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Hydrocarbon Biomarkers and Diamondoid Hydrocarbons from Late Precambrian and Lower Cambrian Rocks of the Katanga Saddle (Siberian Platform)

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Abstract—This study presents geochemical data on organic-rich rock samples collected from Riphean—Lower Paleozoic strata (potential source rocks) of the southern Siberian Platform and compositional data on hydrocarbon biomarkers (steranes, terpanes, *n*-alkanes, 12- and 13-methylalkanes, isoprenanes) and diamondoid hyrocarbons from core samples collected from the Kulindinskaya-1 well, which was drilled by RN-Exploration in 2012 within the Katanga saddle.

Keywords: geochemistry, source rocks, hydrocarbon biomarkers, diamondoid hydrocarbons, Riphean, Vendian, Cambrian, Siberian Platform, Katanga saddle

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The southern part of the Siberian Platform actually has many large oil and gas fields that are confined to the crestal parts of the Nepa–Botuoba and Baykit anteclises (Fig. 1). The construction of the East Siberia–Pacific pipeline system has opened a new stage of appraisal of petroleum potential of the area and renewed interest in the study of major trends in the formation and spatial distribution of hydrocarbon accumulations, prediction and evaluation of new significant oil and gas discoveries.

The geological structure of the Siberian Platform can be divided into two structural levels, a lower structural level, represented by the crystalline basement composed mostly of Archean and Early Proterozoic metamorphic rocks and granitoids, and an upper structural level, represented by a sedimentary platform cover consisting of weakly dislocated sedimentary rocks ranging in age from Lower Riphean (from Mesoproterozoic) to Cenozoic (Malich et al., 2002). The platform cover can be further subdivided into two large structural stages. The lower stage includes partly faulted terrigenous, argillaceous and high-carbonaceous (Filiptsov and Starosel'tsev, 2009) and carbonate rocks of Riphean age and (Malich et al., 2002; Mel'nikov et al., 2008). The total thickness of the Riphean strata varies considerably, reaching 6 km and more in the axial parts of rift-like troughs (Irkineeva—Kotui and others), and thin or pinch out completely at the crest of the Nepa–Botuoba anteclise (Bazhenova et al., 2011; Malich et al., 2002). The upper structural stage of the platform cover comprises a series of sedimentary successions that are almost not faulted or folded and range from Vendian to Cenozoic in age and from 0 to a few kilometers in thickness (Kontorovich et al., 2009). This section containing no major unconformities is represented by various sedimentary and volcano–sedimentary units at the Lower Triassic structural level (Malich et al., 2002).

Sediments from different parts of the Riphean section overlie a pre-Vendian erosional surface of deformed upper sedimentary beds of the Siberian Platform where they underwent the processes of supergene disintegration, chemical and physical weathering and karstification. This significantly improved reservoir properties of Riphean rocks in the vicinity of the pre-Vendian erosional surface.

Petroleum systems (PS) in the southern part of the Siberian Platform are paragenetically related to the Riphean strata, Vendian terrigenous—carbonate strata, and Cambrian salt-bearing carbonate strata (Kerimov et al., 2015a,b,c,d; Kerimov et al., 2016 a,b; Kerimov and Rachinsky, 2016; Mel'nikov et al., 2008, Filiptsov and Starosel'tsev, 2009; Kerimov et al., 2015a; Kerimov et al., 2016; Rachinsky and Kerimov,

