Upper Eocene coal and coaly shale in the Central Myanmar Basin: Origin of organic matter and the effect of weathering

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Extensive coals and coaly shales were deposited at the western margin of the Central Myanmar Basin (CMB) during the Late Eocene. Carbon, nitrogen and sulfur (CNS) elemental analyses, vitrinite reflectance, Rock-Eval pyrolysis, biomarker analyses and $\delta^{13}\text{C}$ measurements were made of eight coal and coaly shale samples from the Pondaung and Yaw Formations. Moderate weathering was recognized in three coaly shales, as manifest by microscopic cracks, holes, and rim structures in vitrinite. A sample pair of fresh and weathered coaly shale collected from the same seam showed that HI, Pr/Ph ratio, and concentration of PAHs and long chain *n*-alkanes ($>n\text{-C}_{20}$) bonded to kerogen in the coaly shales decreased drastically due to weathering, while OI increased. These results suggest that hydrocarbon generation potential of the coaly shale as an oil/gas source rock decreased to about one tenth by the weathering, whereas the free *n*-alkanes, biomarkers such as steranes and triterpanes, and $\delta^{13}\text{C}$ ratios of kerogen were not affected.

Facies change from the coaly shale layers (Phase-I) to the coal layers (Phase-II) was accompanied by variations in origin of the organic matter. The source of the organic matter was mainly terrestrial herbaceous vegetation and/or aquatic plants, deposited in oxic to oxygen-poor peat swamps associated with an estuarine/fluviol-deltaic setting. Phase-I is relatively rich in gymnosperm biomarkers such as retene and 1,7-dimethylphenanthrene (DMP), whereas Phase-II shows an increase in angiosperm proxies such as oleanane content and oleanane/ C_{30} hopane ratio, with $\delta^{13}\text{C}$ values ranging from -24.6‰ to -26.5‰ . Based on HI values of unweathered samples, Phase-I is characterized by type II-III kerogen, while Phase-II contains type III kerogen. Higher values of HI, Pr/Ph ratios, $n\text{-C}_{29}/n\text{-C}_{19}$ alkane ratios (>1.5), and higher concentrations of conifer-derived 1,7-DMP in Phase-I suggest a significant contribution from material of resinous vascular plant origin. High Fluoranthene/(Fluoranthene+Pyrene) ratio in the uppermost coal suggests occurrence of wildfire, probably related to a dry climate experienced in Phase-II. Unweathered CMB samples show good source rock quality, with potential generation of liquid/gas hydrocarbons.

Keywords: Upper Eocene, coal and coaly shale, depositional environment, weathering, paleovegetation, Central Myanmar Basin

INTRODUCTION

Several seams of coal and coaly shale in Late Eocene formations are exposed in the Central Myanmar Basin (CMB) (Bender, 1983). These thin carbonaceous seams are correlated with coal deposits in the Eastern Highlands, the Inner-Myanmar Tertiary Basin and the Arakan Coastal Area, where there is a limited commercial production. However, no detailed geochemical studies have been made of the Late Eocene coals and coaly shales. Furthermore, the organic geochemical characteristics of Myanmar sedimentary/carbonaceous materials have not yet been investigated in any depth. A report by Curiale *et al.* (1994) on the CMB oil family is the only available published work on organic geochemistry in Myanmar that we are aware

of. Based on the composition of hydrocarbons, these authors speculated that the Eocene coals in the CMB were one of the sources for the oils/gas found within the basin. Nevertheless, source rock analyses to determine hydrocarbon generation potential of sediments including coals and/or coaly shales have not yet been made.

In this study, we focus on the characteristics of Late Eocene coals and coaly shale-bearing formations deposited along the western margin of the CMB (Fig. 1). Although the coal seams in the western margin of the CMB are generally thin (5–150 cm), several thicker seams have been found in the central part of the Inner-Myanmar Tertiary Basin (IMTB) (Bender, 1983). The Late Eocene sequence in the CMB is divided into two lithostratigraphic formations; the Pondaung sandstone (Lowermost Late Eocene) and the overlying Yaw shale (Uppermost Late Eocene). We found four coals and four coaly shale beds in the Pondaung and Yaw formations (Fig. 1a) during a detailed geological survey of our study area in the west-

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Fig. 1. (a) Location of the study area in the Central Myanmar Basin (CMB) and Salin sub-basin (CMB and S boundaries from Pivnik *et al.*, 1998), and (b) lithostratigraphic column of the study area in the western margin of the CMB, Myanmar.

ern part of the CMB (Fig. 1b). Vitrinite reflectance, Rock-Eval pyrolysis, elemental analyses, biomarker analyses and $\delta^{13}\text{C}$ measurements were made of these Late Eocene coals and coaly shales. The effects of tropical weathering on the characteristics of coals and coaly shales were also studied. This is the first detailed organic geochemical description of the carbonaceous rocks of Myanmar.

GEOLOGICAL SETTING AND STRATIGRAPHY

Geomorphologically, Myanmar can be sub-divided into four major tectonic provinces: (1) Shan Plateau (Eastern Highlands), (2) Central Cenozoic Belt, (3) Western Fold Belt (i.e., the Indo-Myanmar Ranges and Arakan

Yoma) and (4) the Arakan Coastal Plain, from east to west (Fig. 1a). In early Tertiary time, the Shan Plateau formed a land mass in the east, and the Arakan Yoma comprised a narrow strip of land in the west (Chhibber, 1934). The long and narrow lowland between the two was known as the IMTB or Burmese Gulf (Bender, 1983). The IMTB was underlain by Mesozoic deep-water sediments and older metamorphic basements, and was occupied by the sea until the Late Tertiary period, by which time it had filled with Tertiary clastic sediments (Chhibber, 1934; Bender, 1983).

The CMB, which has been known as the Salin sub-basin (Fig. 1a), formed as a subsiding sub-basin within the IMTB. The total stratigraphic thickness of the CMB

Fig. 2. Outcrop photographs of (a) unweathered coal and (b) weathered coaly shale; photomicrographs of (c) A1-92, illustrating thin micro-fissures in vitrinite (d) A2-54, dark oxidized rims along irregular micro-fissures in vitrinite (e) A2-52A, showing dark oxidized rims along irregular micro-fissures in vitrinite particles.

succession is very thick, up to 19 km (Tun, 1968; Myint and Soe, 1977). The great depth of the CMB is considered to be due to tectonic subsidence (i.e., oblique subduction of the Indian oceanic plate underneath the Myanmar micro-plate) by sediment loading (Hall and Morley, 2004). The CMB is a complex fore-arc/back-arc basin, and is situated between a major right-lateral strike slip fault in the east, and the obliquely converging India plate beneath the Myanmar (Burma) micro-plate to the west (Pivnik *et al.*, 1998).

The Tertiary succession in Myanmar is characterized by fluvio-deltaic sediments in the north, and by transgressive shallow marine shales in the south (Stamp, 1934). The CMB is one of the largest petroliferous onshore sub-basins within the IMTB. Hydrocarbon seeps occur locally along both the eastern and western margins of the CMB. Tertiary coal seams are widely distributed in the CMB and other sub-basins of the IMTB. Coals of Eocene age can be found in Kyaukset village in Minbu district, and in Tazu and Letpanhla villages in Pakokku district within the CMB. Coal seams in this district generally vary from 1.2 to 2.1 m in thickness (Bender, 1983).

