153–304, and δ^{13} C-CH₄ (–44.8%). Epigenetic HCGs of igneous rocks with the average MM_{HC}, Kwet, and Ktr parameters of 16.68 g/mol, 7.3% and 16, respectively, have a magmatogenic nature and δ^{13} C-CH₄ values from –27.5 to –25.4‰ in the ESS, according to [8, 9].

In general, the formation of HCGs composition and concentrations in bottom sediments of the LSZ is controlled by the additivity rules, i.e., subsequent accumulation of HCGs of different genesis in sediments, with the predominance of gas phase and geochemical parameters of higher gas-saturated syngenetic or epigenetic gas source.

The established values of the minimum, maximum, and average concentrations of HCGs in the bottom sediments of the LSZ at the development areas of diverse gas sources in some cases exceed the anomalous parameters of CH_4 and ΣC_2-C_5 of 0.05 and 0.001 cm³/kg established for sediments of the East Arctic seas [15].

The maximum average CH_4 concentrations in the LSZ area are typical of the bottom sediments on the areas of potential development of gas hydrate reservoirs and carbonaceous formations (Table 3).

Minimum CH_4 values are typical of igneous rocks, gas reservoirs, modern sediments, and inferred condensate gas reservoirs, while the intermediate values characterize other gas sources.

The maximum average $\Sigma C_2 - C_5$ concentrations in sediments were found on the distribution areas of inferred oil and condensate reservoirs, while the minimum concentrations occur in the present-day sediments, igneous rocks, gas reservoirs, and peat lands. Intermediate values were determined in the bottom sediments within the development areas of solid bitumens, gas hydrates, inferred condensate-gas reservoirs, and carbonaceous formations.

It was established that concentrations of methane and the total of its homologs in the bottom sediments of the studied area vary within 0.0017-8.3047 and 0.00001-0.02593 cm³/kg and, on average, are 0.1690and 0.00218 cm³/kg, which are 3.4 and 2.2 times higher than the anomalous parameters. Thus, the distribution of their concentrations significantly differs in synclinal, anticlinal, and monoclonal structures (Table 4).

The maximum average CH_4 concentrations are observed in the bottom sediments of synclinal folds (troughs and graben rift); the minimum values were found in those of monoclinal folds (structural terraces) and saddles, and intermediate, in the anticlinal folds (rises and horsts). The average ΣC_2 - C_5 concentrations in sediments of troughs and saddles are 2– 3.5 times higher than those of rises, horsts, and structural terraces (Figs. 3a, 3b; Table 4).

The distribution of HCGs anomalies in LSZ sediments depends on the geostructural position. The anomalous concentrations of CH₄ and ΣC_2-C_5 were found in sediments of the eastern (up to 0.5378 and 0.00647 cm³/kg) and western (up to 0.4580 and 0.00362 cm³/kg) limbs and arched part of the Kotelnichesky Rise (up to 0.0615 and 0.00403 cm³/kg). The CH₄ and ΣC_2-C_5 anomalies in sediments of the southwestern limb of the Rezhetnikovskoe Rise reach 0.6275 and 0.00644 cm³/kg. The formation of anomalies of methane and its homologs of 0.1549 and 0.01019 cm³/kg in sediments of the Sannikovskaya saddle is observed in its central part.

The low CH_4 and ΣC_2 – C_5 concentrations and the absence of their anomalies in sediments of the East Laptev rise and horsts (Table 4) likely indicate the pre-

