IN-RESERVOIR FRACTIONATION AND THE ACCUMULATION OF OIL AND CONDENSATES IN THE SURMA BASIN, NE BANGLADESH

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ABSTRACT

Petroleum in the Surma basin, NE Bangladesh (part of the Bengal Basin) ranges from waxy crude oils to condensates. The origin and source rocks of these hydrocarbons were investigated based on the distributions of saturated and aromatic hydrocarbons in 20 oil samples from seven oil- and gas fields. The relative compositions of pristane, phytane and adjacent n-alkanes suggest that the source rock was deposited in a non-marine setting. The abundance and similar distribution of biphenyls, cadalene and bicadinanes in most of the crude oils and condensates indicates a significant supply of higher-plant derived organic matter to the source rocks. Maturity levels of the crude oils and condensates from the Surma basin correspond to calculated vitrinite reflectance (Rc) values of 1.0–1.3%, indicating hydrocarbon expulsion from the source rock at a comparatively high maturity level. The Rc values of oils from the Titas field in the southern margin of the Surma basin are relatively low (0.8–1.0%). Some oils were severely biodegraded. The similar distribution of diamondoid hydrocarbons in both biodegraded and non-biodegraded oils indicated similar types of source rocks and similar maturity levels to those of oils from the Surma basin. The Oligocene Jenam Shale and/or underlying non-marine deposits located at greater depths may be potential source rocks. The diversity of the petroleum in the Surma
basin was likely due to evaporative fractionation, resulting in residual waxy oils and lighter condensates which subsequently underwent tertiary migration and re-accumulation. Evaporative fractionation due to modification of the reservoir structure occurred during and after the Pliocene, when large-scale tectonic deformation occurred in and around the Bengal Basin.

Key words: Sylhet trough, Surma basin, Bangladesh, Titas gasfield, evaporative fractionation, condensates, waxy oils, naphthalene, diamondoid, reservoir rock, tertiary migration.

INTRODUCTION

Surma basin, a sub-basin of the Bengal Basin located in NE Bangladesh, evolved from a pre-Oligocene passive continental margin to a foreland basin associated with the Indo-Burman and Himalayan mobile belts. The basin is filled by thick (12–16 km) Late Mesozoic and Cenozoic sedimentary succession (Johnson and Alam, 1991). Hydrocarbons (primarily gas and gas-associated condensates) have been discovered in the Mio-Pliocene Surma Group, but the source of these hydrocarbons remains ambiguous. Although a number of geochemical studies have been performed (Hiller and Elahi, 1984; Alam and Pearson, 1990; Ahmed et al., 1991; Ismail and Shamsuddin, 1991; Shamsuddin and Abdullah, 1997; Curiale et al., 2002; Hossain et al., 2009; Takeda et al., 2011; Farhaduzzaman et al., 2013), few studies have evaluated the source rocks in the Surma basin based on the geochemical characteristics of the crude oils and condensates.

The oils and condensates in the Surma basin may have been generated by source rocks in the Oligocene Jenam Formation which was deposited in a deltaic environment (Ahmed et al., 1991). Condensates from the southern margin of the Surma basin are thought to be sourced from marine-influenced terrestrial organic matter (Ahmed et al., 1991). Alam and Pearson (1990) reported the occurrence of bicadinanes and oleanane in waxy oils from the basin, and suggested that the bicadinanes were formed from the polymerized resin of Dipterocarpaceae or similar angiosperms during source rock maturation. Curiale et al. (2002) also suggested
corresponding differences in source rock organic matter between oils from the central and northern Surma basin and those from the south, on the basis of the \( n \)-alkane distribution, pristane/phytane ratio and the presence and abundance of bicadinane. Migration-induced compositional variation in Bangladesh condensates and increasing wax content with depth have also been suggested (Curiale et al., 2002). These studies suggested that prospective source units are the Oligocene Jenam Formation in the central Surma basin and the Miocene Bhuban Formation in the south.

However, previous studies did not include detailed investigations of low molecular weight hydrocarbons, cyclic saturate hydrocarbons or polycyclic aromatic hydrocarbons which can be used to characterize crude oils and condensates, and their sources. In the present study, potential source rocks for the crude oils and condensates in the Surma basin were investigated based on their geochemical characteristics, including distributions of low molecular weight saturate and aromatic hydrocarbons, \( n \)-alkanes, diamondoids, alkynaphthalenes, biphenyls and phenanthrenes.

**Study area and regional geology**

Bangladesh is bordered by the Indian shield to the NW and west, the Shillong Plateau to the north, the Indo-Burman Ranges to the east, and the Bay of Bengal in the south (Fig. 1). The present-day platformal shelf of western Bangladesh was originally part of eastern Gondwana together with India, Australia, and Antarctica (Shamsuddin and Abdullah, 1997). India began to separate from Australia and Antarctica during the Early Cretaceous, drifting north and then NE, and finally colliding with the Asian plate (Shamsuddin and Abdullah, 1997; Curiale et al., 2002; Imam, 2005).

