Biodegradation characteristics of oil shale in the Permian Lucaogou Formation in the southeast of the Junggar Basin, China: implications from organic geochemistry

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Abstract. The Permian Lucaogou Formation oil shale in the southeast of the Junggar Basin is the oldest oil shale in China. In this study, the biodegradation characteristics of the oil shale are investigated based on organic geochemistry. The analytical results show that the biodegraded oil shale contains large humps of unresolved complex mixtures (UCMs) and is depleted of n-alkanes and acyclic isoprenoids (pristine (Pr), phytane (Ph), iC_{18}, iC_{17}, iC_{16}, iC_{15} and iC_{14}). Furthermore, C_{14}–C_{16} bicyclic terpanes are completely preserved. It can be concluded that the biodegradation level of the Lucaogou Formation oil shale is PM1–PM2. In addition, the values of aromatic hydrocarbon proxies (methyldibenzothiophene/dibenzothiophene (MDBT/DBT), dimethyldibenzothiophene/methyldibenzothiophene (DMDBT/MDBT), 1,2,7-trimethylnaphthalene/1,3,6-trimethylnaphthalene (1,2,7-TMN/1,3,6-TMN) and 1,2,4-trimethylnaphthalene/1,6,7-trimethylnaphthalene (1,2,4-TMN/1,6,7-TMN)) also reflect a low biodegradation level. The well-preserved TMN derivatives indicate that the biodegradation level of the Lucaogou Formation oil shale is below PM3.

Keywords: biodegradation characteristics, Lucaogou Formation oil shale, organic geochemistry, Junggar Basin.

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1. Introduction

Understanding and interpreting biodegradation in organic geochemistry is a difficult issue. Biodegradation can affect the structure and composition of organic matter in sediments, significantly changing its properties [1–3]. The distribution of saturated and aromatic hydrocarbons can also be influenced by biodegradation. Thus, the accuracy of determining the source of organic matter and evaluating its thermal maturity, as well as migration of oil may also be distorted because of biodegradation [4]. Generally, biodegradation is controlled by many factors such as microbe population, nutrient supply, oxygen content, pH and temperature [3]. Different influencing factors may lead to different biodegradation levels because the resistance of various organic geochemical materials to the process is different [2, 3]. Thus, biodegradation in sediments is important to be researched.

The Permian Lucaogou Formation oil shale in the Junggar Basin is the oldest oil shale in China. It was formed during the period of 255–260 Ma [5, 6]. The quality of the oil shale is very good, its total organic carbon (TOC, %) ranges from 0.5 to 20.17%, averaging 8.49%. The Lucaogou Formation oil shale in the Junggar Basin has been systematically studied using organic geochemistry, elemental geochemistry and sedimentary geology [7–9]. However, the biodegradation characteristics of the oil shale have not been reported yet. In this work, the evidences of biodegradation from organic geochemistry are analyzed, which will be of great significance for the assessment of the Lucaogou Formation oil shale.

2. Geological setting

The Junggar Basin is one of the most important oil shale basins in the northwest of China (Fig. 1a). It is situated next to the Santanghu and Turpan-Hami basins [10] (Fig. 1b). The study area of this research is located in the southeast of the Junggar Basin. A lot of complex fault and anticline structures are widely developed in the sediments in the study area because of intensive tectonic activities during the Mesozoic and Cenozoic. In addition, the Carboniferous, Triassic, Jurassic, Permian, Neogene and Quaternary strata are widely exposed in the study area (Fig. 1c).

