

## Hydrocarbon Composition and Typification of Mongolia Oils Based on Their Mass Spectral Data

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### Abstract

Results of mass spectrometer determination of the comprehensive group content and molecular-mass distribution of hydrocarbons of 30 various structural types in lower Cretaceous oils of East Gobi and Tamsag oil rich provinces of Mongolia have been presented. Hydrocarbon types of oils according to three current ways of classification have been determined; factors responsible for the distinctions in types of oils being determined by these ways have been found.

### INTRODUCTION

Commercial reserves of oil are discovered in lower Cretaceous terrigenous depositions in the territory of East Gobi (Tsagan-Els, Zumbayan fields) and Tamsag (Tamsagbulag field) oil and gas rich provinces (OGP) of Mongolia. The basic geological-geochemical characteristics of these OGP have been described in works [1, 2], and general physicochemical properties of the made oils and the average structural parameters of their malthe ingredients (oils and resins) have been described in our work [3]. Hydrocarbons (HC) that represent the main body of Mongolian oils still have not been adequately explored: only specific samples of Zumbayan and Tamsagbulag oils became the objects of a detailed research [4, 5].

To complete this gap, we have found the comprehensive hydrocarbon composition of several typical oils from horizons that lie at the depths of 1000–1400 m in Tsagan-Els and Zumbayan fields, and in the range 2300–2500 m in the Tamsagbulag field. Results of this research are generalized in this paper.

### EXPERIMENTAL

Samples of oils, upon removal of traces of water and mechanical impurities from them, were divided into basic groups of ingredients (oils,

resins, asphaltenes) according to standard procedures [6, 7]. Asphaltenes were precipitated by dilution the oil sample with 40-fold volume of *n*-hexane. Deasphaltened oils were spread onto ASC silica gel, the mixture was loaded into a Soxhlet apparatus, and then extraction of oils was conducted with *n*-hexane, and extraction of resins, with benzene + ethanol mixture (1 : 1).

The extracted wide lube cuts were analysed with the use of MX-1310 mass spectrometer at a direct injection of sample in the ionisation chamber at 200 °C and at the energy of ionising electrons of 12 eV. The recorded spectra were processed following a semigraphical technique that had been developed in the Institute of Petroleum Chemistry, Siberian Branch of the Russian Academy of Science [8].

Concentrations of aliphatic HCs of normal and isoprenoid structures were determined by a reported gas chromatographic method that was described for example in [9]. The combination of chromatographic and mass spectrometer data made possible a separate determination of fractions of *n*-alkanes, isoprenanes, and non-isoprenoid isoalkanes as oil constituents.

### RESULTS AND DISCUSSION

As it was demonstrated previously [3], all studied oils of Mongolia, especially those from Tamsagbulag that occur deeper, are poor in

heteroelements. Their mass fraction of sulphur does not exceed 0.2 %. The nitrogen content in oils from Tamsag OGP is equally low; it shows a rise to 0.46 mass % only incidentally in East Gobi objects. Oxygen content of these oils does not exceed 1 and 2 mass %, respectively. In this relation, Tamsagbulag oils contain the less amount of resinous-asphaltene matter (RAM) (5–7 mass %) by comparison to those from East Gobi (9–12 mass %); they possess a smaller density (830–845 vs. 860–900 kg/m<sup>3</sup> at 20 °C) and a smaller average molecular mass (270–380 vs. 300–460 amu). All the studied oils are of high-wax nature, with the mass fraction of solid HCs of no less than 11 %.

According to classification with their concentration of *n*-alkanes and isoprenanes [9], all the described oils are rated in the A<sup>1</sup> type. If these oils are classified according to the ratio of their fractions of carbon atoms that are located in paraffin and naphthenic molecular structures (the  $f_p/f_n$  ratio) of all ingredients of oil in general, as it has been suggested in the work [10], then all the oils from East Gobi OGP are of methanoic or methane-naphthenic nature. From oil of the Tamsagbulag field, oil that occurs at the depth down to 2380 m falls into methane-naphthenic type according to the same attribute.

