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Geochemistry of Neoprotherosoic organic matter in the southeast of the Siberian platform

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Abstract. In the Neoproterozoic of the Aldan-Maya depression, rocks of the Malga, Tsipanda and Kumakh Formations (MF, TF and KF) are rich in organic matter (OM) and can be classified as source rocks. High generation potential is characteristic of the MF and TF (267–511 mg HC/g TOC). The thermal maturity level of OM corresponds to mesocatagenesis substage (T_{max} is 438–443 ºC). Based on the biomarker distribution, the biological precursors of the OM in these formations are: prokaryotes, including cyanobacteria (hopanes, acyclic biomarkers) and to a lesser extent eukaryotes (the presence of steranes) that lived in the Precambrian (the presence of 12- and 13-monomethylalkanes) marine basin (absence of continental biomarkers, the distributions of acyclic alkanes and steranes) with clayey sedimentation (high content of diasteranes). Due to the absence of steranes in some samples (paucity of eukaryotes in the source OM), the MF is likely to have been partly deposited before the emergence of eukaryotes. Most samples have elevated concentrations of low-molecular tricyclanes ($2C_{19-20}/C_{23-26}>1$), which can be attributed to the specificity of the source biota, although it is not typical of the marine OM. A rare homologous series of 2,7-dimethylalkanes that has been recently found by other researchers in some Precambrian strata is present in several samples of the MF and KF and totally absent from all samples of the TF. This may indicate the differences in its biota, depositional environments or its evolution during diagenesis and catagenesis. A distinctive feature of the TF is the high content of ethylcholestanes ($C_{29}/C_{27}=2.5$).

Keywords: Siberian Platform, Precambrian, oil source formations, organic geochemistry, biomarker hydrocarbons

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Introduction

The relevance of research in the organic matter rich source rocks and molecular studies of bitumens and naphthides from Riphean deposits on the Siberian Platform is corroborated, in particular, by the need to use reliable input data for petroleum system analysis, as well as for evaluation of petroleum resources and factors controling petroleum entrapment and leakage (migration out of the reservoir). Most of the Precambrian and Lower Cambrian oils of the Siberian Platform, thick organic matter (OM) rich Neoproterozoic (Riphean) strata in its periphery and margins are considered to be oil-prone (Nepa-Botuoba anteclise..., 1986; Kontorovich et al., 1994a, b, 1996, 1999, etc.). However the fact that the organic matter from Precambrian deposits is dominantly overmature provides additional challenge to interpretation of geochemical results and findings, to the extent of their inappropriateness for genetic and catagenetic diagnostics. Precambrian formations containing organic matter unaffected by metamorphism are widespread in the eastern Siberian Platform and are therefore of great interest in respect to their study by the organic geochemistry methods. Neoproterozoic formations containing moderately transformed OM and localized within the Aldan-Maya depression (southeastern Siberian Platform) were chosen to be the object of this study. Our geochemical study encompasses the succession of formations (Decisions of the All-Union Stratigraphic..., 1983) which include: the Kerpyl Group (Malga and Tsipanda Formations) (Middle Riphean), the Lakhanda Group (Kumakh, Mil'kon, Nel'kan, Ignikan Formations) (Upper-Riphean), and the Uy Group (Kandyk Formation); their ages are 1100–1000 Ma, 1000–850 Ma, and 850–650 Ma, respectively (Khomentovskiy, 1996). This study complements previous efforts to investigate the Precambrian organic matter in the Aldan-Maya depression (Bazhenova et al., 1981; Matvienko, Sobolev, 1984; Sobolev, 1987;

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Bazhenova, 2010; Mozhegova, 2010; Bazhenova et al., 2011, 2014; Dakhnova et al., 2013, 2014; Parfenova et al., 2014; Safronov et al., 2015; Shiganova et al., 2015; Suslova et al., 2015, 2017; Sobolev et al., 2017; Parfenova, Suslova, 2019, etc.).

The composition of biomarker hydrocarbons of bitumens contained in rocks is function of the biota composition, depositional setting, processes of diagenesis and catagenesis, and fluids in mixed migration routes. This research results attest to a significant influence of the migration phenomena on the composition of bitumens in regional Riphean strata.

Research object and methods

The Malga, Tsipanda, Kumakh, Mil'kon, Nel'kan, Ignikan, and Kandyk Formations outcropping in the Maya riverbanks were sampled at different times within the Aldan-Maya depression in the southeastern sector of the Siberian Platform and analyzed at the Laboratory of Petroleum Geochemistry at the Institute of Petroleum Geology and Geophysics, Siberian Branch of the Russian Academy of Sciences (IPGG SB RAS). The geochemical studies involved determination of concentrations and isotopic signatures and pyrolytic characteristics of organic carbon (TOC), and concentrations of bitumens soluble in chloroform (b_{chl}), along with the analysis of bitumen group composition, and hydrocarbons (HCs) composition using gas-liquid chromatography and chromatomass spectrometry among other methods. All the methods are traditionally used at the IPGG SB RAS Laboratory of Petroleum Geochemistry and their detailed description is provided in a recent publication (Suslova et al., 2017), where the data obtained from Rock-Eval pyrolysis and carbon isotope analyses of the organic matter were compared with the characteristics of saturated biomarker HCs obtained by chromatography-mass spectrometry to determine the origin of sources, diagenetic environments, and transformation of the organic matter during catagenesis. Some results of the analyses of this collection were reported as separate fragments in the proceedings of several conferences (Timoshina et al., 2010; Kontorovich et al., 2011a, 2011b, 2012, 2013; Timoshina et al., 2015), and are integral to comprehensive interpretations of the geochemical indicators.

Results and discussion

The studied collection included several organic carbon-rich (TOC ≥1) samples, specifically: mudstone (2 samples, 5.5 and 13.6 % per rock) and siltstone (2 samples, 2.5 and 3.7 % per rock) from the Malga Fm; mudstone from the Tsipanda Fm (3.3 % per rock); mudstones (1.0 and 1.1 % per rock) from the Kumakh Fm and sandstone from the Kandyk Fm (1.2% per rock); other samples contained OM < 1% per rock (Table 1).

The ^{12}C isotope-rich organic matter was established in the Malga Formation ($\delta^{13}C$ = (−33.2) – (−31.5) ‰) (Suslova et al., 2017), as well as in the Kumakh and

No.	Age	Group	Formation	Rock	TOC, % per rock	$\delta^{13}C$, ‰	Pyrolysis				
							mgHC/g rock		HI, mg CH/g TOC	T_{max}, ºC
							S1	S2		
1	R_2	Kerpyl	Malga	mudstone	5.5	–	1.1	28.1	511	441
2				maltha	–	–	–	–	–	–
3				kerit	–	–	–	–	–	–
4				limestone	0.6	–	–	–	–	–
5				siltstone	3.7	–	–	–	–	–
6				siltstone	2.5	–	–	–	–	–
7				mudstone	13.6	–	–	–	–	–
8				mudstone	0.1	–	–	–	–	–
9				mudstone	0.1	–	–	–	–	–
10				mudstone	0.4	–	–	–	–	–
11			Malga-Tsipanda	limestone	0.02	–	–	–	–	–
12				limestone	0.5	–	0.1	0.5	111	443
13			Tsipanda	limestone	0.7	–	0.3	2.6	395	442
14				limestone	0.8	–	0.1	2.0	267	443
15				mudstone	3.3	–	0.1	14.5	434	430
16	R_3	Lakhanda	Kumakh	mudstone	1.0	−32.9	0.1	0.7	68	442
17				mudstone	1.1	−32.6	0.1	0.8	71	438
18			Milkon	limestone	0.03	–	–	–	–	–
19				dolomite	0.2	–	0.1	0.4	205	430
20			Nelkan	dolomite	0.03	–	–	–	–	–
21			Ignikan	limestone	0.1	−30.6	3.9	10.2	161	444
22		Uy	Kandyk	dolomite	0.1	–	–	–	–	–
23				dolomite	0.1	–	0.03	0.1	148	450
24				limestone	0.1	–	0.1	0.2	142	456
25				limestone	0.1	–	0.1	0.2	191	443
26				sandstone	1.2	–	0.1	2.8	230	432

Table 1. Concentrations, pyrolysis and isotopic composition of organic carbon in Riphean rocks of the Aldan-Maya depression

Ignikan Formations ($\delta^{13}C$ = (−32.9) – (−30.6) ‰) (Table 1). The generation potential (hydrogen index HI >160 mg HC/g TOC) was estimated as high in several samples of the studied collection which includes: 1 sample of the Malga mudstone (TOC >1), all of the Tsipanda samples (among them 1 sample with TOC >1), the Mil'kon dolomite (TOC <1), the Ignikan limestone (TOC <1), 1 sandstone (TOC >1) and 1 limestone sample (TOC <1) from the Kandyk Fm (Table 1, Fig. 1).