The study area is a sedimentary depression formed during the Lucaogou Formation sedimentary period. High-quality oil shale in large amounts was widely developed during that period [11, 12]. The major rock types of the Lucaogou Formation are oil shale, mudstone, calcareous mudstone, dolomite, calcareous siltstone, siltstone, fine-grained sandstone and medium-grained sandstone. Based on the continental sequence stratigraphy division principles, the Lucaogou Formation is divided into two complete third-order cycles, which correspond to the lower and upper member, respectively [7–9].
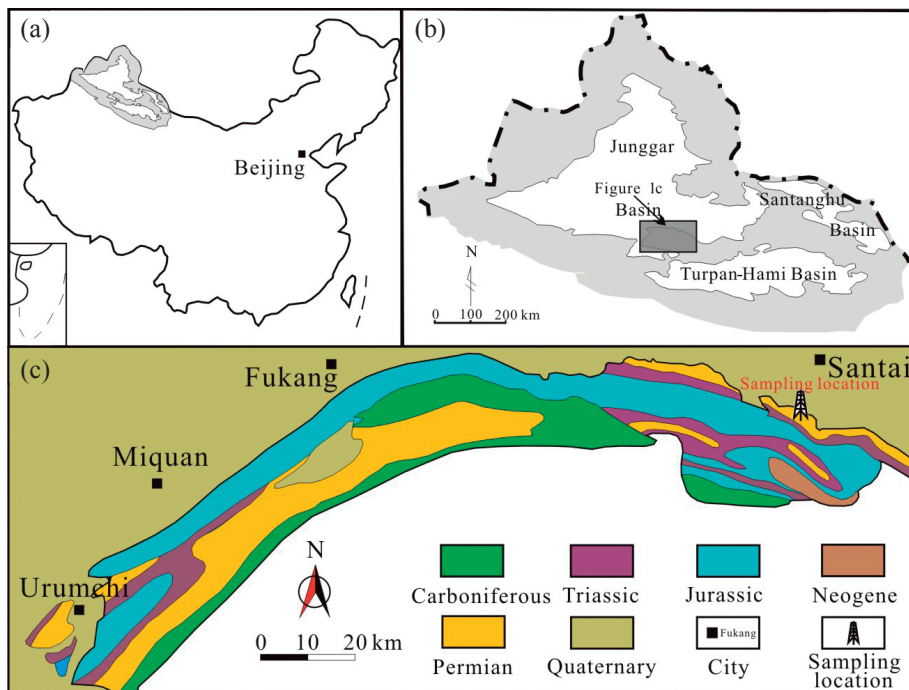
Fig. 1. The location and stratigraphic column of the Lucaogou Formation in the Junggar Basin, NW China: (a) the location of the northwestern areas in China; (b) the location of the northern foot of Bogda Mountains (modified from [12]); (c) the location of the JZK well in the study (modified from [9]).

3. Material and tests

A total of 10 oil shale samples were collected from the JZK well in the Junggar Basin. The well reveals the Permian Lucaogou Formation stratum in the southeast of the basin, NW China (Fig. 1c). All the samples were separated into saturated and aromatic hydrocarbons. Firstly, the organic matter (OM) was extracted from powdered rocks with the ASE 300 Dionex Accelerated Solvent Extractor. The extracted organic matter (EOM) was separated into saturated and aromatic hydrocarbons. Then, the fractions of said hydrocarbons were analyzed by gas chromatography-mass spectrometry (GC-MS) on a Leco Pegasus 4D Time-of-Flight Gas Chromatograph interfaced to an Agilent 7890A Gas Chromatograph. The mass spectrometer (MS) was operated at 200 °C in EI-mode at a 70 eV ionization energy. The fragmentograms of *n*-alkanes and acyclic isoprenoids (m/z 57), bicyclic terpanes (m/z 123), methyldibenzothiophene (MDBT; m/z 198), dibenzothiophene (DBT; m/z 184), dimethyldibenzothiophene (DMDBT; m/z 212) and trimethylnapthalene (TMN; m/z 170) were obtained in this work. All the test data are presented in Table.

Table. Saturated and aromatic hydrocarbons from the Lucaogou Formation oil shale in the Junggar Basin

