



Characterisation of oil and potential source rocks in the northeastern Song Hong Basin, Vietnam: indications of a lacustrine-coal sourced petroleum system

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Abstract

Oil discovered in the B10-STB-1x well in the northeastern Song Hong Basin, Vietnam, has typical lacustrine-coaly geochemical features, indicating the existence of a principally lacustrine sourced petroleum system with minor contributions from a coal source. The presence of lacustrine source rocks in the basin is indicated by distinct high-amplitude seismic reflectors in undrilled half-grabens and outcrops of Oligocene immature mudstones and humic coals at Dong Ho and on Bach Long Vi Island. The mudstones from Dong Ho generally have a TOC content of 8–17 wt.% and HI values > 500 mg HC/g TOC. The organic matter is principally composed of fluorescing amorphous organic matter and liptodetrinite; alginite with *Botryococcus*-morphology is present. The organic matter corresponds to type I kerogen. The Bach Long Vi Island mudstones contain 2–7 wt.% TOC and have HI values from 200–700 mg HC/g TOC. The organic matter is largely similar to that in the Dong Ho mudstones, but in addition may contain some marine organic material. The B10-STB-1x oil shows features common to both the Dong Ho and Bach Long Vi Island mudstones, but was probably principally generated from a source similar to the Dong Ho mudstones with contributions from higher land plant organic matter. A prominent activation energy peak dominates the activation energy distributions of the mudstones, and upon artificial maturation (hydrous pyrolysis) the mudstones show extensive hydrocarbon generation over a narrow temperature range. Hydrocarbon expulsion has been estimated to occur at a vitrinite reflectance of $\sim 0.75\%R_o$. The source of the B10-STB-1x oil may be located in the Thuy Nguyen Graben, and maturity modelling shows that the lacustrine mudstones will be thermally mature in this graben.

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1. Introduction

Palaeogene syn-rift successions of many Asian Cenozoic basins include lacustrine carbonaceous mudstones, which were deposited in lakes in grabens and half-grabens, and coals and coaly mudstones. These Palaeogene lacustrine mudstones, coals and coaly mudstones together with Miocene coals/coaly mudstones, form good to excellent source rocks in Southeast Asia (e.g. Noble et al., 1991; Wang and Sun, 1994; Williams et al., 1995; Todd et al., 1997; Lee and Watkins, 1998;

Wan Hasiah, 1999; Curiale et al., 2000; Peters et al., 2000; Petersen et al., 2001). Although coal and other type III kerogen sourced oil fields are numerous, significant oil reserves in Southeast Asia are generated from lacustrine organic facies (Katz, 1990; Sladen, 1997; Todd et al., 1997). More than 20% of the hydrocarbon production worldwide is probably related to lake-associated deposits (Bohacs et al., 2000).

The Beibuwan and Pearl River Mouth basins on the southern Chinese shelf and the Cuu Long Basin located on the eastern Vietnamese shelf are examples of prospective basins, where the dominant source rock is Palaeogene lacustrine mudstones (e.g. Areshev et al., 1992; Wang and Sun, 1994; Todd et al., 1997; Zhu et al.,

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1999; Lee et al., 2001; Huang et al., 2003) (Fig. 1). Further to the southeast, in the Vietnamese Nam Con Son Basin, paralic carbonaceous mudstones and coastal plain coals are suggested to be the primary source rocks, based on biomarker data from oil and gas-associated liquid samples (Todd et al., 1997) (Fig. 1). Others, however, suggest that lacustrine source rocks may have contributed to the hydrocarbon accumulations (Canh et

al., 1994; Lee et al., 2001). The Vietnamese Cuu Long and Nam Con Son basins have been explored since the 1980's. Oil is produced from fractured basement reservoirs, such as in the White Tiger (Bach Ho) and Dragon (Rong) fields of the Cuu Long Basin, and sandstone reservoirs, as in the Big Bear (Dai Hung) Field of the Nam Con Son Basin (Canh et al., 1994; Dien, 1995; Lee et al., 2001).



Fig. 1. The geological basins offshore Vietnam. The Beibuwan and Pearl River Mouth basins are Chinese.

1.1. The Song Hong Basin

Two Vietnamese basins remain largely unexplored, the undrilled Phu Khanh Basin offshore central Vietnam, and the northeastern Song Hong Basin, in which only a few wells had been drilled (Fig. 1). The Cenozoic basin-fill succession of the greater Song Hong Basin (also called the Bac Bo, Yinggehai or Red River Basin) is considered to be dominated by type III kerogen, and the Song Hong Basin is mainly regarded as being gas-prone (Hao et al., 1995, 1998; Chen et al., 1998; Nielsen et al., 1999). Condensate and gas tests from the 103-TH-1x well and shows of early mature waxy oil in the 102-CQ-1x well in the northern Song Hong Basin, between the Son Chay Fault and the Son Lo Fault Zone, have typical terrestrial geochemical signatures (Fig. 2). This includes high Pr/Ph ratios and the presence of oleanane, and these hydrocarbons were probably generated from Miocene coals and coaly shales (Total, 1990; Geochem Group Limited, 1994; Andersen et al., 1999). In the Hanoi Trough, the onshore extension of the Song Hong Basin, wells have also encountered hydrocarbons, including the Tien Hai gasfield that was discovered as early as 1975. However, the hydrocarbons in the Hanoi

Trough may also have been generated from mature lacustrine mudstones, in addition to Miocene coals (Nielsen et al., 1999). This is indicated for example by the oil show in the GK63 well (Fig. 2), which displays a number of geochemical features suggesting a lacustrine source with some contribution from higher land plant organic matter. These include abundant long-chain *n*-alkanes, Pr/Ph~2, a low concentration of steranes, a low sterane/hopane ratio, a low concentration of tricyclic triterpanes, and low concentrations of oleanane and gammacerane in the oil (GEUS, unpublished data). In addition, several observations may indicate the presence of prolific lacustrine mudstone and coal source rocks in rift-lake successions in grabens and halfgrabens in that part of the Song Hong Basin that lies northeast of the Son Lo Fault zone (Fig. 2). Highly oil-prone but thermally immature lacustrine mudstones crop out on a river bed at Dong Ho on the Vietnamese mainland northeast of the Son Lo Fault zone. On Bach Long Vi Island, southeast of Dong Ho in the Gulf of Tonkin, immature source rocks crop out onshore and below sea-level (Dien et al., 1999) (Fig. 2). The outcrop at Dong Ho, an inverted halfgraben, consists of faintly laminated to structureless, grey to black and dark brown, carbo-

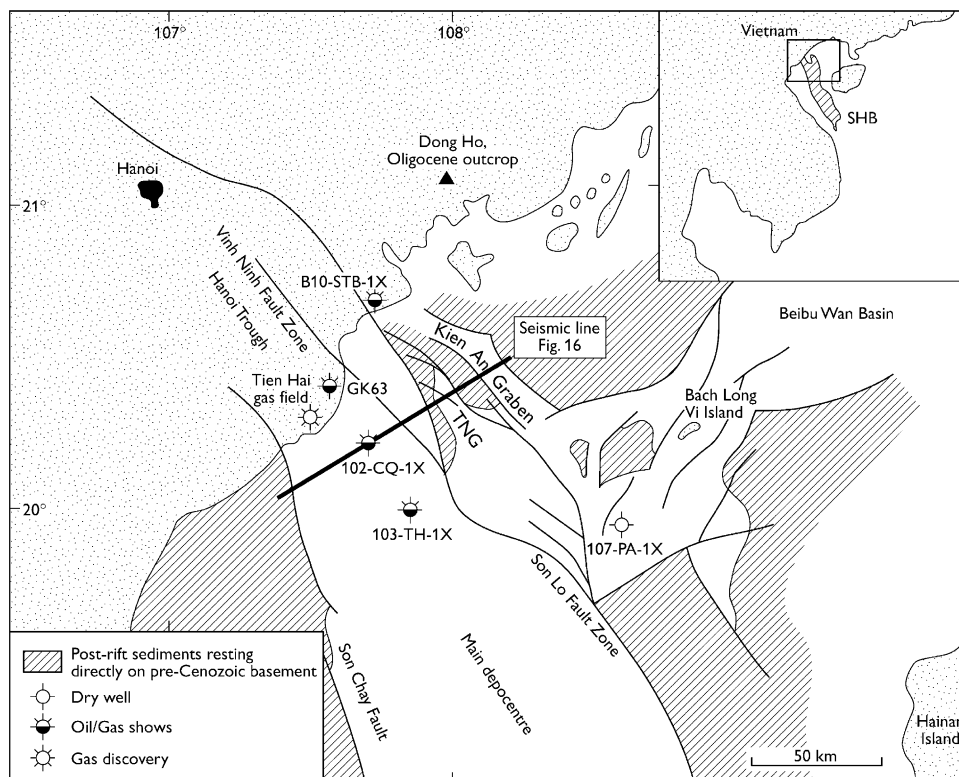


Fig. 2. The structural outline of the northern part of the Song Hong Basin and the Hanoi Trough. The northeastern Song Hong Basin is located north of the Son Lo Fault Zone. The Oligocene outcrops studied in this paper are from Dong Ho and Bach Long Vi Island, and the studied oil from the B10-STB-1x well. TNG: Thuy Nguyen Graben.; SHB: Song Hong Basin.

naceous mudstones interbedded with sandstones and coal seams up to 15 cm thick. The age is considered to be Oligocene, and a preliminary study confirmed a Tertiary age since a high oleanane content was detected in the deposits (Moldowan et al., 1994; Petersen et al., 1998). A recent study on a limited number of coal and mudstone samples showed that the mudstones were deposited in oxygen-deficient, principally fresh water lakes (Petersen et al., 2001). The investigated lacustrine mudstones contain 7–17 wt.% total organic carbon (TOC). The primary organic material is fluorescing amorphous organic matter (AOM), which may reach 82 vol.%, and detrital liptinite which may reach 27 vol.% (Petersen et al., 2001). Alginite with a morphology similar to the fresh to brackish water *Botryococcus* algae is present (up to 9 vol.%) in the fluorescing AOM matrix of the mudstones, and the AOM is considered to be mainly derived from degraded alginite. Subordinate proportions of huminite (up to 11 vol.%) from higher land plants are also present. The organic matter of the mudstones thus corresponds mainly to types I and II kerogen, but the organic composition is variable. This is reflected by the Hydrogen Index (HI) values, which range from 472–690 mg HC/g TOC, and the S₂ yields, which range from 37.96–116.62 mg HC/g rock. Activation energy (E_a) distributions dominated by a large principal E_a-peak and artificial maturation by hydrous pyrolysis of the mudstones show that they, in accordance with the dominant type I kerogen composition, will generate significant proportions of liquid hydrocarbons over a restricted temperature range (Petersen et al., 2001). The investigated coal samples showed that the coals were formed in fresh water mires. The coals are composed of up to 87 vol.% huminite, up to 12 vol.% liptinite and subordinate proportions of inertinite and mineral matter (Petersen et al., 2001). The huminite reflectance values of 0.37–0.41%R_o indicate that the organic matter is immature. HI ranges from 200–242 mg HC/g TOC, and the coals are considered to have a oil- and gas-generative potential (Petersen et al., 2001). The start of the oil window (i.e. start of oil expulsion; “effective oil window”) for the coals has been determined to occur at a maturity corresponding to a vitrinite reflectance of approximately 1.03–1.15%R_o (Petersen, 2002). However, recently analysed coal samples from new outcrops at Dong Ho yield much higher HI values (GEUS, unpublished data), which suggest that for these coals the start of the oil window occurs at lower maturity.

Asphaltic hydrocarbons in interbedded sandstones at Dong Ho are thought to have been generated and migrated from deeply buried shales similar to the immature mudstones exposed at the outcrop (Traynor and Sladen, 1997). Regional occurrence of the lacustrine mudstones in the northeastern part of the Song Hong Basin is suggested by mapping of undrilled Palaeogene

halfgrabens (Rangin et al., 1995; Andersen et al., 1998, 1999), where several seismic sections show syn-rift successions with distinct, continuous high-amplitude reflectors. Such a seismic facies pattern is often interpreted as lacustrine shale-prone units (e.g. Andersen et al., 1998, 1999).