When in more dipped layers, naphthenic structures become a little dominating over paraffin structures in their abundance in molecules, and now these oils are rated in the naphthene-methane type.

Table 1 gives the group hydrocarbon composition of the studied Mongolian oils that has been determined from results of mass spectrometry analysis. Molecular mass distribution (MMD) of members of each homologous series of HC in typical oils of Mongolia is described in our works [4, 5] and it was taken into account in creation of the procedure to process the mass spectra [8].

*n*-Structured isomers that comprise 50–80 % of total alkanes play the leading role among aliphatic HCs in these oils. Isoprenanes comprise a minor group of HCs; their mass fraction in total alkanes does not exceed 9.3 %. The fraction of isoalkanes of alternate non-isoprenoid structure changes in a wide range (12–42 %). It appears that significant part of these HCs should be monomethyl-substitution isomers, that prevail in the composition of oil isoalkanes ([9], etc.).

Molecular mass distributions of *n*-alkanes and non-isoprenoid isoalkanes in the Mongolian oils include all members of the sequence to C<sub>40</sub>–C<sub>41</sub> and they exhibit higher than usual concentrations of all HCs that contain from 11–12 to 25–27 carbon atoms in their molecules (maxima fall on C<sub>17</sub> in East Gobi oils and on C<sub>15</sub> in Tamsagbulag oils).

Naphthenic HCs is the most abundant class of ingredients of the described oils; their mass fraction in oils amounts to 33–37 %. Alkylmonocyclanes dominate among naphthenes (52–58 % of total HCs in this class); fractions of other alicyclic compounds decrease quickly with an increase in the number of rings in their molecules.

Mass fraction of aromatic HCs in samples of the studied oils may amount from 26 to 30 %. They are presented for the most part by compounds with a benzene cycle in their molecules. Mass fraction of these compounds comprises 54–62 % from total arenes in oils of the Tsagan-Els area and up to 69 % in oils that occur more deeply in Zunbayan and Tamsagbulag fields. The combined content of bi- and polyarenes in Zunbayan and Tamsagbulag oils decreases with increasing number of aromatic cycles in the molecules. The relation is violated in oils of the Tsagan-Els area due to the high concentration of tetraarenes.

From monoarenes, alkylbenzenes dominate in all oils (37–44 mass % from the total of these HCs). Fractions of benzobicyclanes are anomalously reduced and fractions of benzotricyclanes are increased for the most part of the oils (especially in the Tsagan-Els field) on the background of a decreased concentration of naphthenobenzenes with the increasing number of rings in their molecules. Benzotricyclanes are most likely to be dehydroaromatized derivatives of biosynthesized steroids.

Biarenes in the studied oils of Mongolia are represented by derivatives of naphthalene and fluorene. The derivatives of naphthalene that contain up to three naphthenic rings in their molecules comprise 73–85 % of the total content of these HCs. Among naphtheno-naphthalenes and among benzopolycyclanes, fractions of tetracyclic compounds (naphtho-bicyclanes) that apparently are also genetically akin to oil steranes are appreciably higher than usual.

TABLE 1

Group hydrocarbon composition of the studied oils from various fields of Mongolia, mass %