The three samples localized in the region of Type III kerogen (Fig. 1) may account for high catagenesis conditions reducing the value of HI or/and admixture of younger migrated solid bitumens. The sample collection used in this study, which primarily focuses on biomarker HCs, has only 1 sample from the Malga Formation studied by Rock-Eval pyrolysis, however results for other samples from the Malga Formation (their bitumens are not investigated) suggest high generation potential of the Malga Fm organic matter (Fig. 1 shows these samples as unnumbered dots). Besides, higher HI values characteristic of the Malga Fm samples are practically independent of TOC concentrations (Fig. 2).

The obtained pyrolytic and biomarker characteristics allowed to infer that the maturity level of the Malga organic matter corresponds to mesocatagenesis grades MC_1^1-MC_1^2, which might reach MC_2 in the deepest buried zones (Matvienko, Sobolev, 1984; Sobolev, 1987; Dakhnova et al., 2013; Bazhenova et al., 2014; Suslova et al., 2015, 2017, Chalaya et al., 2015). Judging by the pyrolytic characteristics (T_{max} – temperature of maximum of the S2 peak of HC yield, the kerogen part of the OM), it can be inferred that the samples of this collection are suitable for diagnostics (TOC >1 %), and that the organic matter from the Malga and Kumakh Formations transformed during MC_1^2 (T_{max} = 438–442°C), while maturity level of OM from Tsipanda and Kandyk Formation is lower. Given the presence of bitumen is possible in some samples, the HI values may be overestimated, and T_{max} underestimated.

High concentrations of bitumens (b_{chl} > 0.2 % per rock) were determined in 3 samples from the Malga Formation and in Kandyk Fm sandstone (Table 2), however, their high bitumen coefficient β (9–16 %) indicates the epigenetic origin of bitumen, except the organic carbon- and autochthonous bitumen-rich Malga Fm mudstone (β = 2.1 %). Despite the fact that abundance of bitumen is observed throughout the Meso- and Neoproterozoic section of the Uchur-Maya region (Parfenova and Suslova, 2019), the remaining samples of the studied collection are bitumen-depleted (b_{chl} < 0.1% per rock, an average 0.02% per rock), being either generally allochthonous (β > 10 %) or mixed (5 % < β < 10 %), while autochtonous (β < 5 %) bitumens were encountered in 3 mudstone samples from the Malga Fm, in 1 limestone from the undifferentiated Malga and

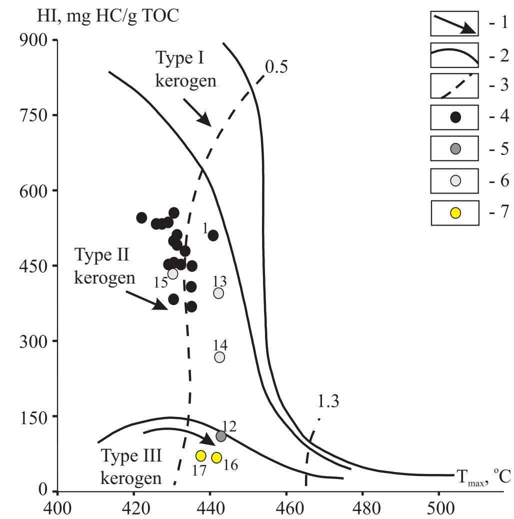

Fig. 1. HI-T_{max} diagram for the organic matter of potential hydrocarbon source of the Maya Rv.: 1 – the direction of variations in HI and T_{max} values during catagenesis, 2 – the lines delimiting the maximum value of the hydrogen index for three types of organic matter (I – sapropelic lacustrine, II – sapropelic marine, III – terrestrial associated with higher land plants), 3 – R^o_{vt}, isolines delimiting the main zone of oil generation; Formations: 4 – Malga, 5 – Malga and Tsipanda, 6 – Tsipanda, 7 – Kumakh; the numbers near the circles correspond to the samples numbering in Table 1

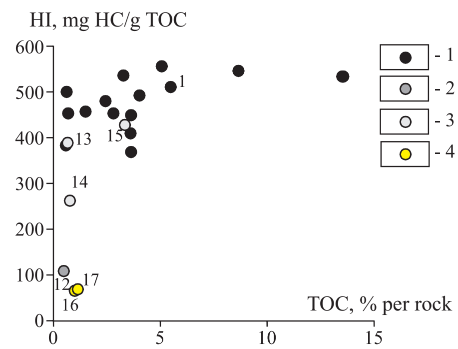

Fig. 2. A relationship between TOC concentrations and hydrogen index (HI) values for the organic matter of potential source rocks from outcrops of the Maya Rv.: Formations: 1 – Malga, 2 – Malga and Tsipanda, 3 – Tsipanda, 4 – Kumakh; the numbers near the circles correspond to the samples numbering in Table 1

Tsipanda Fms, in all of the samples from the Tsipanda and Kumakh Fms and in 1 Kandyk Fm dolomite.

According to this research results (Table 2) and Bazhenova (Bazhenova et al., 2014), the Malga Fm bitumens are ^{12}C-rich ($\delta^{13}C_{bit}$ = (−33.4) – (−31.7) ‰). Samples from the Malga and Tsipanda Formations are the most asphaltene-rich (>10%), while the remaining bitumens are represented primarily by hydrocarbons and resins (Fig. 3).

In the *n*-alkanes series identified from the gas-liquid chromatogram, the maxima account mainly for nC_{17-19}, (Table 3, Fig. 4) in only 3 samples,with the maximum being shifted to a region of high-molecular compounds

No	b_{chl}, % per rock	β, %	Group composition, % per bitumen: Saturated HCs	Aromatic HCs	Resins	Asphaltenes	Sum of resins and asphaltenes	$\delta^{13}C$, ‰
1	–	–	6.5	28.0	32.4	33.0	65.4	−32.7
2	–	–	28.9	22.0	20.7	28.5	49.2	−31.7
3	–	–	3.9	7.9	25.1	63.1	88.2	−33.4
4	0.07	8.4	24.1		58.8	17.1	75.9	–
5	0.44	9.0	17.6	18.1	46.4	17.9	64.3	–
6	0.33	10.1	14.6	12.8	50.0	22.6	72.6	–
7	0.38	2.1	11.0	24.3	52.5	12.3	64.7	–
8	0.001	1.5	27.1	6.8	66.2	not determ.	66.2	–
9	0.018	11.0	45.0	9.5	43.9	1.6	45.5	–
10	0.002	0.4	28.7	7.8	63.5	not determ.	63.5	–
11	0.002	6.4	17.1	2.3	80.6	not determ.	80.6	–
12	0.014	2.1	26.5	6.8	66.7	not determ.	66.7	–
13	0.033	3.8	9.2	16.1	42.7	32.1	74.8	–
14	0.029	2.8	12.2	14.2	46.6	27.1	73.7	–
15	0.094	2.1	10.4	17.5	60.9	11.3	72.2	–
16	0.027	2.1	41.1	12.0	45.1	1.9	46.9	–
17	0.017	1.1	19.7	25.0	48.6	6.2	54.8	–
18	0.003	7.3	31.0	12.9	56.0	not determ.	56.0	–
19	0.036	12.9	38.0	3.1	48.3	10.6	58.9	–
20	0.002	5.3	31.0	13.7	55.4	not determ.	55.4	–
21	0.03	16.3	62.4	13.4	21.9	2.4	24.3	–
22	0.005	4.8	45.4	5.1	49.5	not determ.	49.5	–
23	0.011	11.3	50.8	6.4	39.7	3.1	42.8	–
24	0.011	6.8	67.8	3.0	26.0	3.2	29.2	–
25	0.013	10.9	39.4	13.7	47.0	not determ.	47.0	–
26	0.26	16.0	47.4	7.9	44.4	0.2	44.6	–

Table 2. Concentrations, group and carbon isotopic composition of bitumens in Riphean rocks from the Aldan-Maya depression. The numbers marked yellow denote bitumens and allochthonous bitumens with admixture according to the bitumen coefficient β (> 5 %).