In terms of structure, Bangladesh can be divided into a stable shelf in the NW and west, underlain by Precambrian continental crust; and a deep basin in the south and east in which there is a very thick sedimentary succession (Uddin and Lundberg, 2004). The deep basin area can be further divided into an unfolded platform flank in the west, and to the east the Chittagong-Tripura fold belt (Shamsuddin and Abdullah, 1997). The NE–SW trending Hinge Zone separates the stable shelf and deep basin (Fig. 1), and may mark the transition from continental to oceanic crust.
The Bengal Basin had a complex sedimentary and tectonic history between Eocene and Recent times. Rapid uplift of the Himalayas to the north and the Indo-Burman Ranges in the east, slow subsidence of the western platform area and rapid subsidence of the foredeep in the south took place simultaneously. The Bengal delta, the largest delta in the world, formed in response to a huge influx of detrital material derived from the rising Himalayas and transported by the Ganges and Brahmaputra rivers (Johnson and Alam, 1991). Large-scale tectonic movements occurred along the east-west trending Dauki thrust fault (Fig. 1) during the Pliocene and in the Himalaya and Indo-Burman Ranges, resulting in the formation of anticlinal folds and structural traps (Alam, 1989; Imam and Hussain, 2002).

The Surma basin, also known as the Sylhet trough, is located in NE Bangladesh (Fig. 1) and basin evolved from a pre-Oligocene passive continental margin to a foreland basin associated with the Indo-Burman and Himalayan fold belts. The basin is bounded by the Dauki Fault and Shillong Plateau to the north, the Indian platform to the west and the Chittagong-Tripura fold belt of the Indo-Burman Ranges to the east; it is separated from the Patuakhali depression by the Tangail-Tripura High to the south (Shamsuddin and Abdullah, 1997). The basin has a thick sedimentary fill (up to 12–16 km) comprising Upper Mesozoic to upper Cenozoic deposits (Johnson and Alam, 1991).

**Geology of the Surma basin**

Wells drilled in the Surma basin have not penetrated sedimentary strata older than Oligocene. At the base of the sedimentary succession (Fig. 2), the Paleocene–Early Eocene Tura Sandstone Formation has been encountered by wells in western Bangladesh and is composed of freshwater to mixed-marine sandstones and shales within coal seams. The formation is thought to be equivalent to the Jalangi Formation of West Bengal. During the Middle–Late Eocene, an extensive marine transgression over the shelf area resulted in deposition of the nummulitic Sylhet Limestone in the west and open-marine shale-rich facies in the east (Alam, 1989; Shamsuddin and Abdullah, 1997). The top of the Sylhet Limestone represents the deepest seismic reflector in the Surma basin (Hiller and Elahi, 1984). The overlying Eocene Kopili
Formation in the Surma basin passes gradually from open-marine up to estuarine facies (Banerji, 1981).

The overlying pre-Oligocene Disang Group can be divided into lower and upper units in the Naga Hills of India and the eastern Surma basin (Rao, 1983). The Sylhet Limestone and Kopili Shales are thought to be equivalent to the upper portion of the Disang Group (Shamsuddin and Abdullah, 1997).

Oligocene rocks of the Barail Group crop out on the eastern and northern margins of the Surma basin near the Dauki Fault zone and Oligocene rocks have been encountered in the subsurface by wells Rashidpur #2 and Atgram #IX (Johnson and Alam, 1991). Most of the stable shelf area was exposed during a marine regression during the Oligocene, and the Barail Group is interpreted as a marine to estuarine deposit in the foredeep area (Alam, 1989). Rocks of the Barail Group crop out over a wide area in the NE Surma basin having an off-lap relationship with the underlying Kopili Formation (Banerji, 1984). The Barail Group comprise sediments derived from the rising eastern Himalayas, which were deposited in environments ranging from fluvial to deltaic and pro-deltaic from north to SE (Johnson and Alam, 1991).

The subsidence rate of the basin during the Miocene increased markedly in response to loading caused by encroachment from the west of the Indo-Burman Ranges, associated with the eastward subduction of the Indian plate beneath the Burmese plate. The Surma Group was deposited during this time period and is composed of sediments mostly derived from the eastern Himalayas (Johnson and Alam, 1991). The top of the Surma Group, known as the “Upper Marine Shale”, provides a prominent seismic marker and stratigraphic reference level in the Surma basin, and represents the last marine incursion (Hiller and Elahi, 1984).

The Surma basin subsided rapidly during the Miocene–Plio-Pleistocene, resulting in the deposition of the fluvial Tipam and Dupi Tila Formations. South-directed overthrusting of the Shillong Plateau contributed to this subsidence. The absence of Pliocene and younger strata from the Shillong Plateau suggests that it formed an uplifted block during the Pliocene (Johnson and Alam, 1991).
SAMPLES AND METHODS

In total, 20 crude oil/condensate samples (Table 1) were collected from seven oil and gas fields in the Surma basin (Fig. 1): Beanibazar, Fenchuganj, Kailash Tila, Sylhet, Rashidpur, Habiganj and Titas. The four samples from the Titas gas field represent the southern margin of the basin.

All the samples were analysed by whole oil gas chromatography and gas chromatography/mass spectrometry (GC and GC/MS) using HP6890 and HP6890/HP5973 instruments equipped with a fused silica capillary column DB35MS (30 × 0.25 mm, J&W). The oven temperature of the GC was initially programmed to 40ºC and held isothermally for 4 minutes; it was then programmed to 300ºC at a rate of increase of 5ºC/min and held isothermally for 19 min. The oven temperature of the GC/MS was initially programmed to 40ºC and held isothermally for 5 minutes; subsequently, it was programmed to 300ºC at a rate of increase of 4ºC/min and held isothermally for 5 minutes. Helium was used as carrier gas for GC and GC/MS with a flow rate of 1.5 ml/min and 0.6 ml/min, respectively. Identification of individual compounds was performed using a mass spectra data library together with the retention times in published literature. Relative concentrations of \( n \)-alkane and polycyclic aromatic hydrocarbons (PAHs) were determined from peak areas of the whole oil flame ionization detector (FID) and total ion current (TIC) chromatograms by GC and GC/MS. Quantification of \( n \)-alkanes and PAHs was performed using chromatograms obtained by the whole oil analysis to avoid the evaporative loss of components in the laboratory. Saturate fractions were separated using silica gel (60, Merck) column chromatography and analysed by GC and GC/MS. The relative abundance of acyclic isoprenoids (pristane and phytane) and adamantanes were precisely calculated based on peak areas in the FID and TIC chromatograms. Considering the availability of published literature (e.g. Trolio et al., 1999; Mrkic et al., 2011), a DB5HT fused silica capillary column (30 × 0.25 mm, J&W) was also used for GC/MS analysis to identify the isomers of the PAHs and cyclic saturated hydrocarbons.