		CH ₄ , cm ³ /kg		$\Sigma C_2 - C_5$, cm ³ /kg			
Geostructures	min	max	av.	min	max	av.	
Structural terraces (26)	0.0017	0.6723	0.0613	0.00001	0.00825	0.00132	
Blagoveshchensk (10)	0.0034	0.6723	0.1330	0.00001	0.00825	0.00200	
Shelon (16)	0.0017	0.0553	0.0165	0.00004	0.00806	0.00090	
Rises, horsts (17)	0.0022	0.6275	0.1036	0.00003	0.00644	0.00126	
Reshetnikovskoe (2)	0.0637	0.6275	0.3456	0.00005	0.00644	0.00348	
Kotelnicheskoe (9)	0.0048	0.4580	0.1158	0.00003	0.00424	0.00145	
East Laptevskoe (3)	0.0022	0.0055	0.0036	0.00011	0.00019	0.00015	
Belkovsky, Stolbovsky (3)	0.0025	0.0080	0.0058	0.00008	0.00062	0.00031	
Troughs, graben-rifts (61)	0.0022	8.3047	0.2488	0.00003	0.02593	0.00271	
Novosibirsky (7)	0.0050	0.1058	0.0349	0.00006	0.01849	0.00320	
Anisimovsky (18)	0.0024	0.0659	0.0172	0.00003	0.02593	0.00168	
Belkovsky–Svyatonossky (9)	0.0068	0.3620	0.0552	0.00016	0.00424	0.00194	
Omoloisky (11)	0.0029	0.1524	0.0337	0.00004	0.02313	0.00354	
Tastakhsky (2)	0.2302	0.6900	0.4601	0.00299	0.00899	0.00599	
Northern Omoloisky (14)	0.0022	8.3047	0.9677	0.00007	0.01966	0.00307	
Sannikovskaya saddle (6)	0.0030	0.1549	0.0641	0.00005	0.01019	0.00486	
Studied area (110)	0.0017	8.3047	0.1690	0.00001	0.02593	0.00218	

Table 4. Concentrations of HCGs in bottom sediments of geostructures of the studied area

Bold type shows anomalous concentrations of HCGs, number of samples is shown in parentheses.

dominance of degassing in these structures, but the gas sampling of these geostructures was insufficient to make unambiguous conclusions.

In sediments of the Shelon and Blagoveshchensk terraces, the distribution of anomalies of methane 0.0507 and 0.5378–0.6723, and its homologs of 0.00115–0.00806 and 0.00100–0.00825 cm³/kg (Figs. 3a, 3b) is related to the migration of HCGs along diagonal and transverse fault zones, including the South Anyui suture, which cut across Mesozoic gas and coal–gas formations.

Maximum CH₄ anomalous concentrations of 2.3056–8.3047 cm³/kg—and anomalies of ΣC_2 –C₅— up to 0.00682 cm³/kg were established on the western limb of the Northern Omoloi graben rift in the junction zone of the Eurasian and North American lithospheric plates within the river paleovalley on the areas with maximum thickness of Quaternary sediments.

The formation of CH_4 and ΣC_2-C_5 anomalies up to 0.6904 and 0.00899 cm³/kg is observed in the bottom sediments of the closure of synclinal folds of the Tastakh trough, 0.3620 and 0.00424 in the bottom sediments of the Belkovsky–Svyatonossky trough, 0.0923 and 0.01849 in the bottom sediments of the New Siberian islands, 0.0570 and 0.02593 in the bottom sediments of the Anisimovsky trough, and 0.0547 and 0.00 104 cm³/kg in the bottom sediments of the Omoloi fold. The anomalous concentrations of methane and its homologs up to 0.0659 and $0.00165 \text{ cm}^3/\text{kg}$, respectively, were discovered on the limbs of the Anisimovsky trough, 0.1524 and 0.02313 in the Omoloi trough, 0.4580 and 0.00362 in the Belovsky–Svyatonossky trough, and up to 0.6904 and $0.00899 \text{ cm}^3/\text{kg}$ in the New Siberian Islands (Figs. 3a, 3b).