| Hydrocarbons                    | Tsagan-Els, hole No.        |       |       | Zunbayan,<br>hole ZB-1 | Tamsagbulag,<br>hole No. 3 |
|---------------------------------|-----------------------------|-------|-------|------------------------|----------------------------|
|                                 | 142                         | 1410  | 14    |                        |                            |
|                                 | Average occurrence depth, m |       |       |                        |                            |
|                                 | 1007                        | 1170  | 1290  | 1372                   | 2440                       |
| <i>n</i> -Alkanes               | 14.38                       | 14.58 | 15.59 | 19.34                  | 18.40                      |
| Isoprenanes                     | 1.71                        | 2.19  | 1.80  | 1.82                   | 2.72                       |
| Other isoalkanes                | 9.06                        | 12.08 | 4.62  | 2.92                   | 8.28                       |
| <i>Isoalkanes (total)</i>       | 10.77                       | 14.27 | 6.42  | 4.74                   | 11.00                      |
| <i>Alkanes (total)</i>          | 25.15                       | 28.85 | 22.01 | 24.07                  | 29.41                      |
| Monocyclanes                    | 21.56                       | 18.98 | 20.09 | 18.75                  | 18.69                      |
| Bicyclanes                      | 8.96                        | 8.50  | 7.70  | 7.06                   | 8.87                       |
| Tricyclanes                     | 3.63                        | 3.34  | 3.44  | 6.56                   | 6.70                       |
| Tetracyclanes                   | 1.78                        | 1.25  | 1.81  | 1.38                   | 1.29                       |
| Pentacyclanes                   | 0.99                        | 0.62  | 2.01  | 0.90                   | 0.66                       |
| <i>Naphthenes (total)</i>       | 36.92                       | 32.64 | 35.05 | 34.65                  | 36.22                      |
| Alkylbenzenes                   | 3.47                        | 5.19  | 6.41  | 8.38                   | 7.63                       |
| Benzomonocyclanes               | 4.54                        | 2.75  | 5.05  | 4.29                   | 5.15                       |
| Benzobicyclanes                 | 0.41                        | 1.35  | 0.49  | 2.93                   | 2.18                       |
| Benzo-tricyclanes               | 2.91                        | 3.33  | 3.62  | 2.68                   | 2.46                       |
| Benzo-tetracyclanes             | 1.14                        | 1.31  | 1.37  | 1.60                   | 1.09                       |
| <i>Monoarenes (total)</i>       | 16.15                       | 13.93 | 16.94 | 19.88                  | 18.52                      |
| Alkyl-naphthalenes              | 1.14                        | 1.55  | 1.02  | 0.90                   | 1.42                       |
| Naphthomonocyclanes             | 0.72                        | 1.39  | 0.75  | 0.85                   | 0.80                       |
| Naphthobicyclanes               | 0.85                        | 1.18  | 0.95  | 1.31                   | 0.93                       |
| Naphthotricyclanes              | 0.93                        | 0.78  | 0.54  | 0.94                   | 0.67                       |
| <i>Naphthalenes (total)</i>     | 3.65                        | 4.90  | 3.26  | 3.99                   | 3.83                       |
| Alkylfluorenes                  | 0.46                        | 0.52  | 0.41  | 0.22                   | 0.26                       |
| Monocyclanofluorenes            | 0.51                        | 0.50  | 0.22  | 0.40                   | 0.19                       |
| Bicyclanofluorenes              | 0.17                        | 0.83  | 0.35  | 0.13                   | 0.22                       |
| <i>Fluorenes (total)</i>        | 1.13                        | 1.85  | 0.98  | 0.74                   | 0.68                       |
| <i>Biarenes (total)</i>         | 4.78                        | 6.75  | 4.24  | 4.74                   | 4.50                       |
| Alkyl- (Ph + A)                 | 0.49                        | 0.38  | 0.61  | 0.44                   | 0.27                       |
| Monocyclano- (Ph + A)           | 0.59                        | 0.43  | 0.75  | 0.49                   | 0.91                       |
| Bicyclano- (Ph + A)             | 0.27                        | 0.21  | 0.53  | 0.71                   | 0.44                       |
| <i>Ph + A (total)</i>           | 1.35                        | 1.02  | 1.89  | 1.64                   | 1.62                       |
| Alkylbenzofluorenes             | 0.25                        | 0.20  | 0.17  | 0.29                   | 0.19                       |
| Monocyclanobenzofluorenes       | 0.07                        | 0.05  | 0.12  | 0.13                   | 0.09                       |
| <i>Benzofluorenes (total)</i>   | 0.32                        | 0.25  | 0.29  | 0.42                   | 0.28                       |
| <i>Triarenes (total)</i>        | 1.67                        | 1.27  | 2.18  | 2.06                   | 1.90                       |
| Alkylpyrenes                    | 0.18                        | 0.16  | 0.11  | 0.21                   | 0.25                       |
| Monocyclanopyrenes              | 0.12                        | 0.04  | 0.05  | 0.16                   | 0.10                       |
| <i>Pyrenes (total)</i>          | 0.30                        | 0.20  | 0.16  | 0.37                   | 0.35                       |
| Alkylchrysenes                  | 0.13                        | 0.06  | 0.10  | 0.15                   | 0.09                       |
| Monocyclanochrysenes            | 1.99                        | 2.15  | 2.00  | 0.41                   | 0.75                       |
| <i>Chrysenes (total)</i>        | 2.11                        | 2.21  | 2.10  | 0.56                   | 0.83                       |
| Alkyldibenzofluorenes           | 0.51                        | 0.52  | 0.41  | 0.33                   | 0.36                       |
| <i>Tetraarenes (total)</i>      | 2.91                        | 2.93  | 2.67  | 1.27                   | 1.44                       |
| Perylenes + benzopyrenes        | 0.80                        | 0.87  | 0.86  | 0.43                   | 0.34                       |
| Picenes                         | 0.27                        | 0.27  | 0.30  | 0.32                   | 0.20                       |
| <i>Pentaarenes (total)</i>      | 1.08                        | 1.14  | 1.16  | 0.74                   | 0.54                       |
| <i>Arenes (total)</i>           | 26.59                       | 26.02 | 27.19 | 28.69                  | 26.91                      |
| It has been identified total of | 88.69                       | 87.51 | 84.25 | 87.41                  | 92.53                      |
| The ratio A/N                   | 0.79                        | 0.88  | 0.63  | 0.70                   | 0.81                       |