RESULTS AND DISCUSSION

Petroleum geochemical characteristics of the oils
Biodegradation
Among the 20 samples, three (Rashidpur #1, Habiganj #1, and Habiganj #3+4) were found to be severely biodegraded. In these three samples, \(n\)-alkanes, acyclic isoprenoids, naphthalenes, biphenyls and phenanthrenes were completely removed and a large unresolved complex mixture (UCM) was detected (Fig. 3). Diamondoids (mainly adamantane and alkyladamantanes) were detected as major compounds in these three samples (Fig. 3). The diamondoid hydrocarbons are thought to be generated by the rearrangement of polycyclic hydrocarbons under thermal stress (Wingert, 1992). They are resistant to further thermal stress and biological degradation compared to other petroleum components such as straight-chain and branched alkanes (Wingert, 1992).

The compositional characteristics of these biodegraded oils represent level 3–5 on the biodegradation scale (Grice et al., 2000; Wenger et al., 2002). These severely biodegraded oils were derived from shallow reservoirs less than 1500 m deep (Table 1).

Traces of \(n\)-alkanes (C\(_7\)–C\(_{11}\)) and lower molecular weight aromatic hydrocarbons (toluene and xylenes) were detected in samples Beanibazar #1 + 2 (light), probably due to the effects of water washing and biodegradation. Except for these four biodegraded oil samples, the other 16 samples were considered to be non-biodegraded oils.

Source rock organic matter
The distribution of \(n\)-alkanes in oils from the Surma basin in general range from C\(_7\)–C\(_{35}\) with maxima at around C\(_{10}\)–C\(_{13}\) (Table 2, Figs. 4 and 5). Variations in \(n\)-alkane distribution were observed, particularly in Fenchuganj #2 (3068.5–3083.5 m) and Sylhet #7 (2020–2033 m). These two oils are waxy at room temperature and rich in long-chain (>C\(_{20}\)) \(n\)-alkanes, with maxima at around C\(_{25}\)–C\(_{27}\) and C\(_{22}\) in Fenchuganj #2 and Sylhet #7, respectively, suggesting a higher land plant origin (Tegelaar et al., 1989). Oils from Beanibazar #1+2 (heavy) well were also rich in long-chain \(n\)-alkanes.
All the oils and condensates contained abundant aromatic hydrocarbons such as benzene, toluene, xylenes, naphthalenes, cadalene, biphenyls and phenanthrenes. Biphenyls are bicyclic aromatic hydrocarbons, probably generated from a phenol coupling reaction (Alexander et al., 1994a; Trolio et al., 1996). Alkylbiphenyl distribution in the studied samples were very similar, with 3-methylbiphenyl being highly abundant compared to 4-methylbiphenyl (Fig. 6). Biphenyls are often detected in coal extracts (Mair and Mayer, 1964; Yew and Mair, 1966; White and Lee, 1980), suggesting that the crude oils and condensates in the Surma basin may have originated from organic matter derived from coal or coaly shale. However Cumbers et al. (1987) identified alkylbiphenyls in extracts from Middle Cambrian sedimentary rocks, demonstrating that biphenyls are not exclusively derived from higher plants.

Naphthalene (N) and alkynaphthalenes such as methylnaphthalenes (MNs), dimethylnaphthalenes (DMNs) and trimethylnaphthalenes (TMNs) were detected in all the samples (Fig. 7). 2-MN was abundant and decreased in the order MNs>DMNs>N>TMNs, except in oils from Fenchuganj #2 (3068.5–3083.5 m), Sylhet #7 (2020–2033 m) and Beanibazar #1+2 (heavy), where a decreasing order DMNs>MNs>N>TMNs or DMNs>MNs>TMNs>N was observed. The methylated naphthalene distributions may be influenced by the effects of source, thermal stress and biodegradation (Van Aarssen et al., 1999). Diversity in the relative distribution of naphthalene and alkylated naphthalenes was observed. Marine oils tend to be rich in tetramethylnaphthalenes (TeMNs) compared to MNs or N (TMNs>DMNs>TeMNs>MNs>N), whereas terrestrial oils are poor in TeMNs compared to MNs or N (DMNs>MNs>TMNs>N>TeMNs) (Zhang et al., 2010).

Alkynaphthalenes in all the samples in the present study tended to be rich in DMNs and MNs compared to TMNs, similar to the alkynaphthalene distribution in terrestrial oils described by Zhang et al. (2010). 1,2,5-TMN was detected at trace levels or was not detected. In contrast, 1,2,7-TMN was detected in all the study samples, although it was not fully separated from other compounds; it is thought to co-elute with 1,6,7-TMN and 1,2,6-TMN (Mrkic et al., 2011). Strachan et al. (1988) proposed that 1,2,7-TMN is derived directly from the structural degradation of an oleanane-type triterpenoid, which is a typical angiosperm biomarker. They reported increased relative abundances of 1,2,5-TMN and 1,2,7-TMN in immature Cretaceous and
younger sediments associated with type III organic matter, and the relative abundance of these two isomers decreased with increasing maturity. In contrast, sediments older than Cretaceous with type III kerogen contain a higher relative abundance of 1,2,5-TMN and lower relative abundance of 1,2,7-TMN, suggesting a relationship to angiosperm evolution since the Early Cretaceous.

