Geochemical Characteristics of Methane from Sediments of the Underwater High Posolskaya Bank (Lake Baikal)

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Abstract—The component and carbon isotope compositions were studied in the hydrocabon gases from sediments of the underwater high Posolskaya Bank (Lake Baikal). It was established that sediments of this Baikal area contain methane of microbial (C_1/C_2 >16000; $\delta^{13}C$ 70 ± 3‰) and thermocatalytic (C_1/C_2 <100; $\delta^{13}C$ −46 ± 3‰) origin. Some samples represent a gas mixture of thermocatalytic and microbial origin. This gas is characterized by $\delta^{13}C$ of methane varying from −60 to −70‰ and contains a significant amount of ethane. The main homolog of methane in the thermocatalytic and mixed gas is ethane. Owing to biodegradation, propane and butanes are present in trace amounts.

INTRODUCTION

Baikal is the only freshwater lake, whose sediments contain gas hydrates (GH). They were found for the first time in the deep-water borehole BDP-97 core (depths 121 and 161 m) (Kuz'min et al., 1998) and then in the subsurface sediments of Malen'kii mud volcano (Klerx et al., 2003). These finds stimulated search for gas seeps on the lake floor, i.e., areas of possible occurrence of gas hydrates in the subsurface bottom sediments. As a result, several areas with gas seeps were identified on the Lake Baikal floor (Granin et al., 2010), and sediments of most of them contained GH (Khlystov et al., 2013).

The discovery of subsurface GH pools promoted their comprehensive research, including works on studying the Baikal methane, which is the main component of GH (Kalmychkov et al., 2006; Hachikubo et al., 2010).

Since the publication of these works, new geochemical information on the Baikal methane has been accumulated. In this communication, we report the results of studying methane from the Posolskaya Bank sediments. The great interest in this object is primarily related to the intense release of gas bubbles from the lake floor (Granin et al., 2010). Two types of gas hydrates of cubic structure, CS-1 and CS-2, are present in the subsurface sedimentary beds. At present, in addition to the Posolskaya Bank, CS-2 gas hydrates

were found only in sediments of mud volcanoes of the submarine Kukui canyon (Hachikubo et al., 2009; Kalmychkov et al., 2006; Khlystov et al., 2013; Kida et al., 2006).

GEOLOGICAL STRUCTURE

The Posolskaya Bank (PB) is located in the Selenga River delta. According to the multichannel seismic profiling, the crystalline basement of the PB is a large normal fault-bounded inclined block overlain by sedimentary sequence approximately 3 km thick. The PB is extended along the northeastern direction and traced over 40 km in the relief of the Southern Basin of Baikal. The southern slope of the PB coincides with the boundary of the Posolsky normal fault, while its northern slope is bordered by the Peschanaya normal fault (Scholz and Hutchinson, 2000).