Note. Here and in Table 2: A – alkanes, N – naphthenes, Ph – phenanthrenes.

From triarenes, derivatives of phenanthrene, anthracene, and benzofluorene are present in the oils. Compounds of the first two structural types are present in the oils in much greater quantities as compared to benzofluorenes, the fraction of which is no more than 20 % of total triarenes. Isomeric phenanthrenes (Ph) and anthracenes (A) cannot be separately determined by means of mass spectrometry; therefore, Table 1 specifies their combined concentrations (Ph + A). An assumption can be made that HCs with phenanthrene nucleus in the molecules are the predominant type of triarenes in the oils in point too as it was evidenced previously in oils of other OGP [9]. Among the (Ph + A) HCs, mono- or, much less often, bicyclane derivatives dominate in all Mongolian oils, and among benzofluorenes, those alkyl-substituted dominate.

The availability of derivatives of pyrene, chrysene, and dibenzofluorene in the Mongolian oils has been found. Monocyclanochrysenes that are akin in their skeleton to pentacyclic triterpanes (gopanes) have been detected in greatest quantities in all oils. These HCs comprise 68–75 % from total tetraarenes in oils of the Tsagan-Els area; although their fraction in Tamsagbulag oils that occur deeper drops down to 52 %, and even to 32 % in oil of the Zunbayan field. Oils of the Tsagan-Els field differ from others in terms of their very low relative content of pyrenes (6–10 % from total tetraarenes *vs* 24–30 % in oils of the Zunbayan and Tamsagbulag fields).

Pentaarenes in oils are represented by alkyl-substituted picenes, as well as by perylenes and benzpyrenes that cannot be distinguished in their mass spectra. Picenes amount to ~25 % of all pentaarenes in oils of Tsagan-Els; and their fraction in the remaining oils under investigation rises to 38–43 %. It appears that perylene derivatives that have been revealed in significant amounts previously in alternative oils (to take an example, in Sakhalin oils) play the dominating role among pentaarenes [11].