Sample No.	$Pr/n\text{-}C_{17}$	$Ph/n\text{-}C_{18}$	$(Pr+Ph)/(n\text{-}C_{17}+n\text{-}C_{18})$	CPI_{13-19}	CPI(1)	OEP(1)	MDBT/ DBT	DMDBT/ MDBT	1,2,7-TMN/ 1,3,6-TMN	1,2,4-TMN/ 1,6,7-TMN
J1-3	5.98	5.20	5.65	0.94	1.04	1.00	2.32	1.08	0.10	0.11
J1-4	3.91	2.81	3.35	0.77	1.00	0.95	2.39	1.52	0.09	0.11
J1-15	2.84	4.00	3.30	1.02	1.06	1.04	4.41	1.93	0.12	0.11
J1-23	2.31	2.13	2.22	0.93	1.02	0.96	2.34	1.55	0.11	0.13
J1-32	6.46	4.46	5.50	0.92	0.95	0.75	1.20	1.27	0.11	0.20
J1-40	1.82	3.18	2.37	0.98	0.93	0.78	0.93	5.77	0.11	0.23
J1-41	0.50	0.55	0.53	1.03	1.09	1.03	0.12	1.51	0.10	0.14
J1-25	0.67	0.26	0.48	0.91	1.06	0.95	1.41	1.27	0.07	0.11
J1-30	0.85	0.78	0.82	0.90	1.08	1.04	2.57	1.41	0.07	0.16
J1-31	0.36	0.29	0.32	0.92	1.12	1.09	3.72	1.55	0.05	0.16

Note: Pr – pristane; Ph – phytane; MDBT – methyldibenzothiophene; DBT – dibenzothiophene; DMDBT – dimethyldibenzothiophene; TMN – trimethylnaphthalene. $OEP(1) = (n\text{-}C_{21} + 6\, n\text{-}C_{23} + n\text{-}C_{25})/(4\, n\text{-}C_{22} + 4\, n\text{-}C_{24})$, $CPI(1) = 2\,(n\text{-}C_{23} + n\text{-}C_{25} + n\text{-}C_{27} + n\text{-}C_{29})/[n\text{-}C_{22} + 2\,(n\text{-}C_{24} + n\text{-}C_{26} + n\text{-}C_{28}) + n\text{-}C_{30}]$, $CPI_{13-19} = [n\text{-}C_{13} + 2\,(n\text{-}C_{15} + n\text{-}C_{17}) + n\text{-}C_{19}]/[2(n\text{-}C_{14} + n\text{-}C_{16} + n\text{-}C_{18})]$.

4. Results

4.1. Unresolved complex mixtures

The "baseline hump" which can be observed in the biodegraded oil shale samples is attributed to unresolved complex mixtures (UCMs). Generally, biodegradation can promote the decomposition of *n*-alkanes, alkylcyclohexanes, isoprenoids, steranes and terpaneoids. The cycloalkane homologues that strongly resist degradation are the major constituents of UCMs. These complex compounds can be verified in gas chromatography (GC) chromatograms as a "baseline hump". The organic compound types of UCMs amount to 25000 [3, 13]. UCMs can be clearly seen in total ion chromatograms (TICs) (Fig. 2a), suggesting that the Lucaogou Formation oil shale has undergone biodegradation. In addition, it is worth noting that hopanes are highly abundant in the Lucaogou oil shale samples (Fig. 2a).

4.2. *n*-Alkanes and acyclic isoprenoids

The *n*-alkanes and isoprenoids from saturated hydrocarbons are the most sensitive components to biodegradation [3, 14]. The peaks of *n*-alkanes of some samples have been lost in chromatograms. Both the short-chain carbon number ($< n\text{-}C_{16}$) and long-chain carbon number ($> n\text{-}C_{19}$) species show a significant decrease, which suggests their higher susceptibility to biodegradation compared to the middle-chain carbon number homologues (Fig. 2b). Thus, the presence of *n*-alkanes indicates that the Lucaogou Formation oil shale has suffered biodegradation. In addition, the abundance of acyclic isoprenoids also reflects biodegradation. Generally, the specific acyclic isoprenoids exhibit stronger resistance to biodegradation than *n*-alkanes. The acyclic isoprenoids would be partially consumed by degradation microorganisms when organic matter undergoes more intensive biodegradation [3]. The distribution of acyclic isoprenoids in some oil shale samples shows pristine (Pr), phytane (Ph), iC_{18}, iC_{17}, iC_{16}, iC_{15} and iC_{14} to be more abundant than the adjacent *n*-alkanes (Fig. 2b), suggesting that the Lucaogou Formation oil shale has experienced biodegradation. This is consistent with the results obtained for UCMs and *n*-alkanes.

