

Organic geochemical characteristics of Mae Teep coal deposits, Thailand

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ABSTRACT: The organic geochemical data presented for sixteen rock samples including nine coals, five shales and two mudstones from Mae Teep coal mine located in Lampang province. The total organic carbon (TOC) content of the coal samples ranges from 30.12–73.71 wt.% whereas shale and mudstone values range between 4.81 and 19.49 wt.%. The extractable organic matter (EOM) of all studied samples, which is yielded from bitumen extraction, values between 313.3–9764 ppm representing good to excellent hydrocarbon-generation potential. The thermal maturity and organic matter type of the samples were determined from organic geochemical data. The thermal maturity of the samples is consistent with their being of immature stage. Gas chromatograms obtained from the samples are dominated by long chain n-alkanes and high Carbon Preference Index (CPI) values, indicating terrestrial organic matter input. The biomarker parameters obtained from mass spectrometer data on m/z 191 and m/z 217 indicate that the depositional environments are divided into three main environments: (1) coal seams are deposited in aquatic terrestrial environment under relatively oxic condition; (2) oil shale are deposited in aquatic terrestrial environment under relatively anoxic condition and; (3) mudstones are interpreted to be organic-lean sediment supply in terrestrial environment.

KEYWORDS: biomarker, depositional environment, Mae Teep, thermal maturity

INTRODUCTION

Coal is the one of important domestic fuels in Thailand and is used for coal-fired power plants to generate electricity in the country. Mae Teep basin is located some 20 km south of Ngao district and about 80 km northeast of Lampang province and covers area of about 30 km² [1] (3 km wide and 10 km long). It is one of Cenozoic intermontane basins containing coal deposits with NE-SW trending structure and is currently operated as coal mine (Fig. 1). The coal reserve at Mae Teep coal is estimated at about 10.115 million tons [2]. The coal-bearing sequence, which reaches a thickness of more than 100 m, overlies a tuffaceous sandstone [3]. Cenozoic sedimentary basins have been found to be widely distributed in several parts of Thailand, especially in northwestern Thailand. These basins occur in isolated fault-bound graben and half-graben intermontane basins trending in north-south [1]. It was formed in result of extensional regime influenced by the collision of the Indian Plate and Eurasian Plate which initiated in the Oligocene [4].

The Tertiary sediments are often covered by Quaternary sediments and vary from unconsolidated to consolidated sediments including sand, clay, conglomerate, sandstone, claystone, mudstone, diatomite, coal and oil shale [1,5]. In Thailand, investigation of coal has been studied on physical characteristics such as depositional system from stratigraphy, petrography and proximate and ultimate analysis for coal rank and property [6–8]. There are a few organic geochemistry studies on Cenozoic coals in Thailand [6,9]. Thus, the organic geochemistry investigation of coal in Thailand is very limited and needed in order to get a better understanding of coal deposits in Thailand. The aim of this study is to evaluate coal rank and depositional environment within Mae Teep coal deposits based on organic geochemical data.

MATERIALS AND METHODS

Geologically, Mae Teep basin can be described and divided into two sequences including (1) Tertiary coal-bearing sequence consisting of limestone, mudstone, coal and carbonaceous mudstone with minor