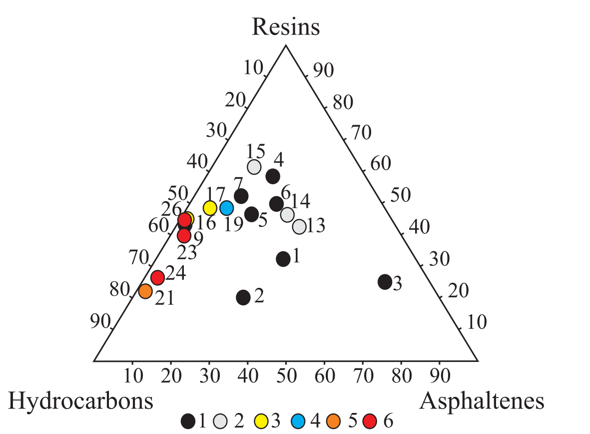

Fig. 3. Trigonogram for group composition of Riphean bitumens from the Maya Rv. outcrops, Formations: 1 – Malga, 2 – Tsipanda, 3 – Kumakh, 4 – Mil'kon, 5 – Ignikan, 6 – Kandyk (The samples numbers correspond to the sample numbering in Table 2)

nC_{23-27} represented by maltha from the Malga Formation, the OM-depleted limestone from the undifferentiated Malga and Tsipanda Formations and the Kandyk Fm dolomite. In the bitumen extracted from limestone of the undifferentiated Malga and Tsipanda Formations odd HCs distinctly prevail over even HCs, which is reflected in CPI (2.0), and therefore, the presence of admixed younger immature terrestrial bitumens is not ruled out. The concentrations of even nC_{28} and nC_{30} in the Mil'kon Fm dolomite being elevated, this suggests either immature carbonate-rich parent organic matter or super-saline environments, as well as high catagenesis conditions (Tissot, Velte, 1981; Peters et al., 2007). In the case of the carbonate sample, the pyrolysis results indicate an immature OM, however bitumen is impregnated by allochthonous terrestrial admixture, admitting therefore availability of other reason. Low but identifiable amounts (5–9 % per acyclic hydrocarbons) of 12- and 13-monomethylalkanes were found in the samples from the Kumakh and Ignikan Formations (Table 3) (Timoshina et al., 2010; Kontorovich et al., 2011a, 2011b, 2013), with the maximum (up to 15%) recorded in the Kumakh Formation (Parfenova and Suslova, 2019). These compounds are viewed as inherent to marine algae and Precambrian cyanobacteria (Petrov, 1984; Peters et al., 2007, etc.). Previously, 12- and 13-monomethylalkanes were detected in oils from Riphean – Lower Cambrian reservoirs of the Siberian Platform (Petrov, 1984; Kontorovich et al., 1996, 1999; Kontorovich et al., 2005; Timoshina, 2004, 2005), and in oils and bitumens of Vendian oil source rocks from Oman (Grantham et al., 1988; Kim, 2004). Earlier, 12- and 13-monomethylalkanes were identified in the Malga Formation (Chalaya et al., 2015), and in its few samples included in the studied collections, only in trace amounts (Table 3). Their concentrations are ranked as low in Upper Riphean Neryuen and Kandyk Formation and Vendian Sardana Formation in Ust-Mayskaya-336

No.	Maximum in n-alkanes	Maximum in acyclic isoprenoids	Ratios of acyclic hydrocarbons					12-,13-monomethyl-alkanes, % per acyclic HC	The presence of 2,7-dimethylalkane series
			Pr/Ph	Pr/n-C_{17}	Ph/n-C_{18}	n-C_{27}/ n-C_{17}	CPI		
1	C_{16}	C_{19}	1.8	0.3	0.2	0.1	1.0	–	
2	C_{23}	C_{20}	0.7	0.5	0.3	11.6	1.2	1.0	
3	–	–	–	–	–	–	–	–	–
4	C_{17}, C_{19}	C_{19}	1.5	0.3	0.2	0.2	1.0	3.8	–
5	C_{16}	C_{19}	2.4	0.3	0.2	0.1	1.0	2.9	–
6	C_{17}	C_{19}	2.0	0.4	0.2	0.2	1.0	2.9	–
7	C_{16}	C_{18}, C_{19}	2.1	0.2	0.1	0.1	1.1	1.9	–
8	C_{17}	C_{20}, C_{19}	1.0	0.3	0.4	0.1	1.2	–	+
9	C_{16}	C_{19}	1.3	0.4	0.6	0.2	1.0	–	–
10	C_{18}	C_{20}	0.8	0.3	0.3	0.1	1.3	–	+
11	C_{27}	C_{20}	0.6	0.5	0.8	1.4	2.0	–	+
12	C_{17}	C_{19}	1.3	0.5	0.4	0.2	1.1	–	+
13	C_{17}	C_{19}	1.4	0.3	0.2	0.1	1.0	–	–
14	C_{19}	C_{20}, C_{19}	1.0	0.2	0.2	0.2	1.0	–	–
15	C_{18}	C_{19}	1.8	0.3	0.2	0.1	1.1	–	–
16	C_{17}	C_{18}	1.4	0.2	0.2	0.1	1.1	7.3	+
17	C_{17}	C_{19}	1.3	0.3	0.3	0.1	1.1	9.0	+
18	C_{17}	C_{19}	1.2	0.4	0.3	0.1	1.1	–	+
19	C_{16}	C_{19}	1.3	0.4	0.5	0.1	0.6	–	+
20	C_{17}	C_{19}	1.1	0.4	0.5	0.3	1.4	–	+
21	C_{19}	C_{19}, C_{20}	1.0	0.2	0.2	0.4	1.0	5.0	–
22	C_{25}	C_{19}, C_{20}	1.0	0.2	0.2	1.4	1.0	–	–
23	C_{17}	C_{19}	1.3	0.2	0.1	0.4	1.0	–	–
24	C_{18}	C_{19}	1.5	0.2	0.1	0.4	1.0	–	–
25	C_{18}	C_{20}, C_{19}	1.0	0.2	0.2	0.4	1.0	–	+
26	C_{17}	C_{19}	1.9	0.5	0.3	0.1	1.1	–	–

Tab. 3. Acyclic saturated biomarker hydrocarbons in Riphean bitumens from the Aldan-Maya depression. The yellow color indicates the sample numbers of bitumens containing an allochthonous admixture according to the bitumen coefficient β (> 5 %).

well (Sobolev et al., 2017), and as trace amounts in the Nelkan rocks (Parfenova, Suslova, 2019).

Low isoprenoid HC to n-alkane ratios Pr/nC_{17} and Ph/nC_{18} observed in all bitumens containing acyclic HC (Table 3) are representive of good preservation of hydrocarbons from biodegradation processes. The Pr/nC_{17} and Ph/nC_{18} ratios are low even in maltha, which suggests that it has a residual-migration origin. Pristane abundance is greater than phytane in most of bitumens, which is twice as more in the Malga Fm (4 samples), Tsipanda (1) and Indican (1); while in 5 samples from the Malga Fm (1), Tsipanda Fm (1), Ignikan Fm (1) and Kandyk Fm (2) pristane and phytane are found to be in equal concentrations (Table 3). Only in 3 bitumen samples from the Malga and Tsipanda Formations phytane prevails over pristane, which is indicative of sapropelic HC. A homological series of 2,7-dimethylalkanes is observed in small concentrations in 10 samples from the collection representing the following Formations: Malga (samples 8 and 10), Malga and Tsipanda (samples 11 and 12), Kumakh (samples 16 and 17), Mil'kon (samples 18 and 19), Nel'kan (sample 20) and Kandyk (sample 25). These are almost all of the studied Formations (except Tsipanda and Ignikan). Figure 5 shows the total ion current chromatogram and mass-chromatograms at m/z 71 and m/z 127 (sample 10, the Malga Formation).

The absence of these compounds in samples from the Tsipanda Formation can be explained by its biota uniqueness, depositional conditions or evolution in diagenesis and catagenesis. Previously, these compounds were identified by Kashirtsev in Vendian mudstones of the Marnino Formation (the Sayan region) (Kashirtsev et al., 2009) and in the organic matter from the Kumakh and Nelkan Formations (Parfenova, Suslova, 2019). In all of the cases, only homologues with an even number of carbon atoms in the molecule have been found.

Among polycyclic isoprenanes, terpanes identified at m/z 191 are significantly higher in all of the samples, than steranes identified at m/z 217 (steranes / terpanes ≤ 0.3 (Table 4)), which indicates a dominantly prokaryotic source of the initial OM (Peters et al., 2007).