Molecular mass distributions of HCs of all observed structural types in the studied Mongolian oils are in complete agreement with previously found features [8] that consist in the fact that alkanes, except for isoprenanes, and HCs that contain 1–2 cycles each in their mole-

cules (namely, mono- and bicyclanes, alkylbenzenes and benzomonocyclanes, alkyl naphthalenes and alkylfluorenes) form in oils extensive, monotonous, unimodal series ranging up to  $C_{40}$ – $C_{41}$  with similar positions of the main maxima and thus they include long alkyl substituents in their molecules.

Tetra- and pentacyclic HCs form narrow series that begin with the lowest theoretically possible member ( $C_{17}$  and  $C_{21}$ , respectively) and that terminate with  $C_{35}$  compounds. These include successions of tetra- and pentacyclanes, benzotri- and benzotetracyclanes, naphthobi- and naphthotricyclanes, mono- and dinaphthenophenanthrenes. The maximum content is usually typical of  $C_{27}$ – $C_{30}$  and  $C_{29}$ – $C_{31}$  members in the respective series of tetra- and pentacyclic HCs. These HCs together with monocyclanochrysenes are genetically related with a high probability to biosynthetic steroids and triterpenoids. Hydrocarbons with three-to-five condensed aromatic cycles in their molecules (alkylphenanthrenes, derivatives of pyrene, picene, of benz- and dibenzfluorenes) are present in the oils in the form of narrow unimodal series of homologues that are restricted with a small number (5–6) of the lowest members.

HCs that comprise total of 84–93 % from their mass or 96–98 % from the mass of lube cut of oil have been identified and quantitatively determined as the constituents of the oils. Non-identified ingredients appear to be hetero-organic compounds or HCs that contain more than five cycles in their molecules. These HCs in small proportion have been detected previously in oils of other regions [9].

According to [8], as judged from the ratio of combined concentrations of alkanes and naphthenes in oils in general (0.63–0.88), all the Mongolian objects should be classed with naphthene-methane type. Meanwhile, the assignment of the majority of them to other types, namely to methane-naphthenic or even to methane type, has been made previously on the basis of the ratios of the fractions of carbon atoms in aliphatic and alicyclic molecular structures [3]. It is apparent that these discrepancies are determined by the great abundance of monocyclanes and monoarenes with long alkyl molecular fragments in the oils.

TABLE 2

Group content of petrol fractions C<sub>7</sub>–C<sub>11</sub>, mass %

| Hydrocarbons                         | Tsagan-Els, hole No. |       |       | Zunbayan,<br>hole BZ-1 | Tamsagbulag,<br>hole No. 3 |
|--------------------------------------|----------------------|-------|-------|------------------------|----------------------------|
|                                      | 142                  | 1410  | 14    |                        |                            |
| <i>n</i> -Alkanes                    | 10.94                | 8.02  | 21.57 | 19.71                  | 15.28                      |
| Isoprenanes                          | 3.78                 | 3.06  | 2.11  | 1.59                   | 4.34                       |
| Other isoalkanes                     | 17.33                | 28.16 | 11.29 | 20.83                  | 24.14                      |
| <i>Isoalkanes (total)</i>            | 21.11                | 31.22 | 13.40 | 22.42                  | 28.48                      |
| <i>Alkanes (total)</i>               | 32.05                | 39.24 | 34.97 | 42.13                  | 43.76                      |
| Alkylmonocyclanes                    | 34.43                | 35.95 | 49.12 | 37.43                  | 25.03                      |
| Alkylbicyclanes                      | 12.74                | 13.71 | 13.52 | 3.64                   | 11.64                      |
| Alkyltricyclanes                     | 1.31                 | 0.04  | 0.50  | 0                      | 0                          |
| <i>Naphthenes (total)</i>            | 48.48                | 49.66 | 63.14 | 41.07                  | 36.67                      |
| Alkylbenzene                         | 4.82                 | 4.79  | 9.03  | 13.65                  | 10.66                      |
| Benzomonocyclanes                    | 13.13                | 3.47  | 2.54  | 2.33                   | 6.12                       |
| Alkyl naphthalenes                   | 1.52                 | 1.89  | 1.93  | 0.82                   | 2.79                       |
| <i>Arenes (total)</i>                | 19.47                | 10.15 | 13.50 | 16.80                  | 19.57                      |
| <i>Overall yield of the fraction</i> | 25.12                | 11.10 | 11.62 | 13.20                  | 8.52                       |
| The ratio A/N                        | 0.66                 | 0.79  | 0.55  | 1.03                   | 1.19                       |

Note. For designation, see Table 1.

