UDC 547.912:543.544:543.51

DOI: 10.15372/CSD2019124

Composition of Aromatic Hydrocarbons in Gas Condensates of Western Siberia

G. S. PEVNEVA[1], N. G. VORONETSKAYA[1], M. V. MOZHAYSKAYA[1], A. K. GOLOVKO[1,2],
E. A. FURSENKO[2], V. A. KASHIRTSEV[2]

[1]*Institute of Petroleum Chemistry, Siberian Branch, Russian Academy of Sciences,*
Tomsk, Russia

E-mail: pevneva@ipc.tsc.ru

[2]*Trofimuk Institute of Petroleum Geology and Geophysics, Siberian Branch, Russian Academy of Sciences,*
Novosibirsk, Russia

(Received August 27, 2018; revised February 27, 2019)

Abstract

This paper presents the results of the studies of cyclic aromatic compounds composition in gas condensates of Maloyamalskoye and Myldzhinskoye gas fields of Western Siberia. The composition of aromatic compounds in gasoline cuts of gas condensates was analyzed by GC method. Data on group composition of C_7–C_{10} aromatic compounds in gasoline cuts demonstrate that the gas condensates of Maloyamal'skoye gas field are enriched with toluene (C_7) and xylenes (C_8) while in the samples of Myl'dzhinskoye gas field the contents of trimethyl- (C_9) and tetramethyl benzenes (C_{10}) are high. The composition of aromatic compounds of benzene, naphthalene and phenanthrene series in the non-fractionated condensates was studied using GCMS method. It has been shown that alkyl benzenes of condensates are represented predominately by low-molecular compounds (C_7–C_{10}), content of long-chain *n*-alkylbenzenes and *n*-alkyltoluenes is very low. In addition to alkyl-substituted benzenes the naphthene-substituted benzenes (methyl- and dimethyltetralines) are identified in gas condensates. The relative concentrations of cadalene, retene, ethyl benzene, 1,2,3- and 1,2,4-trimethylbenzenes are used as genetic factors on the composition of aromatic compounds. It has been established that humic organic matter contributed to the formation of the studied fluids. The evidences of that are: higher than usual values of Prystane/Phytane ratio; high content of xylene; increased content of 1,2,5- and 1,2,7-trimethylnaphthalenes and the presence of aromatic hydrocarbons – biomarkers (cadalene, retene). Parameters calculated from the composition of alkyl benzenes and alkyl naphthalenes determine the thermal maturity of condensates. According to the values of ratio (*m*- + *p*-xylene)/*o*-xylene, naphthalenic indexes (MNR, DNR2 and TNR6) and low content of long-chain C_{11+} alkyl benzenes the catagenic maturity level of the studied fluids corresponds to the main gas generation phase.

Key words: condensates, composition, alkyl benzenes, alkyl naphthalenes, alkyl phenanthrenes, geochemical parameters, Western Siberia

INTRODUCTION

Investigation of the molecular composition of biomarkers, both hydrocarbons and heteroatomic compounds present in petroleum, is a key to understanding the nature of the initial organic matter (OM) and the conditions of its fossilization and subsequent geochemical evolution. The information about the distribution and composition of aromatic hydrocarbons (HC) is used in geochemical generalization to reconstruct the conditions of the formation of petroleum composition to a much lower extent than the information on saturated hydrocarbon biomarkers. This is connected, first of all, with the complexity of the composition and analysis of aromatic fractions, and second, with

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poor understanding of the mechanisms of formation and transformation of aromatic HC at the stages of dia- and catagenesis.

These HC are not synthesized directly by living organisms but are formed from the initial oil-source matter (biopolymers) through thermochemical transformations *via* dehydration, cyclization and aromatization [1–5]. The composition of arenes in naphthides is controlled by the type of initial OM and the conditions of its accumulation, geological, geochemical and depth-related factors [6–9], which leads to their diversity in petroleum and condensates.

Investigation of the composition of aromatic HC is necessary because not all types of light petroleum and condensates contain high-molecular hydrocarbon biomarkers, and in the case of their low concentrations and/or high catagenic maturity of fluids, when biomarkers reach the equilibrium values, mass spectra become low-informative.

To evaluate the degree of maturing with respect to the composition of alkyl arenes, geochemical parameters based on the relations between β- and α-methyl isomers of benzenes, naphthalenes and phenanthrenes are used. During thermal maturing, the formation of thermodynamically more stable β-isomers occurs due to the isomerization of initially formed alkyl arenes [10–13].