4.3. Bicyclic terpanes

C_{14}–C_{15} bicyclic terpanes can act as indicators of biodegradation [15]. These compounds can be revealed by m/z 123 mass chromatograms. They can also be tested in some oil shale samples. C_{14}–C_{15} bicyclic terpanes are well preserved in the biodegraded oil shale samples (Fig. 2c), which suggests the low level of biodegradation of the Lucaogou Formation oil shale.
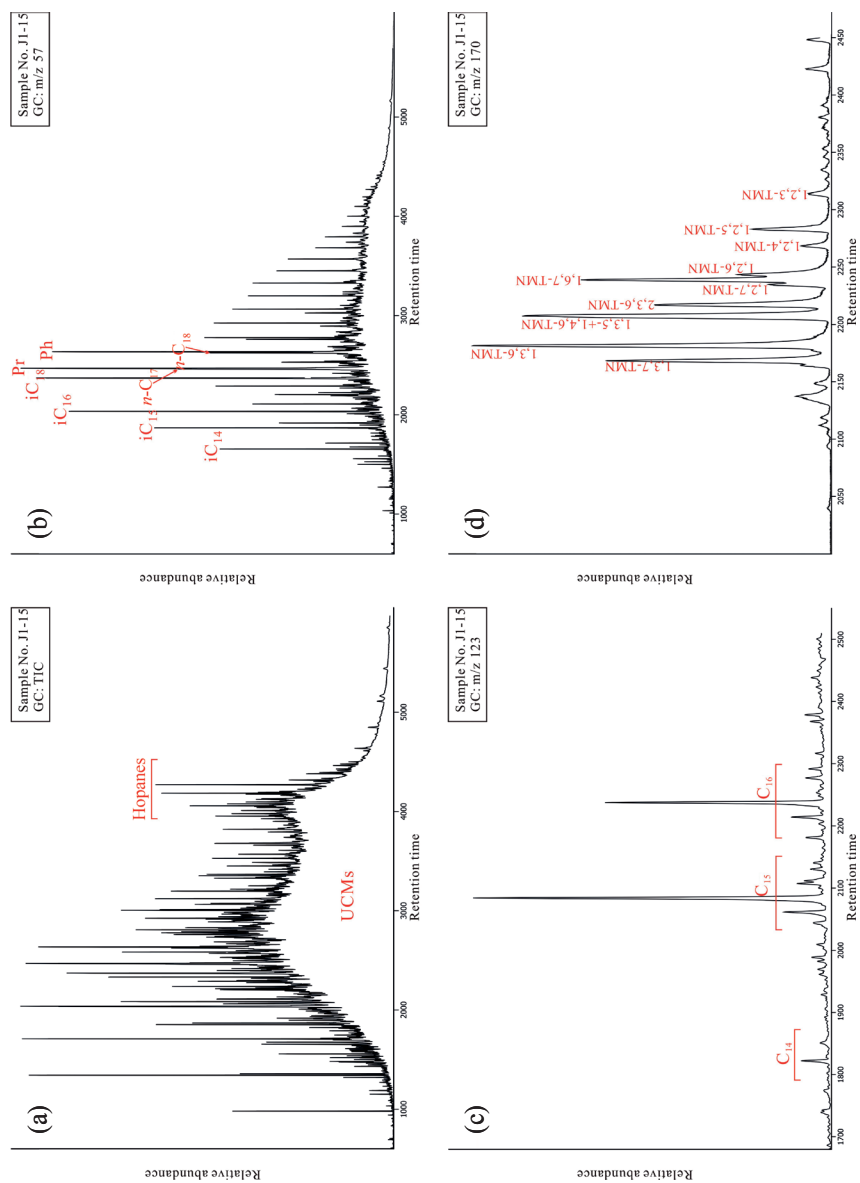
Fig. 2. The total ion chromatograms and mass chromatograms of the Lucaogou Formation oil shale: (a) total ion chromatograms showing UCMs and multiple hopanes of the oil shale; (b) mass chromatograms m/z 57 showing the distribution of acyclic isoprenoids (iC$_{15}$, iC$_{16}$, iC$_{17}$, iC$_{18}$, Pr and Ph) and n-alkanes (n-C$_{17}$ and n-C$_{18}$) of the oil shale; (c) mass chromatograms m/z 123 showing the complete C$_{14}$–C$_{16}$ bicyclic terpanes of the oil shale; (d) mass chromatograms m/z 170 showing the complete trimethyl-napthalenes of the oil shale.