The presence of 1,2,7-TMN in all the oil samples suggests that a part of the source organic matter was derived from higher land plants. A very low concentration or absence of 1,2,5-TMN might be due to the higher maturity level of the oil samples. Only the oils from Titas field contained a detectable amount of 1,2,5-TMN (Fig. 7). Titas oils were also rich in cadalene compared to the other oils, indicating a significant contribution by vascular land plants to the source rock (Bendoraitis, 1974; Radke et al., 1984; Simoneit, 1986). However, apart from the Titas oils, the other oil samples are comparatively poor in cadalene. This could be due to the higher maturity level of the oils in the north to central Surma basin compared to the Titas oils, because cadalene can be thermally decomposed at higher maturities as suggested by Alexander et al. (1994b). By contrast, abundant bicadinanes in oils and condensates from the Surma basin suggest a significant input of vascular land plants to the source rocks (Alam and Pearson, 1990; Curiale et al., 2002; Takeda et al., 2011)

**Depositional environment**

The relative abundance of pristane and phytane can provide useful information on the palaeo-depositional environment of the source rock and the type of source organic matter. Pr/ph and pr/nC\textsubscript{17} ratios of oils from the Surma basin ranged between 5.3–11.9 and 0.8–2.1, respectively (Table 1), suggesting a terrestrial depositional environment. On a cross plot of pr/nC\textsubscript{17} versus ph/nC\textsubscript{18} (Fig. 8), all the oil samples plot within the field corresponding to an oxic environment, and organic matter derived from humic Type III kerogen. The Titas oils, however, plot closer to the field indicating a mixed kerogen type (Type II+III), suggesting a possible contribution of algal organic matter to the source rock for these oils.

**Maturity of the oils**
The maturity levels of the crude oils and condensates in the Surma basin can be assessed by the calculated vitrinite reflectance (Rc) estimated by the methylnaphthalene ratio (MNR), trimethylnaphthalene ratio-2 (TNR-2) (Radke et al., 1984, 1994), methylphenanthrene ratio (MPR) and methylphenanthrene index-1 (MPI-1) (Radke et al., 1984). Calculated Rc values based on MNR, TNR-2, MPR and MPI-1 ranged from 1.0–1.3%, 0.8–1.0%, 1.1–1.3%, and 0.9–1.3%, respectively (Table 3). The maturity indices MPR and MPI-1 could not be determined for the biodegraded oils and condensates because they are very poor in alkylphenanthrenes. By contrast, MNR and TNR-2 were determined for most of the oils and condensates except for the biodegraded oils. Both Rc values calculated from MNR and TNR-2 varied consistently, although the Rc value from MNR generally tended to be 0.2 to 0.3% higher than the Rc value from TNR-2. The differences in Rc values determined from MNR and TNR-2 could be due to the rapid rate of temperature increase in the Surma basin. The rate of temperature increase since the Pliocene is estimated to be approximately 15 to 20°C/million years based on the stratigraphic age of Miocene-Pliocene boundary and the bottom-hole temperatures (95 to 121°C) of boreholes Beanibazar #1X, #2 and Atgram #1X in the Surma basin. Among the investigated oil samples, Rc values from alkylnaphthalene indices for the Titas oils were significantly lower than those from the Beanibazar, Fenchuganj, Kailash Tila, and Sylhet wells in the north to central Surma basin (Table 3). The Rc values from MNR of the Titas oils were about 1.0%, whereas the Rc values of the other oils were about 1.2–1.3% (Table 3). Rc values from TNR-2 of Titas oils were about 0.8%, also significantly lower than the Rc values from TNR-2 of the other oils (about 0.95–1.0%).

**Diamondoids in the severely biodegraded oils**

High concentrations of diamondoid hydrocarbons were detected in the severely biodegraded condensates, and adamantane and alkyladamantanes were present as major compounds (Fig. 3). The distributions of adamantane and alkyladamantanes in both biodegraded and non-biodegraded oils from the Surma basin were similar (Fig. 9), suggesting that all the crude oils/condensates in the study area originated from a similar type of source rocks rich in terrigenous organic matter. According to Chen et al. (1996), the relationship between the methyladamantane index (MAI%) and the Rc in non-biodegraded hydrocarbons can be used to evaluate the maturity levels of
severely biodegraded oils. The MAI (%) of the severely biodegraded oils was also almost identical to those of the non-biodegraded oils, demonstrating that both the biodegraded and non-biodegraded oils are at a similar maturity level (Fig. 10).

**Possible source rocks and timing of oil expulsion**

The Atgram #1X well, the deepest well (4961 m) drilled so far in Bangladesh, is located in the NE of the Surma basin (Fig. 1). An approximately 260 m thick section of the Oligocene Jenam Shale was drilled at this location (Hiller and Elahi, 1984). The TOC content of mudstones from the Jenam Shale (mean Ro = ca. 0.65%) ranged from 0.6 to 2.4%, and that of the overlying Miocene Bhuban Shale (mean Ro = ca. 0.5) was 0.2–0.7% (Shamsuddin and Abdullah, 1997). The comparatively low maturity level of the potential source rocks in the Jenam and Bhuban Formations may be because the wells were drilled on an anticlinal structure, or because of the basin margin location. Thus source rocks in the adjacent synclinal area may be more mature.