1.2. Aim of this paper

This paper focuses on the northeastern part of the Song Hong Basin. A number of potential source rock samples from Bach Long Vi Island and two samples from the classical river bed outcrop at Dong Ho were collected and studied. In addition, an oil sample collected from the Devonian carbonate basement at a depth of 1220–1230 m in the Anzoil B10-STB-1x well was analysed for this study (Fig. 2). The aims of the study are (1) to investigate the hydrocarbon generative potential of the Oligocene rocks from Bach Long Vi Island and Dong Ho, (2) to examine the generative characteristics and the onset of the oil window (oil expulsion) for these rocks, and (3) to determine the source of the B10-STB-1x oil. The results of this study also attempt to confirm a lacustrine-coal sourced petroleum system in the northeastern Song Hong Basin.

2. Methods and samples

The onshore and submarine outcrops on Bach Long Vi Island (samples designated DCN, Table 1) consist of mudstone, siltstone, fine-grained sandstone and coal (Dien et al., 1999). The submerged sediments and parts of the subaerial sediments are of Oligocene age and are included in the Phu Thuy Chau Formation. Five samples were collected from 4.0 m to 22.6 m below sea-level on Bach Long Vi Island. In addition, a mudstone and a coal sample were collected from the classical Dong Ho riverbed outcrop. Under the microscope none of the samples show signs of weathering (oxidation), such as oxidation rims or reduced fluorescence intensity of the organic matter. Also, rock extracts show no signs of evaporation or biodegradation. The samples (whole rock) were in accordance with international standards (Taylor et al., 1998) crushed to a grain size of 63 µm to 1 mm, embedded in epoxy, and ground and polished to make them suitable for maceral analysis by means of reflected light microscopy. A Zeiss incident light microscope equipped with fluorescence-inducing blue light was used for the petrographic analysis. A total of 500 macerals and minerals were counted in each sample to obtain percentage data (Table 1).

All samples were subjected to Rock-Eval pyrolysis using a Vinci Rock-Eval 6 analyser. A Leco CS-200 was used for total organic carbon (TOC) content and total sulphur (TS) determinations (Table 1). The samples

Table 1
Maceral and kerogen type composition (vol.%), TOC and TS contents (wt.%), T_{\max} ($^{\circ}\text{C}$), S_1 and S_2 yields (mg HC/g rock), and Hydrogen Index values (mg HC/g TOC)

Composition	DCN I-2-97	DCN II-1-97	DCN II-2/1-97	DCN III-3-97	DCN IV-2/1-97	Dong Ho 7	Dong Ho 8
Textinite	0	0	0.2	0	0	0	2.0
Textoulminite	0	0.4	0.6	0	0	0	1.0
Eu-ulminite	0	0	0.4	0	0	0	0.4
Attrinite	1.8	4.6	9.4	1.6	1.8	6.6	0.6
Densinite	0	3.0	2.2	0	0.6	0.6	46.6
Porigelinite	0	0	0	0	0	0	5.6
Corpogelinite	0	0	0.6	0	0	0	3.2
Gelinite	0	0.2	1.4	0	0	0	9.6
Sporinite	0.2	0.4	0	0	0	3.2	3.0
Cutinite	0	0	0	0	0	0.6	7.2
Resinite	0	0	0.2	0	0	0	0.8
Alginite	1.0	1.0	1.2	0.6	0.8	4.0	0
Liptodetrinite ^a	4.8	3.0	4.0	6.4	5.4	16.0	11.2
AOM ^b	6.6	2.0	10.8	45.4	38.4	56.4	0
Inertodetrinite	0	0	0	0	0.2	0	0.8
Funginite	0	0	0	0	0	0	1.0
Pyrite	1.6	1.6	1.0	0.8	0.2	2.4	0
Other minerals	84.0	83.8	68.0	45.2	52.6	10.2	5.8
HUMINITE	1.8	8.2	14.8	1.6	2.4	7.2	69.0
LIPTINITE	12.6	6.4	16.2	52.4	44.4	80.2	23.4
INERTINITE	0	0	0	0	0.2	0	1.8
MINERALS	85.6	85.4	69.0	46.0	52.8	12.6	5.8
Type I kerogen	12.4	6.0	16.0	52.4	44.6	76.4	0
Type II kerogen	0.2	0.4	0.2	0	0	3.8	23.4
Type III kerogen	1.8	8.2	14.8	1.6	2.4	7.2	69.0
Type IV kerogen	0	0	0	0	0.2	0	1.8
TOC	1.91	1.48	4.47	1.90	1.89	10.02	57.12
Total sulphur	0.35	0.13	0.20	0.33	0.08	0.53	0.40
T_{\max}	430	429	427	432	433	427	421
S_1	0.19	0.03	0.24	0.12	0.07	1.93	4.97
S_2	7.77	2.89	12.42	8.76	7.36	60.68	179.09
Hydrogen Index	407	195	278	462	390	606	314

^a Liptodetrinite included in type I kerogen, except for the Dong Ho 8 coal where it is included in type II kerogen.

^b AOM: Fluorescing amorphous organic matter; in general intimately associated with mineral matter, but counted together with liptinite/type I kerogen.

were solvent-extracted with a dichloromethane/methanol mixture (93:7 vol./vol.) using a Soxtec apparatus. Asphaltene were removed from the extracts and from the B10-STB-1x oil by precipitation in *n*-pentane, and the maltenes were fractionated by MPLC (Radke et al., 1980) (Tables 2 and 3). A triterpane concentrate of the B10-STB-1x oil was prepared as described in Nytoft and Bojesen-Koefoed (2001). The concentrate was separated into 34 fractions using reverse phase HPLC. A Vydac 201 TP, 5 μm , 250 \times 4.6 mm column was used with acetone/acetonitrile (80:20 vol./vol.). Saturated hydrocarbons from a Bach Long Vi Island sample (DCN II-1-97) were separated into 59 fractions without prior concentration of triterpanes.

The saturated fractions of the extracts and the B10-STB-1x oil were analysed on a Hewlett Packard 5890

gas chromatograph (GC) fitted with a 25 m HP-1 WCOT column. Gas chromatography/mass spectrometry (GC/MS) of the saturated fractions was carried out using a Hewlett Packard 5890 series II gas chromatograph connected to a Hewlett Packard 5971A quadrupole mass spectrometer. The saturated fractions were analysed in selected ion monitoring (SIM) and full-scan mode, although fractions from HPLC separation were only analysed in full scan mode. A 25 m HP-5 column was used for all samples. In addition, the B10-STB-1x oil and a triterpane concentrate from the oil were analysed using columns of similar polarity (ZB-5 or BPX-5) as well as polar columns (ZB-50 and ZB-wax). Further details on this procedure can be found in Nytoft et al. (2002). GC/MS/MS analysis was carried out using an Agilent 6890N gas chromatograph connected to a

Table 2

Extract yields (mg SOM/g OC), percentages of asphaltenes, saturates, aromatics and polar compounds, Pr/Ph ratios and CPI values of source rock samples

Composition	DCN I-2-97	DCN II-1-97	DCN II-2/1-97	DCN III-3-97	DCN IV-2/1-97	Dong Ho 7	Dong Ho 8
Extract yield	111	102	74	91	78	71	85
Asphaltene	83.3	51.8	48.9	57.7	41.0	38.7	61.9
Saturates	13.3	15.7	14.0	11.8	18.1	12.8	4.4
Aromatics	6.7	7.0	7.3	11.8	3.4	7.3	2.9
Polars	80.0	77.4	78.8	76.5	78.5	79.9	92.6
Pr/Ph ratio	0.60	1.06	1.69	0.89	1.05	2.55	3.97
CPI	1.57	1.04	1.37	1.55	1.29	2.43	4.64

Table 3

Composition of solvent extracts of artificially matured source rocks and the B10-STB-1x oil

Composition	DCN I-2-97	DCN III-3-97	Dong Ho 1		Dong Ho 6		B10-STB-1x oil
	330 °C/72 h	330 °C/72 h	330 °C/72 h	345 °C/72 h	330 °C/72 h	345 °C/72 h	
% Asphaltene	25.0	45.7	29.0	27.8	40.9	20.2	2.1
% Saturates	23.3	30.0	30.1	48.7	28.1	49.7	69.8
% Aromatics	10.0	12.5	12.4	15.1	15.4	14.6	15.2
% Polars	66.7	57.5	57.4	36.1	56.5	35.7	15.0
Pr/Ph ratio	1.30	1.80	2.94	2.74	3.07	2.69	3.36
CPI	1.18	1.13	1.26	1.19	1.25	1.18	1.07

Waters (Micromass) Quattro Micro GC tandem quadrupole mass spectrometer. A 30 m HP-5MS column (30 m × 0.25 mm × 0.10 µm) was used. The temperature program was 30 °C/min from 70 to 100 °C, 4 °C/min from 100 to 308 °C and finally 8 min at 308 °C.

Hydrous pyrolysis was carried out in stainless steel HPLC columns. The columns were filled with 0.7–1.0 g of ground sample mixed with water and thoroughly stirred to remove any air. Two samples from Bach Long Vi Island (DCN I-2-97, DCN III-3-97) were artificially matured for 72 h at 240 °C, 270 °C, 300 °C and 330 °C. In addition, two mudstone samples from Dong Ho (Dong Ho 1 and Dong Ho 6) previously matured up to 330 °C (Petersen et al., 2001) were matured at 345 °C/72 h. These two samples are dominated by fluorescing AOM, liptodetrinite and *Botryococcus*-like alginite (Petersen et al., 2001). Dong Ho 1 has a TOC of 6.48 wt.% and an HI of 586 mg HC/g TOC, whereas Dong Ho 6 has a TOC of 16.89 wt.% and an HI of 690 mg HC/g TOC. The hydrous-pyrolysed samples were afterwards subjected to TOC determination, Rock-Eval pyrolysis, and extraction and separation followed by GC analysis.

3. Results and discussion

3.1. Depositional environment

The Dong Ho 8 sample is a humic coal (TOC = 57.12 wt.%; Table 1) dominated by huminite, though with a

substantial proportion of liptinite, mainly in the form of liptodetrinite, cutinite and sporinite (Table 1). The overall organic matter composition for this sample corresponds to type III kerogen with a subordinate proportion of type II kerogen. The high proportion of huminite in the Dong Ho 8 coal sample combined with the absence of pyrite and the low contents of inertinite (1.8 vol.%; Table 1) and TS (0.40 wt.%; Table 1) suggest that the coal accumulated in waterlogged fresh water mires with restricted oxygen availability and a low frequency of wildfires. Such a depositional environment conforms with previous interpretations of the humic coals that crop out at Dong Ho (Petersen et al., 2001). The Pristane/*n*C₁₇ versus Phytane/*n*C₁₈ plot also indicates a peat-forming depositional environment (Fig. 3).

The Dong Ho 7 mudstone sample has a TOC content of 10.02 wt.% and it contains a high proportion of liptinite, principally fluorescing AOM, followed by liptodetrinite and alginite with *Botryococcus*-morphology (Table 1). Huminite is the second most important organic matter type, while the content of mineral matter is considerably lower than in the samples from Bach Long Vi Island. In terms of kerogen type, the organic matter primarily corresponds to type I kerogen followed by a minor proportion of type III kerogen (Table 1). Previously investigated carbonaceous mudstones from Dong Ho represent deposition in terrestrially influenced, low-energy, oxygen-deficient, principally fresh water lakes, which occasionally may have been subjected to marine influence (Traynor and Sladen, 1997; Petersen et

al., 2001). The results from the Dong Ho 7 mudstone are consistent with such a depositional environment (Tables 1 and 2): the high proportions of fluorescing AOM intimately associated with a very fine-grained mineral matrix and terrestrial liptinite; the presence of alginite with *Botryococcus*-morphology; the presence of huminite; the odd-over-even *n*-alkane predominance (CPI=2.43); a Pr/Ph ratio of 2.55; and the pyrite and TS contents of 2.4 vol.% and 0.53 wt.%, respectively. The association of AOM and alginite with *Botryococcus*-morphology suggests that the AOM was derived from degradation of algal remains. In the Pristane/ nC_{17} versus Phytane/ nC_{18} plot, the carbonaceous mudstones from Dong Ho plot within or close to the field of mixed type II/III kerogen (Fig. 3).