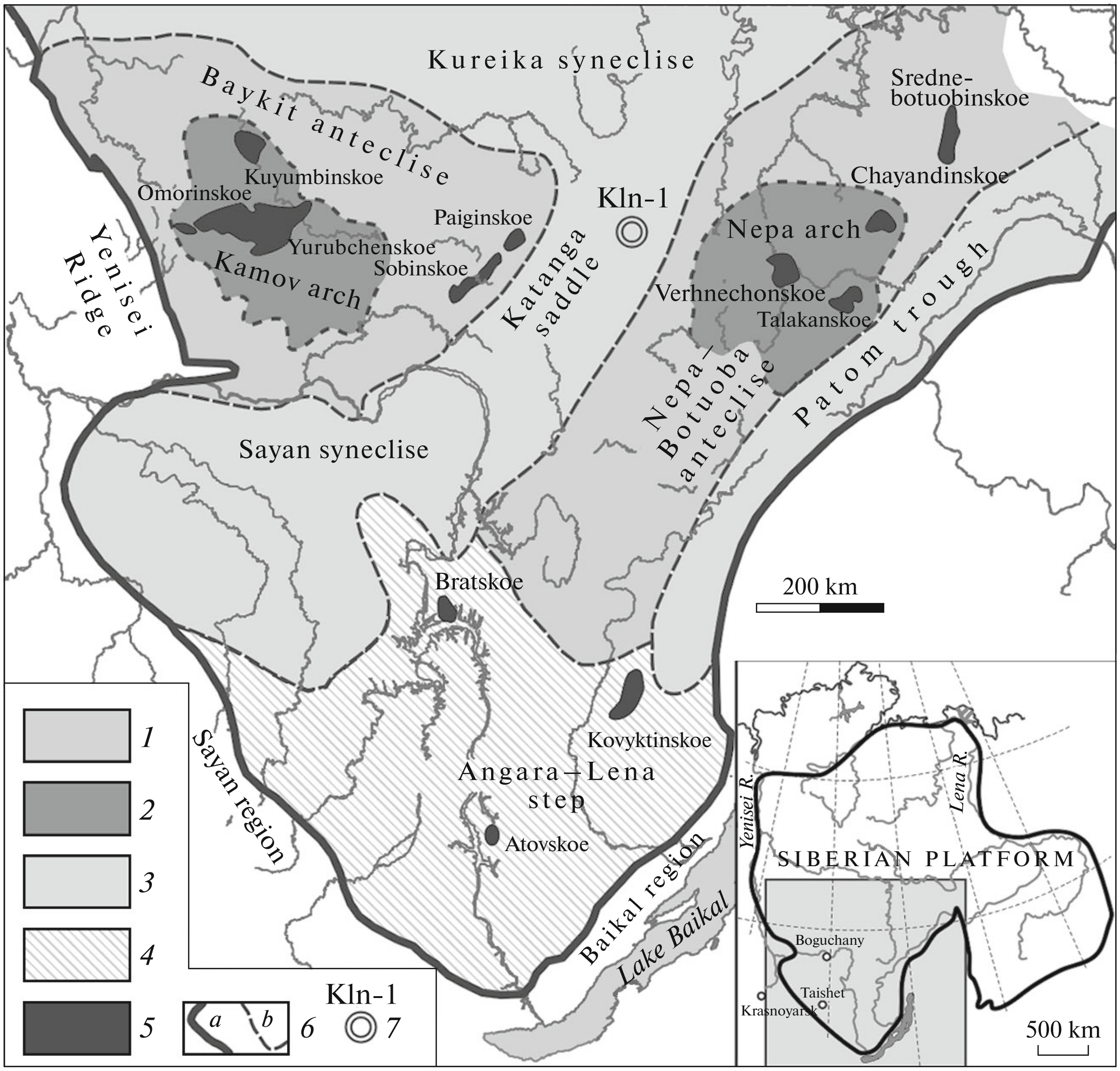

Fig. 1. Sketch map showing major structural elements of the sedimentary cover in the southern part of the Siberian Platform, modified from Mel'nikov et al. (2008), Filiptsov and Starosel'tsev (2009), Howard et al. (2012). *1-2*—positive structures: *1*—anteclises, *2*—anteclise crests; *3*—negative structures: syneclises and saddles; *4*—Angara–Lena step; *5*—oil, gas, and gas/condensate fields; *6*—geologic boundaries: Siberian Platform (*a*) and major structural elements of the sedimentary cover (*b*); *7*—location of Kulindinskaya-1 (Kln-1) well.

2015; Howard et al., 2012). Riphean high-carbonaceous formations containing up to 13.62% C_{org} (Filiptsov and Starosel'tsev, 2009) are the main source rocks in these PS, whereas the main reservoirs are represented by Riphean rocks resting on the pre-Vendian erosional surface that underwent disintegration, chemical and physical weathering and karstification at the pre-Vendian erosional stage, as well as onlapping Vendian high-porosity (and high-permeability) terrigenous beds of the basal part of the Vendian–Cenozoic sedimentary cover of the Siberian Platform. At the same time, regional seals are resented by salt-bearing rocks forming the topmost part of the Upper Vendian section and several layers in the Lower Cambrian section of the Vendian—Cenozoic sedimentary cover.