Abundant organic matter derived from higher land plants (dominated by angiosperms) was deposited during the Middle Eocene–Early Miocene, during deposition of the thick sedimentary sequences of the Jaintia and Barail Groups (Hossain et al., 2009). TOC content (0.15 to 0.9%), Rock-Eval pyrolysis data (S1 and S2) and the maturity level of the Miocene rocks (Ro = ca. 0.45–0.76%) (Shamsuddin and Khan, 1991; Farhaduzzaman et al., 2012) suggest the lower generation potential and lower maturity of the Middle Miocene–Pleistocene Surma, Tipam and Dupi Tila Groups.

Hiller and Elahi (1984) suggested a geothermal gradient ranging from 1.8 to 2.2°C/100 m and non-balanced heat flows in the Surma basin, because of the very young and rapid burial history. The maximum Ro in the Early Miocene Bhuban Formation in Beanibazar #1X was approximately 0.58% at a depth close to the total depth (TD) at 4109 m. Ro increased to 0.7% in the Middle Oligocene Jenam Formation in well Atgram #1X at a depth close to the TD at 4961 m, near the threshold of the oil window. In addition, an Ro value of 0.73% at 3810 m in the Lower Bhuban Formation in well Atgram #1X was recorded, but is considered to be due to the reworking of older vitrinite (Hiller and Elahi, 1984). However a relatively constant Ro of approximately 0.45–0.65% has been observed in the Surma and Barail
Groups in the Surma basin (Shamsuddin and Khan, 1991; Shamsuddin and Abdullah, 1997). This suggests a similar thermal maturity level for rocks in the drilled section in the Surma basin, or the reworking of vitrinite macerals with similar Ro values. The calculated vitrinite reflectance values of the Titas oils (Rc=0.8–1.0%) and other oils from the Surma basin (Rc=1.0–1.3%) were significantly higher than those of the Oligocene and Miocene rocks.

Major reservoir rocks are present in the Bhuban and Bokabil Formations of the Miocene–Early Pliocene Surma Group. Considering the lower maturity level of the sedimentary rocks in the Surma Group, the maturity levels as indicated by the Rc values of the oils are thought to show the timing of oil expulsion from the source rocks. On the basis of the time–temperature and Ro relationship (Suzuki et al., 1993), the geothermal gradient of 1.8–2.2°C/100 m in the Surma basin (Hiller and Elahi, 1984; Ismail and Shamsuddin, 1991), and the estimated Rc values of the oils and condensates (0.8–1.3%), the burial depth of the source rock at the time of hydrocarbon expulsion was estimated to be 5000–6000 m for the Titas oils and 6000–7000 m for the other oils in the north to central Surma basin.

The Oligocene Barail Group were reported at depths of 4700–4961 m and 4010–4595 m in the Atgram #1X and Rashidpur #2 wells, respectively (Johnson and Alam, 1991). Sedimentary strata older than the Oligocene Barail Group have not yet been drilled in the Surma basin. In the NW part of the basin, the cumulative thickness of the Late Eocene Kopili Formation and the Oligocene Barail Group was estimated to be about 1400 m, although it varies within the basin (Johnson and Alam, 1991). According to a study of fossil assemblages by Banerji (1984), the depositional environment of the lower and upper divisions of the Kopili Shale Formation in Meghalaya (northern Surma basin) gradually changed from an open-marine to brackish and almost freshwater estuarine environment. In contrast, the Jenam Formation of the Barail Group was deposited in a fluvio-deltaic environment (Johnson and Alam, 1991). The TOC of the Middle Oligocene Jenam Shale is 0.6–2.7% (Shamsuddin and Abdullah, 1997), suggesting significant source rock potential. However, the mean Ro value of the Middle Oligocene Jenam Shale is about 0.65% (Shamsuddin and Abdullah, 1997), which is lower than the estimated maturity levels of Rc=0.8–1.0% for the Titas oils and Rc=1.0–1.3% for the other oils in the north to central Surma basin. Although the
geochemical data-set for the Oligocene and older sedimentary sequences is limited, the Jenam Shale in the depocentre and/or older sedimentary rocks rich in land-derived organic matter are interpreted to be source rocks for the oils and condensates in the Surma basin.

**Evaporative fractionation of the oils**

Source and maturity parameters of oils and condensates from the Surma basin suggest that all the oils (except the Titas oils) were generated from a similar source rock with significant amounts of terrigenous organic matter. The Titas oils are inferred to be derived from terrigenous source rocks with a significant contribution of marine organic matter. The maturity of the Titas oils is significantly lower than that of the oils from the north to central Surma basin. Except for the Titas samples, the oils show limited maturity levels as indicated by Rc values ranging from 1.0 to 1.3%. However, a significant diversity in the distribution of \( n \)-alkanes has been observed in the oils from the Surma basin (Figs. 4 and 5.).

The \( n \)-alkane distribution in two oil samples, from 2578.6–2581.6 m and 2768–2781 m depths, in well Fenchuganj #2 ranged from \( C_7 \)–\( C_{27} \) with a maximum at around \( C_{10} \)–\( C_{11} \). Deeper oils from the same well, from 3068.5–3072.5 m and 3074.5–3083.5 m, showed an \( n \)-alkane distribution ranging from \( C_7 \)–\( C_{35} \) with a maximum at around \( C_{25} \)–\( C_{27} \) (Fig. 4). The latter oil samples are waxy at room temperature and are dark yellow in colour. A similar variation in the distribution of \( n \)-alkanes was observed in two oils from well Sylhet #7 (Fig. 4). In this well, oils from 1874–1886 m and 1901–1908 m showed \( n \)-alkane distributions in the range of \( C_7 \)–\( C_{22} \) with a maximum at \( C_{10} \). Oil from a depth of 2020–2033 m contains \( n \)-alkanes ranging from \( C_7 \)–\( C_{35} \) with a maximum at around \( C_{17} \)–\( C_{22} \); this oil is waxy at room temperature and has a dark yellow colour.