The thickness of sedimentary bed on the bank slope decreases from the foot of the northwestern slope to its summit. Its uppermost part shows traces of erosion and sediments here are represented by the sandy−gravelly varieties (*Vysokorazreshayushchaya ...,* 2004). Core recovered by the deep-water borehole BDP-99 (lake depth at the drilling point is 202 m, borehole depth is 350 m) on the northwestern slope of the PB consists of three units. The upper unit (0−120 m) is represented by alternating biogenic and terrigenous
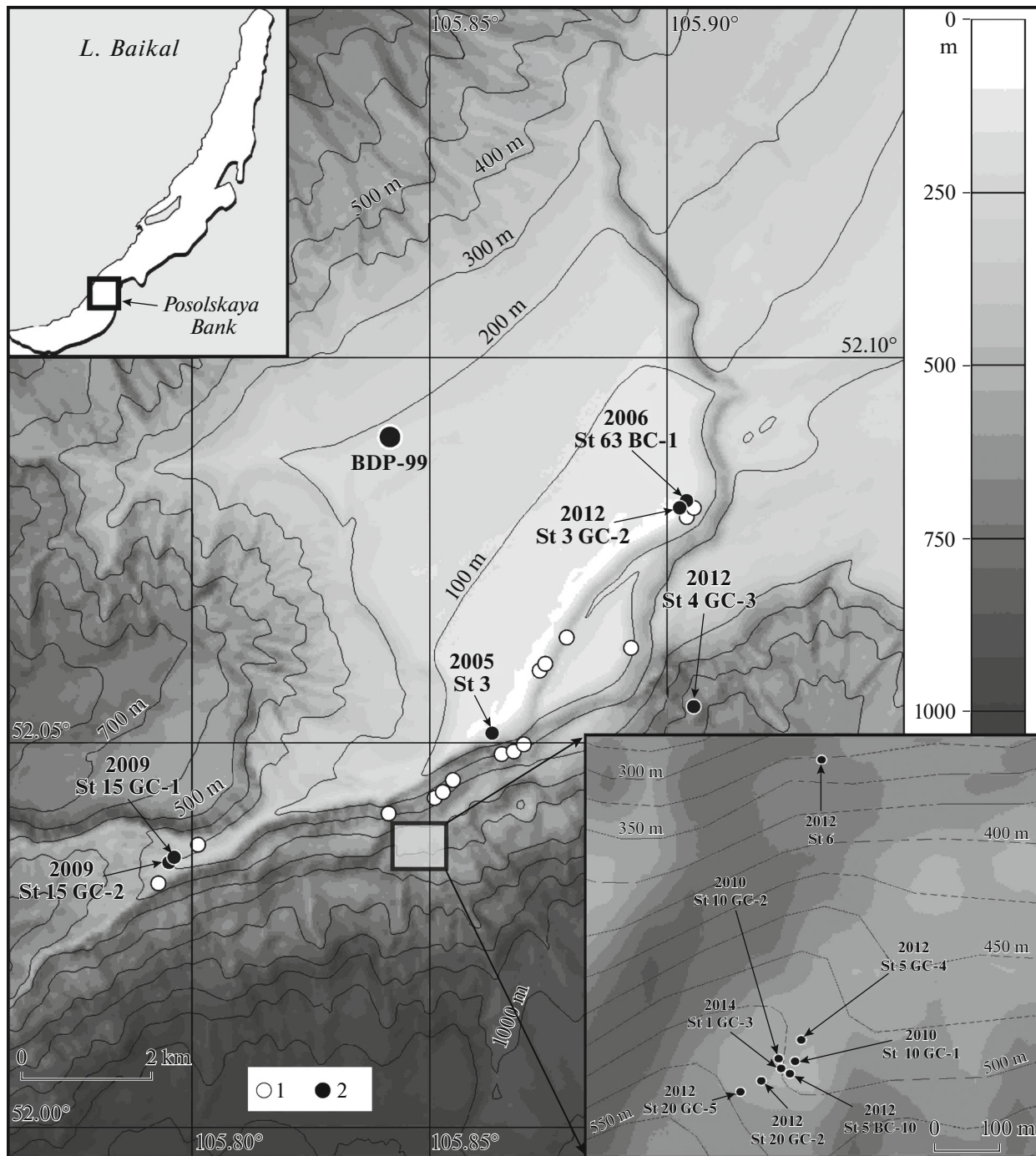
Fig. 1. Location of seeps (Granin et al., 2010) (1) and sediment sampling stations (2) within Posolskaya Bank. Inset shows the position of the Posolskaya Bank.

muds. The middle unit (120−230 m) consists of homogenous silty clays. The lower unit (230−325 m) is represented by silty clays intercalated with turbidites (BDP-99 Baikal Drilling Project Members, 2005; *Vysokorazreshayushchaya ..., 2004*).

Numerous gas seeps were found in the vicinity of the PB crest in its southeastern part (Fig. 1) (Granin et al., 2010). According to (Naudts et al., 2012), gas is

delivered to its discharge site near the crest through the permeable inclined sedimentary beds from the bank foot near the western coast. The cited authors believe that the migration of gas to the crest along the southern PB-bounding fault is less probable, since this pathway can be overlain by GH accumulations.