The present study area is located in the western margin of the CMB (Fig. 1). The lower part of the upper Eocene Pondaung Formation (Fig. 1b) is composed mainly of thick-bedded to massive sandstones interbedded with thinly-bedded grey shales, siltstones, and coal seams. The thickness of sandstone in this formation reflects a sea level regression (Chhibber, 1934). Seams of brownish to black coals 15–150 cm in thickness occur in the upper part of the Pondaung Formation in the study area. The Pondaung Formation is interpreted to have accumulated in swampy brackish-water conditions and in near-shore fluvial environments, as evidenced by fossils and sedimentary facies (Myint and Soe, 1977). Black coal layers *ca.* 1 m in thickness occur in the lower part of the overlying upper Eocene Yaw Formation. The Yaw Formation (Fig. 1b) consists mainly of nodular, bluish-grey shales and fine-grained sandstones with coal seams.

SAMPLES AND METHODS

Samples

Four coals (samples A1-92, A1-113, A1-114, and A2-

55) and four coaly shales (A2-54, A2-52, A2-52A, and A1-120) were collected from Late Eocene Pondaung and Yaw outcrops along the western margin of the CMB (Fig. 1b). The four coal seams were deposited after sedimentation of underlying coaly shale layers. This sedimentary sequence was first found in the present study. Therefore, the sedimentary facies are divisible into two phases: a lower part consisting of the coaly shales (Phase-I) and an upper part containing the coal layers (Phase-II) (Fig. 2). Each coal and coaly shale seam ranged from 15 to 150 cm in thickness. Based mainly on microscopic observation, we detected signs of weathering in three of the coaly shale samples (A2-54, A2-52A and A1-120).

Elemental analysis (C, N and S)

Each sample was prepared by air drying, cleaned with ethanol to remove any surface contamination, and powdered in an iron pestle and mortar. Powdered samples were placed in silver capsules, pre-treated with 1M HCL to remove carbonate, and then dried at room temperature. Samples dried at 110°C for 45 min. were then placed in tin capsules and sealed. Total carbon, sulfur and nitrogen contents were determined using an EA1108 Elemental Analyzer (FISONS Co. Ltd.). Ash contents of the coals were determined by heating in a muffle furnace at 850°C for 2 hours.

Microscopic observation and vitrinite reflectance

Vitrinite reflectance (R_o) measurements were made on all samples. Coal and coaly shale chips (ca. 2 mm) were embedded in an epoxy resin which was allowed to set for 24 hrs. The mounts were then polished using water-lubricated silicon carbide paper of different grades (#320, #600), and finished to a highly reflecting surface using finer grade alumina powders. Reflectance measurements were made under oil immersion in 546 nm reflected light, using a Lambda Vision-OLYMPUS microscope equipped with a TFCAM7000F-LA100USW spectrograph system.

Rock-Eval pyrolysis

Rock-Eval analysis was carried out to evaluate the potential hydrocarbon generation and maturity of the Late Eocene samples, using a Rock-Eval 6. Powdered whole rock samples (6–100 mg) were heated from 300°C to 650°C in a helium flow. The amounts of hydrocarbons released from organic matter were then used to determine hydrogen index (HI mg HC/g TOC), oxygen index (OI mg CO₂/g TOC) and temperature of hydrocarbon maximum generation (T_{max}). This analysis was performed by JAPEX Research Center.

Biomarker analyses and pyrolysis-GC-MS

Extractions were made of coals and coaly shale samples weighing ca. 1.5–5 g, depending on their total or-

ganic carbon contents. Extracts were made in a Soxhlet extractor, using a 9:1 mixture of dichloromethane (DCM) and methanol (CH₃OH) for 120 hrs for the coals (23.1–57.6 wt% TOC) and 72 hrs for the coaly shales (2.08–8.58 wt% TOC). Activated copper was used to remove elemental sulfur from the solvents during the extraction. After a first evaporation of the solvent, the extracts were transferred from the flask to a beaker by dissolution with a little DCM-methanol solvent, and then dried overnight in a draft hood at room temperature. The dried extracts were then dissolved in *n*-hexane and placed on a thin layer chromatographic (TLC) plate for compound class separation. The aliphatic and aromatic fractions from TLC were analyzed using a GC-MS (QP2010: Shimadzu Co. Ltd.) equipped with a 30 m capillary column (*m/z* 50–850; DB-5MS, 0.25 mm i.d., 0.25 μm film (5% phenyl methylpolysiloxane) thickness: Agilent Technologies), using electron impact ionization (70 eV). The GC oven temperature was held at 50°C for 5 min, programmed from 50 to 300°C at a rate of 8°C min⁻¹, and then held at 300°C for 30 min. Helium was used as the carrier gas. *n*-Tetracosane, cholestane and polycyclic aromatic hydrocarbons (PAHs) Solution Mix (Accu Standard Inc., Z-013-17) were used as external standards for differing compound groups in the hydrocarbon fractions. Individual compounds were identified by comparison of retention times with standard mixtures and mass spectra with previous literature and published work from our laboratory. Identification of PAHs was performed by comparison of GC retention times, mass spectra with published data and standard PAHs. In this study, the PAHs quantified were cadalene (Cad), phenanthrene (Phe), fluoranthene (Fla), pyrene (Py), benzo (a) anthracene (BaAn), chrysene/triphenylene (Chry+Tpn), benzo[fluoranthene] (Bfla), benzo[e]pyrenes (BePy), benzo[a]pyrenes (BaPy), perylene (Pery), indeo[1,2,3-*cd*] pyrene (InPy), benzo (ghi) perylene (BgHiP), coronene (Cor) and retene (Ret). Selected PAHs were monitored at *m/z* 183 (Cad), *m/z* 178 (P, An), *m/z* 202 (Fla, Py), *m/z* 291 (Ret), *m/z* 228 (BaAn, Chry), *m/z* 252 (Bfla, BePy, BaPy, Pery), *m/z* 276 (InPy, BgHiP), and *m/z* 300 (Cor). Relative abundances (%) and concentrations (μg/g TOC) were calculated by comparing the TIC (total ion current) chromatogram area to the standard area.

For Py-GC-MS, samples were pyrolysed at 600°C (30 sec.), after removal of free hydrocarbons and non-hydrocarbons using the 9:1 DCM-methanol solvent, using a double-shot pyrolyser (PY-2020D) connected to a GC-17A gas chromatograph coupled with a mass spectrometer (*m/z* 35–850; Shimadzu GC-MS QP5050A). The GC oven was operated with a 30 m fused silica column (DB-5MS, 0.25 mm i.d., 0.25 μm film (5% phenyl methylpolysiloxane) thickness: Agilent Technologies). Helium was used as the carrier gas. The GC oven tem-

Table 1. (a) TOC and TS contents, C/N and C/S ratios, vitrinite reflectance, Rock-Eval pyrolysis parameters and carbon isotope values for Upper Eocene coals and coaly shales from the Central Myanmar Basin, Myanmar. Coal A1-92 is from the Yaw Formation; all others from Pondaung Formation, in ascending stratigraphic order from left to right. (b) Molecular concentrations and ratios.