Terpanes are often dominated by hopanes; the hopane/tricyclane ratio <1 describe only 2 bitumens from the Malga Formation, bitumens from the Kumakh, Ignikan and Kandyk Formations (Table 4, Fig. 6). Even though high concentrations of hopanes in comparison with tricyclanes may denote contamination by younger terrestrial bitumens or / and low catagenesis conditions, these may equally have resulted from the migration processes in a weakly permeable medium (Kontorovich, Timoshina, 2009), since tricyclane molecules are more migratory. High concentrations of hopanes are found in most samples, including most autochthonous bitumens,

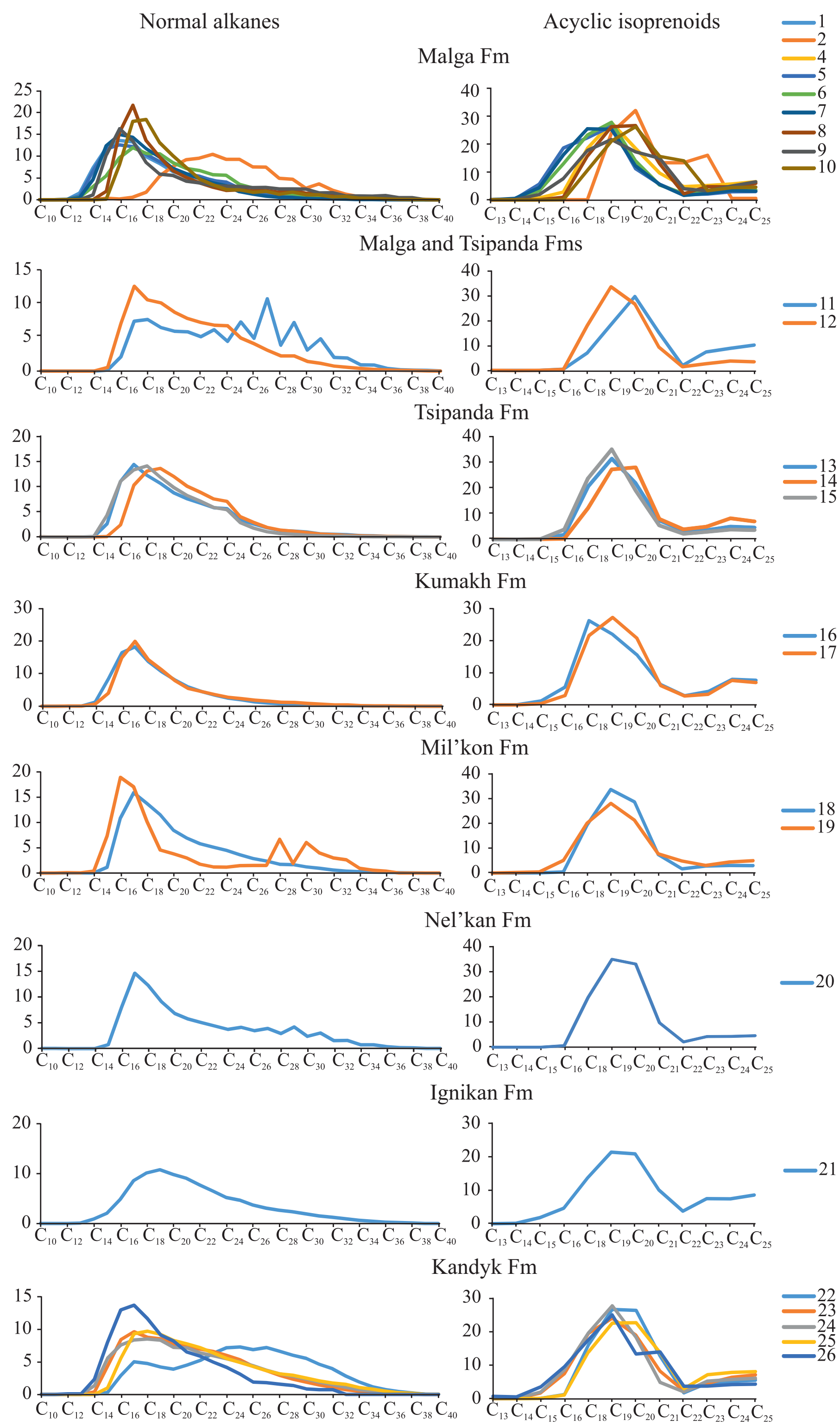

Fig. 4. Acyclic biomarker hydrocarbons in Riphean bitumens from the Maya River outcrops (sample numbers correspond to the sample numbering in Table 3)

which may indicate a dominantly bacterial parent OM. In a hopane series, C_{30} hopane prevails in most of the samples (Fig. 7). The reason why three samples show a maximum at C_{31}, is unclear but most likely it is associated with the specific characteristics of OM, since all the three samples are OM-rich: the Malga Fm kerite and organic carbon- and autochthonous bitumens-rich mudstones from the Tsipanda and Kumakh Formations. Concentrations of C_{31-35} homohopanes tend to decrease with increasing molecular weight (Fig. 7), while the C_{35}/C_{34} homohopanes ratio < 1 (Table 4) indicate transformations of the sedimentary organic matter during diagenesis in reducing environments without high content of free hydrogen sulfide in sediment and bottom waters (Peters, Moldowan, 1993). In two samples from the Malga Fm have C_{35}/C_{34} >1, which implies that an excess of sulfur in the sediment may have occurred oftentimes. The C_{27} trisnorneohopane to C_{27} trisnorhopane ratio

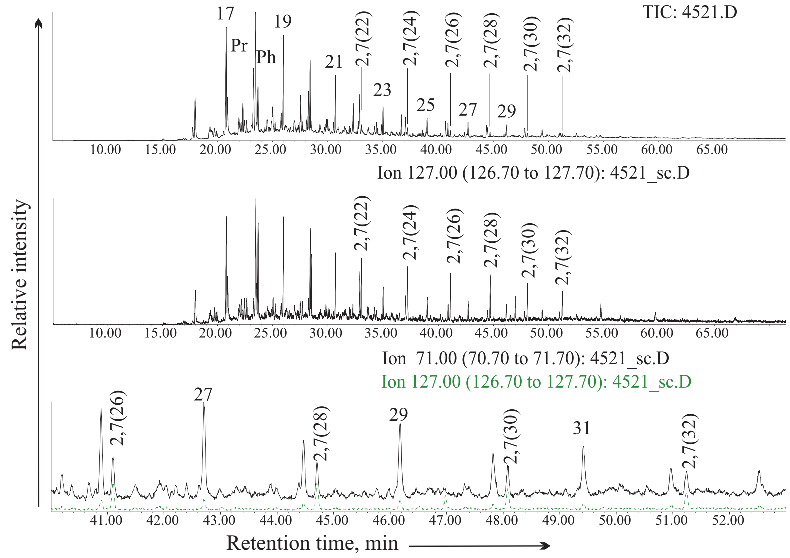

Fig. 5. Total ion current chromatogram and mass fragmentograms for terpanes (m/z 71 and m/z 127) in bitumen (sample No. 10, the Malga formation) (17-31 are n-alkane peaks, Pr and Ph are pristane and phytane, 2,7(22)-2,7(32) are peaks of 2,7-dimethylalkanes

No	Steranes: Dia/Regul	Steranes: C_{29}/C_{27}	Steranes: $C_{29}20S/C_{29}20(S+R)$	Steranes: $C_{29}\beta\beta(20S+20R)/C_{29}\alpha\alpha20R$	Steranes/ pregnanes	Steranes/ terpanes	Terpanes: Ts/Tm in hopanes	Terpanes: C_{35}/C_{34} in homohopanes	Terpanes: $2C_{19-20}/C_{23-26}$ in tricyclanes	Terpanes: Hopanes C_{27-35}/ tricyclanes C_{19-31}
1	0.4	1.4	0.6	1.8	–	–	0.5	0.5	1.4	3.7
2	0.3	1.6	0.4	0.5	22.9	0.1	0.3	0.3	0.6	15.1
3	0.2	1.0	0.4	2.5	27.2	0.3	1.1	2.0	0.2	7.6
4	0.4	1.1	0.4	2.8	12.6	0.2	0.6	0.6	0.9	4.3
5	–	–	–	–	–	–	0.8	0.5	1.8	4.0
6	–	–	–	–	–	–	0.8	0.4	1.6	4.1
7	–	–	–	–	–	–	0.5	0.5	2.2	2.5
8	0.5	1.4	0.4	2.3	5.3	0.2	0.8	0.9	0.5	2.5
9	0.4	1.6	0.4	1.1	9.5	0.1	3.4	0.7	0.8	0.8
10	0.4	1.0	0.3	1.8	1.5	0.2	0.8	1.1	1.1	0.6
11	0.5	1.3	0.4	1.9	6.9	0.3	0.9	0.9	0.4	2.2
12	0.4	1.3	0.3	1.5	5.2	0.1	1.2	0.5	1.4	2.4
13	0.4	1.7	0.5	0.7	4.6	0.02	0.3	0.5	1.7	4.6
14	0.4	1.5	0.5	1.2	3.5	0.03	0.7	0.5	1.8	4.0
15	0.4	2.5	0.4	1.9	6.5	0.02	0.2	0.5	2.0	4.6
16	0.5	0.9	0.4	1.8	1.4	0.1	0.6	0.5	2.9	0.5
17	0.4	1.0	0.4	2.5	1.6	0.1	0.6	0.5	2.2	0.6
18	0.5	1.1	0.4	2.5	2.8	0.2	1.7	0.8	1.1	1.3
19	0.3	1.8	0.6	2.5	15.9	0.1	4.2	0.5	0.2	1.3
20	0.5	1.2	0.4	2.4	7.4	0.2	1.0	1.0	0.5	2.3
21	0.5	0.8	0.5	2.2	2.3	0.04	16.5	0.6	1.4	0.5
22	0.5	1.0	0.4	3.0	2.8	0.2	1.8	0.9	1.3	0.9
23	0.7	0.7	0.5	2.6	3.7	0.1	21.7	0.8	0.5	0.4
24	0.5	1.1	0.4	3.0	4.6	0.1	8.3	0.7	1.8	0.5
25	0.5	1.1	0.4	2.0	2.6	0.1	13.5	0.5	1.3	0.5
26	–	–	–	–	–	–	6.2	–	1.1	0.4

Table 4. The ratios of saturated cyclic biomarker hydrocarbons in Riphean bitumens (the Aldan-Maya depression). Note: the numbers in yellow color represent the sample numbers of bitumens and bitumens containing admixture of allochthonous bitumens, according to the bitumen coefficient β (> 5 %).