Gas was sampled on the PB crest, as well as on its southwestern and southeastern slopes (Fig. 1). Special

attention was focused on the southeastern slope, since signs of subsurface GH were found there at a depth of about 500 m during the manned MIR-2 submersible dive in 2009 (Fig. 1). Later, the GH samples were collected by gravity corers. At present, this is the single PB area with GH seeps found in the near-bottom sediments (Khlystov et al., 2013). However, the development of sandy–gravelly sediments on the PB summit hampered sampling by the gravity corer.

Gas was also collected from seeps on the southeastern slope of PB (lake depth 325 m, 2012 St 6). The methane flux from the lake floor is sufficiently high and gas bubbles reach the surface. This gas seep was previously detected by collaborators of the Hydrophysical Laboratory, Limnological Institute, Siberian Branch, Russian Academy of Sciences (http://hydro. lin.irk.ru/bdseep).

We also adopted the data in (Kuz'min et al., 2001) pertaining to the study of gas seepage from the borehole BDP-99 core (gas sampling was performed down to a depth of 120 m).

TECHNIQUE

The works were carried out onboard the R/V G.Yu. Vereshchagin. Sampling of bottom sediments using the gravity corer (GC) (core length up to 5 m) and the Bentos benthic corer (BC) provided the recovery of core (up to 1.5 m long) with undisturbed upper bed. Coordinates of sampling stations were determined during the corer liftoff from the ground (accuracy of the coordinate determination is ~50 m).

When the core was uplifted to the surface, samples of gas and GH (if present) were collected for isotope studies and the on-board study of methane distribution across the sediment section. Not more than five gas samples were taken for isotope studies. The core sampling interval for constructing the profiles of methane concentration was 5–10 cm in the upper horizons and up to 20–40 cm in the gas-saturated zone.

Hydrocarbon gases (HCG) in the sediment were determined by the head-space (vapor phase) analysis method (Bol'shakov and Egorov, 1987). The HCG concentration in the gas phase was analyzed on EKHO-PID chromatograph with plasma–ionization detector (2-m absorption column with the inner diameter of 2 mm, Porapak sorbent, isothermal regime, T 100°C).

The carbon isotope composition in methane and ethane was determined using Thermoelectron complex equipped with Delta V Advantage mass spectrometer and Trace GC Ultra gas chromatograph (Laboratory of Isotope Geochemistry and Geochronology, Geological Institute, Russian Academy of Sciences). Values of $\delta^{13}C$ are given in per mille (‰) relative to the V-PDB standard using carbon dioxide obtained by the decomposition of IAEA C-O-1 carbonate standard in 100% orthophosphoric acid with a Gas-Bench-II system. The $\delta^{13}C$ measurement accuracy is ± 0.2‰.

RESULTS AND DISCUSSION

Table presents the data obtained during study of the component composition and isotope characteristics of hydrocarbon gases from PB sediments.

The main component of the gas dissolved in pore waters is methane (90.1–99.9 vol % of the total HCG). Its homologs are mainly represented by ethane (C_2), whose content reaches 9.91% (table). Propane and butane (C_{3+}) occur in trace amounts and were determined only qualitatively in most of the studied samples. Therefore, the C_{3+} contents are not reported in table. The value of $\delta^{13}C-C_1$ varies from −69.6 to −42.4‰, while $\delta^{13}C-C_2$ from −27.4 to −23.3‰, averaging −25.9‰ (table).

Figure 2 shows the Bernard diagram, which makes it possible to ascribe methane to definite genetic type. It is seen that the studied samples contain methane of the microbial and thermocatalytic origin. Some samples represent a mixture of methane of different genesis and fall into the field of a mixed gas.

The microbial ($\delta^{13}C-C_1$ from −71.0 to −67.3‰) methane is present in the borehole BDP-99 core (Kuz'min et al., 2001) and contains a small (21.6–60.9 ppm) admixture of ethane. We believe that this gas lacks or contains only insignificant admixture of thermocatalytic gas. Therefore, in general, it best characterizes the microbial methane directly formed in PB sediments. Actually, as mentioned above, rocks at the deep-water borehole drilling site down to a depth of 350 m are mainly made up of the pelite-size sediments, which complicate gas influx from the deeper horizons of bottom sediments.