The organic matter composition of the Bach Long Vi Island samples shows great variability, but the organic matter is primarily composed of liptinite in the form of fluorescing AOM, which is intimately associated with the mineral matrix, followed by liptodetrinite (Table 1). Alginite with a morphology similar to the extant *Botryococcus*-type is present. Some huminite is also present. The content of mineral matter is high in all samples and the TOC content varies from 1.48–4.47 wt.%. Compared to the Dong Ho samples, the Bach Long Vi Island samples have considerably lower TOC and higher mineral matter contents (Fig. 4; Table 1). The organic matter principally corresponds to type I kerogen, followed by type III kerogen (Table 1). Thus,

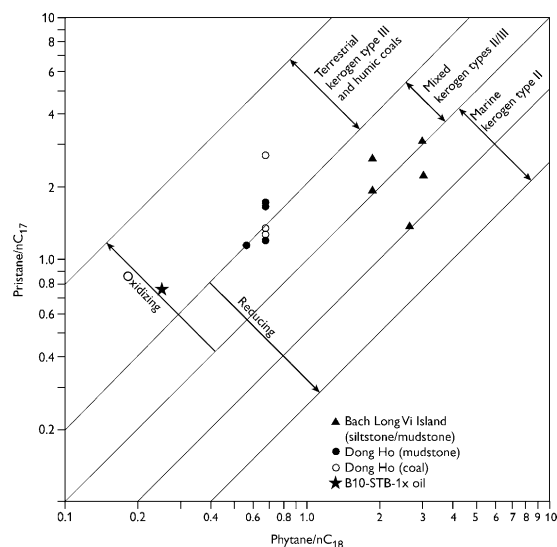


Fig. 3. A Pr/ nC_{17} versus Ph/ nC_{18} plot of the B10-STB-1x oil and the Bach Long Vi Island and Dong Ho samples [Dong Ho data from Petersen et al. (2001) included]. The plot suggests similarity between the Dong Ho samples and the source rock for the B10-STB-1x oil. A weak marine influence on the depositional environment is suspected for the Bach Long Vi Island samples, but this can not be documented.

the samples from Bach Long Vi Island are, in some aspects, similar to the Dong Ho mudstones, but are significantly different in others.

The recognition of alginite with *Botryococcus*-morphology and the low TS contents (0.08–0.35 wt.%; Table 1) suggest a fresh water depositional environment for the Bach Long Vi Island samples. The TS contents correspond to values measured in recent fresh water mires such as the Changuinola peat in Panama (<0.5 wt.% TS) and the Batang Hari River in Sumatra, Indonesia (0.1–0.7 wt.% TS) (Cameron et al., 1989; Phillips and Bustin, 1996). Other parameters, however, may suggest a weak marine influence on the depositional environment. The occurrence of pyrite in all samples may be related to the activity of sulphate-reducing bacteria during saline water introduction (e.g. Cohen et al., 1984; Brown and Cohen, 1995). Also, in contrast to the immature mudstones from Dong Ho, the *n*-alkane distributions of the immature Bach Long Vi Island samples are light-end skewed with low wax and a low odd-over-even *n*-alkane predominance (CPI ranges from 1.04–1.57) (Figs. 5–7; Table 2). This may indicate an overall weaker terrestrial influence on the depositional environment of the Bach Long Vi Island samples than the Dong Ho samples. Extracts from immature terrestrial organic matter normally yield a waxy, heavy-end skewed *n*-alkane distribution. Terrestrially derived oils are also typically waxy. A weaker terrestrial influence on the Bach Long Vi Island samples is therefore also evident by comparing the hydrous-pyrolysed samples (Figs. 5–7). The symmetrical *n*-alkane distribution of the Bach Long Vi Island samples clearly differs from the broad, slightly bimodal and very waxy *n*-alkane distributions of the Dong Ho samples. The Bach Long Vi Island samples are differentiated from the Dong Ho samples based on the Pristane/ nC_{17} versus Phytane/ nC_{18}

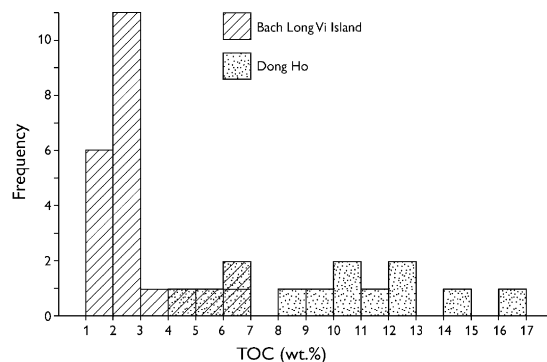


Fig. 4. TOC contents of the samples from Bach Long Vi Island and Dong Ho (excluding coals) illustrate, that the Dong Ho mudstones generally have a TOC content > 8 wt.%, whereas the Bach Long Vi Island samples generally contain between 1–3 wt.% TOC. Data from Dien et al. (1999), Petersen et al. (2001) and unpublished GEUS-data included.

plot, where they plot in the fields of mixed type II/III kerogen and marine type II kerogen (Fig. 3). This could suggest that part of the fluorescing AOM in the Bach Long Vi Island samples may be derived from marine algae (and thus corresponds to marine type II kerogen). In that regard, dinosteranes derived from dinoflagellates (a marine plankton) have been identified by analysis in full scan mode of HPLC fractions from the Bach Long Vi Island DCN II-1-97 sample, which has the most complex sterane distribution of the samples. Dinosteranes constitute only a minor proportion compared to total 4-methylsteranes.

The different organic facies in the lacustrine mudstones are related to varying depositional conditions in the lakes (e.g. Talbot, 1988; Katz, 1990; Bohacs et al., 2000). Carroll and Bohacs (2001) have recently presented a three-fold classification of lacustrine organic facies. According to this classification, the samples from

Dong Ho and Bach Long Vi Island may belong to the 'algal-terrestrial organic facies' and 'algal organic facies', respectively. The former facies is typically characterised by TOC contents of approximately 1–10 wt.%, mixed types I /III kerogen, and common association with fluvial deposits and coals. The latter facies includes laminated mudstones with abundant alginite and homogeneous, fluorescent AOM, both belonging to type I kerogen.

3.2. Source rock potential

The S_2 yields and HI values of the Bach Long Vi samples are in the range 2.89–12.42 mg HC/g rock and 195–462 mg HC/g TOC, respectively, while the Dong Ho 7 mudstone has an S_2 of 60.68 mg HC/g rock and an HI of 606 mg HC/g TOC (Table 1). The Dong Ho 8 coal has an HI value of 314 mg HC/g TOC. The source

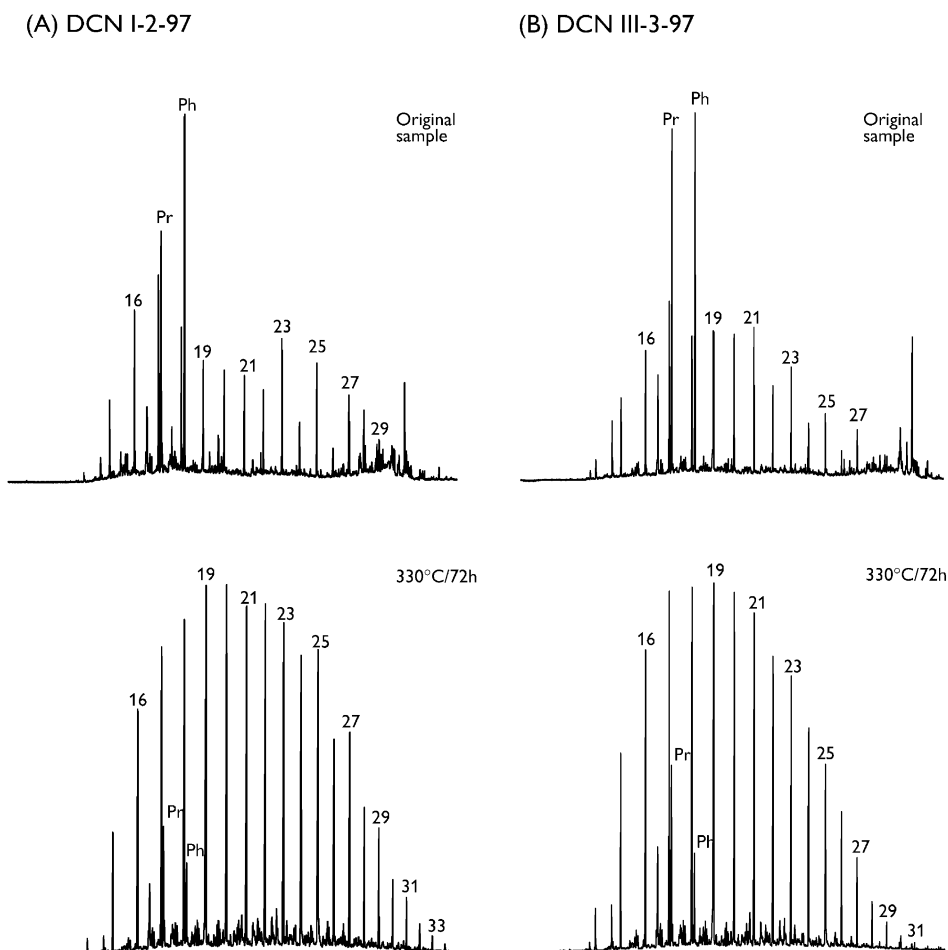


Fig. 5. Gas chromatograms of the (A) DCN I-2-97 and (B) DCN III-3-97 samples from Bach Long Vi Island (Table 1). The original immature samples have light-end skewed, bimodal n -alkane distributions. Note the Pr/Ph ratios are < 1 . The samples hydrolysed at 330 °C/72 h have unimodal n -alkane distributions that extend to at least nC_{33} and are nearly symmetrical centred at nC_{19-20} , with Pr/Ph > 1 .

rock potential of the samples in this study is thus highly variable (Fig. 8; Table 1).

The source potential of the Dong Ho 8 coal is considerably higher than that detected for previously analysed samples from Dong Ho (HI = 200–242 mg HC/g TOC; Petersen et al., 2001). Dien et al. (1999) reported HI values up to 387 mg HC/g TOC for coals from Bach Long Vi Island. The coals are considered to possess an oil and gas generative potential (Petersen et al., 2001).

Previously studied lacustrine mudstones from Dong Ho yield HI values up to 690 mg HC/g TOC, while the studied samples from Bach Long Vi Island yield a maximum HI value of 462 mg HC/g TOC. Dien et al.

(1999), however, reported HI values up to 702 mg HC/g TOC from Bach Long Vi Island (Fig. 8). This indicates that the lacustrine deposits from Dong Ho and Bach Long Vi Island in general are highly oil-prone source rocks.