4.4. Trimethylnapthalene

Parameters relating to aromatic hydrocarbons can also be used to ascertain the degree of biodegradation. Previous studies demonstrated that some of these parameters can be used to evaluate the biodegradation extent of sediments. Trimethylnaphthalene (TMN) is a useful proxy for biodegradation [15]. The abundance and distribution of TMN derivatives can be revealed by m/z 170 mass chromatograms. Several TMN derivatives have been observed in the Lucaogou oil shale samples. The fact that the compounds are well preserved (Fig. 2d) suggests the low level of biodegradation.

5. Discussions

5.1. Biodegradation characteristics

The plot of pristane/n-C$_{17}$ vs phytane/n-C$_{18}$ can be used to distinguish the biodegradation degree of sediments [16]. The biodegraded oil shale samples are located in the biodegraded sediments area and the unbiodegraded oil shale samples are in the unbiodegraded sediments area (Fig. 3a). The phytane/n-C$_{18}$ and pristane/n-C$_{17}$ values of biodegraded oil shale samples range from 1.82 to 6.46 (averaging 3.89) and from 2.13 to 5.20 (averaging 3.63), respectively, while those of unbiodegraded samples vary between 0.26 and 0.78 (averaging 0.47) and from 0.36 to 0.85 (averaging 0.60), respectively.

The carbon preference index (CPI) of n-C$_{13–19}$ and CPI(1) are parameters used to evaluate the thermal maturity and biogradation of sediments [17]. The plots of CPI$_{13–19}$ vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$), CPI(1) vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$) and odd-even predominance (OEP)(1) vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$) are usually used to distinguish the biodegradation degree of sediments [17, 18]. The (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$) values of biodegraded and unbiodegraded oil shale samples are respectively in the range of 2.22–5.65 (averaging 3.73) and 0.32–0.82 (averaging 0.54). In these three plots, the biodegraded oil shale samples are located in the biodegraded sediments area, while the unbiodegraded oil shale samples are in the unbiodegraded sediments area (Figs. 3b–d). The CPI$_{13–19}$, CPI(1) and OEP(1) values of oil shale samples, which range respectively from 0.90 to 1.03 (averaging 0.93), from 0.93 to 1.12 (averaging 1.04) and from 0.75 to 1.09 (averaging 0.96), suggest the low degree of biodegradation. Thus, it can be concluded that the high values of (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$) are due to biodegradation [3, 19].