Thermocatalytic methane is present in sediments on the southeastern slope of the PB at a depth of 500 m (Fig. 1). It contains significant admixture of C_2 (table, Fig. 2). Its maximum concentrations (9.91 and 9.04%) were recorded in gas samples taken in the cores 2010 St 10 CG-1 and 2010 St 10 CG-2, respectively. The values of $\delta^{13}C-C_1$ vary from −50.0 to −42.4‰, averaging −46.0‰. The $\delta^{13}C$ values of ethane fall in the range from −24.9 to −27.2‰, which indicates its thermocatalytic origin (Milkov, 2005; and others).

The C_2 content reaches 3% in GH CS-1 and 14% in GH CS-2 (table). As in gas dissolved in pore waters, C_{3+} occurs in small amounts. The values of $\delta^{13}C-C_1$ in the gas hydrate vary from −51.3 to −44.0‰, while $\delta^{13}C-C_2$ values are comparable with that of gas dissolved in pore waters, averaging −25.7‰.

Virtually complete absence of C_{3+} admixture in the thermocatalytic methane from PB sediments indicates that it represents the product of secondary alterations, which significantly modify the component composi-
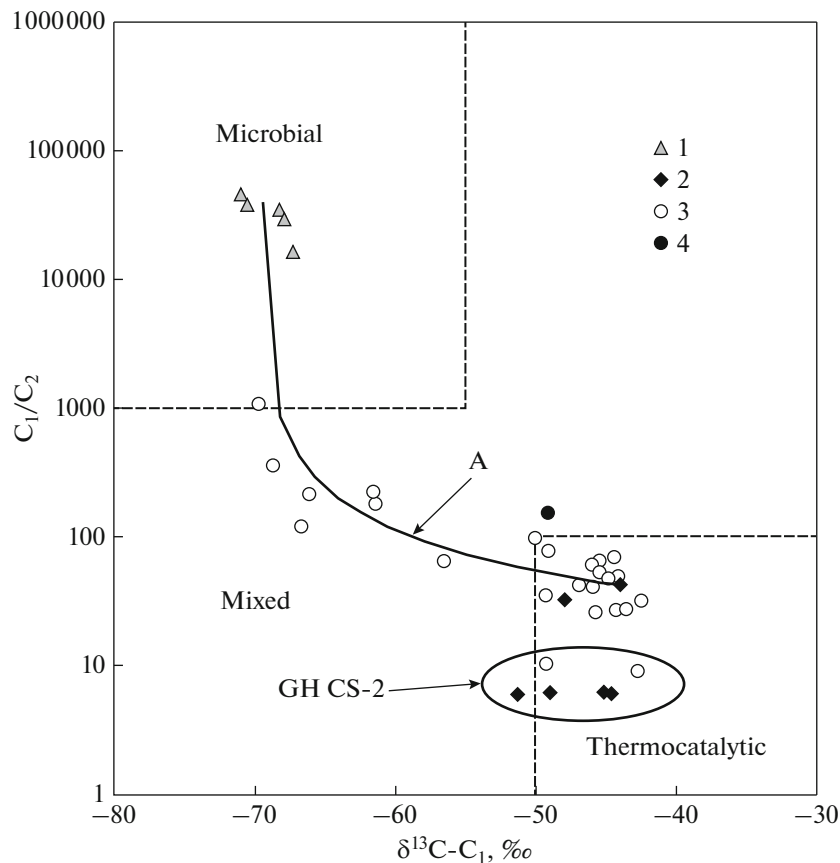
Fig. 2. Genetic varieties of methane from sediments of Posolskaya Bank, after (Bernard et al., 1976).
(1) Borehole BDP-99; (2) gas hydrates; (3) gas dissolved in pore waters; (4) gas bubbles.
(A) Mixing curve of microbial methane (average $\delta^{13}C$-C_1 and C_1/C_2 in gas from the BDP-99 borehole core) and thermogenic methane (average $\delta^{13}C$-C_1 and C_1/C_2 in gas and sediments from the southern slope of PB).

tion of initial gas. In our opinion, the most probable reason for the low C_{3+} content is biodegradation, which can be assisted under anaerobic conditions of Baikal by actinobacteria and some species of gammaprotobacteria that are widespread in the sedimentary sequence of the lake (Zemskaya et al., 2015; and others).