Group Sample	Phase-I coaly shales				Phase-II coals			
	*A1-120	*A2-52A	A2-52	*A2-54	A2-55	A1-114	A1-113	A1-92
(a) TOC, TS, C/N, C/S, R_o, Rock-Eval pyrolysis parameters and carbon isotope values								
TOC (wt%)	7.18	2.08	8.58	7.36	31.3	57.6	23.1	26.3
TS (wt%)	0.08	0.13	1.14	0.48	0.19	0.09	0.47	3.95
C/N	19.8	16.0	30.6	26.3	39.9	66.8	39.3	49.7
C/S	95.4	16.1	7.5	15.5	167	612	49.0	6.60
R_o (%)	0.36	0.30	0.39	0.33	0.42	0.26	0.28	0.29
T_{max} (°C)	442	451	425	435	429	413	421	422
S_1 (mg HC/g)	0.07	0.01	0.23	0.03	0.71	0.36	3.86	1.92
S_2 (mg HC/g)	1.87	0.25	16.4	1.46	52.0	57.3	32.6	37.4
HI (mg HC/g TOC)	26	12	191	20	166	100	141	142
OI (mg CO ₂ /g TOC)	144	212	28	137	44	40	107	73
PI	0.04	0.04	0.01	0.02	0.01	0.01	0.11	0.05
$\delta^{13}C$ (‰ vs. PDB)	-24.6	-25.7	-26.0	-24.5	-25.5	-24.8	-26.5	-26.5
(b) Biomarker compositions								
Pr/Ph	0.24	0.69	9.09	0.23	4.63	0.71	3.16	2.37
Pr/ <i>n</i> -C ₁₇	4.75	7.34	5.44	1.87	9.86	4.80	7.81	7.03
CPI	1.76	2.25	2.22	1.29	2.09	1.46	1.16	2.11
<i>n</i> -C ₂₉ / <i>n</i> -C ₁₉	5.41	1.84	1.73	0.07	0.67	0.85	0.35	1.26
<i>n</i> -C ₂₀₋₂₅ /total <i>n</i> -alkanes	0.32	0.37	0.32	0.69	0.47	0.65	0.48	0.55
Sterane C ₂₉ 20S/(20S+20R)	0.03	0.12	0.17	0.06	0.07	0.28	0.37	0.27
Ts/(Ts+Tm)	0.33	0.46	0.56	0.41	0.45	0.54	0.65	0.37
Oleanane/C ₃₀ hopane	0.12	0.20	0.10	0.05	0.06	0.20	0.84	0.17
Sterane C ₂₉ /(C ₂₇ +C ₂₈ +C ₂₉)	0.74	0.68	0.63	0.64	0.58	0.42	0.45	0.27
Sterane C ₂₇ /(C ₂₇ +C ₂₉)	0.09	0.24	0.22	0.08	0.25	0.41	0.44	0.28
Homohopane index	0.01	0.04	0.01	0.01	0.01	0.24	0.04	0.05
Moretane/C ₃₀ hopane	0.87	0.71	0.50	0.68	0.76	0.32	0.37	0.87
Hopane/sterane	66.7	43.7	26.8	186	64.9	1.55	15.7	6.03
Sterane/hopane	0.02	0.02	0.04	0.01	0.02	0.65	0.06	0.17
Hopane (μg/g TOC)	9.61	4.45	36.3	14.2	23.3	0.22	9.80	2.38
Oleanane (μg/g TOC)	0.15	0.15	0.82	0.08	0.20	0.01	1.56	0.06

*Weathered sample.

R_o = vitrinite reflectance.

PI = production index: $S_1/(S_1+S_2)$.

Pr/Ph = pristane/phytane.

CPI = carbon preference index.

$CPI = 1/2[(C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{24}+C_{26}+C_{28}+C_{30}+C_{32}) + (C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{26}+C_{28}+C_{30}+C_{32}+C_{34})]$.

Total *n*-alkanes = $sum(C_{16} + C_{17} + \dots + C_{38} + C_{39})$.

Ts/(Ts+Tm) = 22,29,30-Trisnorhopane/(22,29,30-Trisnorhopane + 22,29,30-Trisnorneohopane).

Homohopane index = hopane C₃₅/hopane (C₃₁-C₃₅).

perature was programmed from 40 to 300°C (held for 12 min) at 15°C min⁻¹. The interface was set at 280°C.

Carbon isotopes of the organic matter (OM)

Pulverized samples and oxidized copper were placed in quartz tubes, taken to vacuum conditions, and then heated at 900°C for 2 hours. The stable carbon isotope ratios of the CO₂ extracted from these samples were determined by Geoscience Laboratory Inc. (Chikyu Kagaku Kenkyusho).

RESULTS

Total organic carbon (TOC), total sulfur (TS), C/N ratios, and C/S ratios

Total organic carbon contents of the upper Eocene coals and coaly shales range from 2.08 wt% to 57.6 wt% (Table 1a; Fig. 3a). Sulfur contents are slightly elevated in Phase-I coaly shale A2-52 and in Phase-II coal A1-92, at 1.14 wt% and 3.95 wt%, respectively (Fig. 3b), but the remainder of the samples are sulfur-poor (<0.47 wt%).

Fig. 3. Total organic carbon (TOC) and total sulfur (TS) contents, total organic carbon/total nitrogen ratio (C/N), T_{max} , vitrinite reflectance (R_o), and differing distributions of hydrogen index (HI), oxygen index (OI), pristane/phytane (Pr/Ph), and total PAHs for the Upper Eocene coals and coaly shales. Circled data points indicate weathered samples. Arrows show slight changes in elemental, T_{max} and vitrinite reflectance data and significant changes in HI, OI, Pr/Ph and total PAHs between weathered and unweathered samples in the same horizon.

All C/N ratios are high, with a range from 16.0 to 66.8 (Fig. 3c), as are C/S ratios (range 6.6–612) (Table 1a).

Microscopic investigation and vitrinite reflectance

During microscopic investigation, irregular microfissures and dull oxidation rims were observed on the edges of oxidized vitrinite particles (Figs. 2d and 2e) in three coaly shales (A2-54, A2-52A, A1-120). Vitrinite reflectance (R_o) values are relatively uniform, ranging from 0.26 to 0.42% (Fig. 3e; Table 1a).

Rock-Eval pyrolysis

Free hydrocarbon contents (S_1), the amount of kerogen-bonded hydrocarbon yield (S_2), and the carbon dioxide of organic origin (S_3) expelled during pyrolysis are listed in Table 1a. All samples exhibited elevated pyrolysis S_2 yields, with a range of 0.25–57.3 mg HC/g. The Phase-II coals are characterized by moderate Hydrogen Index (HI) values of 100–166 mg HC/g TOC, whereas the Phase-I coaly shales have more variable HI of 12–191 mg HC/g TOC (Fig. 3f; Table 1a). Oxygen Index (OI) values are lower in unweathered samples and higher in weathered samples (Fig. 3g; Table 1a). T_{max} (temperature at maximum evolution of S_2 hydrocarbons) values range between 413°C and 451°C (Fig. 3d; Table 1a), and Production Index (PI) values vary from 0.01 to 0.11 (Table 1a).

The modified van Krevelen diagram (HI-OI diagram)

shows a mixture of Type II and Type III kerogens in the OM of fresh samples (Fig. 6).

n-Alkanes, biomarkers and PAHs

Biomarkers are useful as source indicators to distinguish between marine and continental land plants, and for reconstruction of paleo-environments (Peters and Moldowan, 1993; Meyers, 1997). Biomarker data for all samples are listed in Table 1b.