Six oil samples from Kailash Tila gas field were characterized by a similar \( n \)-alkane distribution ranging from \( C_7 \)–\( C_{28} \) with a maximum at \( C_{10} \) (Fig. 5), and varied from colourless to light yellow. Four samples from Titas also showed a similar \( n \)-alkane distribution ranging from \( C_7 \)–\( C_{25} \) with a maximum at around \( C_{11} \)–\( C_{13} \) (Fig. 5) and likewise varied from colourless to light yellow. The colourless oil sample from
Beanibazar #1+2 (heavy) contained $n$-alkanes ranging from $C_7$–$C_{31}$ with a maximum at $C_{10}$ (Fig. 5).

API gravities of the studied oils were 26.2 to 48.3°, except for two waxy oils from Fenchuganj #2 and Sylhet #7 (Takeda et al., 2011). API gravities of waxy oils from 3068.5–3072.5 m and 3074.5–3083.5 m depths in Fenchuganj #2 and 2020–2033 m in Sylhet #7 were reported to be 17.2° and 28.3°, respectively (Ahmed et al., 1991).

These variations, particularly in the oils from the north to central Surma basin, cannot be interpreted to be due to differences in source rock type and/or maturity levels, because the oils are derived from similar terrigenous source rocks and show similar maturity levels. After the original accumulation of oil in a reservoir, the oil composition can be modified as a result of processes such as thermal degradation, deasphaltening, water washing and tertiary migration. As detailed above, waxy oils are generally located at greater depths than lighter condensates. This difference in distribution may suggest that evaporative fractionation of the reservoired oil has occurred, followed by upward migration of lighter hydrocarbons.

Lighter hydrocarbons may be selectively vaporized and expelled from a reservoir rock due to changes in reservoir temperature and pressure resulting from tectonic activity. These lighter hydrocarbons may migrate to a neighboring reservoir to form a gas-condensate accumulation. Residual oil remaining in the reservoir will then become heavier and waxy. Thompson (1987) demonstrated evaporative fractionation in laboratory experiments which investigated the nature of the products (condensates and residual oil) and the original oil. The experiments showed that progressive evaporation can result in oils which are lighter, more paraffinic and less aromatic than the original oils. In addition, the depletion of lower molecular weight $n$-alkanes and a change in the dominant $n$-alkanes from lighter to heavier in the residual oil were observed. Thompson (1987) proposed a plot of aromaticity (toluene/$nC_7$) versus paraffinicity ($nC_7$/methylcyclohexane) to illustrate the progressive evaporative fractionation process.

The aromaticity and paraffinicity of the oil samples from the Surma basin were determined, based on the relative abundances of toluene, methylcyclohexane and $nC_7$. 

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and aromaticity was plotted versus paraffinicity (Fig. 11). Waxy oils from wells Fenchuganj #2 and Sylhet #7 were characterized by higher aromaticity compared to condensates which had accumulated in reservoir units overlying the waxy oil reservoir. Condensates from Beanibazar #1+2, which have the lowest API gravity (48.3°), showed the lowest aromaticity and highest paraffinicity among all the oil samples. Oils from Kailash Tila and Titas plotted between the waxy oils and the condensates from Beanibazar #1+2. The significantly different aromaticity and paraffinicity observed in similar terrigenous oils with similar maturities strongly suggest that evaporative fractionation has taken place following initial oil accumulation in the reservoir. Tertiary migration and re-accumulation of lighter hydrocarbons is expected to have taken place during and after the formation of traps and anticlinal folds in the Pliocene.

The API gravity of oils in the Surma basin increases with decreasing reservoir depth, corresponding closely to changes in the aromaticity and paraffinicity of the oils. This trend was especially evident in the Fenchuganj #2 and Sylhet #7 wells.

The Tipam and Dupi Tila Formations are characterized by thick fluvial to deltaic sedimentary successions deposited during rapid subsidence of the Surma basin due to overthrusting of the Shillong Plateau during the Pliocene (Johnson and Alam, 1991). Large-scale tectonic movements along the Dauki Fault and in the Himalaya and Indo-Burman Ranges occurred during the Pliocene, resulting in the formation of the present-day basin architecture (Alam, 1989; Imam and Hussain, 2002). Structural dislocation of pre-existing reservoirs occurred in the Surma basin as a result of this Pliocene tectonism, and evaporative fractionation, tertiary migration and re-accumulation of hydrocarbons resulted in the compositional diversity of petroleum which is observed. The occurrence of many oil and gas seepages in the Surma basin suggests that tertiary migration continues at the present day.

CONCLUSIONS

Twenty oils and condensates from seven gas fields in the Surma basin, Bangladesh, were analysed and were found to have highly diverse \(n\)-alkane distributions. The relative abundance of pristane, phytane and adjacent \(n\)-alkanes and abundant
biphenyls, cadalene, and bicanadines suggested that most of the oils and condensates were derived from source rocks rich in terrigenous organic matter. A cross-plot of pr/nC$_{17}$ versus ph/nC$_{18}$ for oils from the Titas field, however, suggested a possible contribution of algal organic matter. Maturities of the oils and condensates were estimated based on the distributions of alkynaphthalenes and alkylphenanthrenes, and showed that oils can be classified into two groups. Oils from wells Beanibazar, Fenchuganj, Kailash Tila and Sylhet in the north to central Surma basin are characterized by relatively high Rc values (1.0–1.3%), whereas Rc values of oils from the Titas well in the southern margin are 0.8–1.0%. Oil samples from the Rashidpur and Habiganj wells were severely biodegraded. However, diamondoid hydrocarbons were identified in both the biodegraded oils and non-biodegraded samples. The distribution of adamantanes and the methyladamantane index of the severely biodegraded oils were almost identical to those of non-biodegraded oils, indicating a similar type of source rocks and similar maturity levels of all the oils in the north to central Surma basin.