The biodegradation is accompanied by the preferential removal of *n*-alkanes (propane, *n*-butane, and others) (Katz et al., 2002; Vandrer et al., 2007). However, methane, ethane, and *iso*-alkanes are reworked by microbial community to a lesser extent. Gas unaffected by bacterial reworking in the methane−*n*-pentane series shows a monotonous, smooth progressive increase of $\delta^{13}C$ (James and Burns, 1983). During the bacterial decomposition of *n*-alkanes, this regularity is disturbed, since reaction products are enriched in ^{12}C isotope, while residual components are enriched in ^{13}C isotope. Consequently, the corresponding curve has a broken shape, which is an indicator of the biodegradation of hydrocarbon gases.

Due to the low admixture of C_{3+} in the methane dissolved in pore waters, as well as its low contents in gas samples taken in sediments by the head-space degassing technique, we failed to determine the isotope composition of homologs in PB sediments. Therefore, the corresponding measurements were only performed for the gas hydrate.

The analysis performed by Hachikubo using the technique described in (Hachikubo et al., 2015) allows the determination of $\delta^{13}C$ at homolog concentrations <0.1% with respect to methane. The results of $\delta^{13}C$ determination in methane homologs in the gas obtained by the decomposition of CS-1 and CS-2 gas hydrates collected at 2012 St 20 are shown in Fig. 3. The values of $\delta^{13}C$ of hydrocarbon gases in CS-1 and CS-2 gas hydrates are virtually identical. It is also seen that propane, *n*-butane, and *n*-pentane are anomalously enriched in ^{13}C relative to ethane and *iso*-alkanes. This fact indicates that *n*-alkanes were subjected to the bacterial reworking. The GH formation is not accompanied by significant fractionation of carbon isotopes (Sassen and MacDonald, 1997). There-

Composition and isotope characteristics of hydrocarbon gases and gas hydrates (HG) from sediments of Posolskaya Bank

Station no.	Interval, cm	$\delta^{13}C$, ‰		C_1/C_2	C_2, %
		C_1	C_2		
2005 St 3 BC-1	45	−66.6	−	118	0.84
2006 St 63 BC-1	65	−61.4	−	178.51	0.56
2009 St 15 GC−1	260	−66.0	−	209.96	0.47
2009 St 15 GC-2	180	−61.5	−27.2	221.1	0.45
2010 St 10 GC-1	10	−50.0	−24.9	96.2	1.03
2010 St 10 GC-1	50	−45.4	−26.3	64.09	1.54
2010 St 10 GC-1	90	−44.1	−26.1	47.89	2.05
2010 St 10 GC-1	120	−42.6	−26.4	9.09	9.91
2010 St 10 GC-1	GH CS-2	−49.0	−27.6	6	14.29
2010 St 10 GC-2	30	−49.2	−26.8	34.9	2.79
2010 St 10 GC-2	60	−45.6	−26	25.74	3.74
2010 St 10 GC-2	120	−49.1	−26	10.06	9.04
2010 St 10 GC-2	GH CS-2	−51.3	−27.4	6	14.29
2012 St 3 GC-2	160	−68.7	−	1072.88	0.09
2012 St 4 GC-3	230	−69.6	−	352.7	0.28
2012 St 5 GC-4	GH	−44.0	−	42.7	2.29
2012 St 5 BC-10	20	−49.0	−	75.8	1.30
2012 St 5 BC-10	40	−46.9	−	41.95	2.33
2012 St 5 BC-10	60	−45.9	−	39.83	2.45
2012 St 5 BC-10	80	−44.2	−27.4	26.45	3.64
2012 St 5 BC-10	98	−43.5	−	27.63	3.49
2012-01 St 6	Gas bubble	−49.1	−	154.41	0.64
2012 St 20 GC-2	GH CS-2	−44.6	−	6.00	14.29
2012 St 20 GC-5	GH CS-2	−45.2	−27.2	6.20	13.89
2012 St 20 GC-5	GH CS-1	−45.9	−	32.40	2.99
2014 St 1 GC-3	10	−44.3	−	69.57	1.42
2014 St 1 GC-3	25	−46.0	−	59.7	1.65
2014 St 1 GC-3	40	−45.4	−	53.4	1.84
2014 St 1 GC-3	60	−44.7	−	47.41	2.07
2014 St 1 GC-3	160	−42.4	−	31.98	3.03