(Ts/Tm) varies widely – from very low values (<0.5), gravitating towards the Malga and Tsipanda Fms, to very high values (>5, up to 21.7) in the Ignikan and Kandyk Fms. The increase may be associated with a terrestrial (in the case of bitumen migration from younger rocks) source of the organic matter (Waples, Machihara, 1990) and/or oxidizing conditions during diagenesis (Moldowan et al., 1986), and/or transformations during high catagenesis conditions (Petrov, 1994). It may also serve as evidence of a high carbonate content of sediment in the OM accumulation basin (Philp, 1985; Rüllkötter, Marzi, 1988). The latter reason is probably relevant for carbonates from the Mil'kon, Ignikan, and Kandyk Fms – in this case, bitumen samples 19 and 21, and 23 through

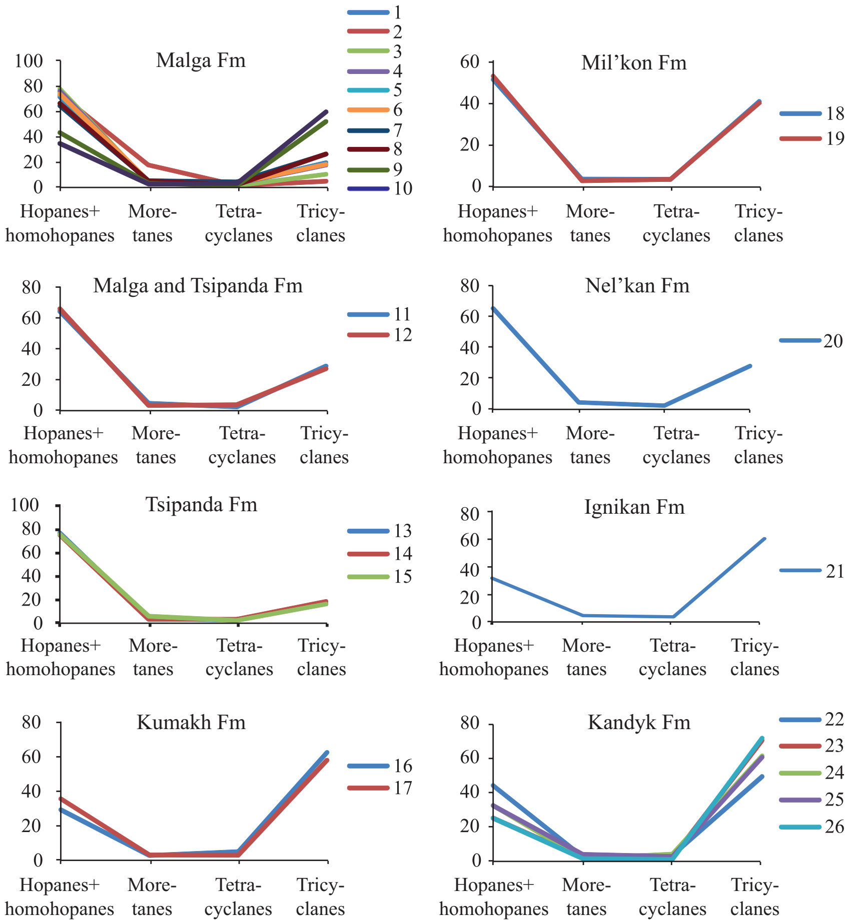

Fig. 6. *Terpanes in Riphean bitumens from the Maya Rv. outcrops (the sample numbers correspond to the sample numbering in Table 4)*

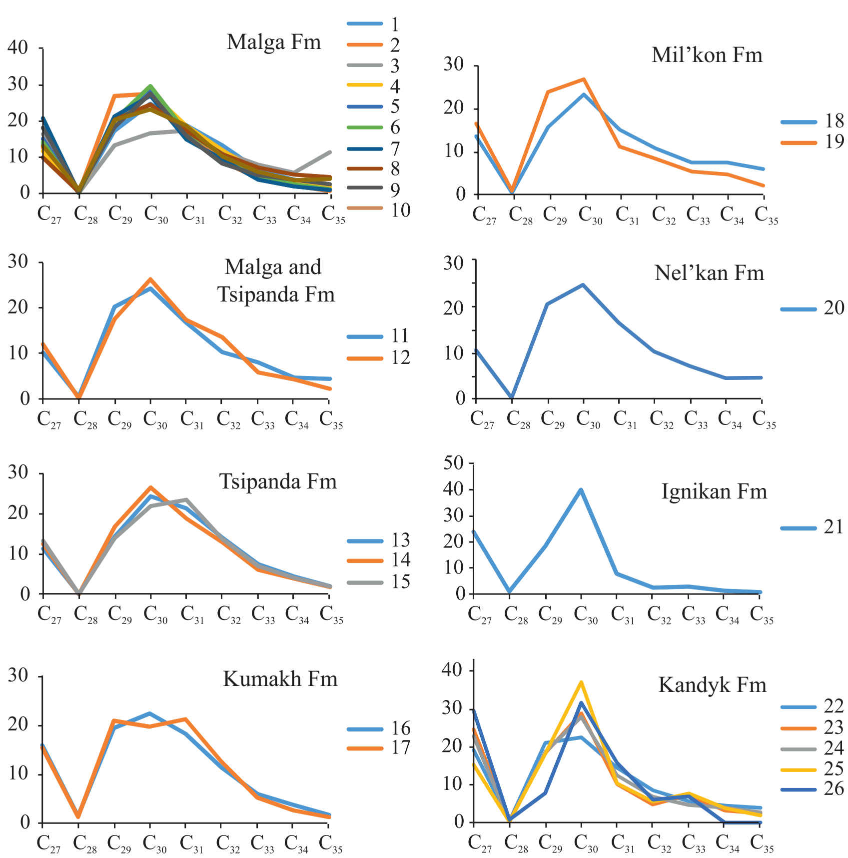

Fig. 7. *Hopanes in Riphean bitumens from the Maya Rv. outcrops (the sample numbers correspond to the sample numbering in Table 4)*

25 can be considered parautochthonous (inherent to their host sequence) rather than allochthonous. However, since the autochthonous (inferred from β) bitumen (Kandyk Formation) has a lower Ts/Tm ratio against allochthonous bitumen, and Ts/Tm >2 is generally found only in samples with β>5 (Tables 2, 4), it would be logical to assume that an increase in Ts/Tm is associated with the migration processes. Elevated gammacerane contents (Table 4) in several bitumens, among them bitumens and limestone in the Malga Fm (1.2–6.2 % per sum of tarpanes), the Kumakh mudstone (1.2 %) and Ignikan limestone (6.8 % per sum of tarpanes), indicate high-salinity waters in the OM accumulation basin (Petrov, 1994). Due to its stability, gammaceran accumulates during biodegradation (Waples, Machihara, 1990, 1991), which may be the cause of its elevated concentrations at least in the Malga kerite. High concentrations of C_{30} 17α-diahopane were found in epigenetic bitumens inherent in the Ignikan Fm (sample 21) and Kandyk Fm (samples 24, 25, 26) (Fig. 8).

While this biomarker is attributed to indicators of continental coal-bearing strata in the literature (Volkman et al., 1983; Philp and Gilbert, 1986), the appearance of 17α-diahopane C_{30} was probably a result of a significant bacterial input to clay-rich sediments deposited in oxic to sub-oxic environments (Peters et al., 2007). The mass fragmentograms of terpanes in bitumens from the Ignikan (sample 21) and Kandyk (samples 24 and 25) limestones (Fig. 8) show a peak marked with a question mark, which may be attributed to a homohopene. Elevated contents of C_{29} 17β,21α-normoretane were registered in the samples from the Ignikan Fm and one sample from the Kandyk Fm (sample 25). High concentrations of moretanes and the presence of homohopene at low contents of 17α, 21β-hopanes (not observed in other samples) may be an indication of contamination of these bitumens with younger, low-mature hydrocarbons (Peters et al., 2007).