Studies revealed a trend of increasing the average HCGs concentrations in bottom sediments with increasing sampling depth (position), which is observed practically in all geostructures and the entire studied area (Table 5). The gradient of increasing the average CH_4 and ΣC_2 – C_5 concentrations in sediments of the studied area with increasing depth from 10–40 to 150–250 cm is 0.00071 and 0.000049 cm³/kg per cm, 0.00189 and 0.000020 in anticlinal folds, 0.00120 and 0.000015 in monoclinal folds, 0.00046 and 0.000034 in synclinal folds, and 0.00086 and 0.000057 cm³/kg per 1 cm in the saddle.

Thus, the maximum values of gradient of methane mass transfer in the LSZ are observed in sediments of anticlinal folds, the minimum values occur in synclinal folds, and intermediate values occur in the monoclinal folds and saddle. Similar maximum parameters of ΣC_2 - C_5 are typical of sediments of synclinal folds and saddle, while the minimum values occur in the monoclinal structures, and intermediate values are typical of anticlinal folds. It should be noted that a sharp increase of the average CH₄ concentrations and a decrease of ΣC_2 - C_5 in sediments of the lowermost sampling horizon (250–325 cm) is likely



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Daramatara	Values of ges geochemical parameters of bottom sediments over their sampling intervals, cm						
Falameters	10-40	40-100	100-150	150-250	250-350		
1	2	3	4	5	6		
Structural terraces							
CH_4 , cm ³ /kg	$\frac{0.0017 - 0.0117}{0.0052(10)}$	$\frac{0.0037 - 0.0359}{0.0171(5)}$	$\frac{0.0063 - 0.0553}{0.0331(5)}$	0.0088-0.6723 0.2152(6)	N.d.		
$\Sigma C_2 - C_5$, cm ³ /kg	$\frac{0.00004 - 0.00050}{0.00022(10)}$	0.00001- 0.00247 0.00078(5)	0.00019-0.00806 0.00227(5)	0.00028-0.00825 0.00283(6)	N.d.		
δ ¹³ C-CH ₄ , ‰	N.d.	N.d.	N.d.	$\frac{-59.056.0}{-57.7(3)}$	N.d.		
C _{org} , %	$\frac{1.82-2.02}{1.92(2)}$	$\frac{1.47}{1.47}$	$\frac{0.93-1.20}{1.08(2)}$	$\frac{0.84 - 1.14}{1.03(3)}$	N.d.		
	I	Rises	, horsts				
CH_4 , cm ³ /kg	0.0025- 0.0654 0.0196(8)	0.0022-0.4580 0.0919(6)	N.d.	$\frac{0.0637 - 0.6275}{0.3511(3)}$	N.d.		
$\Sigma C_2 - C_5$, cm ³ /kg	$\frac{0.00003 - 0.00062}{0.00019(8)}$	0.00011-0.00403 0.00145(6)	N.d.	0.00005-0.00644 0.00374(3)	N.d.		
δ^{13} C-CH ₄ , ‰	$\frac{-78.069.0}{-72.3(3)}$	$\frac{-61.0}{-61.0}$	N.d.	$\frac{-60.949.8}{-55.4(2)}$	N.d.		
C _{org} , %	$\frac{0.98 - 1.98}{1.59(8)}$	$\frac{1.14-1.70}{1.44(3)}$	N.d.	$\frac{0.94 - 1.08}{1.00(3)}$	N.d.		
		Sa	ddle				
CH ₄ , cm ³ /kg	$\frac{0.0030 - 0.0068}{0.0049(2)}$	$\frac{0.0355}{0.0355}$	$\frac{0.0553 - 0.1290}{\textbf{0.0921}(2)}$	<u>0.1549</u> 0.1549	N.d.		
$\Sigma C_2 - C_5$, cm ³ /kg	$\frac{0.00005 - 0.00027}{0.00016(2)}$	0.00213 0.00213	$\frac{0.00806 - 0.00845}{0.00826(2)}$	0.01019 0.01019	N.d.		
δ^{13} C-CH ₄ , ‰	N.d.	N.d.	N.d.	$\frac{-53.0}{-53.0}$	N.d.		
C _{org} , %	$\frac{1.56}{1.56}$	$\frac{0.96}{0.96}$	$\frac{0.91 {-} 0.93}{0.92(2)}$	$\frac{0.89}{0.89}$	N.d.		
		Troughs, g	graben-rifts				
CH_4 , cm ³ /kg	$\frac{0.0024 - 0.0109}{0.0044(10)}$	$\frac{0.0054 - 0.2324}{0.0275(15)}$	$\frac{0.0058 - 0.1058}{0.0277(11)}$	$\frac{0.0050 - 0.3620}{0.0847(14)}$	$\frac{0.0346 - 8.3047}{2.0221(6)}$		
$\Sigma C_2 - C_5$, cm ³ /kg	$\frac{0.00007 - 0.00037}{0.00018(10)}$	0.00006- 0.00299 0.00065(15)	0.00013-0.00433 0.00115(11)	0.00003-0.01966 0.00618(14)	0.0003-0.02593 0.00307(6)		
δ ¹³ C-CH ₄ , ‰	N.d.	N.d.	N.d.	$\frac{-49.8}{-49.8}$	$\frac{-44.8}{-44.8}$		
C _{org} , %	$\frac{1.38 - 1.74}{1.56(2)}$	$\frac{1.01-1.72}{1.30(4)}$	$\frac{0.92 - 1.24}{1.08(3)}$	$\frac{0.62 - 1.32}{0.92(9)}$	$\frac{0.56 - 1.20}{0.92(3)}$		