It was shown that oils from the Baykit anteclise are genetically associated with Riphean source rocks (Bazhenova et al., 2011; Drobot, 1988). However, Vendian terrigenous and carbonate rocks at the base of Vendian–Cenozoic section are found to locally onlap the crystalline complexes in the crestal parts of the Nepa–Botuoba and Baykit anteclises, which host large hydrocarbon accumulations (Malich et al., 2002). This suggests that source rocks are absent directly beneath the reservoir beds and hydrocarbons may have accumulated through lateral migration.

Table 1. Hydrocarbon group-type composition of organic matter from core samples collected in the Kulindinskaya-1 well

Sample no.	Depth, m	Lithology	Age	HC group-type composition of sedimentary OM, wt %		
				saturated HC	aromatic HC	resins
1	1843.78	Bituminous, dolomitic, cryptocrystalline limestones	$Є_{1us}$ Usol'e Fm. (Osa horizon)	52.1	27.6	20.3
2	1954.8	Dolomites, locally calcareous, argillaceous	V-$Є_1$ Tetere Fm. (Ust–Kut horizon)	47.9	22.3	29.8
3	2197.2	Variegated siltstones and mudstones	V_{2vn} Vanavara Fm.	37.5	32.2	30.3
4	2296.4	Mudstones and quartz siltstones	R_{2br} Berei Fm.	45.1	20.8	34.0

Therefore, hydrocarbons accumulated at the crests of the Nepa–Botuoba and Baykit anteclises may have generated either in the Katanga saddle, separating them both spatially and structurally, or in the adjacent syneclises. To verify this assumption, we analyzed the dispersed organic matter (DOM) from Riphean, Vendian, and Cambrian rocks encountered in the Kulindinskaya-1 well (Kln-1 well), which was drilled by RN-Exploration in 2012 within the Katanga saddle. This well has penetrated the entire stratigraphy and intersected the underlying 20 m of the Archean—Early Proterozoic crystalline basement. The lower portion of the drilled section (up to 400 m thick) was cored with nearly 100% core recovery. Core samples collected at different stratigraphic levels were used for DOM analysis.

Samples were first analyzed using the Rock-Eval pyrolysis method (Gordadze et al., 2015; Kerimov et al., 2015 b,c,d,e,f,g,h). The samples collected from the 1834–1846 m interval, which is correlated with the Osa horizon of the Lower Cambrian Usol'e Formation, were classified as "good", "very good", and "excellent" sources rocks based on their C_{org} content (Tissot and Welte, 1981; Kerimov et al., 2014; Peters, 1986). The samples are thermally immature (T_{max} < 435°C) and contain Type II or III kerogen indicating that these rocks did not reach the oil window. The pyrolysis data on immature samples collected from the Osa horizon (Kln-1 well) show good agreement with the results of micropaleontological analysis of core samples collected from the same well at 2188–2201 m (some 350 m below the Osa horizon). The color alteration index (from light yellow to dark orange) of the best preserved organic-walled microfossils indicates that the maximum temperature reached by this fossil-rich interval not higher than 80–100°C (personal communication by V.N. Sergeev, Geological Institute, Russian Academy of Sciences).

A study of biomarkers (*n*-alkanes, isoprenanes, steranes, and terpanes) and diamondoid hydrocarbons (adamantanes and diamantanes) in bitumens extracted from four samples classified as good potential source rocks based on Rock-Eval pyrolysis data was performed to find a genetic link between Precambrian oils from the Baykit anteclise (Kuyumbinskoe, Paiginskoe fields, etc.) and extractable organic matter in rocks of the Katanga saddle penetrated in Kln-1 well.