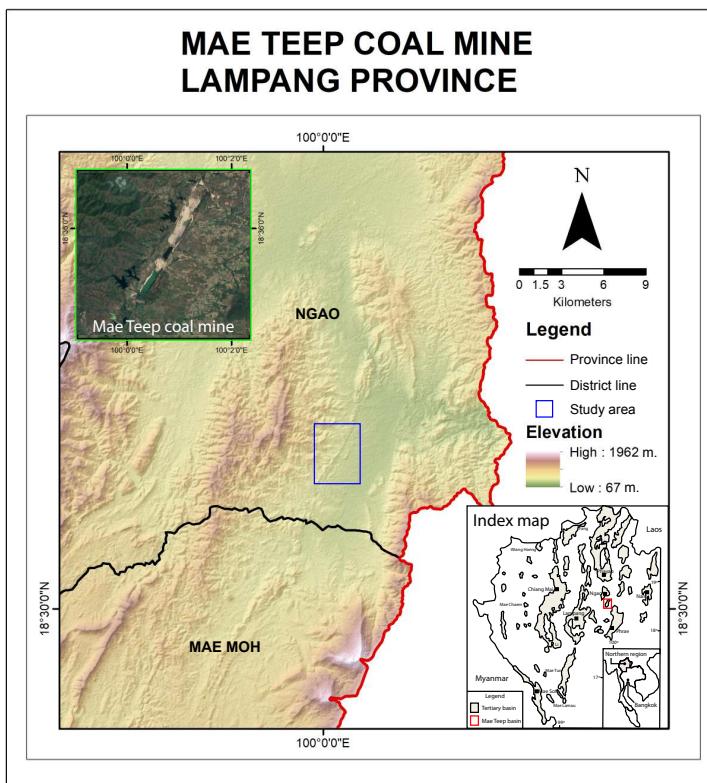


Fig. 1 The map of Mae Teep coal mine in Lampang province, northern Thailand. Blue and green squares display digital elevation model (DEM) and topographic map of Mae Teep coal mine, respectively. Index map shows the distribution of Tertiary coal-bearing basins in northern Thailand in which red square represents Mae Teep basin.

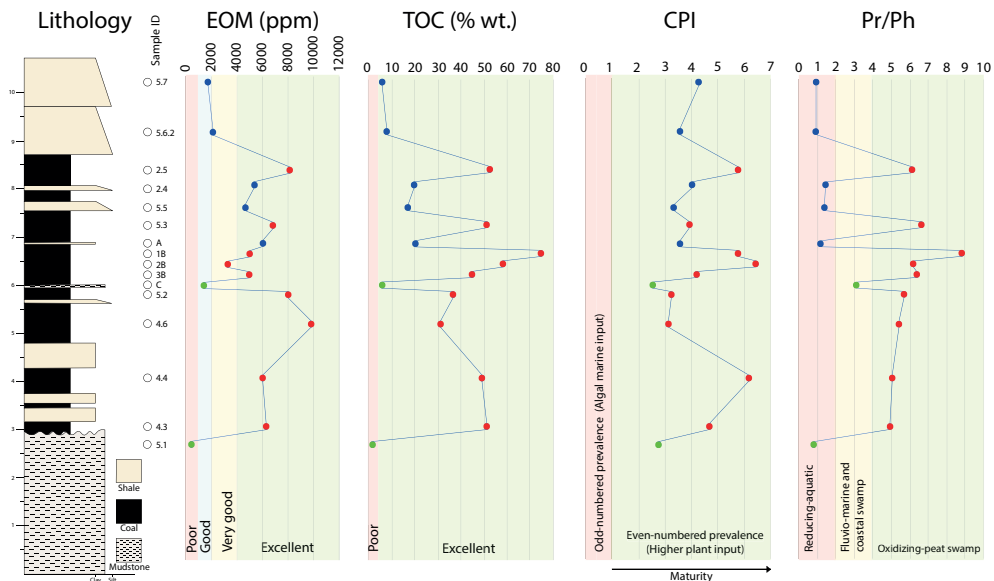


Fig. 2 The lithostratigraphic column measured from the mine wall with profiles of the extractable organic matter (EOM) content, the total organic carbon (TOC) content, carbon preference index (CPI) = $[2(C_{23} + C_{25} + C_{27} + C_{29})] / [C_{22} + [2(C_{24} + C_{26} + C_{28})] + C_{30}]$ and the Pr/Ph ratio. The white circles denote the sample locations (Y-axis). The blue, red, and green dots represent shale, coal, and mudstone, respectively.

oil shale overlying and (2) Quaternary alluvium sequence containing gravel, pebbly sand, muddy sand and sandy clay. This study focuses on Tertiary coal-bearing sequences. The main bituminous to lignite bed, about 5 m thick, is overlain by oil shales and sandstones [3]. The lithostratigraphic column is measured from the mine surface, and hand-picked rock samples are collected to analyze organic geochemical characteristics as presented in Fig. 2.

Sixteen rock samples are collected and classified into three groups including coal, shale and mudstone. In addition, the samples in this study are not taken directly from the mine surface (outcrop wall). Instead, they are collected by digging about 0.10 and 0.30 m into the outcrop to ensure that visibly unweathered material is obtained. The remaining weathered parts of the samples are removed before the next steps. The samples are crushed into pieces by a hammer and a jaw-crusher, respectively. Then, the samples are ground into powders by a cleaned disc mill by grinding with baked quartz sands. The containing glassware and the ball mill are also washed with distilled water and then rinsed with acetone followed by dichloromethane (DCM) in order to prevent contaminations.