(a) *n*-Alkanes and acyclic isoprenoids (Pr/Ph, Pr/*n*-C₁₇, Ph/*n*-C₁₈) The distributions of *n*-alkanes in the upper Eocene coals and coaly shales from the Yaw and Pondaung Formations were identified in the range *n*-C₁₇–*n*-C₃₃ (Fig. 4a). The *n*-alkanes have been classified as short- (<*n*-C₂₀), mid- (*n*-C₂₁–*n*-C₂₆), and long chain (*n*-C₂₇–*n*-C₃₃) homologues. Mid-chain (*n*-C₂₁–*n*-C₂₅) *n*-alkanes with a maximum intensity at *n*-C₂₂ are dominant in Phase-II sample A1-92, while most other samples show predominance in both mid-chain (*n*-C₂₁–*n*-C₂₅) and long-chain (*n*-C₂₇–*n*-C₃₂) *n*-alkanes (Fig. 4a). Carbon preference index (CPI) values were calculated based on the formula $1/2[(C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{24}+C_{26}+C_{28}+C_{30}+C_{32}) + (C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{26}+C_{28}+C_{30}+C_{32}+C_{34})]$ (Peters and Moldowan, 1993). All CPI values are greater than 1, with a range of 1.16–2.25 (Table 1b). With one exception, the samples show a range of quite low to high concentrations of pristane (Pr) and phytane (Ph), from 0.002–17.9 and 0.01–1.96 µg/g TOC, respectively. One coaly

Fig. 4. Ion chromatograms from (a) gas chromatography and mass spectrometry (GC-MS) and (b) pyrolysis-GC-MS of Upper Eocene coals and coaly shales, CMB.

Fig. 5. Biomarker concentrations and ratios of the Upper Eocene coals and coaly shales in the CMB. Circled data points indicate weathered samples.

shale sample (A2-52) has significantly higher values of Pr (17.9 $\mu\text{g/g TOC}$) and Ph (1.96 $\mu\text{g/g TOC}$). Pr/Ph ratios thus range from 0.23 to 9.09 (Fig. 3h; Table 1b). The Pr/ $n\text{-C}_{17}$ values are high (1.87–9.86) and Ph/ $n\text{-C}_{18}$ values are relatively low (0.48–2.96) in all samples.

(b) *Hopanes (Pentacyclic triterpanes)* Pentacyclic triterpanes (m/z 191) originating from bacteria/land plants are significantly enriched. In five samples Tm (17 α (H)-22,29,30-trisnorhopane) is dominant over Ts (18 α (H)-22,29,30-trisnorhopane), whereas in two Phase-II samples (A1-113, A1-114) and one Phase-I sample (A2-52), Ts predominates over Tm. Oleanane/ C_{30} hopane ratios are higher in the Phase-II coals than in Phase-I coaly shales (Fig. 5b; Table 1b). Oleanane is highly enriched in Phase-II coal A1-113, at 1.56 $\mu\text{g/g TOC}$ (Table 1b). In terms of the homohopanes, C_{30} 17 β ,21 α (H)-hopane is most abundant in most samples. One exception is Phase-II coal A1-92, which has a high C_{29} 17 β ,21 α (H)-hopane content. Contents of C_{31} – C_{33} homologues are high in all samples, whereas values of the C_{35} homohopane index are very low (<0.05), except for a value of 0.24 in one Phase-II coal (Fig. 5h; Table 1b). Hopane/sterane ratios are generally higher in the coaly shales than in the coals (Fig. 5g; Table 1b). Values of C_{31} 22S/(22S+22R), C_{32} 22S/(22S+22R), moretane/ C_{30} hopane, Ts/Tm and Ts/(Ts+Tm) range between 0.23 and 0.56, 0.54 and 0.86, 0.32 and 0.87, 0.50 and 1.86, and 0.33 and 0.65, respectively (Table 1b).

(c) *Steranes* Regular steranes are dominant. The C_{28} sterane concentration is significantly greater (63%) than C_{27} sterane and C_{29} sterane in Phase-II coal A1-92, while

the other coals show predominance of C_{29} steranes (42–74%). Most Phase-I coaly shales are enriched in C_{29} steranes (63–74%). Regular sterane C_{29} -20S/(20S+20R) values range from 0.03 to 0.37 overall, and sterane $C_{29}/(C_{27}+C_{28}+C_{29})$ ratios are uniformly greater (0.63–0.74) in the Phase-I coaly shales than in the Phase-II coals (0.27–0.58) (Fig. 5c; Table 1b).

(d) *Polycyclic aromatic hydrocarbons (PAHs)* Aromatic compounds comprising combustion/diagenesis-derived PAHs (Table 2) and their alkylated hydrocarbons are relatively abundant in unweathered samples. Various unsubstituted PAHs were detected, including 2-6 ring compounds such as Cad, Phe, An, Py, Ret, Fla, BaAn, Chry+Tpn, Bfla, BePy, BaPy, Pery, InPy, and BghiP. Total PAH concentration is greatest in Phase-I coaly shale A2-52 (Fig. 3i).

Concentrations of two to five-ring PAHs are relatively high in unweathered samples. Six-ring PAHs such as InPy and BghiP were present at low levels in Phase-II coal A1-114, but were not detected in other coal or in phase-I coaly shale A2-54 (Table 2). Methyl derivatives such as methylphenanthrenes (3-MP, 2-MP, (9+4)-MP, 1-MP) and dimethylphenanthrenes (DMPs) were detected in the coals and the unweathered coaly shale A2-52. 1,7 DMP is more abundant (0.53 $\mu\text{g/g TOC}$) in A2-52 than in the coals (0.03–0.21 $\mu\text{g/g TOC}$), but was not detected in the other three coaly shales (Table 2). The same pattern is seen for perylene, which is abundant in A2-52 (9.86 $\mu\text{g/g TOC}$), present at lower levels in all coals (0.15–0.66 $\mu\text{g/g TOC}$), but was not detected in the remaining coaly shales (Fig.

Table 2. Concentrations of polycyclic aromatic hydrocarbons ($\mu\text{g/g TOC}$) in the Upper Eocene coals and coaly shales of the Central Myanmar Basin, Myanmar. Abbreviations are: BaAn, benzo[a]anthracene; Chry+Tpn, chrysene/triphenylene; Bflas, benzo[fluoranthene]; BePy, benzo[e]pyrene; BaPy, benzo[a]pyrene; Pery, perylene; MP, methyl phenanthrenes; nd, not detected.