Some alkyl arenes inherit (to a definite extent) the structural features of the molecules of their biological precursors, and they are used to establish the genetic relationship. For example, the precursors of alkyl benzenes may be fatty acids, diaromatic carotenoids (*e.g.* isorenieratene), bacteria [14–17]. The potential precursors of diarenes include terpenoid hydrocarbons, cyclic sesquiterpenoids, carotenoids [1, 2, 9, 10, 18, 19]. Relic hydrocarbon biomarker 1,6-dimethyl-7-isopropylnaphthalene (cadalene) is a marker of higher terrestrial plants, mainly angiosperms. Its structure points to the genetic relationship with dicyclic sesquiterpenes – aliphatic (farnesol) and alicyclic (cadinene) [19, 20]. Retene (1-methyl-7-isopropylphenanthrene), the derivative of abietic acid, one of the major components of the tar of conifers, is widely used in geochemical studies to evaluate the genotype of initial OM [19–22].

Interest to the studies of aromatic HC in gas condensates of West Siberia is due to the fact that these fluids contain high concentrations of adamantane HC. Similarly to aromatic HC, they are not present in the initial biomass but they are present almost in all types of petroleum generated by marine or continental OM. They are also present in weakly and strongly transformed oil, both in carbonate and in clay rocks [23–27]. Studies of the features of the composition of aromatic HC of gas condensates promote revelation of the regularities of the conditions under which their deposits are formed.

EXPERIMENTAL

The objects of investigation were condensates of the Maloyamalskoye gas field from the pool in Middle Jurassic deposits (the Tyumen suite, sampling depth 2264 to 2366 m), Myldzhinskoye gas field from the pools in Lower Cretaceous (the Kulomzinskaya suite, sampling depth 2154 to 2197 m), and Upper Jurassic deposits (the Vasyuganskaya suite, sampling depth 2395 to 2423 m). The Maloyamalskoye gas field is situated in the southern part of the Southern Yamal oil and gas-bearing region of the Yamal oil and gas-bearing area [28]. The Myldzhinskoye gas field is situated in the east of the Middle Vasyugan megaswell, at the boundary with the Ust-Tymskaya depression within the Vasyugan oil and gas-bearing territory of the West Siberian oil and gas-bearing province [29].

Analysis of the hydrocarbon composition of non-fractionated condensates was carried out by means of gas chromatography (GC) with a Kristall 2000 M chromatograph equipped with a flame ionization detector and a capillary column 100 m long with the inner diameter of 0.22 mm (phase: polymethylsiloxane). Chromatograms were recorded in the mode of linear temperature programming from 35 to 250 °C, at the initial temperature the isotherm was 13 min, the rate of heating from 35 to 45 °C was 5 °C/min, 15 min isotherm, from 45 to 60 °C – 1 °C/min, 15 min isotherm, from 60 to 250 °C – 2 °C/min, 20 min isotherm. The carrier gas was helium. The treatment of chromatographic data and detailed analysis of hydrocarbons according to the ASTM standards were carried out using the Chromatec Gasoline software.

The composition of saturated and aromatic HC was analysed by means of gas chromatography – mass spectrometry (GC-MS) using a quadrupole system GC-MS-QP5050 Shimadzu with the computer-based system of data asquisition and processing. The chromatograph is equipped with a capillary quartz column DB5-MS 30 m long and the inner diameter 0.25 mm. The analysis was carried out in the mode of temperature programming from 80 to 290 °C with a rate of 2 °C/min, the final temperature was maintained constant for 25 min. The carrier gas was helium. The ionizing voltage was 70 eV, source temperature 250 °C. Data collection and processing in the modes of SCAN and SIM were carried out with the help of the GCMSsolution software. Identification of the compounds was carried out by comparing the recorded mass spectra with the mass spectra present in the NIST and WILEY libraries. The relative content of the compounds of each type was estimated as a ratio of its total intensity to the sum of the areas of all identified compounds.

RESULTS AND DISCUSSION

Hydrocarbons from the gasoline cuts of the condensates are represented by four basic classes: *n*-alkanes, isoalkanes, cyclanes and arenes. In the gasoline cuts of the condensates from the Myldzhinskoye gas field, normal alkanes dominate (Table 1). Their amount exceeds the content of isoparaffins and cyclanes by a factor of 2.0–2.5 and 2.9–3.7, respectively. Among the cyclic saturated HC, cyclopentanes (CP) dominate over cyclohexanes (CHe). With an increase in the depth of occurrence, the content of aromatic HC increases.