Taking into account the time–temperature history of the Surma basin and the estimated maturity levels of the oils and condensates, the burial depth of the source rocks at the major stage of hydrocarbon expulsion is estimated to be between 5000–6000 m for the Titas oils and 6000–7000 m for the other oils in the Surma basin. Although the geochemical data set for the Oligocene and older sedimentary sequences is limited, the Oligocene Jenam Shale in the depocentre and/or older sedimentary rocks rich in terrigenous organic matter are identified as possible source rocks. The aromaticity and paraffinicity of the oils, as evaluated by toluene, methylcyclohexane and nC$_7$, indicated progressive subsurface evaporative fractionation after initial accumulation of oil in the reservoir. This resulted in separate accumulations of relatively light condensates and heavier waxy oils, the latter in underlying reservoir units relative to the former. Tertiary migration is interpreted to have occurred during and after the Pliocene, when pre-existing reservoirs were dislocated by large-scale tectonic activity.

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REFERENCES


Table Captions

Table 1. The general geochemical characteristics of the crude oils and condensates. T, toluene; MCH, methylcyclohexane; Pr, pristane; Ph, phytane.

Table 2. Relative abundance (%) of n-alkane calculated from whole oil FID chromatograms.

Table 3. Maturity parameter estimated from aromatic hydrocarbons distribution. MNR, methylnaphthalene ratio; TNR, trimethylnaphthalene ratio; MPR, methylphenanthrene ratio; MPI, methylphenanthrene index; Rc, calculated vitrinite reflectance; MAI, methyladamantane index.

MNR = (2-MN/1-MN), Radke et al. (1982b), Rc (%) from MNR = (0.17 X MNR + 0.82); TNR-2 = (2,3,6 + 1,3,7-TMN) / (1,4,6 + 1,3,5 + 1,3,6-TMN), Radke et al. (1986); Rc (%) from TNR-2 = {0.4 + (0.6 x TNR-2)}; MPR = (2-MP/1-MP), Radke et al. (1982b), Rc (%) from MPR = (0.99 x log MPR + 0.94); MPI-1 = 1.5 [2-MP + 3-MP] / [P + 9-MP + 1-MP], Radke et al. (1982a), Rc (%) from MPI-1 = (0.6 x MPI-1 + 0.4) for Ro < 1.35; MAI (%) = 1-MA / (1-MA + 2-MA) (%), Chen et al. (1996).
Figure Captions

Fig. 1. Distribution of oil and gas deposits in the Surma basin and major tectonic elements of Bangladesh and surrounding areas modified after Banerji, 1981 and Uddin and Lundberg, 2004.

Fig. 2. Generalized Stratigraphic succession of the Surma basin, Bangladesh with schematic lithology (not scaled), modified after Hiller and Elahi, 1984 and Alam et al., 2003.

Fig. 3. Whole oil TIC chromatograms of the severely biodegraded oils (Habiganj #1; Habiganj #3+4 and Rashidpur #1). Identification of important peaks: 1, adamantane; 2, 1-methyladamantane; 3, 1,3-dimethyladamantane; 4, 1,3,5-trimethyladamantane; 5, 2-methyladamantane; 6, 1,4-dimethyladamantane (cis + trans); 7, 1,3,6-trimethyladamantane; 8, 1,2-dimethyladamantane; 9, 1,3,4-trimethyladamantane (cis + trans).

Fig. 4. Whole oil FID chromatograms of Fenchuganj #2 (2578.6 – 2581 m), Fenchuganj #2 (3068.5 – 3072.5 m, 3074.5 – 3083.5 m), Sylhet #7 (1874 – 1886 m, 1901–1908 m) and Sylhet #7 (2020 – 2033 m). Identification of important peaks: 1, methylcyclohexane; 2, toluene; 3, ethylbenzene; 4, meta + para-xylene; 5, ortho-xylene; 6 + 7, trimethylbenzene; 8, naphthalene; 9, 2-methylnaphthalene; 10, 1-methylnaphthalene; 11, biphenyl; 12, pristane; 13, phytane; 15, phenanthrene.

Fig. 5. Whole oil FID chromatograms of Beanibazar #1 + 2 (heavy), Titas #1, Kailash Tila #2 (2927 – 2930 m). Identification of important peaks: 1, methylcyclohexane; 2, toluene; 3, ethylbenzene; 4, meta + para-xylene; 5, ortho-xylene; 6 + 7, trimethylbenzene; 8, naphthalene; 9, 2-methylnaphthalene; 10, 1-methylnaphthalene; 11, biphenyl; 12, pristane; 13, phytane; 14, cadalene; 15, phenanthrene.