CS-1 and CS-2 are types of the cubic structure of gas hydrates. Dash denotes not analyzed.

fore, we can suggest with high probability that source gas was also subjected to the bacterial reworking.

Methane of a mixed genesis was found on the PB summit (2005 St 3 BC-1, 2006 St 63 GC-1, 2012 St 3 GC-2) and on its southwestern slope (2009 St 15 GC-1, 2009 St 15 GC-2). It is characterized by $\delta^{13}C$ values typical of the microbial methane (−56.4 to −69.6‰) and a high (0.09−1.55%) admixture of ethane, the $\delta^{13}C$ value of which is indicative of thermocatalytic gas. Gas with such characteristics is present not only in PB sediments, but also in other regions of Baikal (Hachikubo et al., 2010; Kalmychkov et al., 2006).
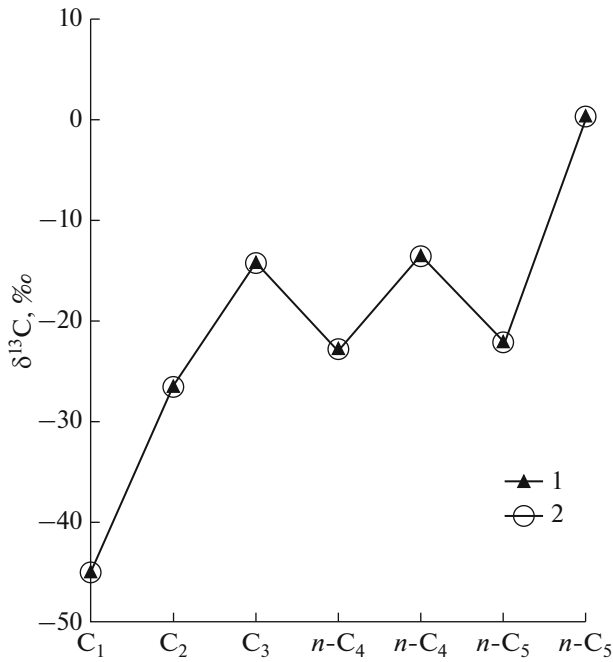
Fig. 3. Carbon isotope composition of methane homologs in gas hydrate (GH) samples CS-1 and CS-2. (1) GH CS-1; (2) GH CS-2.

Methane of such type is formed by the mixing of thermocatalytic and microbial gases. This process takes place in any gas systems along the entire thermocatalytic gas migration path extending from the source

to the bottom surface, leading to the decrease of C_{2+} and $\delta^{13}C\text{-}C_1$ value in the mixture.

It should, however, be noted that the intense formation of methane in Baikal begins already in the uppermost near-bottom sediment (Namsaraev et al., 1995), where mixing is most intense. A wide scatter of $\delta^{13}C$ values and ethane concentrations in the thermocatalytic gas (table, Fig. 2) is mainly related to mixing of the ascending flow with the microbial gas in the near-bottom sediment (Fig. 4).

It is noteworthy that the anaerobic and aerobic oxidation of methane in the subsurface beds of Baikal sediments (Pimenov et al., 2014) leads to enrichment of the residual methane in ^{13}C and increase of C_{2+} in the gas. However, data presented in Fig. 4 indicate that the oxidation of methane in PB sediments does not play a significant role in the formation of isotope characteristics and component composition of HCG.

Figure 2 demonstrates the mixing curve of microbial and thermocatalytic methane, which is well consistent with the obtained experimental data.