The condensates from the Maloyamalskoye gas field differ from those from the Myldzhinskoye gas field by the substantial concentrations of cyclanes with the domination of cyclohexanes over cyclopentanes (see Table 1). The ratio of *n*-alkanes/cyclanes mainly varies within the range 1.0–1.1. A higher content of aromatic HC is detected in the samples from the Maloyamalskoye gas field.

At this, no clear dependence of the changes in the content of aromatic HC with the depth of occurrence was established. The high values of prystane/phytane ratio are the evidence of the continental genesis of the fluids in both gas fields.

Analysis of the group composition of aromatic hydrocarbons C_7–C_{10} of gasoline cuts showed that the condensates from the Maloyamalskoye gas field are enriched with toluene and xylenes, while the samples from the Myldzhinskoye gas field contain high concentrations of trimethylbenzenes (TMB) and tetramethylbenzenes (TeMB) (Table 2). The ratio of the concentrations of toluene to the sum of xylenes in the condensates from the Myldzhinskoye gas field is smaller than 0.1, while in the condensates from the Maloyamalskoye gas field the variation of values is much broader – from 0.5 to 1.2.

The ratio *m*- + *p*-xylene/*o*-xylene is used in geochemical practice as one of the indications of maturing degree. This parameter may be informative for the investigation of condensates because they do not contain high-molecular hydrocarbon biomarkers [27]. The variations of the ratios of *m*- + *p*-xylene/*o*-xylene in the condensates from the Maloyamalskoye gas field depending on the depth of occurrence are insignificant (see Table 2). This parameter is much higher for the samples from the Myldzhinskoye gas field, therefore, these samples may be related to more mature ones in comparison with the condensates from the Maloyamalskoye gas field. In the condensates from the Myldzhinskoye gas field, the total content of *m*- and *p*-xylenes increases substantially with an increase in the depth of occurrence. The content of adamantanes in the condensates from the Myldzhin-

TABLE 1

Group composition of gasoline cuts of gas condensates, mass %

Hydrocarbons	Gas field						
	Myldzhinskoye			Maloyamalskoye			
	Occurrence depth, m						
	2154–2164	2183–2197	2395–2423	2298–2319	2337–2366	2264–2305	2317–2324
	Age of deposits						
	K_{1v}	K_{1v}	J_{3o}	J_2	J_2	J_2	J_2
n-Alkanes	53.0	50.7	54.9	34.4	41.7	36.1	34.8
Isoalkanes	26.4	25.6	22.4	25.0	19.4	23.4	23.3
Cyclanes	15.6	17.5	14.8	33.1	25.2	31.6	33.0
Arenes	5.0	6.2	7.1	7.5	13.7	8.9	8.9
n-Alkanes/cyclanes	3.4	2.9	3.7	1.0	1.7	1.1	1.1
Sum of CP/sum of CHe	1.7	1.7	1.3	0.7	0.6	0.7	0.6
Prystane/phytane	5.0	5.4	3.9	3.2	7.9	10.9	4.7

Note. CP – cyclopentanes, CHe – cyclohexanes.

TABLE 2

Characterization of the condensates with respect to the composition of alkyl benzenes C_7–C_{10}

Hydrocarbons	Gas field						
	Myldzhinskoye			Maloyamalskoye			
	Occurrence depth, m						
	2154–2164	2183–2197	2395–2423	2298–2319	2337–2366	2264–2305	2317–2324
Content, % with respect to the sum of benzenes C_7–C_{10}							
Toluene	0.8	0.9	1.1	32.0	13.7	28.0	28.7
Xylenes	25.3	17.8	14.2	31.3	32.8	33.0	42.1
TMB	29.1	32.1	30.7	27.6	39.8	22.9	15.1
TeMB	44.8	49.1	54.0	9.0	13.7	16.1	14.2
Content, % with respect to the sum of benzenes C_8							
Ethyl benzene	15.2	15.8	23.8	11.1	11.7	11.1	7.7
m-Xylene	48.1	55.4	50.0	55.8	51.2	51.2	49.2
p-Xylene	19.4	20.5	20.8	8.2	11.3	13.9	22.1
o-Xylene	17.2	8.3	5.4	25.0	25.9	23.8	21.0
Geochemical parameters							
Toluene/sum of xylenes	0.04	0.06	0.08	1.15	0.47	0.96	0.84
EB/sum of xylenes	0.2	0.2	0.3	0.1	0.1	0.1	0.1
m- + *p*-xylene/*o*-xylene	3.9	9.1	13.0	2.6	2.4	2.7	3.4
1,2,4/1,2,3-TMB	1.1	3.9	1.8	4.8	4.2	0.3	0.2