In a series of tricyclane, most samples (15 samples) are dominated by low molecular weight C_{21}, and C_{19} in two

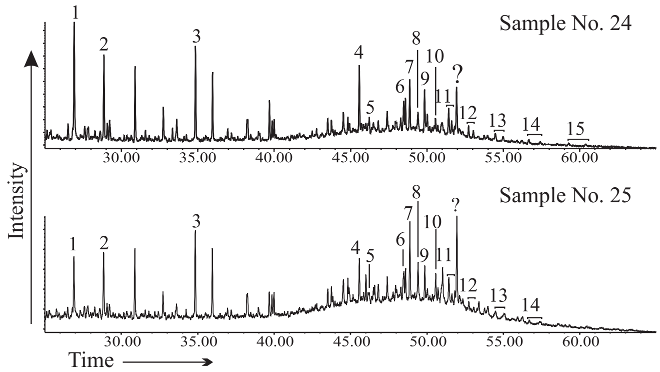

Fig. 8. Mass fragmentograms of terpanes (m/z 191) in limestones (samples 24 and 25) from the Kandyk Formation: 1, 2, 3 – tricyclanes C_{19}, C_{20}, C_{23}, 4 и 5 – trisnorneohopane (Ts) and trisnorhopane (Tm) C_{27}, 6 – adiantane C_{29}, 7 – 17α – diahopane, 8-17β,21α – normoretane C_{29}, 9 – hopane C_{30}, 10 – 17β,21α – moretane C_{30}, 11, 12, 13, 14, 15 – homohopane pairs C_{31}, C_{32}, C_{33}, C_{34}, C_{35}

limestones from the Ignikan and Kandyk Fms (Fig. 9). The maximum at C_{23} characteristic for the marine OM (Kontorovich et al., 1999) was revealed in 3 mudstone samples (the Malga Fm), in one limestone sample (the undifferentiated Malga and Tsipanda Fms), and in one dolomite and one sandstone samples (the Kandyk Fm). In four samples (including the Malga bitumens) anomalous concentrations of high-molecular-weight tricyclanes (C_{31} and C_{28}), with the tricyclane index $2C_{19-20}/C_{23-26}$ <1 (Table 4) are diagnostic of the marine OM (Kontorovich et al., 1999). Low tricyclane index was observed in a few other samples, while it not always coincides with the maximum at C_{23}. In most samples of the collection $2C_{19-20}/C_{23-26}$ >1, which is not typical of HC that formed in the marine sedimentary strata. The reason may lie in the specifics of the parent biota.

Recent studies attribute a time when first eukaryotes came into being to the Neoproterozoic, which is corroborated by the finds of contemporaneous C_{27} steranes (about 800 Ma BP) and solitary sponge-derived C_{28} steranes associated with the pre-Vendian glaciation event, along with the mass emergence of remaining steranes in the narrow pre-Vendian period of 659–645 Ma recognized for the flourishing Ediacaran algal biota (Brocks et al., 2016, 2017). The international geosciences literature provides sufficient evidence of the findings of steranes (Summons, Walter, 1990; Pratt et al., 1991; Bazhenova, Arefiev, 1996; Berney, Pawlowski, 2006) and their absence (Flannery, George, 2014; Blumenberg et al., 2012; Luo et al., 2015) in pre-Neoproterozoic time. In Riphean rocks of the Baykit anteclise, steranes are present in the widest age range, and where they are identifiable, all the C_{27-30} steranes are encountered, with concentrations of C_{27} and C_{29} being remarkably similar (Timoshina, 2005, etc.). Alternatively, in oils, sterane abundancies range from zero to concentrations described by two types of distribution: (i) with approximately equal concentrations of C_{27} and C_{29}; and (ii) with remarkable predominance of C_{29} (Kontorovich et al., 1996, 1999, 2005, 2001; Timoshina, 2020, etc.). The many researchers studying the organic matter from the Malga Formation noted both the presence of steranes (Chalaya et al., 2015) and their absence (Suslova et al., 2017) or trace amounts (Bazhenova et al., 2014; Dakhnova et al., 2014).

In the studied collection, out of 10 samples, steranes are absent from three samples (samples 5, 6 and 7) from the Malga Fm (Table 4), while these samples are interpreted to be organic carbon rich (Table 1). One would assume that the biota of the Malga time might be completely devoid of eukaryotes, whereas steranes in OM-depleted specimens (samples 4, 8, 9, and 10) and bitumens (samples 2 and 3) are allochthonous. The latter, however, are found in autochthonous bitumens (inferred from low concentrations of hydrocarbons and high content of asphaltenes (Table 2)) from the organic carbon-rich Malga mudstone (sample 1) (Tables 1, 4), and were previously identified by other researchers (Bazhenova et al., 2014; Dakhnova et al., 2014; Chalaya et al., 2015). Therefore, it is possible that the Malga time served as the boundary for origination of first eukaryotes. Bitumens in all other samples contain steranes, except allochthonous bitumen from the Kandyk sandstone. This bitumen is probably older (inferred from the absence of eukaryotes in the initial biota) (Brocks et al., 1999; Brocks et al., 2003; Peters et al., 2007) or may have contained too few steranes which were lost during the migration.

Earlier, it was noted that C_{27} dominates in steranes from the Malga Fm, while rocks in the Lakhanda Group exhibit similar contents of C_{27} and C_{29} (Chalaya et al., 2015). In this study, fairly equal concentrations of C_{27} and C_{29} sterans were observed among homologues of C_{27-30} steranes in most samples (Table 4, Fig. 10, 11). This distribution is typical of marine black shale deposits and corresponds to HCs generated by the OM, whose precursors were marine planktonic autotrophic (C_{29} sterane), as well as heterotrophic organisms (C_{27} sterane) (Petrov, 1984). A similar distribution of steranes was observed in Proterozoic oils of the MacArthur basin in Australia, in Vendian oils of the Tishkovskoye field (East European Platform) (Kontorovich et al., 2005), and in extractable bitumens from Upper Proterozoic deposits of the Yenisei ridge and the Baikyt anteclise (Timoshina, 2005). Slightly elevated concentrations of C_{29} (C_{29}/C_{27} in the range of 1.6–1.8) were observed in 3 samples from the Malga and Tsipanda Fms (maltha and samples with TOC <1 %) and in the Mil'kon Fm dolomite (TOC <1%), whereas in the Tsipanda Fm mudstone (TOC >1 %) C_{29} is more than twice higher than C_{27}. At higher TOC, the latter sample contains autochthonous bitumen (judging from β). Therefore, an increase in concentration

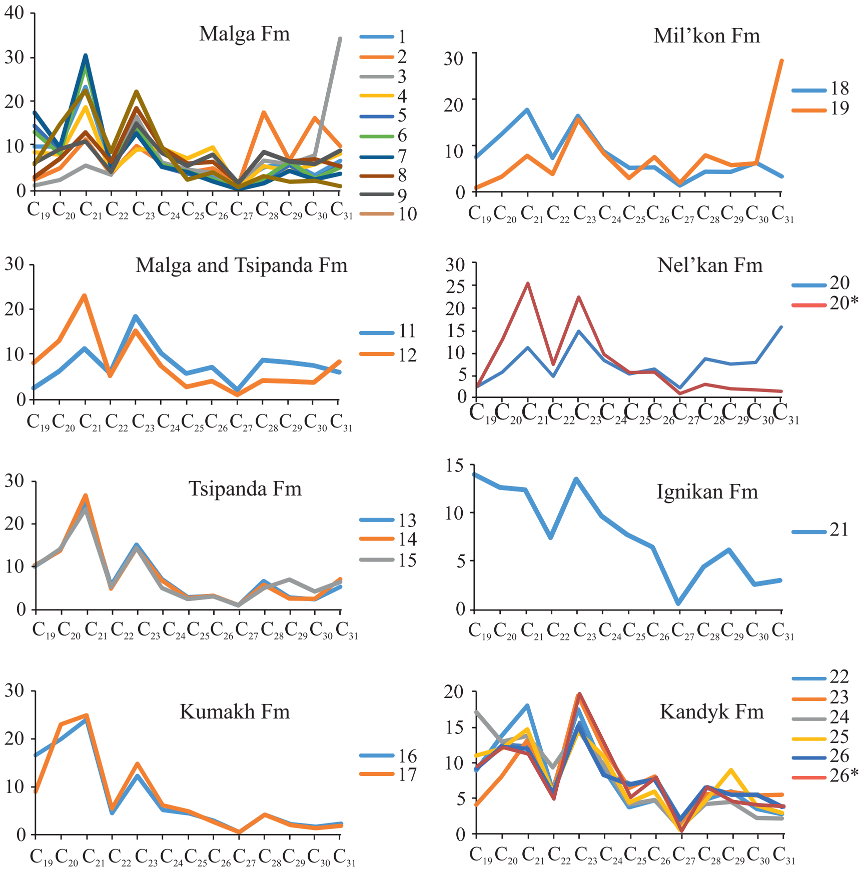

Fig. 9. Tricyclanes in Riphean bitumens from the Maya Rv. outcrops (the sample numbers correspond to the sample numbering in Table 4).