The methods of bitumen extraction (chloroform extraction of organic matter), chromatography-mass spectrometry (analysis of high-molecular-weight hydrocarbon biomarkers: steranes and terpanes, as well as diamondoid hydrocarbons), gas–liquid chromatography (analysis of *n*-alkanes and isoprenanes in the chloroform extracted bitumens), and high-performance liquid chromatography (hydrocarbon group-type analysis of saturated and aromatic HC, resins, and asphaltenes) were described in detail in previous studies (Bazhenova and Alef'ev, 1998; Gordadze, 2002; Gordadze and Matveeva, 2001).

Results and discussion. The results of hydrocarbon group-type analysis of rock samples are shown in Table 1. Most samples are characterized by low and moderate bitumen content (chloroform-extractable bitumen A) (0.02–0.04% in samples 2–4), except for sample 1, which has the high content of chloroform-extractable bitumen A (0.16%). The relative distributions of n-alkanes and isoprenanes (pristane and phytane) are shown in Fig. 2a and b, respectively. The geochemical characterization of the DOM in rocks based on the distributions of n-alkanes, isoprenanes, steranes, and terpanes is shown in Figs. 3a–3c, respectively.

The DOM in rock samples collected from Kln-1 well is characterized by anomalously high C_{13} *n*-alkane concentrations, which vary from 30.9–34.0% in samples 1–3 to 14.6% (n-$C_{13}H_{28}$) in sample 4. The n-C_{13}/(n-C_{12} + n-C_{14}) ratio varies from 1.8–2.8 in samples 1–3 to 1.0 sample 4.

Samples 1–2 show a distinct odd-to-even C_{15} and C_{17} *n*-alkane distribution with the (n-C_{15} + n-C_{17})/2 · n-C_{16} values of 1.1–1.2.

The lowest pristane/phytane ratios in samples 1, 3, and 4 (0.8–1.1) are indicative of reducing marine conditions in sediments, whereas slightly higher Pr/Ph

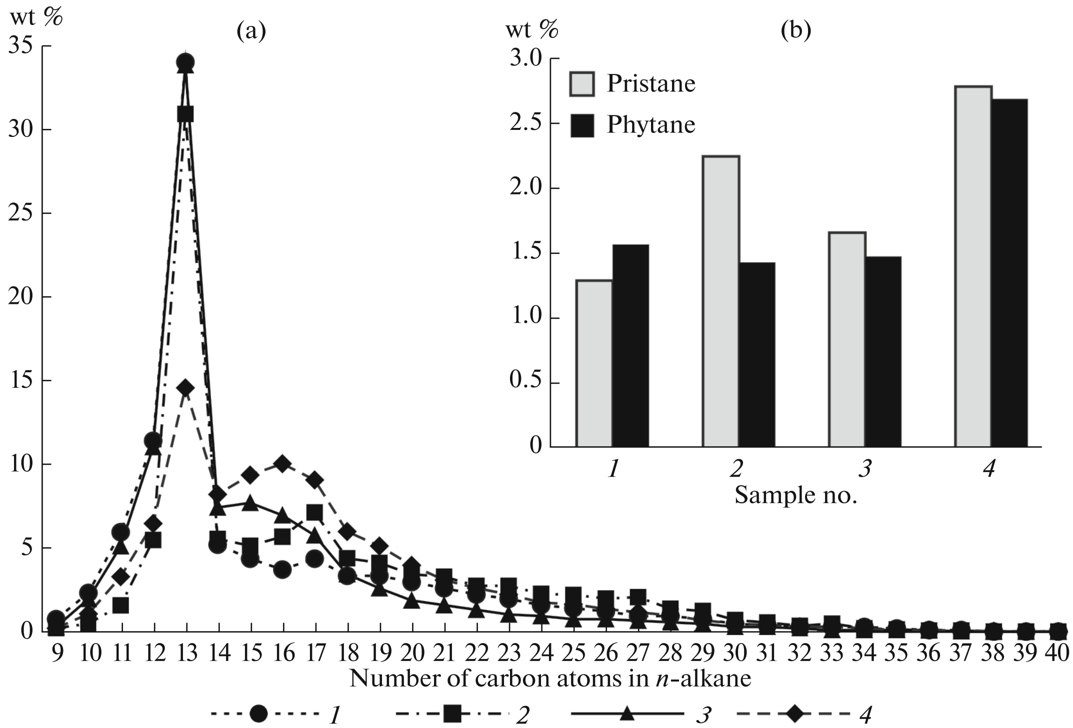

Fig. 2. Relative distribution of n-alcanes (C_9–C_{40}) (a) and isoprenanes (pristane and phytane) (b) in DOM (dispersed organic matter) of rocks. Samples numbers *1–4* correspond to the ones in Table 1.

values (1.6) detected in sample 2 testify to suboxic conditions or thermal alteration of organic matter.