Extractable organic matter

Organic matters within sedimentary rocks can be deformed under certain condition. Bitumen is one product of deposition and maturation of organic matter during organic sediment deposits by diagenesis, catagenesis and metagenesis processes [10] and can be extracted by organic solvent with solid-liquid extraction technique [11, 12]. Thus, extractable organic matter (EOM) is a synonym word for bitumen proposed by geochemists in order to avoid confusion. The bitumen is extracted from 50 g of powdered samples by a mixture of dichloromethane (DCM) and methanol (CH_3OH) (93:7) using a Soxhlet apparatus for 48 h. After the extraction process, the massive solvent is removed from the extract and collected for isolation by rotary evaporator. A concentrate of the solution was fractionated into saturated hydrocarbons, aromatic hydrocarbons, and resin by column chromatography on silica gel recording in part per million (ppm). A chromatographic column (50×2.5 cm) is cleaned by using acetone and dichloromethane, respectively, and followed by packing with silica gel of 40–370 mesh activated for 24 h at 120°C and capped with a few centimeters of alumina. Only saturated hydrocarbon fraction is used in this study. The fractions

of all samples are dissolved in hexanes and analyzed using gas chromatography-mass spectrometry (GC-MS). The GC-MS analysis is performed on an Agilent 7000C GC-MS Triple Quad with a gas chromatograph attached directly to the ion source (70 eV ionization voltage, 100 mA filament emission current, 280°C inlet temperature). Chromatograms were acquired in scanning: 35–700 molecular weight and selected-ion-monitored (SIM) for compound identification and integration. The distribution of organic compounds in the bitumen extracts is monitored by fragmentograms of n-alkane (m/z 85), terpanes and hopanes (m/z 191) and steranes (m/z 217).

Total organic carbon measurement

The pulverized samples are analyzed for total organic carbon (TOC) contents by SHIMADZU TOC analyzer with solid sample module (SSM-5000A) [11]. Detail of the TOC measurement is given in Sleutel et al [13]. The TOC content was measured on sixteen samples of the Mae Teep coal mine and reported in percentage per rock weight (wt.%). Rocks that have TOC values exceeding 0.5 wt.% from shale are considered a hydrocarbon potential source [10, 14]. In this study, hydrocarbon potential source rock assessment is used to confirm depositional environments and coal ranks of the samples.

RESULTS AND DISCUSSION

Quantity of organic matter

The quantitative analysis of the organic matter from the samples is reported as TOC and EOM contents as shown in Table 1. In addition, the quality of the organic matter is used to specify types of organic matters and environments within the study area.

The average TOC value of the samples from the study area is 32.12 wt.% ranging from 4.81–73.71 wt.%. The samples can be classified as excellent hydrocarbon potential at present day by TOC content based on Tissot and Welte [15]. The coal samples exhibit TOC values between 30.12–73.71 wt.% whereas the lowest TOC value (0.87 wt.%) was recorded in mudstone.

The EOM content of the samples from the study area gives values ranging from 313.3–9764 ppm with average 4881 ppm which can be evaluated as good to excellent generative potential for petroleum source rock based on Tissot and Welte [15]. The highest EOM content appears on the coal samples while mudstone has the lowest EOM value.

Table 1 The geochemical results and biomarker ratios of saturated hydrocarbon of the samples from Mae Teep coal mine.