Group Sample	Phase-I coaly shales				Phase-II coals			
	*A1-120	*A2-52A	A2-52	*A2-54	A2-55	A1-114	A1-113	A1-92
Cadalene (Cad) ($\mu\text{g/g TOC}$)	nd	nd	9.12	nd	0.32	nd	0.02	3.10
Anthracene (A) ($\mu\text{g/g TOC}$)	nd	nd	0.24	nd	0.05	nd	nd	1.97
Phenanthrene (P) ($\mu\text{g/g TOC}$)	nd	nd	0.16	nd	0.08	0.42	0.03	0.64
Fluoranthene (Fla) ($\mu\text{g/g TOC}$)	nd	nd	0.20	0.04	0.16	0.81	0.11	1.05
Pyrene (Py) ($\mu\text{g/g TOC}$)	nd	nd	0.93	0.01	0.37	1.24	0.17	0.37
Retene (Ret) ($\mu\text{g/g TOC}$)	0.01	0.05	0.03	0.12	0.16	nd	0.02	nd
BaAn ($\mu\text{g/g TOC}$)	nd	nd	1.76	0.04	0.25	0.40	0.48	0.17
Chry+Tpn ($\mu\text{g/g TOC}$)	nd	nd	0.84	0.01	0.32	0.74	0.17	2.25
Bflas ($\mu\text{g/g TOC}$)	nd	nd	1.81	0.05	0.11	0.85	0.98	0.49
BePy ($\mu\text{g/g TOC}$)	nd	nd	5.50	0.08	0.10	1.16	0.91	0.86
BaPy ($\mu\text{g/g TOC}$)	nd	nd	1.85	0.02	0.04	0.29	nd	0.56
Perylene ($\mu\text{g/g TOC}$)	nd	nd	9.86	nd	0.49	0.26	0.15	0.66
InPy ($\mu\text{g/g TOC}$)	nd	nd	nd	0.06	nd	0.13	nd	nd
BghiP ($\mu\text{g/g TOC}$)	nd	nd	nd	0.10	nd	0.14	nd	nd
Cor ($\mu\text{g/g TOC}$)	nd	nd	nd	nd	nd	nd	nd	nd
1,7 DMP ($\mu\text{g/g TOC}$)	nd	nd	0.53	nd	0.19	0.21	0.03	0.09
3-MP ($\mu\text{g/g TOC}$)	nd	nd	0.32	nd	0.06	0.29	0.02	0.26
2-MP ($\mu\text{g/g TOC}$)	nd	nd	0.41	nd	0.09	0.37	0.07	0.70
9-MP ($\mu\text{g/g TOC}$)	nd	nd	0.46	nd	0.13	0.93	0.03	1.51
1-MP ($\mu\text{g/g TOC}$)	nd	nd	0.75	nd	0.14	0.60	0.05	0.87
Fla/(Fla+Py)	nd	nd	0.18	0.73	0.31	0.40	0.38	0.74
Non-alkylated PAHs ^a ($\mu\text{g/g TOC}$)	nd	nd	13.3	0.42	1.49	6.17	2.83	8.37
Total PAHs ^b ($\mu\text{g/g TOC}$)	0.01	0.05	34.8	0.54	3.07	8.83	3.22	15.6

^aNon-alkylated PAHs = (A+P+Fla+Py+BaAn+Chry+Tpn+Bflas+BePy+BaPy+InPy+BghiP+Cor).

^bTotal PAHs = (Cad+A+P+Fla+Py+Ret+BaAn+Chry+Tpn+Bflas+BePy+BaPy+Pery+InPy+BghiP+Cor+1,7DMP+3MP+2MP+9MP+1MP).

*Weathered sample.

5e; Table 2).

(e) *Sesquiterpenoids, diterpenoids and triterpenoids with non-hopanoid skeletons* Sesquiterpenoid-derived cadalene ($\text{C}_{15}\text{H}_{18}$) is abundant in coaly shale A2-52 and coal A1-92, present in coals A1-113, A2-55, and not detected in the remaining samples (Table 2). Aromatic diterpenoids such as retene (m/z 219) were detected in trace amounts in most samples (Table 2).

(f) *Carbon isotopes of the OM* Phase-I coaly shales have $\delta^{13}\text{C}$ values ranging from -24.5 to -26‰ , and the Phase-II coals vary from -24.8 to -26.5‰ (Fig. 5i; Table 1a).

DISCUSSION

Moderate weathering processes

The compositions of three outcropping coaly shale samples (A2-54, A2-52A, A1-120) have been moderately altered by oxidation and weathering in the humid tropi-

cal climate of central Myanmar, which has a mean annual rainfall of 1000 mm, and an average temperature of more than 27°C . Microscope investigation identified many thin, irregular micro-fissures and dark oxidation rims on the edges of oxidized vitrinite particles (Figs. 2d and 2e). The weathered samples are brownish-black and partly brown in color, and are characteristically more friable than the unweathered coals. This color feature also indicates progressive alteration of coal to soil (Copard *et al.*, 2002). Petsch *et al.* (2000) showed that if carbonaceous shales are permeable, this condition can favour oxidation of the surface of organic matter, which is subsequently degraded during weathering. This feature is supported by the pair of fresh (A2-52) and weathered (A2-52A) coaly shales collected from the same seam in the Pondaung Formation. These show obvious loss of TOC (from 8.6 to 2.1 wt%, respectively) due to weathering. Increased permeability in the coaly shales thus favours

chemical weathering along the micro-fissures.

$T_{\max}$ has been used as a parameter to determine not only maturation, but also the extent of oxidation (Copard *et al.*, 2002). The $T_{\max}$ values of 451°C in the weathered coaly shale (A2-52A) are clearly greater than the 425°C determined for the fresh coaly shale (A2-52) (Table 1a), even though R_o of these samples are similar (0.30 and 0.39%, respectively), showing immaturity. The increased $T_{\max}$ may be caused by a decrease in hydrocarbons weakly bonded to kerogen. The three weathered coaly shales analyzed here are characterized by high $T_{\max}$ values (435–451°C) related to their very high OI values (137–212 mg CO₂/g TOC; Fig. 3g), low Pr/Ph ratios (0.23–0.69; Fig. 3h) and low HI values (12–26 mg HC/g TOC; Table 1a and Fig. 3f). Copard *et al.* (2000) considered that anomalously high $T_{\max}$ (500–575°C) and OI (50–80 mg CO₂/g TOC) values in immature or slightly mature samples showed the effects of oxidation of organic matter at high temperature. The low Pr/Ph ratios may be produced by decreased concentrations due to the effect of water washing, and the loss of light molecular weight hydrocarbons which are partly soluble in water (e.g., Martínez and Escobar, 1995).

The Py-GC-MS compositions of the weathered samples show marked decrease in long chain *n*-alkanes (>*n*-C₂₀) bonded to kerogen (Fig. 4b). This suggests that bonded hydrocarbons may be lost due to change in pH to acidic conditions, because acid hydrolysis could decrease the bonded long fatty acids (e.g., Martínez and Escobar, 1995). The PAHs in the weathered samples (mostly 3- to 5-ring compounds) are either decreased significantly in concentration or are totally lost (Table 2). Pery, Phe, BaPy and BePy are unstable during oxidation (e.g., Kawka and Simoneit, 1990), and all have been reduced or lost in these samples (Table 2). Methyl derivatives of PAHs such as MPs (Table 2) have also been lost from all three weathered samples.

Nevertheless, the fresh and weathered sample pair (A2-52 and A2-52A) shows no notable differences in relative relationship of free *n*-alkane content, biomarkers such as distributions of steranes and triterpanes, or kerogen $\delta^{13}\text{C}$ isotopic ratio (Figs. 4a and 5i), despite the moderate weathering. Therefore, these data could be used as representative markers to identify the origin of organic matter.

Thermal maturity and type of organic matter

The Upper Eocene sediments have R_o values ranging from 0.26 to 0.42% (Table 1a), indicating immature OM. $T_{\max}$ values of 413 to 429°C for the fresh samples also suggests immaturity, in agreement with the immature R_o values (<0.5%). Peters (1986) stated that $T_{\max}$ values of less than 435°C indicated immature OM. The sterane C₂₉ 20S/(20S+20R) ratio is used as a maturity indicator; values here also indicate thermally immature OM (0.03–0.37;

Fig. 6. Modified van Krevelen diagram (HI-OI diagram) showing mixed Type II and Type III kerogens in the Upper Eocene coals and coaly shales, indicating that most are gas-prone. Circled data points indicate weathered samples.

Table 1b). High Carbon Preference Index (CPI) values of more than 1 (1.2–2.3; Table 1b) further indicate immaturity. Moretane/C₃₀ hopane ratios (Table 1b) in the upper Eocene samples vary from 0.32 to 0.87, within the range of 0.15–0.8 that indicates immaturity (Peters and Moldowan, 1993).