 Table 5.
 The isotope-geochemical parameters of HCGs in bottom sediments of geostructures of the studied area over sampling intervals

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Darameters	Values of ges geochemical parameters of bottom sediments over their sampling intervals, cm								
ranneters	10-40	40-100	100-150	150-250	250-350				
1	2	3	4	5	6				
		Studi	ed area						
CH ₄ , cm ³ /kg	$\frac{0.0017 - 0.0654}{0.0087(30)}$	$\frac{0.0022 - 0.4580}{0.0407(27)}$	$\frac{0.0058 - 0.1290}{0.0391(18)}$	$\frac{0.0048 {-} \textbf{0.6900}}{\textbf{0.1327}(24)}$	<u>0.0346-8.3047</u> 2.0221(6)				
ΣC_2 - C_5 , cm ³ /kg	$\frac{0.00003 - 0.00062}{0.00019(30)}$	0.00001- 0.00403 0.00091(27)	0.00013-0.00845 0.00212(18)	0.00003-0.02313 0.00880(24)	0.0003-0.02593 0.00307(6)				
δ^{13} C-CH ₄ , ‰	$\frac{-78.069.0}{-72.3(3)}$	$\frac{-61.0}{-61.0}$	N.d.	$\frac{-60.949.8}{-55.2(7)}$	$\frac{-44.8}{-44.8}$				
C _{org} , %	$\frac{1.38 - 2.02^*}{1.64(13)}$	$\frac{0.96 - 1.72}{1.33(9)}$	$\frac{0.91 - 1.24}{1.05(5)}$	$\frac{0.62 - 1.32}{0.95(16)}$	$\frac{0.56-1.20}{0.92(3)}$				

 Table 5. (Contd.)

* The range of variations in nominator, the average value (number of cases) in denominator; (n.d.) not determined. Bold types distinguishes anomalous concentrations of HCGs.

related to the sampling of the gas-saturated zone of potential areas of gas hydrate metastability [7, 14].

It was established during studies that the average content of organic matter (C_{org}) in LSZ sediments shows a systematic decrease from 1.64 to 0.92% with increasing sampling depth (Table 4) against the background of increasing average concentrations of CH₄, ΣC_2-C_5 , and $\delta^{13}C$ -CH₄, which also confirms the predominance of epigenetic HCGs in bottom sediments, as well as natural gas diffusion and migration from underlying gas sources over biochemical transformations of organic matter. Thereby, it should be noted that an increase of C_{org} contents in sediments of the upper horizon of the Kotelnichesky Rise from 1.5 to 2% is accompanied by the increase of concentrations of syngenetic CH₄ from 0.0180 to 0.0654 cm³/kg.