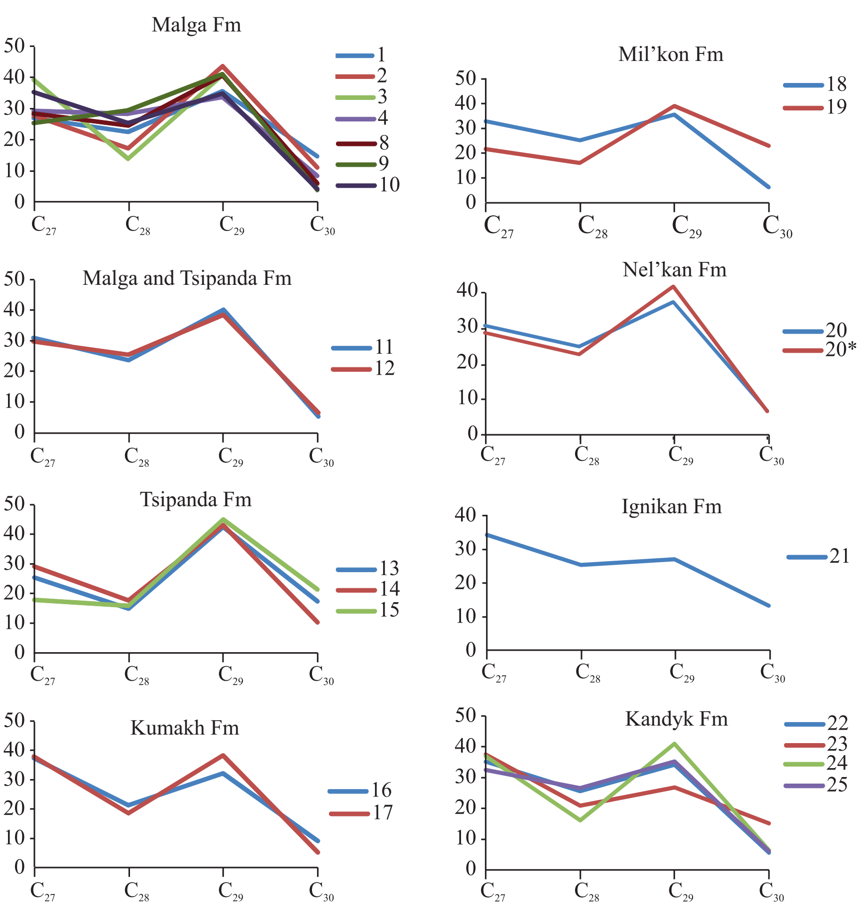

Fig. 10. Steranes in Riphean bitumens from the Maya river outcrops (the sample numbers correspond to the sample numbering in Table 4)

of ethylcholestanes is not associated with contamination by allochthonous bitumens, but rather it is interpreted as a signature of the Precambrian OM noted in most common oils of the Siberian Platform (Petrov, 1984; Kontorovich et al., 1996, 1999, 2005; Timoshina, 2004, 2005). Given that steranes have not been identified, except three samples from the Malga Fm and the Kandyk Fm sandstone (sample 26), they might be originally missing from the parent organic matter (very old) because of the absence of eukaryotes (Brocks et al., 1999, 2003; Peters et al., 2007). Biodegradation of steranes following *n*-alkanes probably resulted in their disappearance, however *n*-alkanes were preserved in bitumens of the Kandyk and Malga Formations. Based on distributions of biomarker hydrocarbons, isotopic and pyrolytic characteristics of the organic matter from the Malga Fm, Tsipanda Fm and Ignikan Fm, all the researchers defined it as the marine OM (Bazhenova et al., 1981, 2014; Matvienko, Sobolev, 1984; Sobolev, 1987; Dakhnova et al., 2013, 2014; Safronov et al., 2015, Suslova et al., 2015, 2017). At this, it was noted that the presence of microfossils in the Malga Fm is consistent with the absence or low content of steranes, since these are mainly the remains of bacteria and cyanobacteria. There are only isolated finds of larger acritarchs, which may be interpreted as representatives of eukaryotes (Suslova et al., 2017). The studied samples contain mainly high concentrations of diasteranes Dia/Regul (0.4–0.7, averaging 0.4 (Table 4)), which is indicative of the accumulation of parent OM in the basin dominated by clayey sediments (Mello et al., 1988; Waples, Machihara, 1990, 1991; Petrov, 1994), being in discord with the lithology of a number of carbonate samples. Bitumens in the carbonate samples are probably either allochthonous or parautochthonous, and diasteranes may have accumulated in them due to some other factor, such as catagenesis. In respect to sterane abundancies, bitumens correspond primarily to low-mature organic matter (maturity coefficient $C_{29}\beta\beta(20S+20R)/C_{29}\alpha\alpha20R<3$), which

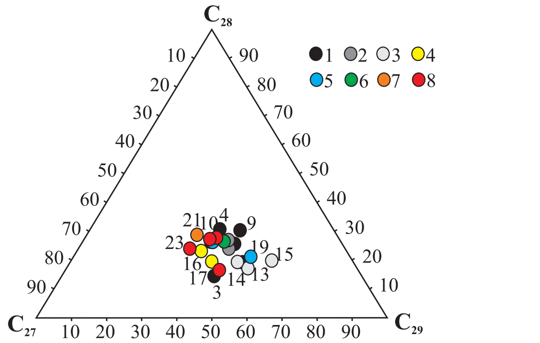

Fig. 11. Trigonogram of steranes in Riphean bitumens from the Maya Rv. outcrops; Formations: 1 – Malga, 2 – Malga and Tsipanda, 3 – Tsipanda, 4 – Kumakh, 5 – Mil'kon, 6 – Nel'kan, 7 – Ignikan, 8 – Kandyk (the sample numbers correspond to the sample numbering in Table 4)

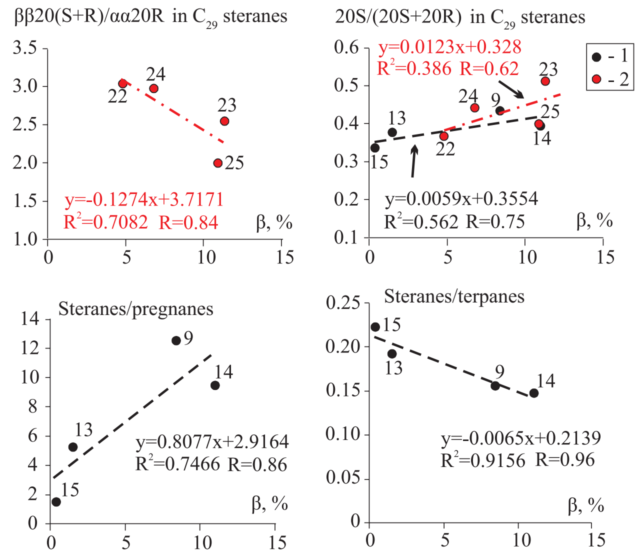

Fig. 12. A relationship between steranes ratios and allochtonous constituent of bitumens in the Riphean organic matter from the Maya Rv. outcrops (formations: 1 – Malga, 2 – Kandyk) with trends (shown by the dotted lines for Malga Fm (black), Kandyk Fm (red)). The sample numbers correspond to the sample numbering in Tables 2 and 4.

is contradictory to the results of Rock-Eval pyrolysis (catagenesis grades MC_1^1-MC_2). However, given very few steranes found in the studied bitumens, and generally low yields of bitumen, except individual samples (with no steranes identified in them), one should apply the sterane parameters with great care.