The HI-OI diagram shows that the unweathered coaly shale is characterized by a mixture of Type II and Type III kerogens (Fig. 6). Hydrocarbon generation potential for these coals and coaly shale may be reasonably good for gas, based on the value of nearly 200 mg HC/g TOC (HI: Fig. 3f). Generally, OM is categorized as Type II (oil prone) on the basis of HI values with 300–600 mg HC/g TOC, Type III (gas prone) with HI of 200–300 mg HC/g TOC, and Type IV (inertinite) when HI is <50 mg HC/g TOC (Peters, 1986).

Origins of organic matter

Change in sedimentary facies from the coaly shale layers (Phase-I) to the coal layers (Phase-II) seems to be accompanied by change in the origin of the organic matter. The coals (Phase-II) have C/N ratios greater than 30 (39.3–66.8), indicating vascular plant sources (Meyers

and Ishiwatari, 1993; Meyers, 1994). C/N ratios of the coaly shales (Phase-I) range from 16.0 to 30.6, and are clearly lower than those of the vascular plant-derived coals.

In general, short chain *n*-alkanes ($n\text{-C}_{12}\text{--}n\text{-C}_{19}$) are derived from plankton (Meyers, 1997), algae and micro-organisms (Cranwell, 1977; Cranwell *et al.*, 1987), whereas long chain *n*-alkanes ($n\text{-C}_{20}\text{--}n\text{-C}_{35}$) are derived from vascular plant waxes (Tissot and Welte, 1984). Dominance in mid-chain *n*-alkanes ($n\text{-C}_{20}\text{--}n\text{-C}_{26}$) can reflect an origin from bacteria and aquatic plants (Davis, 1968; Ficken *et al.*, 2000; Nott *et al.*, 2000). Submerged and floating aquatic plants commonly show maxima at *n*-alkanes C_{21} , C_{23} , and C_{25} (De Souza *et al.*, 2011). In our present study, the relative abundance of $n\text{-C}_{20}\text{--}n\text{-C}_{33}$ alkanes and maxima at $n\text{-C}_{21}$ in Phase-II coals A1-114 and A2-55 and at $n\text{-C}_{22}$ in A1-92 suggest that the OM mainly originated from vascular plants, along with a secondary contribution from aquatic plants and/or bacteria. In contrast, the Phase-I coaly shales clearly exhibit predominance of odd-over-even carbon numbers in wax-derived long-chain *n*-alkanes ($n\text{-C}_{27}\text{--}n\text{-C}_{33}$). The average CPI value of 1.9 of the Phase-I coaly shales is greater than the 1.7 average of the Phase-II coals, suggesting a slightly more abundant contribution of vascular plant waxes in the former relative to the latter. High Pr/Ph ratios in the unweathered coaly shales (9.1 in A2-52; Table 1b) can indicate not only inputs of vascular plant waxes, but also an oxic depositional environment (e.g., Fabiańska *et al.*, 2013). The sterane $C_{27}/(C_{27}+C_{29})$ ratio is higher in Phase-II coals than in phase II coaly shales, suggesting additional planktonic inputs (Table 1b).

Retene has been identified as a biomarker for many vascular plants such as gymnosperms, especially resinous conifers (Noble *et al.*, 1986; Simoneit *et al.*, 1986; Otto *et al.*, 1997, 2003; Otto and Simoneit, 2001; Piedad-Sánchez *et al.*, 2004), whereas triterpenoids are markers of angiosperms (Moldowan *et al.*, 1994; Nakamura *et al.*, 2010). Presence of retene and related compounds in trace concentration in the Phase-II coals can be attributed to a small contribution of gymnosperms to their sedimentary OM. Retene can be transformed to 1,7-DMP (pimanthrene) (Simoneit *et al.*, 1986; Armstroff *et al.*, 2006), which itself originates mainly from pine resin abietic acid (Wakeham *et al.*, 1980; Radke *et al.*, 1982; Simoneit *et al.*, 1986; Budzinski *et al.*, 1997). The low abundances of 1,7-DMP (pimanthrene) in the Phase-II coals (0.03–0.21 $\mu\text{g/g}$ TOC; Table 2) also suggest limited contribution from resinous vascular plants, especially conifers (e.g., Del Rio *et al.*, 1992). In contrast, the higher 1,7-DMP content in coaly shale A2-52 (0.53 $\mu\text{g/g}$ TOC) suggests a greater contribution of resinous materials in Phase-I. Higher oleanane/ C_{30} hopane in the Phase-II coals (Fig. 5b) indicates an abundant contribution of

angiosperm vegetation (Philp and Gilbert, 1986; Peters and Moldowan, 1993; Moldowan *et al.*, 1994). These values do not show absolute contributions, but can indicate relative proportions. Therefore, these results indicate possibilities in Phase-II (coals) after Phase-I (coaly shales) such as an increase of angiosperm and/or a decrease of gymnosperm in absolute plant-production.

The upper Eocene CMB samples studied here are thus characterized by mixed inputs of angiosperm and gymnosperm vegetation. The Phase-I coaly shales reflect mixed inputs of abundant resinous conifers (i.e., gymnosperms) and lesser angiosperm plants, whereas the paleovegetation supplying the Phase-II coals was mainly dominated by angiosperms, with a minor gymnosperm component. Cadalene ($C_{15}H_{28}$) is one of the precursors of the cyclic sesquiterpenoidal hydrocarbons in vascular plant resins (Simoneit *et al.*, 1986; Otto *et al.*, 1997; Killips and Killips, 2005) and can be a product of diagenetic transformation of sesquiterpenoid biomarkers (Simoneit, 2005; cf., Stefanova *et al.*, 2013). The relatively high concentrations of ses- and tri-terpenoid markers (e.g., cadalene and oleanane) in the Phase-II coals thus suggest that their OM mainly contains inputs of angiosperms, with a small contribution from resinous conifers. The Phase-I coaly shales are characterized by higher portions of diterpenoid compounds relative to triterpenoid compound markers, indicating more significant gymnosperm-dominated paleovegetation relative to angiosperms.

Lücke *et al.* (1999) showed that angiosperms had lighter $\delta^{13}\text{C}$ values of about -26‰ relative to gymnosperms (about -24‰); Bechtel *et al.* (2003) also indicated that angiosperms average $\delta^{13}\text{C}$ of -25.5‰ to -26.6‰ , whereas gymnosperm vegetation averages between -22.7‰ and -25.1‰ . Although $\delta^{13}\text{C}$ of atmospheric CO_2 can change during long geologic time spans, our samples were from a rather limited age range in the late Eocene, and the $\delta^{13}\text{C}$ of atmospheric CO_2 would have been roughly constant during the time interval concerned. The coaly shales have $\delta^{13}\text{C}$ values ranging from -24.5‰ to -26.0‰ (Table 1, avg. -25.2‰), suggesting influence of changes in peat-forming vegetation, with both gymnosperm and angiosperm contributions. On the other hand, the Phase-II coals have $\delta^{13}\text{C}$ values ranging between -24.8‰ and -26.5‰ (Table 1a, avg. -25.8‰), suggesting that the depositional environment was more influenced by angiosperm vegetation, with a minor gymnosperm contribution. The two uppermost coals (-26.5‰) clearly show an abundant contribution of angiosperms, whereas the lowermost coaly shale (A1-120, -24.6‰) reflects greater gymnosperm contribution.