Gas obtained by the decomposition of CS-2 gas hydrates and gas dissolved in the pore waters of the core (2010 St 10 CG-1 and 2010 St 10 CG-2) fall into a separate field in the Bernard diagram (Fig. 2). This is consistent with the data reported in (Manakov et al., 2013) suggesting that the Baikal CS-2 gas hydrate is an intermediate product formed during the decomposition of CS-1 gas hydrate and, hence, does not reflect
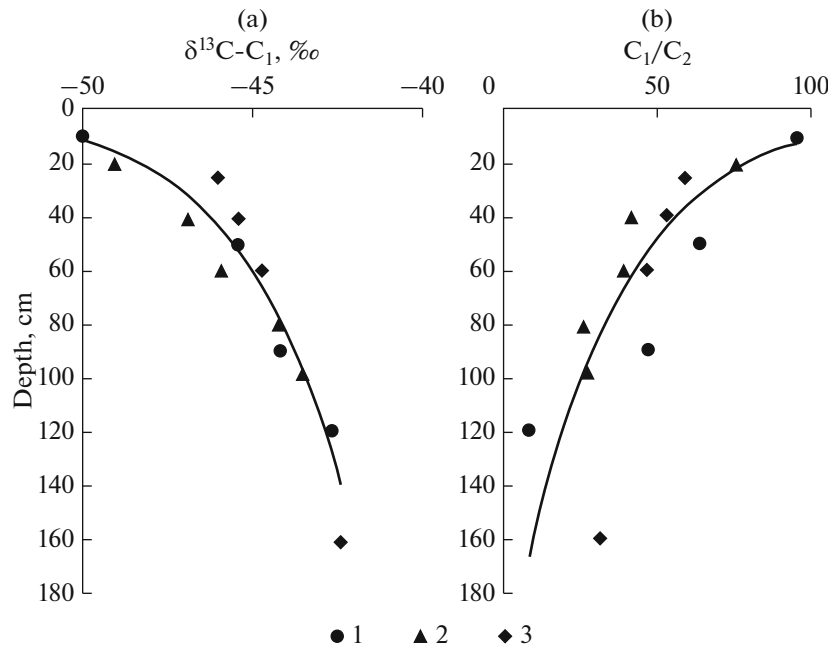

Fig. 4. Variations of $\delta^{13}C\text{-}C_1$ values (a) and C_1/C_2 ratio (b) with sediment depth. (1) 2010 St 10 GC-1; (2) 2012 St 5 BC-10; (3) 2014 St 1 GC-3.
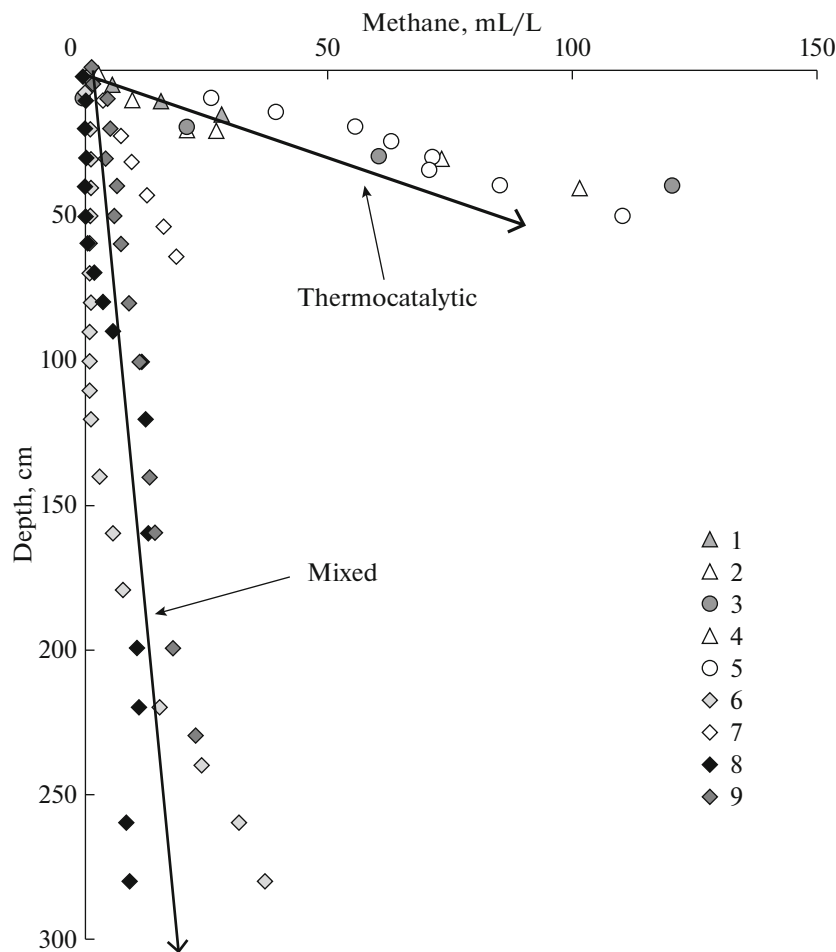
Fig. 5. Concentration profiles of methane from PB sediments. (1) 2010 St 10 GC-1; (2) 2010 St 10 GC-2; (3) 2012 St 5 BC-10; (4) 2012 St 5 GC-4; (5) 2014 St 1 GC-3; (6) 2006 St 63 BC-1; (7) 2009 St 15 GC-1; (8) 2009 St 15 GC-2; (9) 2012 St 4 GC-3.