Lithology	Sample ID	EOM (ppm) ^a	TOC (% wt.) ^b	n-Alkane and isoprenoid				Terpane and triterpane			Regular sterane		
				CPI ^c	Pr/Ph ^d	Pr/C ₁₇	Ph/C ₁₈	Ts/(Ts+Tm) ^e	C ₃₁	22S/(22S+22R)	%C ₂₇	%C ₂₈	%C ₂₉
Coal	1B	4886	73.71	5.72	8.71	3.78	0.50	0.07		0.09	8	27	65
Coal	2B	3170	57.66	6.39	6.09	3.79	0.80	0.12		0.58	10	32	58
Coal	3B	4819	44.00	4.13	6.29	4.31	0.77	0.05		0.28	14	18	68
Coal	2.5	7979	51.86	5.68	6.00	5.08	0.76	0.12		0.30	8	18	74
Coal	4.3	6132	50.04	4.64	4.83	4.31	1.04	0.20		0.65	17	30	53
Coal	4.4	5,829	48.37	6.12	4.91	7.92	1.09	0.16		0.75	18	37	45
Coal	4.6	9764	30.12	3.09	5.30	1.64	0.28	0.10		0.29	15	24	61
Coal	5.2	7852	35.78	3.18	5.56	3.13	0.53	0.11		0.41	15	44	41
Coal	5.3	6725	50.47	3.85	6.54	5.17	0.63	0.16		0.21	10	33	57
Shale	A	5956	19.49	3.52	1.07	3.16	4.28	0.09		0.41	21	12	67
Shale	2.4	5257	18.80	3.95	1.34	7.74	6.37	0.09		0.41	10	25	65
Shale	5.5	4568	16.11	3.27	1.29	2.57	3.32	0.10		0.43	9	25	66
Shale	5.6.2	1998	6.94	3.48	0.81	2.62	3.31	0.12		0.44	20	19	61
Shale	5.7	1569	4.82	4.20	0.82	2.79	3.71	0.08		0.31	20	23	57
Mudstone	C	1277	4.96	2.47	2.98	0.44	0.16	0.07		0.25	8	35	57
Mudstone	5.1	313.3	0.88	2.67	0.69	0.76	0.06	0.20		0.35	13	18	69
Average		4881	32.12	4.15	3.95	3.70	1.73	0.12		0.39	14	26	60

^a Extractable organic matter; ^b total organic carbon content; ^c carbon preference index; ^d pristane/phytane;

^e Ts: C₂₇18 α (H),22,29,30-trisnorhopane and Tm: C₂₇17 α (H),22,29,30-trisnorhopane.

Molecular geochemistry

The molecular geochemistry analysis of organic matters on non-biomarker and biomarker distributions can be used to define depositional environment conditions. The saturated hydrocarbon fraction (aliphatic hydrocarbon) extracted from the samples is analyzed by using GC-MS in order to determine non-biomarkers and biomarkers used for interpreting thermal maturity, depositional environments and types of organic matters.

n-Alkanes and isoprenoids

The examples of gas chromatograms of saturated hydrocarbon fractions from the rock samples in the study area are shown Fig. 3. The saturated gas chromatograms of the samples display a full suite of saturated hydrocarbons between nC₇–nC₃₅ n-alkanes, isoprenoids, pristane (Pr) and phytane (Ph). The n-alkane distribution can be used to indicate type of organic matter which can be divided into three types: short chain n-alkanes (lower than nC₂₀) are represented as microbial input. In addition, middle chain n-alkanes (nC₁₁–nC₂₀) indicate origin of algae and/or bacteria in organic matter. Long chain n-alkanes (more than nC₂₀) are considered input of terrestrial waxes, especially in the nC₂₅–nC₃₃ range [16]. However, short chain n-alkanes could be originated from the alterations of certain higher plants, algae and/or bacteria [17]. A similar n-alkane distribution, i.e. a unimodal distribution pattern, for all samples is observed (Fig. 3). The

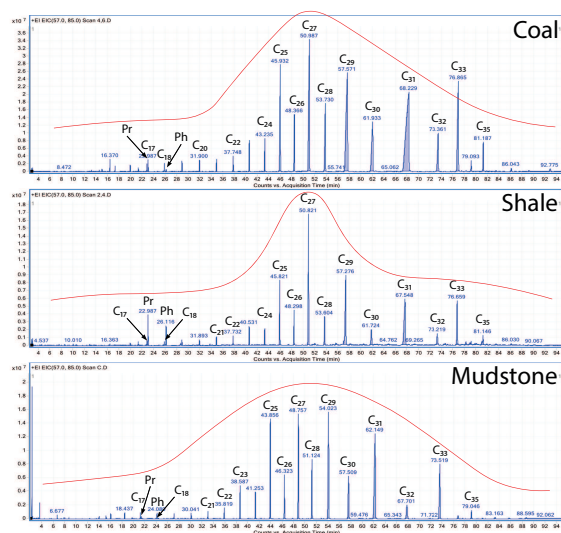


Fig. 3 The examples of m/z 85 chromatogram show the distribution of n-alkanes and isoprenoids. Three represent samples including coal, shale and mudstone show unimodal pattern of long-chain n-alkanes.

maximum peaks for all samples are between nC₂₄ and nC₃₃. The distribution patterns of n-alkanes of the studied samples suggest that organic matters are mainly derived from terrestrial waxes due to the presence of long chain n-alkanes (Fig. 3).