In summary, the results of the outcrop samples from Dong Ho and Bach Long Vi Island suggest two potential source rock types in the northeastern Song Hong Basin: (1) lacustrine fresh water to possibly marine-influenced source rocks dominated by oil-prone type I kerogen, and (2) humic coals dominated by oil- and gas-prone type III kerogen (Table 4). The principal source rocks are lacustrine. It is, however, also evident that the

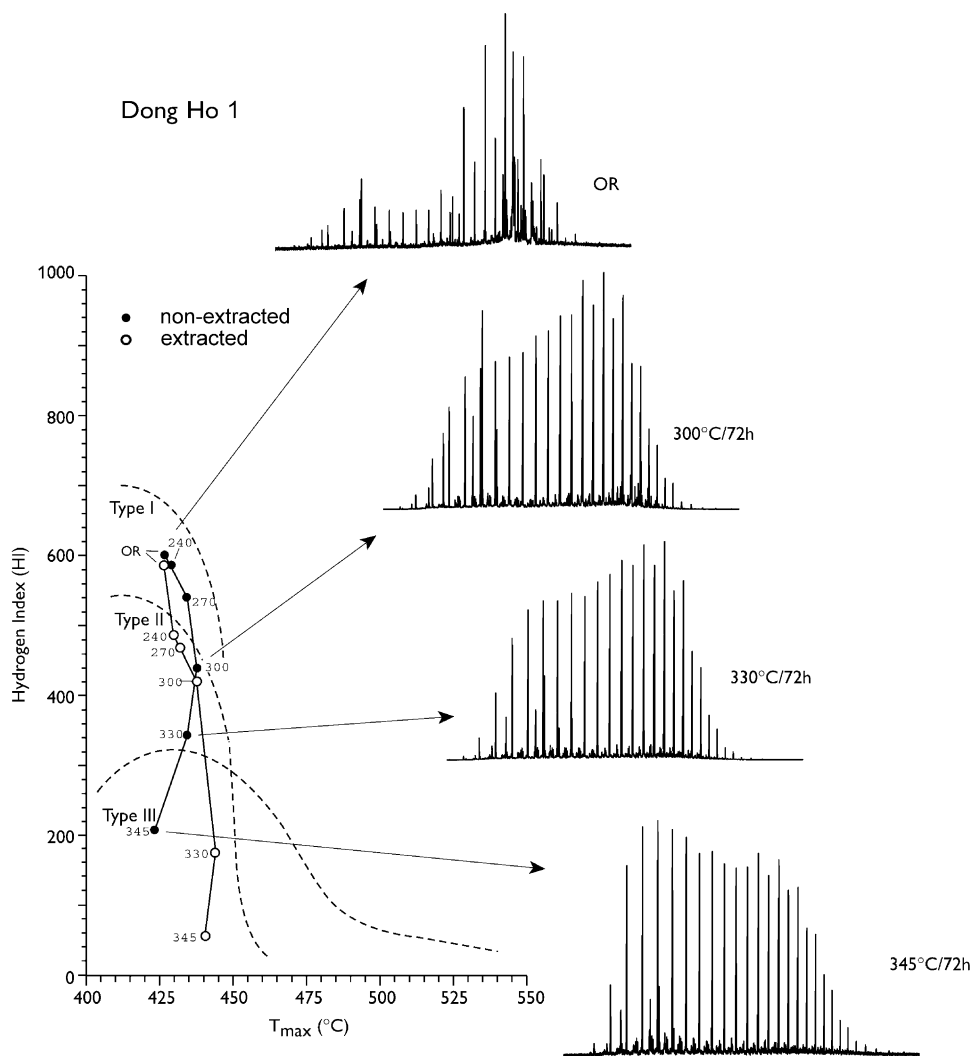


Fig. 6. Stepwise artificial maturation by hydrous pyrolysis of the Dong Ho 1 lacustrine mudstone. The HI decreases significantly, particularly above 300 °C/72 h. The decrease in T_{max} is caused by the generation of labile, extractable heavy hydrocarbons/bitumen during maturation as shown by comparison with the solvent extracted samples. The n -alkane distribution of the extracted saturates from the sample subjected to hydrous pyrolysis at 345 °C/72 h resembles a mature terrestrial-sourced oil. OR: original immature sample; 240, 270, 300, 330, 345: hydrous pyrolysis temperature, °C/72 h.

generative potential of the lacustrine source rocks is highly variable, with HI values down to 195 mg HC/g TOC for the Bach Long Vi Island samples, suggesting a more limited potential for liquid hydrocarbon generation. In general, lacustrine source rocks may show a significant variability in organic facies resulting in varying generation potentials. Such an organic facies variation in lacustrine source rock units is strongly related to primary organic matter productivity versus organic matter preservation, two processes which are linked to the depositional conditions of the different lake types (Katz, 1990, 1995; Katz et al., 1993; Sladen, 1997; Bohacs et al., 2000; Carroll and Bohacs, 2001). These

relationships are, for example, well-known from the varying degrees of preserved organic matter in the lakes of tropical Africa (Talbot, 1988). The Eocene Green River Formation is characterised by a number of organic facies in different stratigraphic units (Ruble et al., 2001). Consequently the HI values reach 900 mg HC/g TOC in intervals dominated by algal organic matter (type I kerogen) and only 100 mg HC/g TOC in more vitrinite-rich (type III kerogen) intervals (Taylor et al., 1998). Similarly the Eocene to Oligocene Pematang Brown Shale source rock in central Sumatra, Indonesia, shows a distinct source quality difference between the lower and upper part, which is related to different

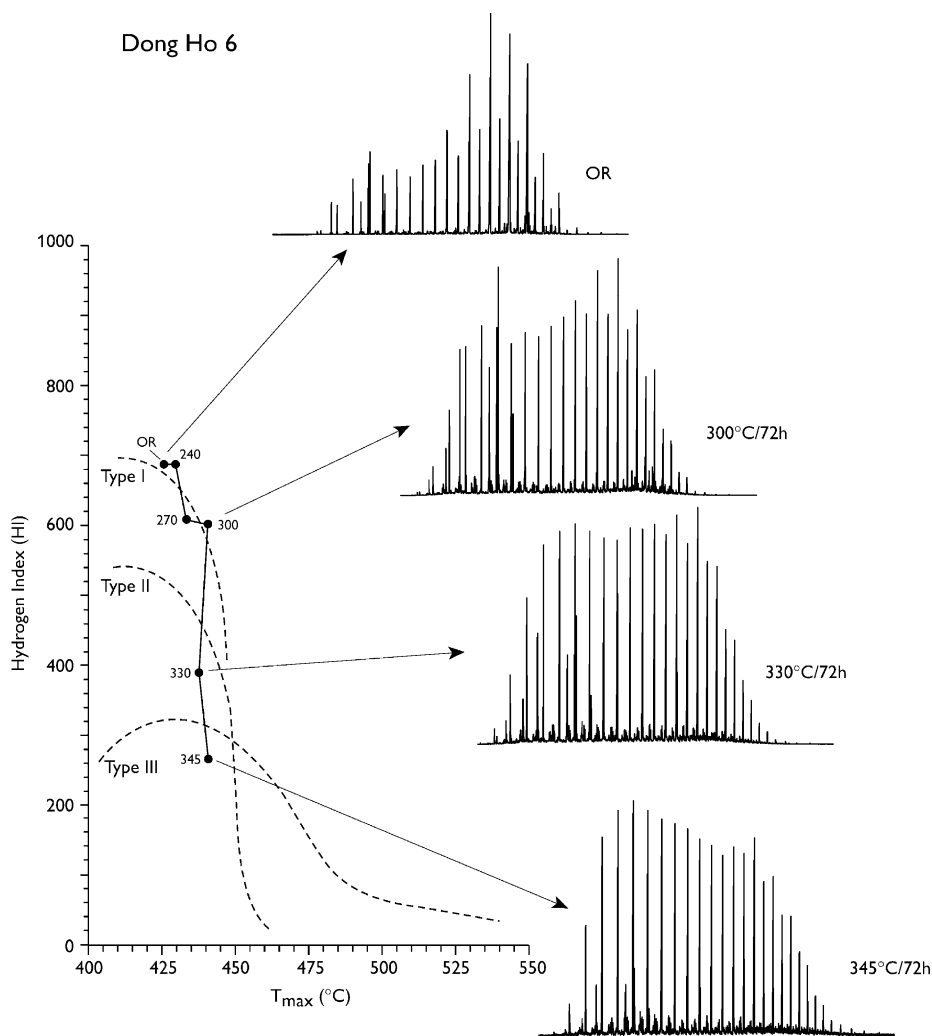


Fig. 7. Stepwise artificial maturation by hydrous pyrolysis of the Dong Ho 6 lacustrine mudstone. The HI shows a significant decrease from 300 to 345 °C/72 h, but virtually no change in T_{max} is observed, suggesting pronounced hydrocarbon generation above 300 °C/72 h. The *n*-alkanes of the extracted saturates from the hydrous-pyrolysed samples show an immature distribution below 300 °C/72 h. At 330 °C/72 h the *n*-alkanes have attained the distribution of an early mature terrestrial-sourced oil, and at 345 °C/72 h the *n*-alkanes resemble a mature terrestrial-sourced oil. OR: original immature sample; 240, 270, 300, 330, 345: hydrous pyrolysis temperature, °C/72 h.

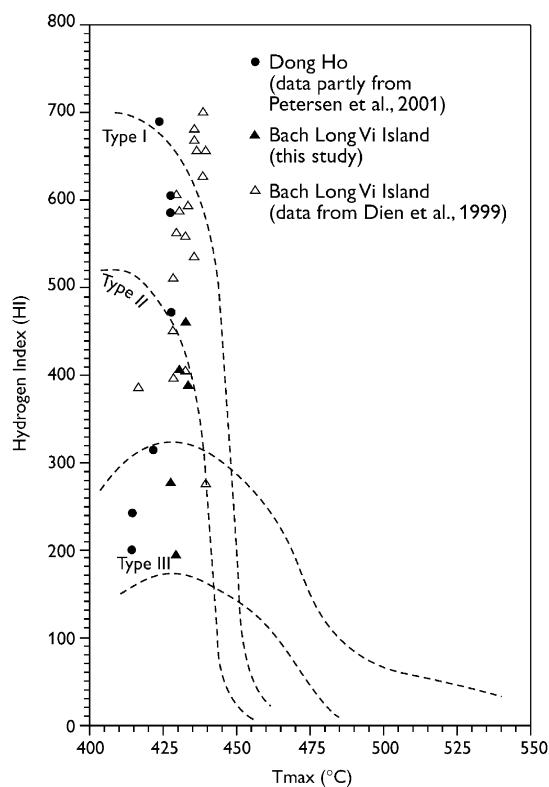


Fig. 8. HI versus T_{\max} plot of the samples from Dong Ho and Bach Long Vi Island. The mudstones generally have an HI greater than 400 mg HC/g TOC with numerous samples having an HI from 600–700 mg HC/g TOC. The humic coals have HIs from 200 mg HC/g TOC to just above 300 mg HC/g TOC.

organic facies and kerogen type content (Katz et al., 1993). A better understanding of the distribution of the lacustrine organic facies in the northeastern Song Hong Basin and accordingly a better assessment of the

hydrocarbon potential therefore is dependant on a more detailed knowledge of the depositional conditions in the lakes during the times of organic-rich mud accumulation.

3.3. Hydrocarbon generation characteristics

The four artificially matured samples follow the maturation path of the types I/II kerogen on the HI versus T_{\max} diagrams, which is in agreement with their organic matter composition (Figs. 6, 7, 9). The DCN I-2-97 sample shows a decrease in HI value from the initial 407 mg HC/g TOC to 152 mg HC/g TOC at the highest hydrous pyrolysis temperature (330 °C/72 h). The change in T_{\max} is small, only 9 °C. The initial decrease in T_{\max} (240 °C/72 h and 270 °C/72 h) is related to generated and extractable bitumen (see description of Dong Ho 1 below). The HI of the DCN III-3-97 sample decreases from an initial value of 462 mg HC/g TOC to 244 mg HC/g TOC (at 330 °C/72 h) during artificial maturation. From 300–330 °C/72 h the HI decreases sharply while T_{\max} shows little change (Fig. 9). Two lacustrine mudstone samples (samples Dong Ho 1 and 6) from Dong Ho were hydrous-pyrolysed up to 345 °C/72 h. Together with the results from previous hydrous pyrolysis experiments (Petersen et al., 2001), it can be shown that the HI values decrease rapidly during artificial maturation, in particular above 300 °C/72 h (Figs. 6 and 7). From 300 °C/72 h to 345 °C/72 h, the change in T_{\max} of Dong Ho 6 is negligible, while T_{\max} of Dong Ho 1 actually decreases above 300 °C/72 h (Figs. 6 and 7). However, extracted hydrous-pyrolysed samples of Dong Ho 1 show a slight increase in T_{\max} and a much more pronounced drop in the HI (Fig. 6). This indicates that generated, extractable bitumen or heavy hydrocarbons influences the remaining generative potential as represented by Rock-Eval derived S_2 . For example, at

Table 4
Summary of source rock characteristics

Locality	Lithotype	Organic matter composition		Kerogen	Hydrogen Index range	Start of the oil window	Principal HC product
		Primary	Secondary				
Dong Ho	Humic coal (TOC: 58–67 wt.%)	Huminite	Liptodetrinite Cutinite Sporinite	Type III	200–314	~1.03–1.15% R_{O^a}	Oil and Gas
	Mudstone (TOC: 4–17 wt.%) ^b	Fluorescing AOM Liptodetrinite	Alginite Sporinite Huminite	Type I/II	472–690	~0.75% R_O	Oil
Bach Long Vi Island	Siltstone & mudstone ^c TOC: 2–5 wt.%)	Fluorescing AOM	Liptodetrinite Alginite Huminite	Type I/II	195–462 ^c		Oil

^a Petersen (2002).