The bitumens from samples 1, 2, and 4 are characterized by the presence of the homologous series of 12- and 13-methylalkanes, indicators of the ancient (Riphean) crude oils from East Siberia (for example, oils from the Kuyumbinskoe, Yaraktinskoe fields, etc. (Bazhenova and Alef'ev, 1998)) and squalane, an irregular isoprenane (2, 6, 10, 15, 19, 23-hexamethyltetracosane).

The absence of 12- and 13-methylalkanes and squalane in sample 3 suggests an origin of bitumens from a younger source, which is not typical of crude oils from ancient strata of East Siberia. This also suggests the existence of different sources for bitumens in sample 3 (Vanavara Formation) and samples 1, 2, and 4.

The good reservoir quality of the Vanavara Formation (sample 3) can explain the presence of younger oils, which may have been generated by and expelled from younger source rocks. However, more samples of the Vanavara Formation rocks are needed for more reliable interpretation of potential source rocks. In addition, an integrated study of all tectonic, geochronothermobarometric, and geochemical aspects is required in order to define appropriate migration pathways and to better evaluate the possible oil–source rock correlation.

Steranes. Crude oils from Precambrian rocks of East Siberia can be best correlated with extractable bitumen from sample 1, as indicated by the regular C_{28}/C_{29} sterane ratio of 0.3 (Fig. 2b), whereas in Precambrian oils from the Kuyumbinskoe field, this ratio is equal to 0.35 (Gordadze, 2002).

In the remaining samples (2–4), the C_{28}/C_{29} sterane ratio is higher, ranging between 0.55 and 0.64, which corresponds to organic matter from Devonian and Carboniferous rocks (Grantham and Wakefie1d, 1988). It should be noted that this index can only be applied to marine organic matter (except for Precambrian rocks), because terrestrial organic matter is characterized by higher C_{29} sterane abundances. However, in this case Pr/Ph will be greater than 3. The $C_{27} : C_{28} : C_{29}$ sterane distributions in samples 2–4 are nearly identical (being on average 31 : 26 : 43%) as compared to sample 1 (18 : 19 : 63%).

Pregnanes (C_{21}–C_{22}) are almost absent in Precambrian oils (Gordadze and Matveeva, 2001). Bitumens from samples 1, 2, and 4 have the lowest $\Sigma(C_{21}–C_{22})/\Sigma(C_{27}–C_{29})$ ratios (average 0.09) as compared to sample 3 (0.24).

The equilibrium maturity coefficients K^1_{mat} and K^2_{mat} are equal to 0.55 and 0.85–0.86, respectively. Although these coefficients in all samples analyzed do not represent equilibrium values (Fig. 3b), the dispersed organic matter in these rocks is interpreted to be within the oil window maturity range. The highest value of K^2_{mat} calculated from C_{29} steranes was