The Influence of Geological Factors

Based on the above-mentioned facts, the formation and distribution of HCGs in the bottom sediments of the LSZ depend on the complex influence of the main geological factors: tectonics, gas saturation of underlying lithotypes and gas sources, stages of catagenesis, organic saturation, depth, thickness of sediments, seismic activity, hydrogeological, geocryological, and other conditions of gas accumulation or their degassing.

The Tectonic Factor

It was established that the average CH_4 and ΣC_2 – C_5 concentrations in the bottom sediments of the studied area systematically increase westward to the junction zone of the Eurasian and North American lithospheric plates from 0.1725 and 0.00230 and from 0.2071 and 0.00272 cm³/kg within the New Siberian-Chukotka and Rauchan–Oloi fold systems to 0.3495 and 0.00305 cm³/kg in the Verkhoyansk–Kolyma system. Thus, the maximum and anomalous concentrations of CH_4 and $\Sigma C_2 - C_5$ (8.3047) and $0.01966 \text{ cm}^3/\text{kg}$) were found in sediments of the Northern Omoloi graben rift, which is in a direct contact with suture zone of the plates. In the same direction, we observe an increase of the degree of the fold and fault deformation of the basement (Figs. 1, 2), which are the main factors of mass transfer and HCGs migration in the bottom sediments and formation of their polygenous composition. The significant uplift of the M surface up to 21-27 km in the lithospheric plate junction [4], thus facilitating deep-seated Earh's degassing, is also an important fact.

As indicated previously, the formation and distribution of anomalous HCGs concentrations in the bottom sediments of LSZ structures depend not only on the tectonic structure, but also on the geostructural position in the arched or trough parts, the closure and limbs of the synclinal and anticlinal folds, frequently with tectonic boundaries, as well as the degree of fault deformations in the monoclinal folds.

Faults depending on their gas permeability, are pathways for gas migration and also gas confining layers facilitating HCGs accumulation in rock complexes. Based on the geological survey and seismic data [3, 4], most longitudinal faults of the region are accompanied by small fold zone and intense rock compaction, which significantly hampers the vertical migration of HCGs, while transverse faults facilitate migration and usually cause rock degassing. Diagonal faults are associated with a thick zone of fold and fractured rocks and also cause a degassing effect, but a sharp change of their strike is accompanied by the formation of zones of the compacted rocks with low gas permeability and serve as gas confining beds [4, 7-9].

Seismic activity plays a dual role in the formation of gas saturation of bottom sediments of the LSZ. The presence of inferred condensate, hydrate-bearing and other gas-bearing underlying rocks, on the one hand, facilitates the HCGs redistribution and inflow in the upper horizons, as well as increasing HC-saturation of sediments during development and activation of earthquakes, and, on the other hand, degases them with gas release into the water column and atmosphere. The relationships between these processes likely depend on the earthquake depth, gas saturation of rocks, and tectonic factor. It was established that bottom sediments within linear seismoactive zone of lithospheric plate junction, which is represented by a chain of earthquake epicenters (Fig. 1), show anomalies of CH₄ 0.0507-8.3047 and ΣC_2 -C₅, 0.00115- $0.0197 \text{ cm}^3/\text{kg}.$

Magmatism

According to the gas sampling of borehole cores, the volcanic rocks of Great Lyakhovsky island have lower CH₄ and ΣC_2 -C₅ concentrations relative to other lithotypes: up to 2.81 and 0.032 cm³/kg. This likely explains the minimum HCGs concentrations in bottom sediments at the development areas of the LSZ magmatic rocks (Table 2) and indicates their low migration hydrocarbon potential against the background of natural degassing of sediments.