^b Unpublished GEUS-data included.

^c Dien et al. (1999) report TOC contents of mudstones up to 7 wt.% and HI values up to 702.

330 °C/72 h the S_2 yields for Dong Ho 1 may be reduced by approximately 73% upon solvent extraction.

The four samples do not show a steady increase in T_{max} and gradual decrease of HI with increasing maturity (i.e. hydrous pyrolysis temperature) as for example shown by humic coals and type III kerogen (e.g. Petersen et al., 2001; Petersen, 2002). In contrast, the lacustrine samples exhibit a dramatic decrease in HI with virtually no change in T_{max} . Such a maturity evolution can be related to the relatively homogeneous organic matter composition of lacustrine deposits (dominance of one bonding energy type), which typically is reflected by a narrow activation energy distribution (e.g. Tissot et al., 1987; Jarvie, 1991; Tegelaar and Noble, 1994; Petersen et al., 2002). The dominant narrow activation energy distribution has the effect, that within a restricted depth interval most energy, derived from the elevated temperature with subsidence, is used to convert the kerogen to hydrocarbons. Only a little energy is left to mature the organic matter. The activation energy distribution dictates that the lacustrine source rocks generate significant amounts of hydrocarbons within a restricted temperature range.

The significant decrease in HI above 300 °C/72 h is thus compatible with lacustrine source rocks, and indicates significant hydrocarbon generation above this hydrous pyrolysis temperature. The increase in S_1 yields (normalised to TOC) is another evidence of considerable hydrocarbon generation at this temperature (Fig. 10). Extract yields also increase significantly, but most important is the change in extract composition from a polar-dominated petroleum to being increasingly dominated by saturated hydrocarbons (Fig. 11; Tables 2 and 3). A prominent change towards higher proportions of saturated hydrocarbons is first noticeable at 330 °C/72 h, particularly for the two Dong Ho samples pyrolysed at 345 °C/72 h. These two samples clearly indicate that the generated hydrocarbon product approaches a composition similar to the B10-STB-1x oil, which contains 69.8% saturated hydrocarbons (Fig. 11; Table 3). Significant hydrocarbon generation above 300 °C/72 h is supported by the *n*-alkane distributions of the gas chromatograms, but the matured Bach Long Vi Island and Dong Ho samples show different distributions of the *n*-alkanes. The gas chromatograms of the saturated fractions of the immature Bach Long Vi Island samples

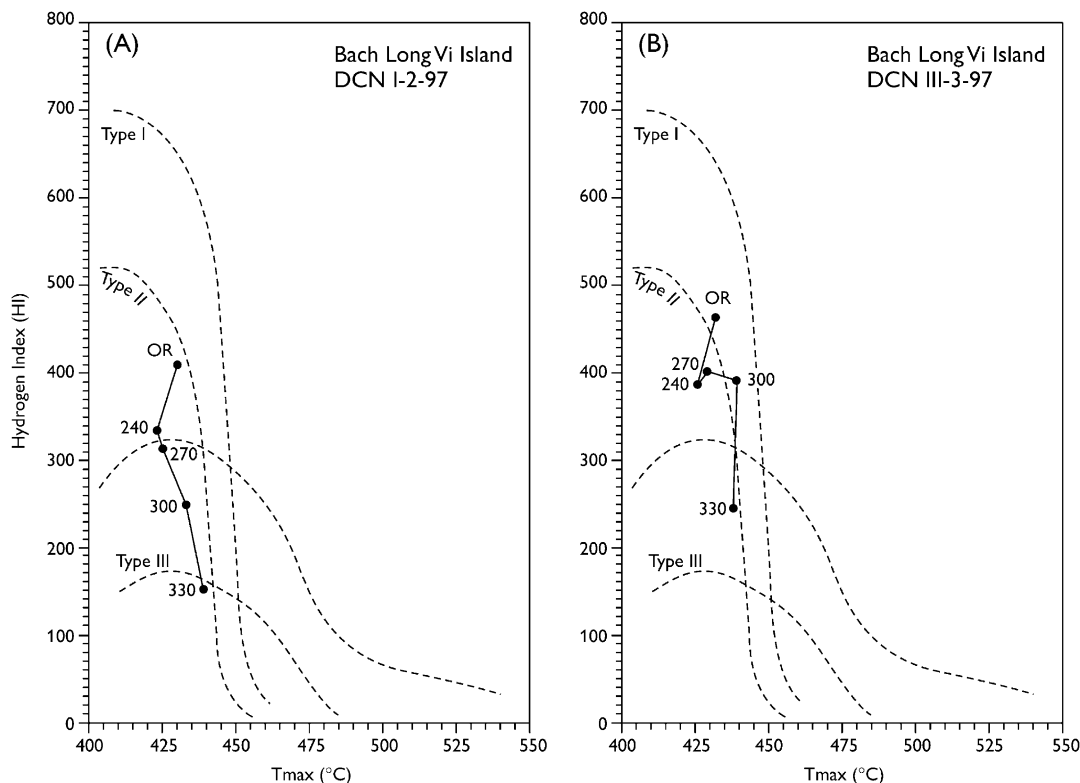


Fig. 9. Stepwise artificial maturation by hydrous pyrolysis of two samples from Bach Long Vi Island. The significant decrease in HI and the minor to negligible increase in T_{max} is related to the organic matter composition, which principally corresponds to types I/II kerogen (see Table 1). OR: original immature sample; 240, 270, 300, 330: hydrous pyrolysis temperature, °C/72 h.

are characterised by high proportions of pristane and phytane (relative to *n*-alkanes), light-end skewed *n*-alkane distributions with a maximum at nC_{17} , an odd-carbon predominance, and *n*-alkanes extending at least to nC_{33} (Fig. 5). In contrast, the most matured samples (330 °C/72 h) show broad unimodal, nearly symmetrical *n*-alkane distributions centred at nC_{19-20} and a decreased odd-carbon numbered *n*-alkane predominance (Fig. 5; Tables 2 and 3). The decrease in *n*-alkane abundance with increasing carbon-chain length above nC_{20} is pronounced, but *n*-alkanes up to at least nC_{33} are recorded (Fig. 5). Pr/Ph ratios increase to 1.30 and 1.80 from less than 1.00. At 300 °C/72 h or lower temperatures the two Dong Ho samples yield distinctly heavy-end skewed

n-alkane distributions with a maximum between nC_{25-31} and with *n*-alkanes extending to about nC_{35} (Figs. 6 and 7). This indicates immaturity. With increasing maturation (300–330 °C/72 h) the samples yield broad heavy-end skewed *n*-alkane distributions with a slight bimodality (in particular Dong Ho 6), which resemble the *n*-alkane distribution of early mature oil. At the final hydrous pyrolysis temperature of 345 °C/72 h the gas chromatograms display broad slightly bimodal but distinctly light-end skewed *n*-alkane distributions that maximise at nC_{17} and extend at least to nC_{36} . Such an *n*-alkane distribution resembles the distribution of mature oil. Ruble et al. (2001) also observed a significant increase in the proportion of saturated hydrocarbons generated from the Green River Shale subjected to hydrous pyrolysis between 330–360 °C/72 h; below this temperature, the main generation product was bitumen.

3.4. The start of the oil window for the lacustrine source rocks

At a hydrous pyrolysis temperature of 330 °C/72 h several important observations can be made: a significant decrease in HI with no or negligible change in T_{max} ; a significant increase in the proportion of saturated hydrocarbons in the pyrolysates; and an *n*-alkane distribution comparable to early mature – mature oils. The amount of generated hydrocarbons (S_1 yields normalised to TOC) increases significantly during maturation, in particular above a hydrous pyrolysis temperature of 300 °C/72 h, with the exception that for

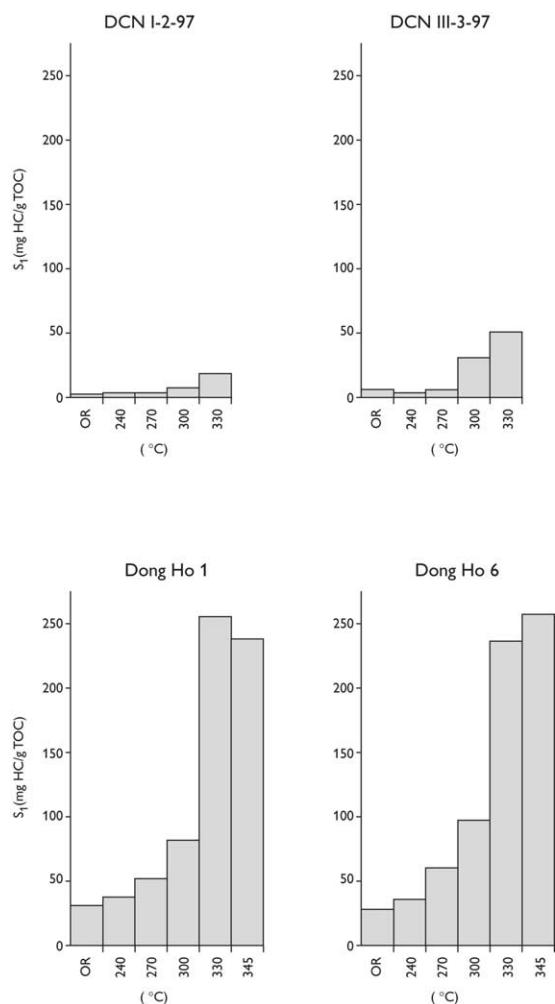


Fig. 10. Evolution in S_1 yields (normalised to TOC) during artificial maturation of the samples from Bach Long Vi Island and Dong Ho. The increase in S_1 yields is particularly pronounced at a hydrous pyrolysis temperature of 330 °C/72 h. Note the decrease in S_1 yields from 330 to 345 °C/72 h for the Dong Ho 1 sample.

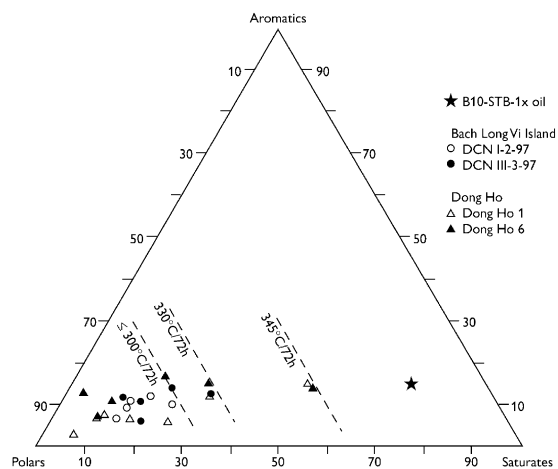


Fig. 11. Plot showing the evolution of the extract composition during artificial maturation. No compositional change is observed at hydrous pyrolysis temperatures < 300 °C/72 h. The proportion of saturated hydrocarbons increases at 330 °C/72 h and in particular at 345 °C/72 h the increase is pronounced. The extract composition of the hydrous-pyrolysed samples seems to approach that of the B10-STB-1x oil, which is strongly dominated by saturated hydrocarbons.