Pristane (C₁₉H₄₀) and phytane (C₂₀H₄₂) are regular isoprenoid hydrocarbons, both of which were derived from the phytol side chain of chlorophyll

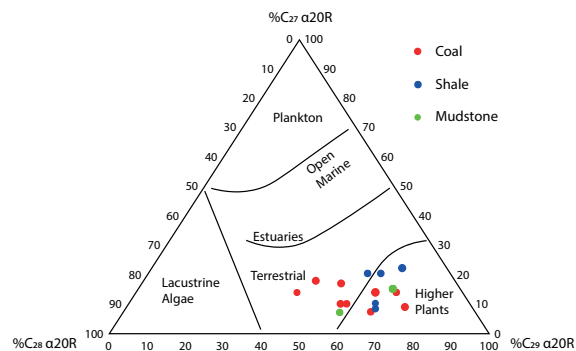


Fig. 4 Ternary diagram of regular steranes ($C_{27}:C_{28}:C_{29}$) shows the relationship between sterane compositions, organic matter input, and depositional environment for the studied samples.

molecule [18]. The phytol can be transformed into pristane or phytane depending on depositional environments. The Pr/Ph ratio of bitumens can be used to indicate depositional environment conditions and source of organic matter [19–21]. Pr/Ph ratio more than 3.0 indicates oxic conditions usually associated with terrigenous organic matter input under relatively oxic condition while Pr/Ph ratio lower than 1.0 represents anoxic conditions commonly associated with hypersaline or carbonate environments [22, 23]. The Pr/Ph ratio between 1.0 and 3.0 suggests intermediate conditions (suboxic conditions) [24]. In addition, Pr/Ph ratios can be divided into three groups including (1) ratio lower than 2.0 indicating reducing-aquatic deposition (including marine, fresh water, and brackish water), (2) intermediate ratio from 2.0–4.0 indicating fluvio-marine and coastal swamp deposition and (3) ratio more than 4.0 indicating peat swamp in an oxidizing environment.

The coal samples show Pr/Ph ratio between 4.83 and 8.71 indicating an oxic condition associated with peat swamp deposits (Fig. 2 and Table 1) while shale and mudstone samples have Pr/Ph ratio between 0.69 and 1.34 indicating an anoxic to sub-oxic condition associated with aquatic deposition (Fig. 2 and Table 1).

Terpanes and steranes

The distributions of terpanes and steranes are analyzed by using GC-MS measuring the ions m/z 191 and m/z 217 mass chromatograms, respectively. Gas chromatograms of terpanes and steranes are shown in Fig. 4.

The organic matter inputs can be identified by

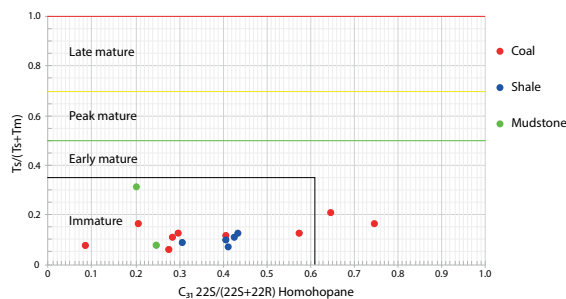


Fig. 5 Cross-plot between the ratios of the $Ts/(Ts+Tm)$ and $C_{31} 22S/(22S+22R)$ homohopane shows that the studied samples fall in immature stage.

using regular sterane distribution. The distribution of regular steranes can be divided into three groups: C_{27} regular sterane represents planktonic marine influenced environment, C_{28} regular sterane indicates yeast, fungi, plankton and algae in freshwater of lacustrine depositional environment and C_{29} regular sterane indicates terrestrial higher plants and brown and green algae [25]. However, C_{29} regular sterane can also be derived from microalgae or cyanobacteria. In addition, some C_{28} and C_{27} regular steranes may derive from freshwater plankton and/or algae [26].

The relative abundance of C_{27} , C_{28} and C_{29} regular steranes from this study is converted to a percentage and plotted in a ternary diagram as shown in Fig. 4 and listed in Table 1. In this study, all samples show similar distributions of the regular steranes and can be interpreted that these samples may derived from the same organic matter source or same depositional environment. The significant amount of C_{29} regular sterane (more than 60%) suggests terrestrial higher plant sources distributed in this area.