Note. TMB – trimethylbenzene, TeMB – tetramethylbenzene, EB – ethyl benzene.

skoye gas field is 1.5–3.0 % with respect to the sum of all the identified compounds, and in the condensates from the Maloyamalskoye gas field their content is much lower (0.4–0.9 %).

In geochemistry, relative concentrations of ethyl benzene, 1,2,3- and 1,2,4-TMB are used as the genetic indices for the composition of low-molecular aromatic C_7–C_{10} hydrocarbons [14, 15, 27, 30]. The primary products of the genesis of sapropel initial OM are most probably ethyl benzene and o-xylene, purely humic OM produces mainly ethyl benzene and *p*-xylene (their concentrations were higher than the equilibrium) [27]. In the condensates from the Myldzhinskoye gas field, ethyl benzene concentration is 15.2–23.8 % with respect to the sum of C_8 benzenes, and in the samples from the Maloyamalskoye gas field it is 7.7–11.1 %, which is higher than the equilibrium concentration (5.9 % at 600 K). In the studied condensates, no clear regularity was revealed for the change of ethyl benzene concentration with the change in the depth of occurrence. Among xylenes, *m*-xylenes are present in the maximal concentration; their concentration is close to or exceeds the equilibrium concentration (50.1 % at 600 K). The decreased ethyl benzene content and the high content of *m*-xylene may be the signs of the continental nature of the initial OM that had generated those condensates [27]. The ratio of the concentrations ethyl bensene/sum of xylenes is equal to 0.2–0.3 for the condensates from the Myldzhinskoye gas field, while for those from the Maloyamalskoye gas field this value is equal to 0.1 (see Table 2).

According to the data reported in [14, 15], precursors of 1,2,3-TMB may be bound non-aromatic carotenoids (for example, β,β-carotenes), which undergo aromatization and/or the loss of methyl groups at the stage of diagenesis, while substantial concentrations of 1,2,4-TMB are the evidence of the presence of plastoquinones in the initial OM. The high values of the ratio 1,2,4-TMB/1,2,3-TMB, varying from 1.1 to 3.9 in the condensates from the Myldzhinskoye gas field, and from 0.2 to 4.8 in those from the Maloyamalskoye gas field, are most probably due to the algal type of the initial OM. In this collection of samples, this parameter varies without any regularity depending on the depth of occurrence within one gas field. Among tetramethylbenzenes (TMB), 1,2,3,5-TeMB dominates in the condensates under investigation.

The composition of the aromatic HC of the series of benzene, naphthalene and phenanthrene in non-fractionated condensates was analyzed by means of GC-MS. Analysis of the data of group composition of aromatic HC showed that the major fraction (from 88 to 95.2 rel. %) in the studied fluids is comprised by monoarenes (Table 3). The amount of diarenes varies within a broad range – from 3.3 to 10.0 %. Triaromatic HC are present in insignificant amounts.