Decreased C_{29} steranes originated from vascular plants (Huang and Meinschein, 1978, 1979) are observed in the Phase-II coals (Fig. 5c), and this also suggests an increase in algae/phytoplankton contribution relative to fungi/ter-

restrial plants. Similarly, fungi-derived perylene decreases from the Phase-I coaly shales to the Phase-II coals (Fig. 5e). In addition, high hopane/sterane ratios (Fig. 5g; Table 1b) can be attributed to inputs of prokaryotic bacteria organisms (Peters *et al.*, 2005; Mrkić *et al.*, 2011). The wide range of hopane concentrations (0.22–36.3 $\mu\text{g/g}$ TOC, Table 1b) in the analyzed samples suggests extensive inputs of bacterial organic matter (e.g., Böcker *et al.*, 2013) or preferential loss of hopanes by degradation. However, average hopane concentrations in the coaly shales (16.1 $\mu\text{g/g}$ TOC) are almost double those of the coals (8.9 $\mu\text{g/g}$ TOC), suggesting that bacterial activity may have decreased during the anoxic conditions (low Pr/Ph ratios in Fig. 3h) of Phase-II coal deposition.

Depositional environments

The ratio of TOC to TS (C/S ratio) in all upper Eocene samples is greater than 6 (Table 1), implying oxic marine or non-marine environments (marine C/S <5; Berner, 1984; Berner and Raiswell, 1984). Three coals (A1-113, A1-114, A2-55) are characterized by low sulphur contents (0.09–0.47 wt%) and very high C/S ratios (49–612), implying that they were deposited in non-marine fresh water environments, such as those associated with fluvial-deltaic settings and coastal plains. The remaining coal (A1-92) has a high sulfur content (3.95 wt%) and moderate C/S ratio (6.6), suggesting deposition in an oxygen-poor environment inundated by seawater fluctuation during peat accumulation (e.g., near-shore and tidal-influenced deltaic plain). The Phase-I coaly shale A2-52 also has a slightly elevated sulfur content (1.14 wt%) and moderate C/S ratio (7.5), and thus its OM may also have been deposited in a peat swamp in a fluvial-deltaic setting influenced by sea level fluctuations. Ts/Tm (22,29,30-Trisnorhopane to 22,29,30-Trisnorneohopane) ratios in these upper Eocene sediments support the above interpretations. Ts/Tm ratios are known to be related to the characteristics of both maturity and contribution of marine organic matter (e.g., Seifert and Moldowan, 1978). Higher Ts/(Ts+Tm) ratios in the upper part of the Phase-II coals (Fig. 5f; Table 1b) suggest an increase in marine OM at that stage. These interpretations are consistent with the proposal of Chhibber (1934) that the Yaw Formation (upper Phase-II) was frequently influenced by marine waters, based on the presence of marine vertebrate fossils. According to Haq *et al.* (1987), cyclic sea level fluctuations of +90 m increasing to +150 m occurred in the period from 36.5 to 39.4 million years ago, consistent with the age of the upper Eocene Pondaung and Yaw Formations.

Low to high Pr/Ph values of the fresh samples (Fig. 3h; Table 1b) in these upper Eocene Formations indicate that the sediments were deposited in oxic to oxygen-poor environments. Pr/Ph ratio is useful as a geochemical in-

dicator of the depositional environment redox condition (Didyk *et al.*, 1978). Low Pr/Ph ratios (<1) are associated with anoxic conditions, values between 1 and 3 indicate sub-oxic conditions, and high Pr/Ph ratios (>3) indicate oxic environments containing inputs of vascular plants (Didyk *et al.*, 1978; Powell, 1988; Peters and Moldowan, 1993). The Pr/Ph ratios in the Phase-II coals (0.71–4.63; Table 1b) suggest a spectrum from oxygen-poor to oxic environments, whereas the single fresh Phase-I coaly shale (9.09 in A2-52) indicates an oxic environment. Low concentrations of pristane and phytane, and relatively high Pr/*n*-C₁₇ ratio (Table 1b) may indicate immature OM (e.g., Dzou *et al.*, 1995; Hughes *et al.*, 1995; Koopmans *et al.*, 1999). For the weathered samples, low concentrations of pristane and phytane and low Pr/Ph ratios are additionally caused by decomposition/evaporation during the weathering. Slightly elevated homohopane oxic-anoxic index values (0.01–0.24) in the Phase-II coals further suggest a range from oxic to oxygen-poor environments (Fig. 5h), because the homohopane ratio has been observed to increase with a decrease in dissolved oxygen (anoxia > 0.2; Waseda and Nishita, 1998). The homohopane index values of the fresh samples show a negative curvilinear negative correlation with Pr/Ph (Fig. 7d). Therefore, we consider the Pr/Ph ratios of fresh coals and coaly shales to be effective as an oxic/anoxic indicator. The C₃₁–C₃₃ homohopanes are enriched in all samples with very low C₃₅ homohopane. The Phase-II coals are slightly richer in C₃₅ homohopane, indicating oxygen-poor fresh water environment (fresh water C/S ratio >5), whereas the Phase-I coaly shale shows oxic condition.

In the unweathered samples the Pr/Ph ratios exhibit negative correlation with *n*-C_{20–25}/total *n*-alkanes (Fig. 7a), and positive correlation with sterane C₂₉/(C₂₇+C₂₈+C₂₉) (Fig. 7b) and HI (Fig. 7c). We suggest that these relationships imply that aquatic plants and vascular plant material with high HI was deposited under oxic conditions. Very high Pr/*n*-C₁₇ values (4.80–9.86; Table 1b) in the unweathered coals and coaly shale imply that they were deposited in inland peat swamps and/or in high-biodegradation conditions, because Pr/*n*-C₁₇ values >1 generally indicate inland peat swamp environments (Amijaya and Littke, 2005) and active biodegradation (e.g., Waseda and Nishita, 1998). All of the studied samples are very likely to have been deposited in inland peat swamps, with associated oxic to oxygen-poor fresh water environments.

Paleoclimate

The Upper Eocene coals and coaly shales contain relatively abundant perylene, which is regarded as a biogenic product thought by some authors to be generated from perylenequinone pigments during early diagenesis

Fig. 7. Cross plots of (a) Pr/Ph vs. $n\text{-C}_{20-25}/\text{total } n\text{-alkanes}$ (b) Pr/Ph vs. sterane $C_{29}/(C_{27}+C_{28}+C_{29})$, and (c) Pr/Ph vs. HI, and (d) Pr/Ph vs. homohopane index, showing hydrocarbon generation, source of OM, and depositional environments for the Upper Eocene coals and coaly shales. Open circle data points indicate weathered samples; shaded zones illustrate the broad correlations in unweathered samples.

Fig. 8. Cross-plot of the non-alkylated hydrocarbons against the total MP (alkylated hydrocarbons) for the unweathered coals and coaly shale.

(Wakeham *et al.*, 1980; Budzinski *et al.*, 1997; Silliman *et al.*, 1998; Jiang *et al.*, 2000). Perylene is also considered to originate from wood-degrading fungi (Grice *et al.*, 2009; Suzuki *et al.*, 2010; Marynowski *et al.*, 2013), and is predominantly found in humid and minimum-oxygen environments (Aizenshtat, 1973). Perylene can therefore be considered to be a useful marker of humid/wet climatic conditions (Grice *et al.*, 2009; Suzuki *et al.*, 2010). In the present study, the Phase-I coaly shale A2-52 contains abundant gymnosperm markers, and is also enriched in perylene (Table 2; Fig. 5e). This perylene could thus have been generated from wood-degrading fungi in a humid climate. Moist conditions also favour the growth of coniferous trees in subtropical rain forests. Perylene concentrations gradually decrease in the Phase-II coals, suggesting that climatic condition gradually changed from humid to relatively dry. Shift of climate from wet (humid) to dry (warm) could also account for the change in origin of the OM from gymnosperm-rich Phase-I (coaly shales) to angiosperm-rich Phase-II (coals).