Thermal maturity and coal rank

The study of thermal maturity is used to understand the change of organic matters in rocks during lithification or diagenetic process. In this study, coal samples are mainly focused to assess the thermal maturity by using terpane biomarker ratios including homohopane isomerization ratio and $Ts/(Ts+Tm)$ ratio. Furthermore, the change of organic matter of coal can be used to define the difference in coal rank that reflects the degree of coalification depending on time of burial depth, temperature and pressure from burial depth. Other factors such as igneous intrusions, tectonics or heat flow in the basin can

play a part for transformation of peat into coal. Thus, thermal maturity can be evaluated relatively to coal rank by using American Society for Testing and Material (ASTM) standards.

C_{31} homohopane 22S/(22S+22R) isomerization ratio is a maturity indicator and is measured from the m/z 191 mass chromatogram. This ratio determines the configuration of homohopane molecules between biological form (22R) and geological form (22S). Ratio ranges from 0–0.6 indicating equilibrium during maturation. Early stage of oil generation values between 0.50 and 0.54. Then, ratio can be reached from 0.57–0.62 indicating oil window maturity [27]. The Homohopane isomerization ratio can be calculated from any or all of C_{31} – C_{35} homohopanes. In this study, C_{31} homohopane is used as biomarker maturity indicator showing 22S/(22S+22R) ratio ranging from 0.09–0.75 for coal samples (Table 1). The ratios suggest that organic matters of coal samples have been reached at immature stage to early oil window (Fig. 5).

The ratio of Ts ($C_{27}18\alpha(H),22,29,30$ -trisorneohopane) and Tm ($C_{27}17\alpha(H),22,29,30$ -trisorhopane), so called Ts/(Ts+Tm), is used to determine thermal maturity stage. Tm is less stable than Ts during catagenesis. Therefore, the high value of this ratio represents low amounts of Tm with higher thermal maturity of organic matter [27]. Organic matter is mature when the values of Ts/(Ts+Tm) ratio are between 0.35 and 0.95 [28]. In addition, the Ts/(Ts+Tm) ratio may also be influenced by variation in lithology, in which carbonate rocks are significantly lower than clay-rich siliclastic source rocks [16, 29]. The studied samples are mainly from siliclastic rocks derived from terrestrial source. Thus, the variation of Ts/Tm ratios appears to be more strongly influenced by maturity instead of source inputs. Ts/(Ts+Tm) ratios from this study range from 0.05–0.16 as presented in Table 1 indicating that the organic matter is in immature stage (Fig. 5). The result of Ts/(Ts+Tm) ratios in this study coincides with homohopane isomerization ratio.

Then, thermal maturation stage from biomarker analysis is compared to ASTM to define coal ranks of the samples. As a result, all studied coal samples are considered high volatile bituminous coal equivalent to the ASTM coal rank.

Depositional environment

The biomarker distributions measured from GC-MS are used to interpret the organic matter source and

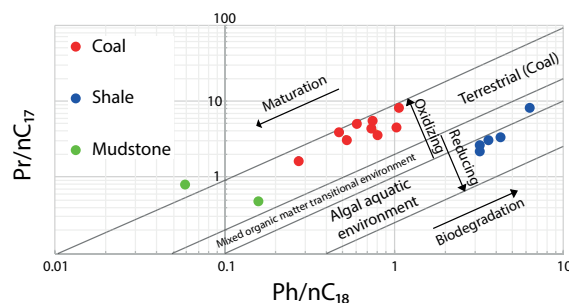


Fig. 6 Log plot between the ratios of phytane to nC_{18} alkane (Ph/nC_{18}) and pristane to nC_{17} alkane (Pr/nC_{17}).

depositional environment in this study. The ion m/z 85, m/z 191 and m/z 217 mass chromatograms are monitored in order to determine the peak area of n-alkanes, terpanes and steranes, respectively, which can be calculated as parameters shown in Table 1 including carbon preference index (CPI), Pr/Ph, Pr/nC_{17} , Ph/nC_{18} and regular steranes.

The samples from Mae Teep coal mine have similar n-alkanes, isoprenoids and steranes distributions. However, the predominance of the long chain n-alkanes and C_{29} regular sterane suggests that most of organic matters were originated from the terrestrial higher plants (Fig. 3).