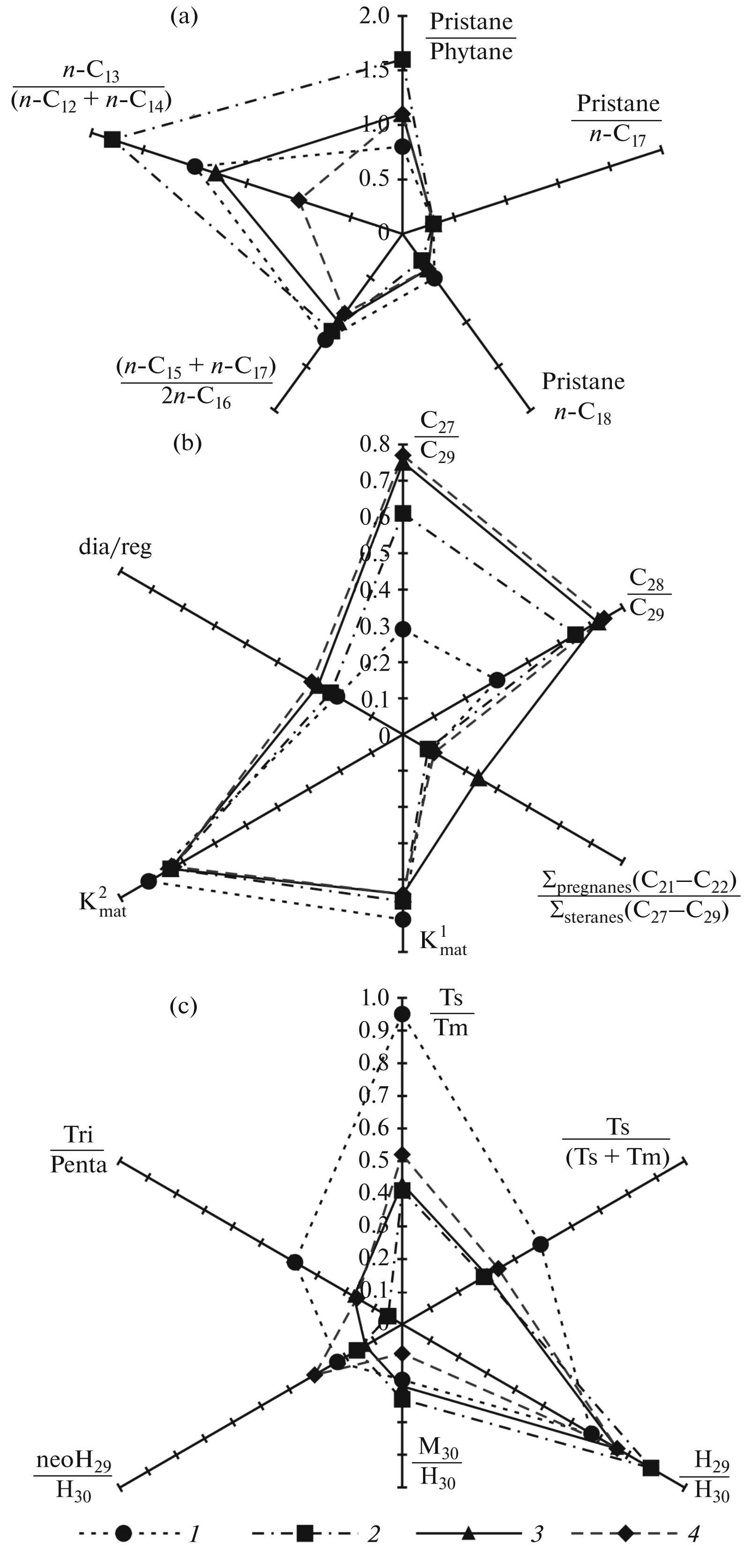

Fig. 3. Geochemical characterization of DOM based on the distributions of (a) *n*-alkanes and isoprenanes, (b) steranes, and (c) terpanes. Samples numbers (*1–4*) are the same as in Table 1.

Table 2. Contents of C_{11}–C_{13} adamantanes and C_{14}–C_{15} diamantanes in sedimentary organic matter (GC–MS data)

Hydrocarbons	Sample no.			
	1	*2*	*3*	*4*
	wt %			
C_{11} adamantanes				
1-methyladamantane	68.2	63.8	59.4	59.0
2-methyladamantane	31.8	36.2	40.6	41.0
C_{12} adamantanes				
1-ethyladamantane	7.0	6.9	5.9	8.0
2-ethyladamantane	11.0	8.3	10.9	13.0
1,3-dimethyladamantane	25.0	22.2	19.8	19.6
1,4-dimethyladamantane (cis)	19.0	22.2	20.8	18.8
1,4-dimethyladamantane (trans)	16.0	21.5	19.8	17.4
1,2-dimethyladamantane	22.0	18.8	22.8	23.2
C_{13} adamantanes				
1-ethyl-3-methyladamantane	17.8	14.8	14.2	18.6
1,3,5-trimethyladamantane	16.5	15.7	11.8	15.3
1,3,6-trimethyladamantane	17.8	25.2	23.5	18.6
1,3,4-trimethyladamantane (cis)	26.0	21.7	24.7	23.7
1,3,4-trimethyladamantane (trans)	21.9	22.6	25.8	23.8
C_{14}–C_{15} diamantanes				
Diamantane	47.3	22.5	33.8	27.0
4-methyldiamantane	23.5	30.5	28.2	27.0
1-methyldiamantane	11.8	22.5	14.1	28.8
3-methyldiamantane	19.4	24.5	23.9	18.0