Migration-controled biomarker indicators

Given that in the study of Precambrian samples, especially those that are highly transformed (or possibly highly thermally transformed), the question as to whether bitumen is autochthonous is very relevant. To this end, relationships between the biomarker parameters and the bitumen coefficient were plotted in order to putatively estimate potential effects of the hydrocarbons migration on their distribution. Acyclic saturated hydrocarbons showed no clear dependence of C_{27}/C_{17}, CPI, and Pr/Ph on β. In the steranes (Fig. 12), the proportion of S-forms increases as compared to R-forms with increasing β in the samples from the Malga and Kandyk Formations, which is probably a result of a greater transformation of allochthonous terrestrial admixtures during catagenesis. The relationship between ββ- and αα-isomers shows an opposite trend for the Kandyk and Tsipanda formations, which suggests a lesser transformation of the introduced organic matter. However, the samples from the Tsipanda Fm are interpreted to be autochthonous. As such, this tendency might be not true, rather it is associated with an error stemming from a paucity of steranes. The pregnanes to steranes ratio in the samples from the Malga Formation is clearly directly proportional to the percentage of bitumens. Pregnanes are probably more migratory than steranes and are capable to accumulate in low permeability strata (Kontorovich, Timoshina, 2009). At this, high concentrations of pregnanes are typical of marine, lagoon depositional environments of the parent high salinity strata (ten Haven et al., 1988), while pregnanes can also accumulate as compared to degrading $C_{27\text{-}30}$ steranes under high catagenesis conditions, and are more resistant to biodegradation than steranes (Peters et al., 2007). The input of bitumens took place probably from deeper buried strata whose accumulation is associated with higher salinity of basinal waters, while hydrocarbons might be biodegraded to some extent.

However, a decrease in the proportion of steranes having eukaryotic organisms as their ancient source (Peters et al., 2007), as compared to terpanes, may indicate an older source. Besides the fact that more compact molecules migrate better, the possible migration can be supported by other lines of evidence, which include: an increase in Ts (against Tm) in terpanes in allochthonous bitumens; slightly reduced C_{35} (against C_{34}) concentrations in homohopanes, which is more pronouncedly observed in the Malga samples; and a slight increase in the number of tricyclanes compared to hopanes against an increase in the bitumen coefficient (Fig. 13). Among possible reasons for the directly proportional relationship between Ts/Tm and the bitumen coefficient are the accumulation of the initial organic matter for allochthonous bitumen in the carbonate sediment (Philp, 1983; Rüllkötter, Marzi, 1988), its diagenesis in an oxidizing environment (Moldowan et al., 1986), and high catagenesis conditions (Waples, Machihara, 1990; van Graas et al., 1990; Petrov, 1994). In the case of allochthonous terrestrial admixture having a lower tricyclane index indicating more marine conditions for accumulation of the initial OM (Kontorovich et al., 1999) or higher catagenesis, a weak decreasing tend in the tricyclane index and affiliated increase in β may relate indirectly to the migration phenomenon. If the admixture contains a lot of tricyclanes due to either more marine source or higher catagenesis, this may as well be related to an increase in the number of tricyclanes compared to hopanes (Petrov, 1994).

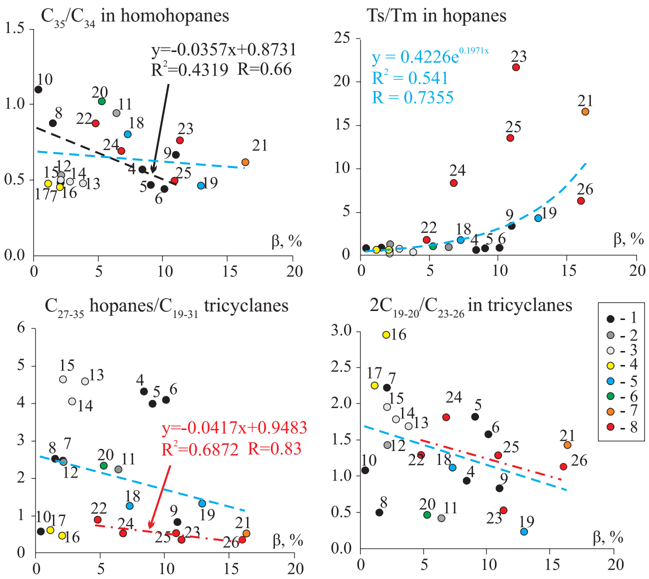

Fig. 13. A relationship between the terpanes ratios and the allochtonous constituent of bitumens in the organic matter from Riphean outcrops of the Maya Rv. (Formations: 1 – Malga, 2 – Malga and Tsipanda, 3 –Tsipanda, 4 – Kumakh, 5 – Mil'kon, 6 – Nel'kan, 7 – Ignikan, 8 – Kandyk) with trends for all the samples (blue dashed line) and separately for the Malga (black dashed line) and Kandyk (red dashed line) Formations (the sample numbers correspond to the sample numbering in Tables 2 and 4)

Thus, there is a distinct connection of the steranes/pregnanes and steranes/terpanes ratios with the migration processes shown for the Malga formation, which contains both allochthonous and autochthonous bitumens. The trend for the steranes/pregnanes ratio may indicate both the input of hydrocarbons (possibly biodegraded) from deeper buried and higher salinity strata during the organic matter accumulation. A decrease in concentration of steranes compared against terpanes with an increase in the bitumen coefficient may be the result of HC input from an older, more eukaryote-depleted source. A rather fuzzy tendency towards a decrease in the tricyclane index with the increasing bitumen coefficient may indicate the input of hydrocarbons whose source was of more marine origin. A significant increase in Ts/Tm in allochthonous bitumens may be a result of the input of HCs from carbonate strata, where the initial OM is exposed to a high catagenesis conditions and has passed diagenesis in an oxidative environment, although the superposition of purely migration effects is also possible. A higher level of catagenesis for migrated hydrocarbons is also reflected in elevated proportion of S-forms compared to R-forms in steranes (Han et al., 2017).

Conclusion

The studied Precambrian rocks from the Aldan-Maya depression are organic carbon-rich (TOC > 1 %) and can be attributed to the oil-prone source mudstones and siltstones of the Malga, Tsipanda and Kumakh Formations. Samples from other studied formations are TOC- and bitumens-depleted, with the latter being either allochthonous or mixed, indicating the migratory nature of oil components in the studied deposits.

The highest generation potential (HI = 267–511 mg HC/g TOC) was observed in the samples from the Malga and Tsipanda Formations. According to T_{max}, the catagenesis of the OM from the Malga and Tsipanda formations corresponds to catagenesis grade MC_1^2, while maturity level of the Tsipanda and Kandyk organic matter is lower. However, due to the possible presence of bitumen in some samples, the HI values may be overestimated and the T_{max} underestimated.

Based on the distribution of biomarker HCs, different prokaryotes are found to have been the biological precursors of the oil-prone Kumakh, Tsipanda, and Malga source rocks, including cyanobacteria (hopanes, acyclic biomarkers) and, to a lesser extent, auto- and heterotrophic eukaryotes (concentrations and distribution of steranes) that lived in the marine (no biomarkers of terrestrial OM, distribution of acyclic alkanes, steranes) Precambrian (12- and 13-monomethylalkanes) basins with clayey sedimentation (high content of diasterans). The Malga Formation probably partially accumulated before the appearance of eukaryotes (the absence of steranes in some samples). The concentrations of low molecular weight tricyclanes ($2C_{19-20}/C_{23-26}$>1) in the Tsipanda and Kumakh bitumens, and in the half of bitumens from the Malga Formation are generally elevated, which is probably associated with specific parent biota, although not being characteristic of the marine OM.

A rare homologous series of 2,7-dimethylalkanes identified in several samples from the Malga Formation and in both samples of the Kumakh Formation, however not registered in any sample of the Tsipanda Formation, may represent distinctions of its biota, its accumulation conditions or evolution during diagenesis and catagenesis. The distinctness of the Tsipanda Formation is also accentuated by high content of ethylcholestanes (C_{29}/C_{27}=2.5) in its carbonaceous mudstone.

Biomarker characteristics similar to those observed in the samples from the Malga, Tsipanda, and Kumakh Formations were dominantly encountered in allochthonous and mixed bitumens of OM-depleted samples from the studied collection (except the Kandyk bituminous sandstone). These are generally characterized by elevated tricyclane index (>1), and finds of 12- and 13-monomethylalkanes in the Tsipanda limestone, and 2,7-dimethylalkanes in carbonates of the Mil'kon and Nel'kan Fms and in one sample from the Kandyk Fm.

In several allochthonous bitumens, the content of trisnorneohopane has significantly increased compared to trisnorhopane (Ts/Tm>3, 6.2–21.7 in samples from the Ignikan and Kandyk Fms), which can be explained neither by high catagenesis conditions nor admixture of younger terrestrial hydrocarbons. This may be associated with diagenetic conditions or migration processes.

In the studied collections, the relationships that in varying degrees are controlled by the migration processes (i.e. to some extent are dependent on the value of bitumen ratio β) include: Ts/Tm in hopanes, C_{27-35} hopanes/C_{19-31} tricyclanes, $2C_{19-20}/C_{23-26}$ in tricyclanes, C_{35}/C_{34} in homohopanes, 20S/20R and ββ20(S+R)/αα20R in C_{29} steranes, steranes/pregnanes, steranes/terpanes. It would be highly logical to investigate possible anomalies of these parameters during migration and their causes using a more extensive and diverse collection.

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