TABLE 3

Group composition of alkyl arenes according to the data of GC-MS

Hydrocarbons	Gas field						
	Myldzhinskoye			Maloyamalskoye			
	Occurrence depth, m						
	2154–2164	2183–2197	2395–2423	2298–2319	2337–2366	2264–2305	2317–2324
	Content, % with respect to the sum of the areas of identified peaks						
Monoarenes	93.63	93.72	88.19	95.16	91.15	93.83	89.78
n-AB, including:	25.33	28.77	29.84	70.42	57.05	68.35	53.22
AB_7–AB_{10}	20.6	25.2	27.3	69.3	55.9	67.5	52.2
AB_{11}–AB_{19}	4.8	3.6	2.6	1.1	1.1	0.8	1.0
n-AT, including:	31.21	29.97	28.29	18.40	34.03	18.26	24.60
AT_7–AT_{10}	21.4	22.5	23.4	17.5	31.7	17.3	22.9
AT_{11}–AT_{19}	9.8	7.5	4.9	0.9	2.3	0.9	1.7
TMB	25.94	24.99	22.18	5.09	0.00	6.00	9.30
TeMB	10.91	9.85	7.60	1.22	0.00	1.18	2.61
Tetralines	0.24	0.15	0.28	0.02	0.07	0.04	0.06
Diarenes	3.34	3.62	10.20	4.40	8.72	5.63	9.14
Naphthalenes	0.23	0.37	2.07	0.92	1.86	0.99	1.58
MN	0.89	0.97	3.97	1.64	3.29	2.12	3.16
DMN	1.00	1.20	2.68	1.30	2.44	1.73	2.82
TMB	0.79	0.70	1.00	0.45	0.93	0.67	1.34
TeMN	0.22	0.18	0.17	0.06	0.13	0.08	0.15
Cadalene	0.11	0.10	0.20	0.003	0.01	0.004	0.01
Fluorenes	0.09	0.10	0.11	0.03	0.05	0.04	0.08
Triarenes	0.02	0.14	0.11	0.05	0.14	0.05	0.16
Phenanthrene	0.02	0.01	0.06	0.02	0.06	0.02	0.06
MP	–	0.02	0.02	0.01	0.04	0.02	0.06
DMP	–	0.03	0.01	0.01	0.02	0.01	0.02
TMP	–	0.04	–	0.002	0.01	–	0.01
Retene	–	0.04	0.02	0.001	0.007	0.003	0.01
Adamantanes	3.01	2.51	1.50	0.39	–	0.49	0.91
	Geochemical parameters						
MNR	1.1	1.1	1.4	1.8	2.1	1.9	1.7
DNR2	0.9	0.6	0.9	1.7	1.7	1.7	1.8
TNR6	1.2	1.0	1.4	1.7	1.5	1.7	2.0
Cadalene/retene	–	2.4	8.9	2.5	1.7	1.5	1.3

Note. 1. MN – methyl naphthalenes, DMN – dimethyl naphthalenes, TMN – trimethyl naphthalenes; MP – methyl phenanthrenes, DMP – dimethyl phenanthrenes, TMP – trimethyl phenanthrenes; MNR = 2-(β-)methyl-/1-(α-)methylnaphthalene; DNR2 = [2,6-(β,β-) + 2,7-(β,β-)DMN]/1,5-(α,α-)DMN; TNR6 = [1,3,7-(α,β,β-) + 1,3,6-(α,β,β-)TMN]/[1,2,5-(α,β,α-) + 1,2,4-(α,β,α-)TMN]. 2. Dash means that the content was not determined.

Among monoarenes, *n*-alkyl benzenes (*n*-AB) (m/z 91), *n*-alkyl toluenes (*n*-AT) (m/z 105), TMB (m/z 119), TeMB (m/z 133) were identified. Pseudo-homologous series of long-chain alkyl benzenes with isoprenoid side chains are not present in the studied condensates. In addition to alkyl substituted benzenes, we also identified naphthene-substituted benzenes in the condensates – methyl- and dimethyltetralines. The amount of these compounds is not large and accounts for 0.1– 0.3 rel. %.

The content of long-chain *n*-AB and *n*-AT C_{11+} is very low in comparison with low-molecular alkyl benzenes C_7–C_{10} (see Table 3). The low content of long-chain AB in the condensates may be due to the degradation of alkyl benzenes with long substituents as a result of the thermal action at the stage of catagenesis with the formation of the components with shorter side chains [14, 15]. According to the literature data, the fraction of long-chain *n*-AB and *n*-AT in petroleum is higher than in gas condensates [27, 31]. In the studied samples, long-chain *n*-AB and *n*-AT are represented by the compounds containing 5 to 11–13 carbon atoms in the side substituents. The mass-fragmentogram reveals the ion *m*/*z* 105 which is characteristic of *n*-AT, and there is a series of triplets corresponding to the homologous series of *meta*-(*m*-), *para*-(*p*-) and *ortho*-(*o*-)methyl isomers, the relative content of which decreases with an increase in the number of carbon atoms in the molecule. The content of *p*-AT is much lower than that of *m*- and *o*-isomers.