Table 3. Relative abundances of adamantanes and diamantanes in organic matter from core samples

Sample no.	Relative abundance of stable isomers, %			
	adamantanes		diamantanes	
	$\frac{1MA}{\sum MA}$	$\frac{1,3\partial MA}{\sum \partial MA}$	$\frac{1,3,5mMA}{\sum mMA}$	$\frac{4MD}{\sum MD}$
1	68	30	20	41
2	64	26	18	39
3	59	24	14	43
4	59	25	19	37

detected in sample 1 (0.81) while it is equal to 0.73–0.74 in samples 2–4. The low dia/reg sterane ratio in samples 1 and 2 (0.21 and 0.23) is associated with carbonate source rocks and that in samples 3 and 4 (0.27 and 0.29) is used as evidence of petroleum generated from argillaceous–carbonate sources.

Terpanes. All the samples analyzed are characterized by generally low maturity-dependent Ts/Tm ratios, ranging from 0.93 in Precambrian oils to 0.45 in samples 2–4, except for sample 1 where this ratio is equal to 0.95 (Fig. 3c).

The ratio of cheilantanes to hopanes (tri/penta) in samples studied varies widely from 0.05 to 0.38. The highest values of this ratio (0.38) are observed in sample 1 while the lowest values (0.13) are found in samples 2–4. This ratio is equal to 0.59 for the Precambrian oils from the Kuyumbinskoe field (Gordadze, 2002).

Diamondoid hydrocarbons (adamantanes and diamantanes). Proto-adamantanes are present in large amounts in samples 2 and 4, only in minor amounts in samples 1 and 3, and are almost absent in samples 2 and 4. The adamantane and diamantane distributions in organic matter in samples 1 and 3 are nearly identical to those of Precambrian oils (Tables 1–3).

CONCLUSIONS

The geochemical study of hydrocarbon biomarkers (steranes, terpanes, *n*-alkanes, 12- and 13-methylalkanes, and isoprenanes) and diamondoid hydrocarbons from Riphean–Lower Paleozoic rocks collected in Kln-1 well shows that organic matter in all the samples analyzed is within the oil window maturity range based on the C_{29} sterane distribution. Because this conclusion is not supported by pyrolysis and micropaleontological data for rocks collected in Kln-1 well, it seems most likely that some bitumen components were derived from the rock matrix while the others (including those within the oil window maturity range) originated from external sources (via migration of oil-rich fluids). Samples 1 and 3 are characterized by an anomalously high relative abundance of n-tridecane, which is normally absent in crude oils. Samples 2 and 4 appear to contain epigenetic OM.

The homologous series of 12- and 13-methylalkanes is present in all samples analyzed, except for sample 3, which is typical of the Precambrian oils from the Siberian Platform. However, based on the C_{27} : C_{28} : C_{29} sterane distribution, organic matter extracted from sample 1 can be assigned a Precambrian age, although this sample was collected from the Cambrian section (Osa horizon of the Usol'e Fm.).

Comparison of biomarker, adamantane, and diamantane distributions in organic matter from the studied rocks shows that the biomarker distribution is sample 1 is almost identical to that of the Precambrian oils from the Kuyumbinskoe field.

Extractable bitumens from the Osa horizon (sample 1) contain exogenous components of organic matter that may have been derived from the source rocks of the Kuyumbinskoe field. Therefore, the geochemical data suggest a considerable similarity in the molecular compositions of extractable bitumens from rocks penetrated in Kln-1 well and Precambrian oils from the Kuyumbinskoe field. These results indicate the possible migration of hydrocarbon fluids to the crest of the Baykit anteclise from the Katanga saddle or via this saddle from adjacent syneclises.

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