Dong Ho 1 the S_1 yield actually decreases from 330 °C/72 h to 345 °C/72 h (Fig. 10). This is an important observation with respect to the definition of the start of the oil window for the studied Vietnamese lacustrine source rocks. The decrease in S_1 yields (normalised to TOC) is anticipated to indicate the start of hydrocarbon expulsion from humic coals and type III kerogen and thus to define the start of the oil window for such source rocks (see Price (1989) and Petersen (2002) for a thorough discussion). Petersen (2002) established a correlation between Rock-Eval derived T_{max} and vitrinite reflectance on the basis of a large naturally matured coal data-set. Although the correlation between T_{max} and vitrinite reflectance was established for humic coals, it is attempted to use the correlation for the type I kerogen dominated Dong Ho 6 sample, which contains 11 vol.% type III kerogen. The T_{max} value at 330 °C/72 h is 436 °C, and by using Eq. (1) ($T_{max} = 51.96\%R_o + 398.39$) in Petersen (2002), the equivalent vitrinite reflectance ($\%R_{eqv}$) is $0.72\%R_{eqv}$. At 345 °C/72 h the corresponding vitrinite reflectance value is $0.80\%R_{eqv}$. In contrast to Dong Ho 1, the Dong Ho 6 sample does not show a decrease in S_1 yields. However, the extraordinarily high potential of Dong Ho 6 combined with the very pronounced drop in HI from 300 °C/72 h to 345 °C/72 h suggest that the amount of generated hydrocarbons exceeds the hydrocarbon expulsion efficiency at this maturity stage. The only limited increase in S_1 yields between 330 °C/72 h and 345 °C/72 h may imply that expulsion has started (Fig. 10). For the two Bach Long Vi Island samples, other factors may influence the expulsion efficiency. The lower amounts of organic matter and the lower proportions of generated hydrocarbons may retard the expulsion efficiency.

In conclusion, the decrease in S_1 between 330 °C/72 h and 345 °C/72 h in Dong Ho 1 combined with the calculated vitrinite reflectances for Dong Ho 6 suggest that the start of the oil window (i.e. oil expulsion) for these type I kerogen dominated lacustrine source rocks lies between $0.72\%R_{eqv}$ and $0.80\%R_{eqv}$ (approximately $0.75\%R_{eqv}$ in average). This is at a considerably higher maturity than the conventional start of the oil window at $0.6\%R_o$. The result is, however, in agreement with other studies of lacustrine source rocks. Tegelaar and Noble (1994) determined the onset of oil generation and peak oil generation for source rocks containing telalginites derived from *Botryococcus braunii* and *Tasmanites* algae (e.g. Australian Torbanite) to occur approximately at 140–150 °C and 150–160 °C, respectively. The corresponding calculated vitrinite reflectance values are approximately 0.96 – $1.13\%R_o$ and 1.13 – $1.31\%R_o$, respectively (Tegelaar and Noble, 1994). For lacustrine lamalginite/fluoramorphinite kerogens (e.g. Green River Shale), Tegelaar and Noble (1994) calculated the onset of oil generation to occur from 0.8 – $0.9\%R_o$ and the peak oil generation to occur between $0.95\%R_o$ and

$1.05\%R_o$. Ruble et al. (2001) also suggested peak oil generation from the Green River Shale to occur at approximately $1.0\%R_o$, while the onset of oil generation starts at $0.75\%R_o$ (Baskin and Peters, 1992). A significantly lower onset of oil generation can be recorded for sulphur-rich lacustrine source rocks, as for example approximately $0.63\%R_o$ for a Miocene sample from the lacustrine Mae Sot Basin in northern Thailand (Tegelaar and Noble, 1994). However, the TS contents of the investigated Dong Ho and Bach Long Vi Island samples do not suggest the presence of sulphur-rich kerogen in these samples. Oil expulsion is considered to commence shortly after the onset of oil generation due to the significant hydrocarbon generation over a narrow temperature range (Figs. 6, 7, 9). For the Green River Shale, for example, the oil window may only span $0.3\%R_o$ (Tegelaar and Noble, 1994).

3.5. The B10-STB-1x oil: evidence for a lacustrine-coal sourced petroleum system

The B10-STB-1x oil contains several characteristic non-marine geochemical signatures. This includes the very low sulphur content of 0.1–0.2 wt.%, a Pr/Ph ratio of 3.36, the abundance of long-chain *n*-alkanes that extends to about nC_{33} , the pronounced content of C_{29} steranes (45%) and the low sterane/hopane ratio of 0.06–0.09 (e.g. Clayton, 1993; Philp, 1994; Hunt, 1996) (Figs. 12–14; Tables 5 and 6). The sterane/hopane ratio for the B10-STB-1x oil was measured as the sum of $C_{27} + C_{28} + C_{29}$ $\alpha\alpha\alpha$, 20R-steranes versus the $\alpha\beta$ -hopane. Because of the low sterane/hopane ratio in the B10-STB-1x oil, several hopanes and some unidentified compounds were visible in the m/z 217 and m/z 218

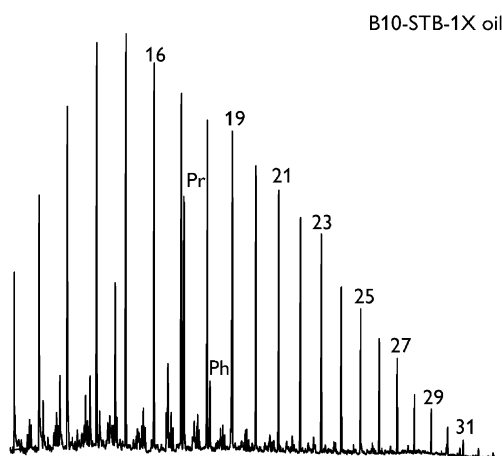


Fig. 12. Gas chromatogram showing the light-end skewed *n*-alkane distribution of the B10-STB-1x oil. The *n*-alkanes extend to at least nC_{33} and the Pr/Ph ratio is 3.36. The geochemical signature is typical of paraffinic oil from a shale source.

mass chromatograms. Similarly, the Bach Long Vi Island and Dong Ho samples had a high content of oleanenes rendering the m/z 217 and especially the m/z 218 mass chromatograms useless for quantification of steranes. We have therefore only used GC/MS/MS-data for the sterane ratios in Table 5. Hence, using GC/MS/MS ($M^+ \rightarrow 217$ and $412 \rightarrow 191$), the ratio was somewhat higher (0.09) than that obtained from ordinary SIM-analysis using m/z 217 and m/z 191 (0.06) (Table 6). The Pr/Ph ratio is significantly higher than those detected for the Bach Long Vi Island samples, whereas it is more similar to the Dong Ho samples, especially the coals (see Petersen et al., 2001). Furthermore, bicadinanes were identified in HPLC fractions of a triterpane concentrate from the oil. The bicadinanes are not very prominent in the m/z 191 mass chromatogram, but they can be seen in the m/z 369 mass chromatogram (Fig. 14).

Bicadinanes are considered a marker from angiospermous dammar-type resins (van Aarssen et al., 1992). The oil contains a moderate amount of $18\alpha(H)$ -oleanane and a minor amount of co-eluting lupane (Fig. 14). Using a polar GC-column (ZB-wax), oleanane and lupane can be separated, and a lupane/(lupane + oleanane) ratio of approximately 0.06 was documented by Nytoft et al. (2002). A low content of $19\alpha(H)$ -taraxastane is also present. The presence of $18\alpha(H)$ -oleanane, lupane and $19\alpha(H)$ -taraxastane is indicative of angiospermous land plant input to the oil (Peters and Moldovan, 1993; Perkins et al., 1995) (Fig. 14). In addition, high contents of $18\alpha(H)$ -oleanane and lupane are usually associated with a Tertiary source, although exceptions do occur (Moldovan et al., 1994). Also, the depositional environment influences the amount of oleanane (Murray et al., 1997). Oleanenes, which are

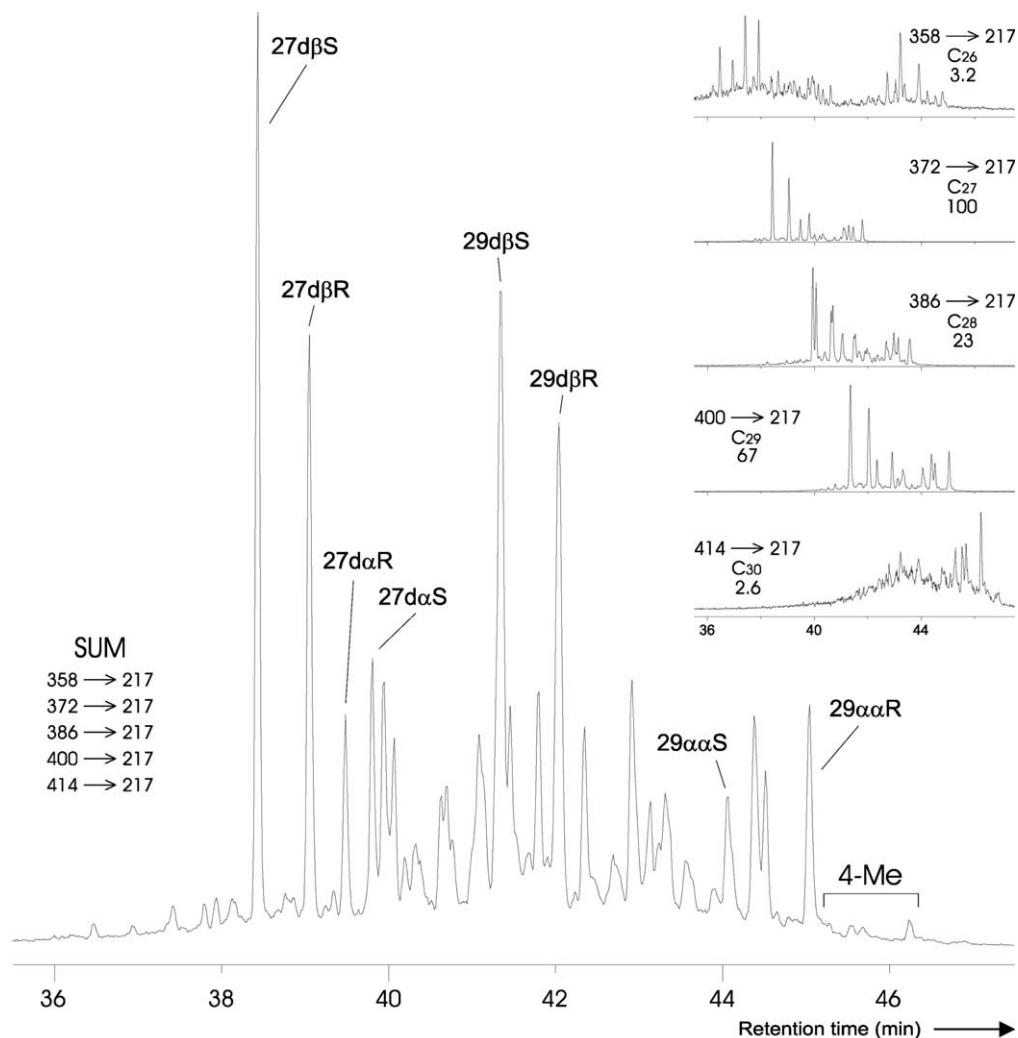


Fig. 13. GC/MS/MS analysis of C_{26} – C_{30} steranes ($M^+ \rightarrow 217$) in the B10-STB-1x oil. The main chromatogram shows the sum of m/z 358, 372, 386, 400 and 414 \rightarrow 217 transitions. Insets show sterane epimer distribution by carbon number.