The gas potential of the underlying lithotypes and gas sources is one of the main factors of HCGs formation and distribution in the LSZ bottom sediments. The formation of the anomalous average CH_4 concentrations are restricted to the areas of potentially gas hydrate and coal reservoirs, inferred oil–gas and condensate reservoirs, peat lands and solid bitumens; those of ΣC_2 – C_5 are confined to the inferred oil and condensate reservoirs, coal and hydrate reservoirs, as well as inferred condensate–gas reservoirs and solid bitumens. On the development areas of other lithotypes and gas sources, these HCGs anomalies are absent (Table 2).

Catagenesis

The study of migration HCGs of coal-bearing formations showed that the average concentrations of CH₄ and ΣC_2 -C₅ are 0.0681 and 0.00081 in sediments from peat lands, 0.1836 and 0.00226 in brown coals (PC₁-PC₂ catagenetic stage), and 0.4922 and 0.00615 cm³/kg in hard coal (MC₁). Thus, with increasing catagenetic stage of the gas source (from PC₁ to MC₁), the bottom sediments of the LSZ show a seven-fold increase of the average HCGs concentrations, which was also established from gas sampling of boreholes (see section Gas Potential).

It should be noted that the above tendency is also overprinted by the variability of the increase of HCGs concentrations from the stratigraphic (from $Q-N_2^3$ to K_1) and established (factual) depths of the indicated sources and bottom sediments (Table 4), as well as the thickness of Quaternary bottom sediments, the maximum values of which were recorded within the river paleovalley of the studied area.

Geocryological and Hydrogeological Conditions

Permafrost rocks and frozen bottom sediments serve as a gas confining layer and prevent natural diffusion and migration of HCGs, "conserve" available HC-gases, lead to the decrease of the gas permeability of the deposit, and, as a result, cause a sharp increase of natural gases concentrations at the permafrosttalik boundary, which is observed in boreholes recovering this zone. In some cases, this is accompanied by gas seeps and emanations. This situation likely takes place in the generation areas of gas hydrate reservoirs in bottom deposits. The stratal groundwaters in the Late Cretaceous-Cenozoic deposits have high contents of CH₄ and $\Sigma C_2 - C_5$ up to 51.70 and 0.011%, respectively, and gas saturation, in general, up to $70 \text{ cm}^3/\text{L}$, which indicates the possible enrichment of bottom sediments in HCGs in the course of their transition from a dissolved to free state at their oversaturation in groundwaters.

CONCLUSIONS

The Laptev–Siberian Sea zone has a complex geological and tectonic structure of Paleozoic, Mesozoic, and Cenozoic carbonate, terrigenous-carbonate, and terrigenous rock complexes, which is complicated by plicative and fault tectonics, high seismic activity, and magmatic activity. The intensity of these processes significantly increases in the junction zone of the Eurasian and North American lithospheric plates.

The HCGs of rocks and bottom sediments of the LSZ contain methane, as well as its saturated and unsaturated homologues up to pentane. The isotope–gas-geochemical parameters of HCGs indicate that the sediments of the LSZ contain both syngenetic and predominant epigenetic (migration) gases of different lythotypes and biochemogenic, metamorphic, and magmatogenic gas sources.

The HCGs composition and concentrations in LSZ sediments are controlled by additivity rules, i.e., subsequent accumulation of HCGs of different genesis in sediments, with the predominance of gas phase and geochemical parameters of the higher gas saturated syngenetic or epigenetic gas source.

The CH₄ and ΣC_2 -C₅ concentrations in the LSZ bottom sediments reach 8.3047 and 0.02593 cm³/kg, which indicates the formation of HCGs geochemical anomalies that exceed the threshold contents by 166 and 26 times, respectively.

The HCGs formation and distribution in sediments depend on the complex influence of geological factors on gas accumulation or degassing.

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CONFLICT OF INTEREST

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

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