Fig. 6. Biphenyl and alkylbiphenyl distribution. BP, biphenyl; MBP, methylbiphenyl.
Fig. 7. Naphthalene and alkynaphthalene distribution. N, naphthalene; MN, methylnaphthalene; EN, ethylnaphthalene; 1, 2,6 + 2,7-dimethylnaphthalene (DMN); 2, 1,3 + 1,7-DMN; 3, 1,6-DMN; 4, 1,4 + 2,3-DMN; 5, 1,5-DMN; 6, 1,2-DMN; 7, 1,3,7-trimethylnaphthalene (TMN); 8, 1,3,6- TMN; 9, 1,3,5 + 1,4,6- TMN; 10, 2,3,6- TMN; 11, 1,2,7 + 1,6,7 + 1,2,6- TMN; 12, 1,2,5- TMN.

Fig. 8. Cross plot of pristane/nC\textsubscript{17} vs phytane/nC\textsubscript{18} for evaluation of the source rock depositional environment.

Fig. 9. Adamantane and alkyladamantane distribution of the severely biodegraded and non-biodegraded oils. A, adamantane; MA, methyladamantane; EA, ethyladamantane.

Fig. 10. Cross plot of methyladamantan index (MAI) (%) vs. calculated vitrinite reflectance (Rc) (%) from the distribution of trimethylnaphthalenes (TNR-2).

Fig. 11. Aromaticity (Toluene/nC\textsubscript{7}) vs. paraffinicity (nC\textsubscript{7}/Methylcyclohexane) of oils from the Surma basin. Evaporative fractionation after the accumulation of oils resulted in the formation of diverse oils, as shown by the whole oil chromatograms.
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* Probable colcued with other compound
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<th>MPR</th>
<th>$R_e$ (%) From MPR</th>
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Fig. 1
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<th>Lithology</th>
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<td>Holocene</td>
<td>Dihing</td>
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<td>Relatively thin subaerial local deposits&lt;sup&gt;b&lt;/sup&gt;.</td>
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<tr>
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<td>Upper Dugi Tila</td>
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<td>Probably braided, fluvial system to meandering river deposits&lt;sup&gt;a&lt;/sup&gt;, lake-river plain&lt;sup&gt;c&lt;/sup&gt;; fluvial-deltaic&lt;sup&gt;d&lt;/sup&gt;, continental-fluvietic&lt;sup&gt;e&lt;/sup&gt;.</td>
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<td>Lower Dugi Tila</td>
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<td>Late Pliocene</td>
<td>Dupi Tila</td>
<td>Girujan Clay</td>
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<td>Pro-delta and delta-front deposits of large mud-rich delta system&lt;sup&gt;a&lt;/sup&gt;; fluvial-deltaic to estuarine&lt;sup&gt;d&lt;/sup&gt;; deltaic to shallow marine&lt;sup&gt;e&lt;/sup&gt;.</td>
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<td>Tipam Sandstone</td>
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<td>Mid-Pliocene</td>
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<td>Fluvial, delta to pro-delta deposits&lt;sup&gt;a&lt;/sup&gt;; marine to estuarine&lt;sup&gt;e&lt;/sup&gt;; deltaic with marine incursions&lt;sup&gt;d&lt;/sup&gt;.</td>
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<td>Early Pliocene-Miocene</td>
<td>Surma</td>
<td>Kopili Shale</td>
<td></td>
<td>Transition of basinal to shallow-marine&lt;sup&gt;a&lt;/sup&gt;; shallow-marine to marine, shallow-marine to carbonate and deltaic to slope deposition (from bottom upwards)&lt;sup&gt;b&lt;/sup&gt;; deep basinal&lt;sup&gt;d&lt;/sup&gt;; marine&lt;sup&gt;e&lt;/sup&gt;.</td>
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<td>Jaintia</td>
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<td>Pre-Paleocene</td>
<td>Undifferentiated sedimentary rocks on the continental basement complex.</td>
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Sand, silt, clay; Clay; Shale; Sandstone; Coal; Limestone; Unconformity; UMS- Upper Marine Shale.

<sup>a</sup>- Johnson and Alam, 1991; <sup>b</sup>- Alam et al., 2003; <sup>c</sup>- Imam, 2005; <sup>d</sup>- Curiale et al., 2002; <sup>e</sup>- Alam, 1989.
Fig. 3

Habiganj #1

Habiganj #3+4

Rashidpur #1
Fig. 6

**Titas #1**
(2617-2769m)

m/z 154+168

**Fenchuganj #2**
(2578.6-2581.6m)

m/z 154+168

**Sylhet #7**
(1874-1886, 1901-1908m)

m/z 154+168

**Kailash Tila #2**
(2927-2930m)

m/z 154+168

Retention Time
Fig. 7

Titas #1 (2617-2769m)

Beanibazar #1+2 (heavy)

m/z 128+142+156+170

Retention Time
Fig. 8

- **Beanibazar- 1+2 (heavy)**
- **Fenchuganj- 2**
- **Titas- 1, 5,8 & 10**
- **Kailash Tila- 2, 2+6 & 4**
- **Sylhet-7**
- **Yufutsu-OKi B**
- **Akebono-SK-1**

Graph showing the relationship between Pristane/nC_{17} and Phytane/nC_{18} with different regions indicating Terrigenous Type III, Mixed Type II/III, and Marine Algal Type II.
Fig. 9

Habiganj #1
(1417-1428 m)
Biodegraded oil

1-MA

m/z 135+136

2-MA

2-EA

1-EA

Rashidpur #1
(1401-1445 m)
Biodegraded oil

1-MA

m/z 135+136

2-MA

2-EA

1-EA

Sylhet #7
(1874-1886, 1901-1908 m)
Non-biodegraded oil

1-MA

m/z 135+136

2-MA

2-EA

1-EA

Titas #1
(2617-2769 m)
Non-biodegraded oil

1-MA

m/z 135+136

2-MA

2-EA

1-EA

Retention Time