All isomers of mono-, di-, tri-, and tetramethyl-substituted naphthalenes were identified in the studied condensates, as well as fluorenes containing two aromatic cycles and one naphthene cycle. The qualitative composition of alkyl naphthalenes is similar in all condensates, but differences are observed in their content (see Table 3). It was established in numerous studies that the differences are due to the type of initial OM and the conditions of sediment accumulation [9, 10, 12, 13, 32]. Analysis of the individual composition of alkyl naphthalenes showed that the compounds prevailing in all condensates are unsubstituted naphthalene, as well as 2- and 1-methyl naphthalenes. An increased content of 1,2,5- and 1,2,7-TMN, as well as the presence of cadalene, point to the participation of humic OM in the formation of these fluids [6], and sediment accumulation proceeded in marine or lacustrine deposits under oxygen-free or sub-oxidative conditions [9]. The amount of cadalene in the condensates from the Myldzhinskoye gas field is 0.1–0.2 rel. %, while in the condensates from the Maloyamalskoye gas field their content is small and equals 0.01 rel. % and less.

The data on the composition of alkyl naphthalenes were used to establish the degree of thermal transformation of the condensates [7, 12, 13, 32]. The values of methyl- (MNR), dimethyl- (DNR2) and trimethylnaphthalene (TNR6) indices >1 point to the predominance of thermodynamically more stable β-isomers over α-isomers, which allows us to conclude that the studied fluids are characterized by a rather high degree of transformation (see Table 3). No clear regularity was revealed in the changes of naphthalene parameters of these fluids depending on the depth of occurrence. Comparing the values of naphthalene indices for two gas fields, one should stress that the condensates from the Maloyamalskoye gas field are more transformed that those from the Myldzhinskoye gas field. However, the evaluation of the degree of transformation on the basis of the composition of the naphthalene fraction is used in geochemical practice in combination with the data on the composition of saturated HC biomarkers.

As mentioned above, the content of triaromatic HC in the studied condensates is insignificant. Nevertheless, practically all samples contain HC with three aromatic cycles – phenanthrene and its methyl-, di- and trimethyl-substituted isomers; 1-methyl-7-isopropylphenanthrene was identified (see Table 3). Among phenanthrene compounds, unsubstituted phenanthrene dominates. In view of the low content of methyl phenanthrenes, a reliable calculation of the geochemical parameter MPI1 = 1.5(2-MP + 3-MP)/(1-MP + 9-MP + P) could not be carried out. This parameter is widely used in geochemistry to determine the degree of transformation. The presence of retene along with cadalene points to the participation of coniferous plants in the formation of the initial OM of fluids [20, 22]. Close values of the ratios of cadalene to retene in different fluids within the boundaries of an oil and gas-bearing province may point to their common genetic source. One can see in the data shown in Table 3 that cadalene dominates over retene in the studied condensates. The ratio of cadalene to retene is higher in the samples from the Myldzhinskoye gas field than in the samples from the Maloyamalskoye gas field.

CONCLUSION

It is established that the qualitative composition of aromatic HC of gas condensates from two gas fields is similar but differences in the content were revealed.

On the basis of the values of the ratio of *m*- + *p*-xylene/*o*-xylene, which is used in geochemical practice as one of the parameters of the degree of maturing, it is established that the condensates from the Myldzhinskoye gas field should be related to more mature ones in comparison with those from the Maloyamalskoye gas field. The low content of long-chain alkyl benzenes AB C_{11+} in the condensates from both gas fields may be due to the degradation of alkyl benzenes with long substituents under thermal action at the stage of catagenesis with the formation of the components with shorter side chains.

A decreased content of ethyl benzene and the high content of *m*-xylene are the signs of the continental nature of the initial OM that had generated these condensates. In addition, an increased content of 1,2,5- and 1,2,7-TMN and the presence of cadalene also point to the fact that humic OM took part in the formation of these fluids, and sediment accumulation proceeded in marine or lacustrine deposits under oxygen-free or suboxidative conditions. The presence of retene along with cadalene points to the participation of coniferous plants in the formation of initial OM of the fluids. Cadalene dominates over retene in the studied condensates, the ratio of cadalene to retene being higher in the samples from the Myldzhinskoye gas field than in the samples from the Maloyamalskoye gas field.

Thus obtained results expand the knowledge on the features of the composition of aromatic HC in gas condensates and contribute into the accumulation of scientific data forming the basis for the development of the theory of the genesis of naphthides.

Acknowledgements

The work was carried out according to the project No. NIOKRT AAAA-A17-117030310199-1 within the framework of the state task.

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