Values of >0.5 for the ratio Fluoranthene/(Fluoranthene+Pyrene) (Fla/(Fla+Py)) are representative indicators of wildfire (e.g., Yunker *et al.*, 2002). Only the uppermost coal (A1-92) has a Fla/(Fla+Py) ratio greater than 0.5 (0.74, Table 2), suggesting the occurrence of combustion in wildfire in this part of the succession. This is compatible with the estimation of dry climate in Phase-II. The other non-alkylated PAHs such as BaAn, Chry, Tpn, Bfla, BePy and BaPy could be diagenetic products formed from original coals and coaly shales, because their concentrations show positive correlation with MP ($R = 0.67$, Fig. 8). Alkyl-substituted PAHs such as MP are largely dominated (Table 2) by those produced by diagenesis rather than by fire (Liu *et al.*, 2005).

Characteristics as gas/oil source rocks

Coal is known to be a source rock with gas-generating potential (Tissot and Welte, 1984). The oxidizing facies can favour a tendency to be gas prone, rather than oil prone. The OM in the Late Eocene sediments of the present study is characterized as immature, as noted above ($R_o = 0.26\text{--}0.42\%$; Table 1a). We determined the potential of hydrocarbon generation as source rocks by pyrolysis S_2 yields, according to Peters and Cassa (1994). The S_2 values are relatively high (16.4–57.3 mg HC/g rock; Table 1a) in the fresh upper Eocene coals and coaly shale, suggesting good to excellent hydrocarbon generating potential.

According to Hunt (1991), hydrogen-rich coals containing resinous plants can generate liquid hydrocarbons, whereas hydrogen-poor coals can generate gas. Although most samples of the present study are probably related to gas-prone Type III and gas/oil-prone Type II and Type III kerogens, the high HI and low OI values for the coals (A1-92, A1-113 and A2-55; Table 1a) and the fresh coaly shale (A2-52) suggest the potential to generate some volumes of liquid hydrocarbons as well as gas. The weathered samples with low HI (12–26 mg HC/g TOC) and very high OI (137–212 mg CO_2 /g TOC) compared to the unweathered samples with high HI (100–191 mg HC/g TOC) and low OI (28–107 mg CO_2 /g TOC) indicate that hydrocarbon generation potential was decreased to about one tenth of the original value by moderate weathering. The weathered coaly shales, therefore, may have had good potential for hydrocarbon generation before weathering. The late Eocene samples show good source rock quality, with potential generation of liquid/gas hydrocarbons. Khin (1991) reported that source rocks in the CMB must lie deeper than *ca.* 3 km, because samples taken from the basin at about that depth have low maturity, with R_o values of 0.5%. Curiale *et al.* (1994) suggested that Paleogene and Early Oligocene source rocks in the deeply-buried parts of the CMB have probably reached the minimum thermal maturity for hydrocarbon generation. Consequently, upper Eocene sediments which are even more favourable for hydrocarbon generation may thus occur in the deeper parts of the basin, compared to those present at its margins.

CONCLUSIONS

- (1) Four coal seams containing 23–58 wt% TOC (Phase-II) were deposited after four coaly shale seams containing 2–8 wt% TOC (Phase-I). Moderate weathering was recognized in three coaly shales (A2-54, A2-52A, A1-120) containing <8 wt% TOC, based on microscopic identification of weathering rims, fine cracks, and hole structures in vitrinite.
- (2) The weathered coaly shale samples are characterized

by very low Pr/Ph (<0.7), high T_{max} (435–451°C), low HI (12–26 mg HC/g TOC), very high OI (137–212 mg CO_2 /g TOC) and decreased long chain *n*-alkanes (> $n\text{-C}_{20}$) bonded to kerogens. These results suggest that hydrocarbon generation potentials of oil/gas source rocks are decreased to about one tenth of the original value by moderate weathering. However, free *n*-alkanes, biomarkers such as steranes and triterpanes, and $\delta^{13}\text{C}$ ratios of kerogen were not affected.

(3) Fresh samples are characterized by a mixture of Type II and Type III kerogens. Hydrocarbon generation potential of these gas-prone coals and coaly shales in the CMB is reasonably good, based on values of almost 200 mg HC/g TOC (HI).

(4) Facies change from the Phase-I coaly shales to the Phase-II coals was accompanied by changes in the origin of the organic matter. The coaly shales contain abundant gymnosperm biomarkers such as retene and 1,7-DMP. Pr/Ph ratios (0.71–9.09) from unweathered samples show a negative correlation with the homohopane index and $n\text{-C}_{20-25}$ /total *n*-alkanes. Pr/Pr ratios also show positive correlation with HI and sterane $\text{C}_{29}/(\text{C}_{27}+\text{C}_{28}+\text{C}_{29})$.

In the Phase-I succession, unweathered Pondaung coaly shale (A2-52) exhibits high Pr/Ph ratio (9.1), moderate Ts/Tm ratio, predominance of C_{29} steranes, high CPI (2.22), slightly enriched sulfur content (1.14 wt%), low C_{35} homohopane index and $n\text{-C}_{20-25}$ /total *n*-alkanes, indicating that it was mainly derived from terrestrial waxy plant materials with a minor input of bacteria and aquatic plants, and deposited in a peat swamp in a coastal plain or deltaic setting under oxic conditions.

The Phase-II coals show abundant contribution of angiosperm vegetation as oleanane content and oleanane/ C_{30} hopane ratio. The coals have low to relatively high Pr/Ph values (0.71–4.63), low to relatively high Ts/Tm ratios, enrichment of C_{29} steranes in Pondaung coals (A1-113, A1-114, A2-55) and high C_{28} sterane in Yaw coal (A1-92), low to high CPI values (1.2–2.1), low to high sulfur contents (0.09–3.95 wt%), C_{35} homohopane index and $n\text{-C}_{20-25}$ /total *n*-alkanes values averaging 0.54. These results imply that the source of the OM was mainly terrestrial herbaceous vegetation and/or aquatic plants, preserved in oxic to oxygen-poor environments in peat swamps associated with an estuarine/fluviol-deltaic setting. The Yaw coal was probably affected by subsequent sea level rise.

(5) $\delta^{13}\text{C}$ values of all samples range from -24.6‰ to -26.5‰ , suggesting variable contributions of gymnosperm and angiosperm vegetation. Angiosperm deposition is especially well-preserved in the two uppermost coals. Concentrations of conifer-derived 1,7 DMP are higher in samples with high HI (Phase-I coaly shale A2-52, and Phase-II coals A1-114 and A2-55). Wax-rich vascular plants are predominant in the Phase-I coaly shales,

based on $n\text{-C}_{29}/n\text{-C}_{19}$ alkane ratios of >1.5 and $n\text{-C}_{20-25}/\text{total } n\text{-alkanes}$ ratio averaging 0.43. These associations suggest that high hydrocarbon generation potential in the coaly shales can be attributed to resinous vascular plant origin.

(6) Higher perylene abundances in Phase-I coaly shales decreasing into the Phase-II coals suggest a slowly alternating shift of climate from wet (humid/seasonal) to dry (warm) in the western margin of the CMB in the upper Eocene, leading to floral changes.

(7) Vitrinite reflectance (R_o : 0.26–0.42%) and biomarker maturity parameters for all samples yield thermally immature organic matter.

(8) A record of significant wildfire ($\text{Fla}/(\text{Fla}+\text{Py}) = 0.74$) was recognized only in the late stage of Phase-II.

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