Aromatic hydrocarbons are effective proxies of biodegradation. The plots of MDBT/DBT vs DMDBT/MDBT and 1,2,7-TMN/1,3,6-TMN vs 1,2,4-TMN/1,6,7-TMN are used to determine the biodegradation degree of sediments [3, 15, 20]. Generally, the resistance of the abovementioned compounds to biodegradation obeys the following order: MDBT > DBT, DMDBT > MDBT, 1,2,7-TMN > 1,3,6-TMN and 1,2,4-TMN > 1,6,7-TMN. Compared to the unbiodegraded sediments, the biodegraded sediments have higher values of MDBT/DBT, DMDBT/MDBT, 1,2,7-TMN/1,3,6-TMN and
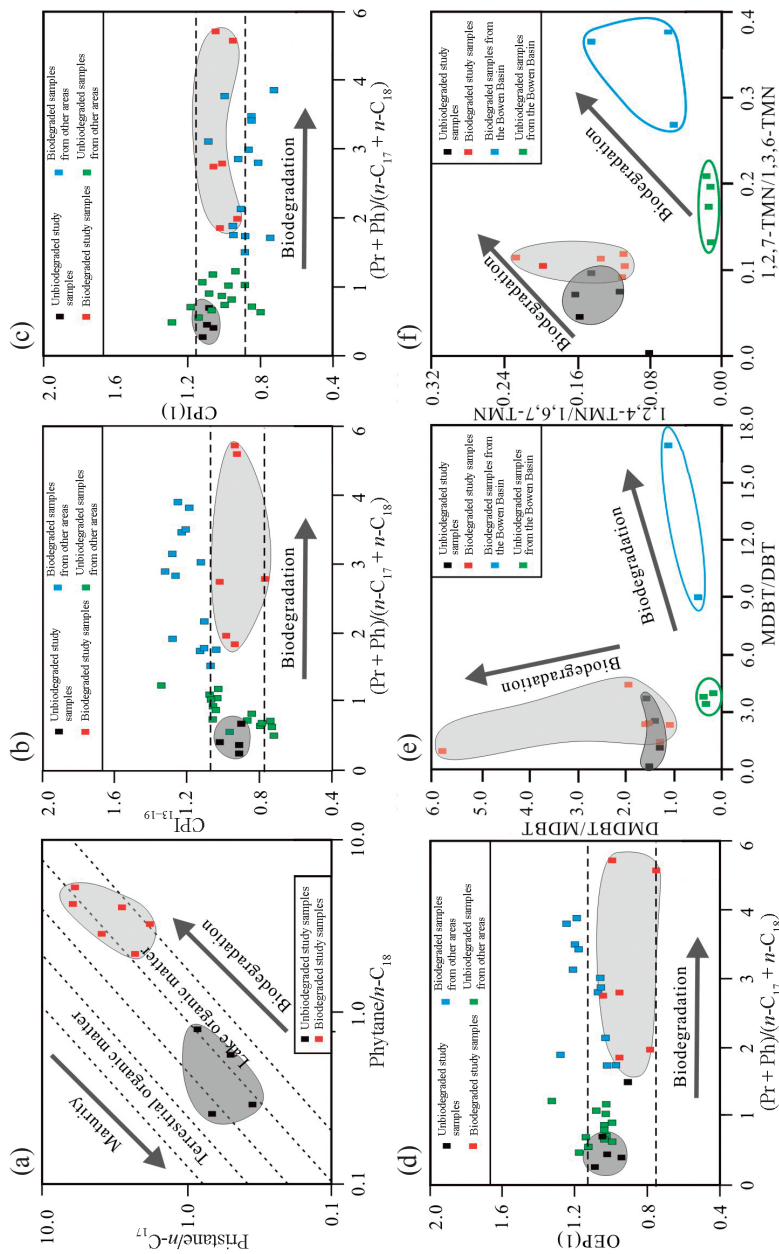
Fig. 3. The cross-plots demonstrating the biodegradation degree of the studied samples: (a) plot of pristane/n-C$_{17}$ vs phytane/n-C$_{18}$; (b) plot of CPI$_{13-19}$ vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$); (c) plot of CPI(1) vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$); (d) plot of OEP(1) vs (Pr + Ph)/(n-C$_{17}$ + n-C$_{18}$); (e) plot of MDBT/DBT vs DMDBT/MDBT; (f) plot of 1,2,7-TMN/1,3,6-TMN vs 1,2,4-TMN/1,6,7-TMN. [(OEP(1) = (n-C$_{21}$ + 6 n-C$_{23}$ + n-C$_{25}$)/(4 n-C$_{22}$ + 4 n-C$_{24}$) and CPI(1) = 2 (n-C$_{23}$ + n-C$_{25}$ + n-C$_{27}$ + n-C$_{29}$)/[n-C$_{22}$ + 2 (n-C$_{24}$ + n-C$_{26}$ + n-C$_{28}$) + n-C$_{30}$] are from [3], CPI$_{13-19}$ = [n-C$_{13}$ + 2 (n-C$_{15}$ + n-C$_{17}$) + n-C$_{19}$]/2(n-C$_{14}$ + n-C$_{16}$ + n-C$_{18}$) is from [18]. (The data for oil shale from the other areas in Figures 3b and 3c and are from [17, 18], the data for the Australian Bowen Basin oil shale in Figures 3e and 3f are from [15]).