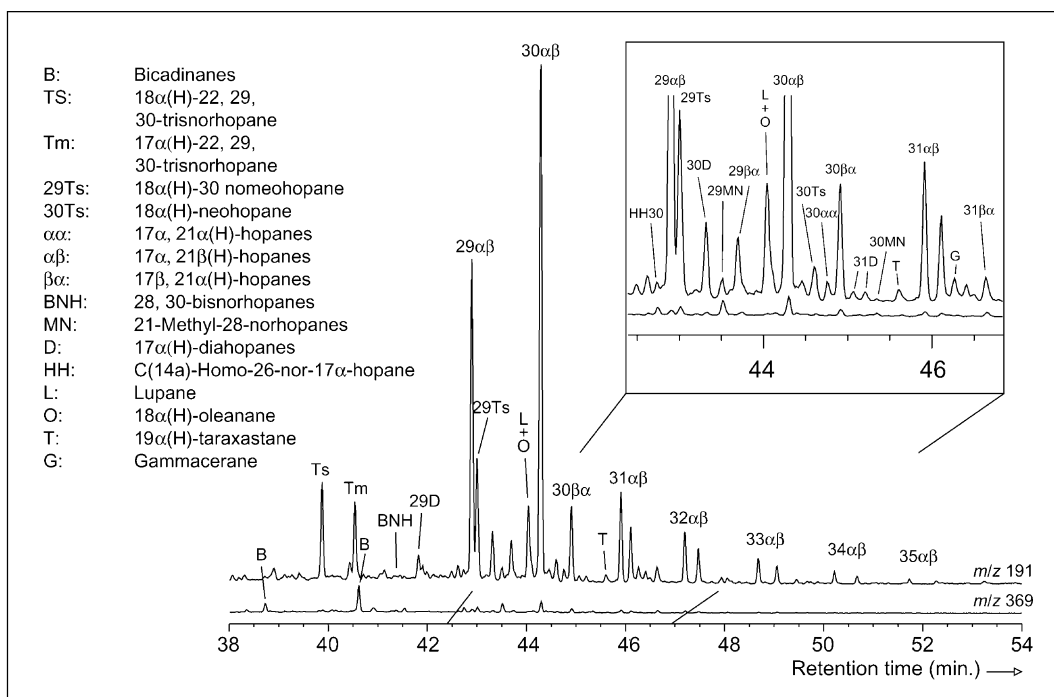


Fig. 14. m/z 191 mass chromatogram of the B10-STB-1x oil showing hopanes and other triterpanes. Bicadinanes and 21-methyl-28-norhopanes can be seen in the m/z 369 mass chromatogram. Terrestrial markers are lupane, oleanane, taraxastane and bicadinane.

Table 5
GC/MS/MS data

Composition	DCN I-2-97	DCN II-1-97	DCN II-2/1-97	DCN III-3-97	DCN IV-2/1-97	Dong Ho 1	Dong Ho 6	Dong Ho 7	Dong Ho 8	B10-STB-1X oil
% 27 reg. ster	33	29	14	34	36	26	23	31	6	41
% 28 reg. ster	23	22	12	13	13	17	29	15	29	14
% 29 reg. ster	44	49	74	53	52	57	48	54	65	45
% Σ 27 ster	31	28	18	36	40	24	24	27	8	37
% Σ 28 ster	25	27	20	18	18	19	26	20	26	20
% Σ 29 ster	45	45	62	46	43	57	50	53	65	44
% Σ 28 4Me	38	42	27	17	27	34	24	16	14	39
% Σ 29 4Me	21	18	21	20	22	15	8	6	17	17
% Σ 30 4Me	40	39	52	64	51	51	69	78	69	44
4Me / (4Me + ster)	0.33	0.46	0.45	0.57	0.56	0.74	0.89	0.93	0.36	0.48
% H30 $\alpha\beta$	89	80	78	91	90	62	62	67	24	89
% H30 $\alpha\alpha$	6	9	6	4	5	1	2	1	4	3
% H30 $\beta\alpha$	4	8	15	4	4	14	11	18	25	8
% H30 $\beta\beta$	1	2	1	1	1	22	25	15	47	0
H31, 22S / (22S + 22R)	0.17	0.14	0.11	0.33	0.30	0.04	0.02	0.02	0.03	0.56
Gammacerane	0.03	0.03	0.02	0.02	0.02	0.00	0.00	0.00	0.00	0.02
BNH	0.02	0.21	0.00	0.02	0.02	0.00	0.00	0.00	0.00	0.006

% 27 reg. ster = C_{27} sterane * 100 / (C_{27} + C_{28} + C_{29} steranes) using only $\alpha\alpha\alpha$, 20R isomers (372 \rightarrow 217, 386 \rightarrow 217, 400 \rightarrow 217). % Σ 27 ster = C_{27} steranes * 100 / (C_{27} + C_{28} + C_{29} steranes) using all isomers including diasteranes (372 \rightarrow 217, 386 \rightarrow 217, 400 \rightarrow 217). % Σ 28 4Me = C_{28} 4-methylsteranes * 100 / (C_{28} + C_{29} + C_{30} 4-methylsteranes) using all isomers (386 \rightarrow 231, 400 \rightarrow 231, 414 \rightarrow 231). 4Me / (4Me + ster) = C_{30} 4-methylsteranes / (C_{30} 4-methylsteranes + C_{29} steranes) using all isomers including diasteranes (414 \rightarrow 231, 400 \rightarrow 217). % H30 $\alpha\beta$ = H30 $\alpha\beta$ * 100 / (H30 $\alpha\beta$ + H30 $\alpha\alpha$ + H30 $\beta\alpha$ + H30 $\beta\beta$) (412 \rightarrow 191). Gammacerane = Gammacerane / (Gammacerane + Σ H30) (384 \rightarrow 191, 412 \rightarrow 191). BNH = Σ 28,30-bisnorhopanes / (Σ 28,30-bisnorhopanes + Σ H30).

precursors of oleananes, have been detected in all Bach Long Vi Island and Dong Ho samples in this study, and in mudstones and coals from Dong Ho in previous studies (Petersen et al., 1998). In a few cases (samples DCN II-I-97 and Dong Ho 1) oleanenes are more abundant than hopanes. However, oleanene and ursene are also present in the B10-STB-1x oil as olean-13(18)-ene, olean-18-ene, olean-12-ene, 18 α (H)-olean-12-ene and urs-12-ene were identified in the HPLC purified fractions. These unsaturated compounds of terrigenous origin are often found in oils (Curiale and Lin, 1991; Curiale, 1995; Peakman et al., 1991; Ukpabio et al., 1994; Eneogwe et al., 2002), where they appear, in most cases, to be inherited directly from the source rock. The presence of the less stable hop-17(21)-enes (C₃₀–C₃₄) may, however, suggest that they could also have been picked up from immature source rocks during migration.

Because of the high content of unsaturated triterpenoids which made the *m/z* 191 mass chromatograms very complex, GC/MS/MS had to be used to get reliable hopane data for the Bach Long Vi Island and Dong Ho samples (Table 5). Hopane isomerisation at C-22: 22S/(22S+22R) is only 0.02–0.04 in the Dong Ho samples and 0.11–0.33 in the Bach Long Vi Island samples consistent with the slightly lower *T*_{max} of the former. The less mature Dong Ho samples also contain a higher proportion of the thermodynamically unstable $\beta\beta$ -hopanes and a relatively high content of $\beta\alpha$ -hopanes (moretanenes). The relative concentration of $\alpha\alpha$ -hopanes (Nytoft and Bojesen-Koefoed, 2001) in the Dong Ho samples is low, whereas three of the Bach Long Vi Island samples have similar concentrations of $\alpha\alpha$ - and $\beta\alpha$ -hopanes.

The oil contains a relatively high content of neohopanes and diahopanes, and other rearranged hopanes (e.g. Moldowan et al., 1991; Farrimond and Telnæs, 1996). In addition to these well-known series of rearranged hopanes, C(14a)-homo-26-nor-17 α -hopanes (Trendel et al., 1993), 21-methyl-28-norhopanes (Nytoft and Bojesen-Koefoed, 2001; Nytoft et al., 2002) and

several minor, still unidentified series of rearranged hopanes were detected. Abundant rearranged hopanes and diasteranes suggest an origin from a clay-rich source, which agrees with samples from both Bach Long Vi Island and Dong Ho. Both deposits also contain abundant neohop-13(18)-enes (C₂₉–C₃₁) and diahop-13-enes which appear to be the immediate precursors of neohopanes and diahopanes respectively (Bojesen-Koefoed et al., 2001; Nytoft et al., 2002).

The distribution of steranes in the Bach Long Vi Island and Dong Ho samples is shown in Table 5. The sterane concentration in the Dong Ho samples (except Dong Ho 6 used for hydrous pyrolysis) was low, whereas the Bach Long Vi Island samples (especially DCN III-3-97 and DCN IV-2/1-97) had a higher concentration. The distribution of steranes in the DCN III-3-97 and DCN IV-2/1-97 samples is very similar to that of the B10-STB-1x oil. Hence, a small contribution from a source rock similar to the Bach Long Vi Island samples could be responsible for the distribution of steranes in the oil if the bulk of the oil was generated from a source rock poor in steranes (such as the Dong Ho mudstones). The oil contains a high proportion of C₂₇ steranes (41%), which is in agreement with an algal-dominated (*Botryococcus*-type) lacustrine source rock, and a very low proportion of C₂₈ steranes (14%), which is also typical of lacustrine-derived oils (Fig. 13; Tables 5 and 6) (Peters and Moldowan, 1993). These characteristics are very similar to the geochemical parameters listed as diagnostic of fresh water, lacustrine-sourced oils in Brazil by Mello and Maxwell (1990). Likewise, the distribution of steranes in the B10-STB-1x oil (Table 5) is very similar to that reported by Holba et al. (2003) for a number of lacustrine oils from Congo and Gabon. Common for these oils is a low content of C₂₈ steranes. The presence of C₃₀ steranes (4-desmethylsteranes) is the most powerful parameter for identifying input from marine organic matter (Peters and Moldowan, 1993); these steranes have not been identified in the oil under study, emphasising its terrestrial origin. Although the mass spectra of 4-methylsteranes have a *m/z* 231 base

Table 6
Characteristics of petroleum derived from different source rock types

Parameter	Source rock type			B10-STB-1x
	Marine	Terrestrial	Lacustrine	
Sulphur	High	Low	Low	0.1–0.2 wt. %
Pristane/Phytane	~2	~3	~1–3	3.36
Steranes/hopanes	High	Low	Low	0.06–0.09a
C ₂₇ –C ₂₉ steranes	High C ₂₇	High C ₂₉	Low C ₂₈	41% C ₂₇ , 14% C ₂₈ , 45% C ₂₉
C ₃₀ steranes	Present	Absent or low	Absent	Absent
TPP ^b	~0.3	~0.3	0.3–1.0	0.65

^a See text for explanation.

^b TPP: tetracyclic polyphenols (Holba et al., 2003).

peak they also contain a small m/z 217 fragment. The small peaks eluting after the C_{29} steranes in Fig. 13 are in fact C_{30} 4-methylsteranes and not C_{30} -steranes which have slightly different retention times. A high ratio of 4-methylsteranes to 4-desmethylsteranes is common in lacustrine oils (Holba et al., 2003). The B10-STB-1x oil has equal concentrations of the two groups of steranes (Table 5) and a tetracyclic polyprenoids (TPP) ratio of 0.65. The B10-STB-1x oil thus plots in the zone “Lacustrine I” of Holba et al. (2003) (their Fig. 4b). The 4-methylsteranes are probably derived from dinoflagellates and the TPP ratio is an indicator of fresh/brackish water algal input (Holba et al., 2000, 2003). The Bach Long Vi Island samples and the Dong Ho 8 sample had similar 4-methylsterane ratios, whereas the remaining Dong Ho samples were dominated by 4-methylsteranes. One dinosterane was identified by full scan analysis of HPLC-fractions from the B10-STB-1x oil and several in HPLC fractions from the Bach Long Vi Island DCN-II-1-97 sample. However, GC/MS/MS showed that dinosteranes were minor compounds in all samples compared to other 4-methylsteranes, consistent with lacustrine organic matter.

Gammacerane was positively identified in the HPLC fractions of the B10-STB-1x-oil in full scan mode, but the content is low. Actually, it is smaller than it appears from the m/z 191 mass chromatogram because of co-elution with a C_{31} neohopane and some minor compounds (Fig. 14). GC/MS/MS data ($412 \rightarrow 191$) showed a gammacerane/hopane ratio of 0.02 (Table 5). Similar ratios were detected in the Bach Long Vi Island samples, whereas gammacerane was absent in the Dong Ho samples. A high concentration of gammacerane is often found in sediments deposited under hypersaline conditions but is not restricted to this type of deposits. Gammacerane is also taken as an indicator of water column stratification (Sinninghe Damste et al., 1995). The low gammacerane content in the oil does not add information to identification of the source rock.