CPI is the ratio of n-alkane distribution containing the relative between odd and even carbon-numbered n-alkanes. CPI can be used to describe maturity, biodegradation and source of organic matter [10]. CPI values above or below 1.0 indicate immature stage while values close to 1.0 represent mature stage [16]. In reducing environment, n-fatty acids, alcohols and phytol are dominant over decarboxylation resulting in predominance of even carbon-numbered n-alkanes over odd carbon-numbered n-alkanes ($CPI < 1.0$), and phytane is dominant over pristane. Whereas oxidizing environment containing more decarboxylation increases odd carbon-numbered n-alkanes over even carbon-numbered n-alkanes ($CPI > 1.0$) and pristane is dominant over phytane [10].

In this study, the high CPI values and Pr/Ph ratio in coal samples indicate that they are deposited in the peat swamp under oxic conditions. Whereas shale samples are considered to deposit in aquatic-terrestrial environment with minor algal and/or bacterial input due to relatively high middle chain n-alkanes compared to those from mudstones and coals in this basin. In addition, low Pr/Ph ratio from shale samples suggests that they are de-

posited under anoxic condition leading to increase the preservation of freshwater algae and bacteria in sedimentary formation. Mudstone samples are deposited in organic-lean aquatic-terrestrial environment under suboxic condition due to moderate Pr/Ph ratio. The moderate values of CPI in shale and mudstone samples are possibly caused by high weathering after the deposition. All samples exhibit similar organic facies from terrestrial environment confirming by Pr/nC₁₇ and Ph/nC₁₈ ratios (Fig. 6).

Generally, in this basin, sediments deposited within oxic condition are sub-bituminous coal. These sediments are thought to indicate a high-water level with low to intermediate depositional energy while sediments deposited within anoxic condition trend to be oil shale (Fig. 7). In addition, the change of oxic to anoxic conditions is considered by the fluctuation of water level due to seasonal variation such as wet and dry periods [30].

Moreover, this study is consistent with the depositional environment in Mae Moh basin under oxic condition, the adjacent basin operated as a coal mine. Mae Teep and Mae Moh basins are dominated by higher plant inputs due to high molecular weight n-alkanes dominant [30] while oil shale in both basins are believed to be deposited in lacustrine environment with organic matter inputs derived from algae due to the presence of low molecular weight n-alkanes [30]. Similarly, oxic-anoxic fluctuation provides oil shale formation on the bottom of lakes and bogs within Mae Moh basin [30].

CONCLUSION

The study of organic geochemical characteristics from Mae Teep coal mine leads to the conclusion that the samples in the study area exhibit high TOC values as well as bitumen (or EOM) content. All studied samples are at immature to early oil window at present day as indicated by non-biomarker and biomarker parameters. In terms of coal rank, thermal maturity can be related to lignite to sub-bituminous from coal samples of this study. The biomarker study of the study area indicates that organic matters in rock samples come from higher plant materials with minor algae and/or bacteria based on Pr/nC₁₇, Ph/nC₁₈ and CPI. The depositional environments of this study area are interpreted to be peat swamp environment under an oxic condition during coal deposits and aquatic-terrestrial environment with an anoxic to suboxic condition during mudstone and shale deposits.

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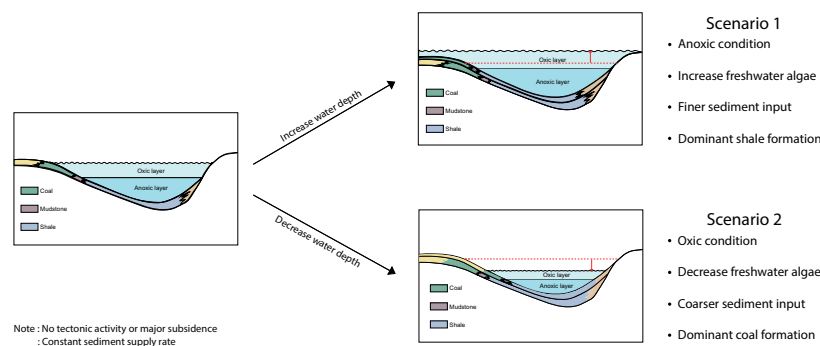


Fig. 7 Schematic diagram of depositional environment change of coal and oil shale in the study area. Scenario 1 is a high-water level with increasing freshwater algae into the basin giving mudstone, shale and oil shale in the study area. Scenario 2 is a low-water level with decreasing higher plant into the basin leading to coal formation.

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