1,2,4-TMN/1,6,7-TMN [15]. The DMDBT/MDBT values of biodegraded oil shale samples range from 1.08 to 5.77, averaging 2.19, while those of unbiodegraded oil shale samples vary between 1.27 and 1.55, averaging 1.43. However, the MDBT/DBT values of all oil shale samples fall into a narrow range, 0.12–4.41. The above values suggest that the Lucaogou Formation oil shale has undergone biodegradation but these are different from the respective figures for the oil shale samples from the Bowen Basin in Australia [15] (Fig. 3e). The 1,2,7-TMN/1,3,6-TMN and 1,2,4-TMN/1,6,7-TMN of Lucaogou oil shale samples are in a narrower range compared to Australian Bowen Basin oil shale samples. The 1,2,7-TMN/1,3,6-TMN values of biodegraded and unbiodegraded oil shale samples vary respectively between 0.09 and 0.12 (averaging 0.11) and from 0.05 to 0.10 (averaging 0.07). The 1,2,4-TMN/ 1,6,7-TMN figures of biodegraded and unbiodegraded oil shale samples range from 0.11 to 0.23 (averaging 0.15) and from 0.11 to 0.16 (averaging 0.14), respectively. The TMN derivatives are completely preserved in oil shale samples. At the same time, several parameter values of biodegraded oil shale samples are higher than those of unbiodegraded ones (Fig. 3f).

5.2. Biodegradation level

Based on the above analysis, the biodegradation level of the Lucaogou Formation oil shale can be assessed. Generally, the biodegradation of oil shale begins with the loss of straight-chain *n*-alkanes, followed sequentially by acyclic isoprenoids, and highly-branched and cyclic saturated hydrocarbons [21]. The loss of these compounds contributes to an increase in UCMs. In addition, some scholars developed a quasi-systematic classification scheme (PM scale) to rank the level of biodegradation with PM1 denoting the lowest level and PM10 the highest level of biodegradation. The PM scale has been widely used in various cases. The initial alteration, substantial depletion and complete elimination of *n*-alkanes correspond to PM1, PM2 and PM3, respectively. The initial alteration, substantial depletion and complete elimination of acyclic isoprenoids correspond respectively to PM2, PM3 and PM4. The initial alteration, substantial depletion and complete elimination of bicyclic terpanes correspond to PM3, PM4 and PM5, respectively [3, 22, 23] (Fig. 4). The analytical results show that the biodegraded oil shale samples contain large humps of UCMs and are depleted of *n*-alkanes and acyclic isoprenoids. It is worth noting that 25-norhopanes are not observed in the Lucaogou Formation oil shale samples. Furthermore, C_{14}–C_{16} bicyclic terpanes are completely preserved. Thus, it can be concluded that the biodegradation level of the Lucaogou Formation oil shale is low, being PM1–PM2. Additionally, the initial alteration, substantial depletion and complete elimination of trimethylnaphthalene derivatives correspond to PM3, PM5 and PM6, respectively [24–26]. The TMN derivatives are completely preserved in the Lucaogou oil shale, indicating that its biodegradation level is below PM3
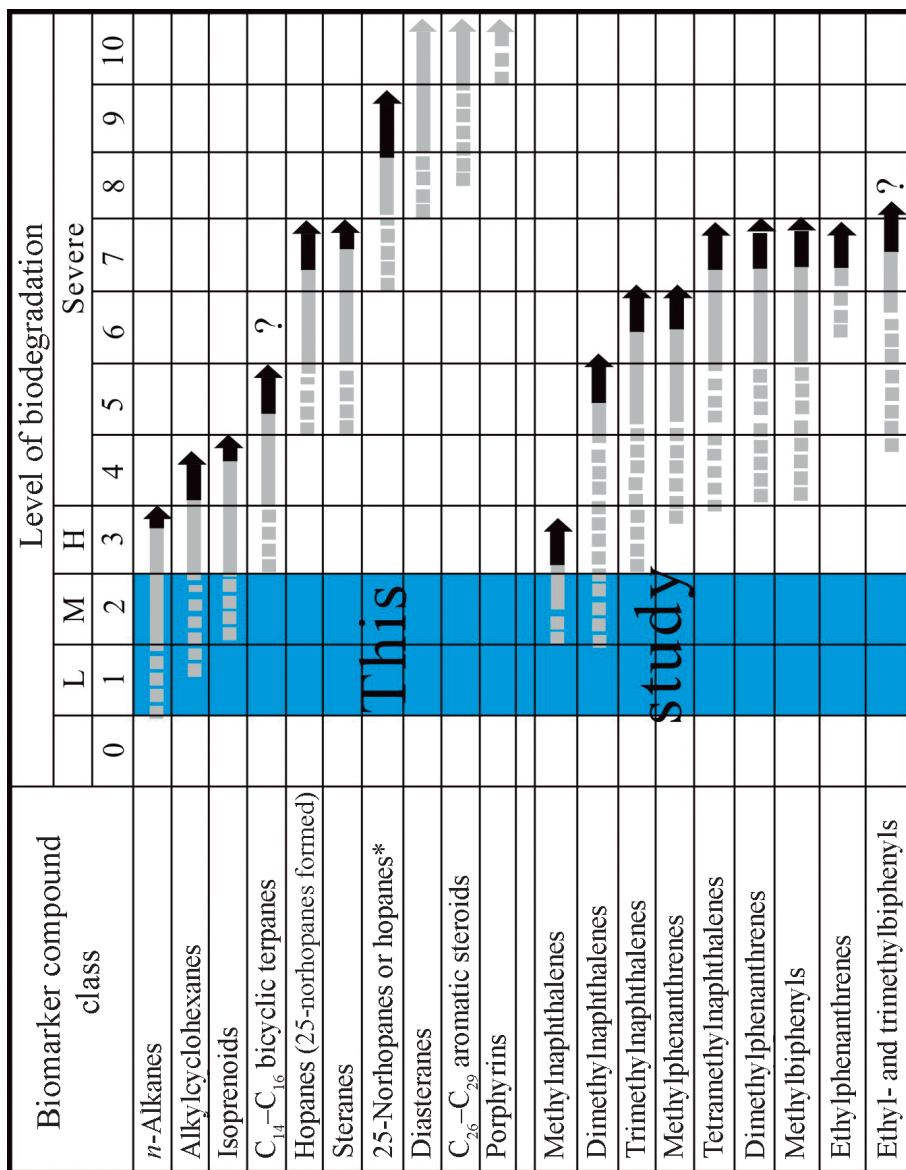
Fig. 4. The biodegradation level of the Lucaogou Formation oil shale with respect to the biomarker biodegradation scale progressing from the initial alteration (dashed lines) to substantial depletion (solid gray lines) and complete elimination (black lines). (L – light; M – moderate; H – high.) * Hopanes degraded without the formation of 25-norhopanes. (The biodegradation range of each parameter is from [3].)