Bisnorhopanes ($\alpha\beta$ and $\beta\alpha$) were identified in the HPLC-purified fractions of the B10-STB-1x oil. However, the content is low (Table 5), and the bisnorhopanes are hardly visible in the m/z 191 mass chromatogram (Fig. 14). Six isomers of bisnorhopane were likewise detected in the HPLC fractions from the Bach Long Vi Island DCN II-1-97 sample (full scan) together with 28,30-bisnorhop-13(18)-ene, which is a possible precursor (Bojesen-Koefoed et al., 2001). The distribution of bisnorhopanes in this sample is shown in Fig. 15. Bisnorhopanes were absent in the Dong Ho samples. The stereochemistry (at C-17, C-18 and C-21) of the bisnorhopanes present in crude oils were determined by Moldowan et al. (1984). All have the $18\alpha(H)$ -stereochemistry. The stereochemistry at C-17 and C-21 is $17\alpha(H),21\beta(H)$, $17\beta(H),21\alpha(H)$ and $17\beta(H),21\beta(H)$. The $\alpha\beta$ and $\beta\alpha$ epimers dominate in crude oils. The

three unidentified epimers which have been detected in the Bach Long Vi Island samples and other immature sediments (Bojesen-Koefoed et al., 2001) appear to be thermally unstable, since they have only been detected in trace amounts in crude oil. In the DCN I-2-97 and DCN II-1-97 samples the unidentified compounds constitute half of the total 28,30-bisnorhopane content but only one fourth in the DCN III-3-97 and DCN IV-2/1-97 samples. The $\beta\beta$ -epimer and two of the unidentified bisnorhopanes coelute with $\alpha\beta$ -30-norhopane and 29Ts. The late eluting bisnorhopane coelutes with $\beta\alpha$ -30-norhopane (normoretane). It is, therefore, necessary to use GC/MS/MS to detect small concentrations of the four late eluting bisnorhopanes.

The oil is extremely paraffinic (70% saturates), which is typical of lacustrine-sourced oils due to the primary contribution of long-chain n -alkanes from the lipid-rich algal organic matter. A high content of saturated hydrocarbons has, for example, been observed in lacustrine source rocks from the Upper Carboniferous in East Greenland (Christiansen et al., 1990). The hydrous pyrolysis experiments of the lacustrine mudstones in the present study demonstrate a considerable increase in paraffinity of the extracts with increasing maturity (Fig. 11). The artificial maturation of the samples from Dong Ho actually shows a compositional development of the extracts towards a composition similar to the B10-STB-1x oil (Fig. 11). In addition, the gas chromatograms of the most artificially matured ($345^\circ\text{C}/72\text{ hr}$) lacustrine mudstones from Dong Ho show an evolution, which with a slightly higher maturity would yield a light-end skewed n -alkane distribution resembling that

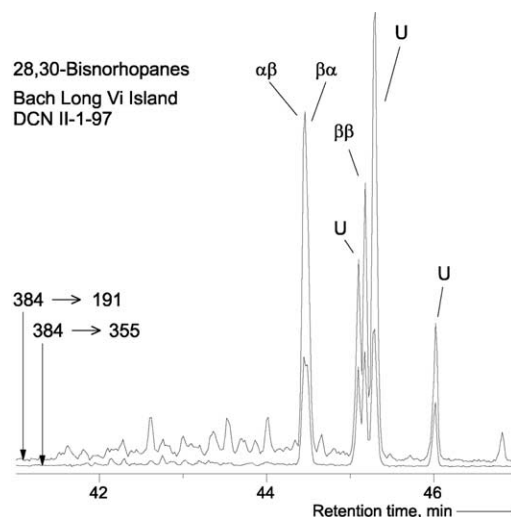


Fig. 15. GC/MS/MS analysis of 28,30-bisnorhopanes ($384 \rightarrow 191$ and $384 \rightarrow 355$) in the DCN II-1-97 sample. $\alpha\beta$: $17\alpha(H), 21\beta(H)$ -28,30-bisnorhopane. $\beta\alpha$: $17\beta(H), 21\alpha(H)$ -28,30-bisnorhopane. $\beta\beta$: $17\beta(H), 21\beta(H)$ -28,30-bisnorhopane. U: 28,30-bisnorhopane, unknown stereochemistry.

of the B10-STB-1x oil (Figs. 6,7,12). This contrasts with the artificially matured Bach Long Vi Island samples, which yield a nearly unimodal *n*-alkane distribution (Fig. 5).

In summary, the B10-STB-1x oil was sourced from lacustrine facies containing algal kerogen with some contribution from terrestrial higher land plants (coal or coaly source). This conclusion is in agreement with Dau et al. (2000). The oil does not contain any strong saline indicators, which could favour a source similar to, for example, the Dong Ho mudstones. However, the oil has characteristics similar to both the Bach Long Vi Island and Dong Ho potential source rocks. The Pr/Ph ratio and the *n*-alkane distribution of the oil point to a Dong Ho type source rock, whereas the sterane distribution of the oil is very similar to the distribution of three of the Bach Long Vi Island samples. As discussed above, the contribution from such a source rock type may be accentuated in the oil if the principal source rock contains a very low sterane content, which is the case for the Dong Ho mudstones. The B10-STB-1x oil composition probably represents an average of the relative contribution from a lacustrine source rock succession with its inherited variation in organic facies and source potential. The quality of the potential Bach Long Vi Island and Dong Ho lacustrine source rocks is rather similar. The significantly higher TOC content of the Dong Ho samples (generally >8 wt.%, Fig. 4), however, would make a source rock similar to such deposits much more important in terms of generated volumes of hydrocarbons. This is also evident from the hydrous pyrolysis experiments, where the artificially matured Dong Ho samples contain considerably higher S_1 yields (Fig. 10). Thus, it is suggested that a Dong Ho type source rock is the principal source for the B10-STB-1x oil.

3.6. Maturity modelling of source rock units in the Kien An and Thuy Nguyen grabens

Distinct, continuous, high amplitude seismic reflections, which are interpreted as lacustrine shale-prone units, have been mapped regionally in the syn-rift successions in the halfgrabens of the northeastern Song Hong Basin (Andersen et al., 1998). This includes the Kien An and Thuy Nguyen grabens south of the Anzoil B10-STB-1x wellsite (Fig. 2). Petersen (2002) has determined that oil expulsion from the humic coals outcropping at Dong Ho occurs at a thermal maturity corresponding to a vitrinite reflectance of approximately 1.03–1.15% R_o . Modelling showed that the coals cannot have attained the necessary maturity in the Kien An and Thuy Nguyen grabens to act as the principal source rocks for liquid hydrocarbons. Recently collected coal samples from another outcrop of the Dong Ho Formation yield, however, much higher HI values (unpub-

lished data, GEUS), which could suggest earlier oil expulsion from these coals. A minor contribution from such coals would agree with the composition of the B10-STB-1x oil. Liquid hydrocarbon expulsion from the lacustrine mudstones at Dong Ho has been estimated to occur at a vitrinite reflectance between 0.72% R_o and 0.80% R_o , averaging approximately 0.75% R_o .

Vitrinite reflectance values in the 102-CQ well have been derived using the kinetic model EASY% R_o (Burnham and Sweeney, 1989; Sweeney and Burnham, 1990) (Figs. 2 and 16). The shale-prone source rock unit is defined between the seismic reflectors Ca350 and Ca380. The result shows that the lacustrine mudstones will only just have reached the start of the oil window in the basal part of the Kien An Graben, whereas the Thuy Nguyen Graben contains a mature source rock section (Fig. 16). The kitchen of the B10-STB-1x oil is thus most likely to be located in the Thuy Nguyen Graben, but a contribution from the Kien An Graben cannot be excluded. The calculated sterane $C_{29} S/(S+R)$ ratio of 0.42 (from GC/MS/MS) for the B10-STB-1x oil corresponds approximately to a vitrinite reflectance of 0.65% R_o (cf. Peters and Moldowan, 1993, their fig. 3.46), which is slightly lower than the lower limit of the estimated start of the oil window. Dau et al. (2000) concluded that the vitrinite reflectance range of the source rock, that generated the B10-STB-1x oil lies between 0.6% R_o and 0.8% R_o .

A location of the source kitchen in the Thuy Nguyen and Kien An grabens necessitates long-distance migration from the kitchen through basement rocks to the well-site. Similar long distance migration of hydro-

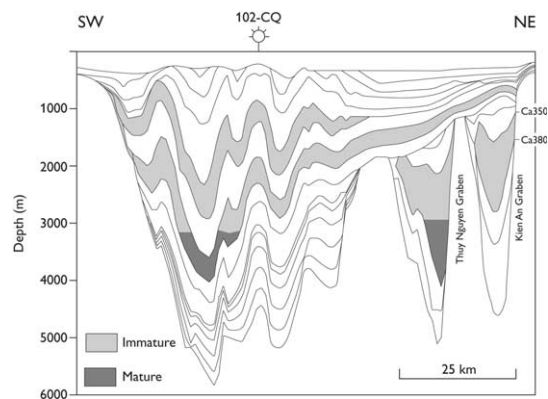


Fig. 16. Modelled vitrinite reflectance values using the kinetic EASY% R_o model (Burnham and Sweeney, 1989; Sweeney and Burnham, 1990) along the seismic line shown in Fig. 2. A vitrinite reflectance value of 0.75% R_o is used as the start of the oil window for the lacustrine mudstones (see Section 3.4). The mudstones in the source rock unit (between the seismic reflectors Ca350 and Ca380) will be mature in the Thuy Nguyen Graben, whereas they will only be mature in the basalmost part of the Kien An Graben.

carbons is known, however, from the Quy Nhon area, central Vietnam, where oil seeps in fractured granitic basement were probably generated in the offshore Phu Khanh Basin, which indicates migration of at least 10 km (Traynor and Sladen, 1997; GEUS, unpublished data).

4. Conclusions

An operating lacustrine-coal sourced petroleum system is suggested in the northeastern Song Hong Basin by:

1. The presence of highly oil-prone lacustrine and oil- and gas-prone humic coal source rocks in the northeastern Song Hong Basin is confirmed in Oligocene outcrops at Dong Ho and on the Bach Long Vi Island. The source rocks are also suggested to be present in the subsurface offshore by continuous, high amplitude seismic reflectors in undrilled Palaeogene syn-rift successions.
2. The mudstones from the two outcrops consist principally of fluorescing amorphous organic matter and liptodetrinite together with structured *Botryococcus*-type alginite (type I kerogen). The mudstones are lacustrine, but the depositional conditions at Bach Long Vi Island may have been slightly more marine-influenced, although this can not be fully documented. The coals consist primarily of huminitic organic matter.
3. The Dong Ho mudstones generally have a TOC content of 8–17 wt.% and HI values > 500 mg HC/g TOC, whereas the Bach Long Vi Island samples contain 2–7 wt.% TOC and yield HI values of 200–700 mg HC/g TOC. The coals yield HI values from about 200 to slightly over 300 mg HC/g TOC.
4. Artificial maturation by progressive hydrous pyrolysis demonstrates that the mudstones will generate large quantities of paraffinic oil within a relatively small temperature range. Hydrocarbon expulsion (start of the oil window) for the Dong Ho mudstones is estimated to occur at a maturity corresponding to a vitrinite reflectance between 0.72% R_o and 0.80% R_o (average ~0.75% R_o).
5. The B10-STB-1x oil was generated from Tertiary lacustrine source rocks with contributions from higher land plant organic matter, probably coals. The oil is early mature and it has geochemical characteristics resembling both the Dong Ho and Bach Long Vi Island mudstones, but was probably largely generated from a source more similar to the Dong Ho mudstones.

6. The source kitchen may be located in the Thuy Nguyen Graben and less likely the Kien An Graben. Maturity modelling shows that, by using a start of the oil window at ~0.75% R_o , the lacustrine mudstones will be thermally mature in the Thuy Nguyen Graben and only mature in the basal part of the Kien An Graben. Humic coals with high HI values may also have been able to reach the oil expulsion threshold.

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