the component composition of methane delivered from the lower sedimentary horizons.

In the case of intensely ascending flows of thermocatalytic gas, the isotope characteristics and component composition are subjected to a lesser mixing. Therefore, reliable estimation of gas characteristics in the source requires the analysis of methane-rich samples taken in or near an operating channel of active gas migration.

Gases taken from seeps directly near the floor are best suitable for these purposes. However, gas bubbles taken at 2012-St 6 from the lake surface are characterized by lower $\delta^{13}C\text{-}C_1$ values (relative to the thermocatalytic methane) and ethane admixture (Fig. 2). In our opinion, this is related to the following fact: the proportions of methane and ethane concentrations in the initial gas are disturbed during the ascent of bubbles to the surface (lake depth at the sampling point 325 m) due to the higher solubility of ethane in water (Hammiot, 1991; and others).

Gas hydrates, whose formation requires high methane concentration in the pore waters, also mark the pathways of active gas migration. This is illustrated in Fig. 5 showing profiles of methane concentration in PB sediments. It is seen that the hydrate-bearing core and the core taken in the GH accumulation area are characterized by high gas content, with the methane concentration reaching 120 mL/L already at a depth of 40 cm. Gas in this core is of the thermocatalytic origin. At less intense methane flow, the corresponding gas samples contain methane of the mixed genesis.

Thus, methane from the GH-bearing core reflects the component composition and isotope characteristics of thermocatalytic gas transported from the lower sedimentary horizons. It should also be noted that the thermocatalytic gas is only partially conserved in hydrates, whereas some of its part reaches the PB summit through numerous fractures of the Posolsky fault and forms methane of the mixed type. The above conclusion is also supported by the presence of bubble gas discharge beyond the GH stability zone on the south-

eastern slope of PB at a depth of 325 m. This fact imposes some constraints on the mechanism of gas migration in PB proposed in (Naudts et al., 2012) and excludes this pathway of thermocatalytic methane migration from the GH stability zone.

It was shown above that the low content of propane and butanes in the thermocatalytic gas is related to their bacterial reworking. However, it is unclear yet whether this is the only cause for such component composition of the thermocatalytic methane from PB sediments. It is quite possible that gas in the source is characterized by initially low concentrations of heavy homologs, and bacterial processes additionally decrease their concentrations.

CONCLUSIONS

The component and carbon isotope compositions of hydrocarbon gases were studied in sediments of the underwater high Posolskaya Bank (Lake Baikal).

The obtained data showed that sediments of this area contain methane of the microbial and thermocatalytic origin. Some samples represent a mixture of the thermocatalytic and microbial gas. Such gas contains the thermocatalytic ethane and a great amount of methane characterized by low $\delta^{13}C$ values typical of the microbial methane.

In the thermogenic and mixed gas, the methane homologs are represented mainly by ethane. Due to biodegradation, propane and butanes occur in trace amounts.

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