(Fig. 4). The values of MDBT/DBT, DMDBT/MDBT, 1,2,7-TMN/1,3,6-TMN and 1,2,4-TMN/1,6,7-TMN also suggest the low biodegradation level of the Lucaogou Formation oil shale. In summary, the analysis of UCMs and saturated and aromatic hydrocarbons indicates that the biodegradation level of the Lucaogou Formation oil shale corresponds to PM1–PM2.

6. Conclusions

Based on the analytical results for unresolved complex mixtures, saturated hydrocarbons and aromatic hydrocarbons, the biodegradation characteristics of the Lucaogou Formation oil shale have been well revealed. The following conclusions can be drawn:

1. The biodegraded oil shale has large humps of UCMs and is depleted of *n*-alkanes and acyclic isoprenoids (Pr, Ph, iC_{18}, iC_{17}, iC_{16}, iC_{15} and iC_{14}). The unbiodegraded oil shale, on the contrary, contains *n*-alkanes and acyclic isoprenoids. In all samples, C_{14}–C_{16} bicyclic terpanes are completely preserved. In addition, the values of MDBT/DBT, DMDBT/MDBT, 1,2,7-TMN/1,3,6-TMN and 1,2,4-TMN/1,6,7-TMN of the Lucaogou Formation oil shale also reflect its low biodegradation level.

2. The discrimination plots of saturated and aromatic hydrocarbons suggest that the Lucaogou Formation oil shale has undergone biodegradation. The analytical results indicate that the biodegradation level of the oil shale is PM1–PM2. The well-preserved trimethylnaphthalene derivatives in oil shale samples also suggest that the biodegradation level of the Lucaogou Formation oil shale is below PM3. These features give evidence of the good preservation conditions for the oldest oil shale in China.

Acknowledgements

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