The obtained mass spectral data make it possible to perform typification of oils also in terms of the group hydrocarbon composition of their petrol fractions according to the most common present-time ways of classification that have been suggested by the authors [12, 13]. As a matter of fact, these ways are identical, and their distinctions consist only in the way they make up the names of oil types. Indeed, data on the composition of fractions that boil away mostly in the range of 70–215 °C are available upon summarizing the concentrations of HC C<sub>7</sub>–C<sub>11</sub>, the range being close to the actual one for usually selected petrols. Table 2 gives the results of these computations.

The combined content of petrol HCs C<sub>7</sub>–C<sub>11</sub> in East Goby oils changes within the limits of 11–13 mass %. This quantity increases up to 25 mass % only in oil from the hole 142 of the Tsagan-Els field that was extracted from the least occurrence depth of the productive layer, which is unusual since the concentration of low boiling HCs in oil, as a rule, increases with the increasing occurrence depth. The fraction of HC C<sub>7</sub>–C<sub>11</sub> in Tamsagbulag oil (8.5 %) appeared to be still lower in spite of its greater occurrence depth.

Low boiling fractions of oils appeared to be much poorer in terms of their aromatic hydrocarbons and much richer in terms of their paraffin hydrocarbons as compared with the same oils in general. This is in agreement with the reported fact that concentrations of aromatic HCs in oil fractions increase with the increasing boiling temperature. However, fractions of naphthenes in petrols that are present in oils of East Goby OGP (in particular, concentrations of monocyclanes, and also those of bicyclanes in Tsagan-Els oils) are much higher than in the same objects in general, *i.e.* in middle and high boiling oil fractions. To put it otherwise, the concentrations of naphthenic HCs in distillate fractions of these oils drop with an increase in their boiling point whereas the reverse tendency was evidenced for the oils studied earlier from alternative oil rich areas.

Based on the analysis of petrol compositions, one more set of names of oil types has been suggested (in terms of [12]). It is evident that they do not agree with previously specified types by virtue of the distinctions of the criteria that have been used when classifying the objects. According to the “petrol” classification scheme, oils of the Tsagan-Els field are considered naph-

thene-methane, and Zunbayan and Tamsagbulag oils are considered methane-naphthenic. It is evident that phenomena of "methanization" of oils with depth are reflected to the full extent on the composition of petrol HCs that are absent, as a rule, from organic matter that is synthesized by living organisms, and they are formed in the course of catagenic transformations of these materials underground.

## CONCLUSION

Group hydrocarbon compositions for typical oils of East Gobi and Tamsag oil rich provinces of Mongolia and petrol fractions they involve have been found. HC with long linear or with weakly branched alkyl chains in their molecules (*n*-alkanes, monomethylalkanes, alkylmonocycloalkanes, alkylbenzenes, etc.) comprise no less than 40–46 mass % of the oils. Molecular-mass distributions of members of these HC homologous series include all ingredients up to C<sub>40</sub>–C<sub>41</sub>. This fact testifies that phenomena of biodegradation of oils exerted no pronounced effect on the processes of formation of their composition underground. Total concentration of these HC rises appreciably with an increase in occurrence depths of productive layers. The high content of these HC was the factor responsible for discrepancies in the determination of hydrocarbon types of oils based on the ratios of methane and naphthene HCs or of average frac-

tions of carbon atoms in aliphatic and alicyclic structures in molecules of oil ingredients.

A distinctive feature of the composition of the studied Mongolian oils is reducing the concentration of naphthenic HCs in oil fractions with an increase in their boiling point. This reducing is uncharacteristic for oils of other oil rich provinces.

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