

OCCURRENCE OF PENTA-AROMATIC HYDROCARBONS IN JAINTIA AND BARAIL GROUP MUDSTONES, NORTHEASTERN BENGAL BASIN, BANGLADESH

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Abstract: In the present experiment, concentration, distribution and sources of penta-aromatic hydrocarbons in seven drill core and outcrop samples from Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh have been studied. Gas chromatography-mass spectrometry (GC-MS) was used to obtain composition details about the sedimentary organic matter (OM). Mudstone samples were found to contain relatively high penta-aromatic hydrocarbon abundances in the lower Jaintia Group than in the overlying Barail Group. High concentration of perylene suggests terrigenous sources and significantly higher content of total organic carbon in the samples. A biogenic origin of perylene therefore indicates oxygen deficient environmental condition for deposition of OM. Perylene over pentacyclic aromatic hydrocarbon isomers regulating *in-situ* diagenetic origin.

Key words: Mudstone, penta-aromatic hydrocarbons, GC-MS, Bengal Basin, Bangladesh

সারাংশ: বর্তমান গবেষণায় উত্তরপূর্ব বেঙ্গল বেসিনের জয়ন্তিয়া এবং বরাইল গ্রুপের কদম শিলায় পেটা-অ্যারোম্যাটিক হাইড্রোকার্বন বৈশিষ্ট্য নিয়ে আলোচনা করা হয়। গবেষণায় দেখা যায় যে, বরাইল গ্রুপের কদম শিলায় চাইতে জয়ন্তিয়া কদম শিলায় পেটা-অ্যারোম্যাটিক হাইড্রোকার্বনের পরিমাণ বেশি। এসব পললে বিদ্যমান বেশি পরিমাণ পেরিলিন মহাদেশীয় উৎস অঞ্চল এবং অপেক্ষাকৃত কম অক্সিজেন সম্পন্ন পরিবেশের ইংগিত প্রদান করে। তবে পেটাসাইক্লিক অ্যারোম্যাটিক হাইড্রোকার্বন আইসোমারের তুলনায় বেশি পরিমাণ পেরিলিন অবক্ষেপণ পরবর্তী স্থানীয় পরিবর্তন নির্দেশ করে।

Introduction

Polycyclic aromatic hydrocarbons (PAHs) in sediments and sedimentary rocks have been studied by many authors (Aizenshtat, 1973; Wakeham et al., 1979, 1980; Jiang et al., 1998, 2000; Grimalt et al., 2004; Grice et al., 2007; Hossain et al., 2013a). PAHs are widely distributed in soils (Youngblood and Blumer, 1975; Blumer and Youngblood, 1975), lake sediments (Laflamme and Hites, 1978; Giger and Schaffner, 1978; Wakeham et al., 1980; Gschwend and Hites, 1981; Kawamura et al., 1987), recent marine sediments (Aizenshtat, 1973; Youngblood and Blumer, 1975; Hites et al., 1977; Wakeham et al., 1979; Gschwend and Hites, 1981), river particulates as well as estuary sediments (Giger and Schaffner, 1978; Hites et al., 1980; Brown and Maher, 1992), and peats or coals (Aizenshtat, 1973; Venkatesan, 1988; Garrigues et al., 1988; Hossain et al., 2013b).

Penta-aromatic hydrocarbons especially benzo [*b,j,k*] fluoranthene (Bflas), benzo [*e*] pyrene (BePy) and benzo [*a*] pyrene (BaPy) are common in recent and ancient sediments (Baumard et al., 1998; Jiang et al., 1998, 2000; Readman et al., 2002; Bakhtiari et al., 2009). This hydrocarbon has been originated from biological precursors through diagenesis in continental and marine OM (Aizenshtat, 1973; Laflamme and Hites, 1978; Hites et al., 1980; Tan and Heit, 1981; Tan

et al., 1996; Jiang et al., 1998; Grice et al., 2009). Perylene is a penta-aromatic hydrocarbon being derived from aquatic and continental organic matter in anoxic environments (Aizenshtat, 1973; Wakeham et al., 1979; Silliman et al., 2000; Suzuki et al., 2010).

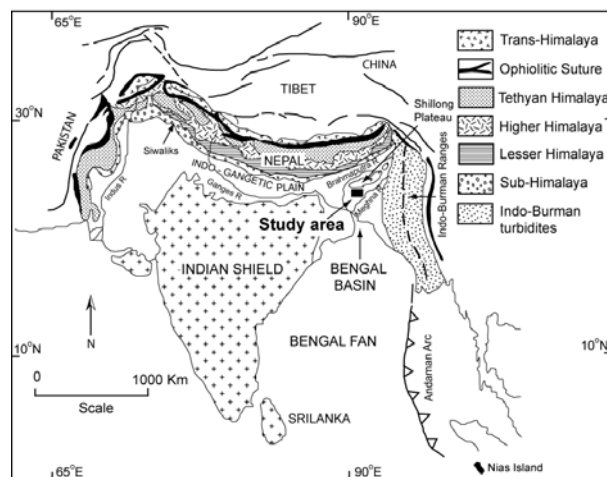


Fig. 1 Geological map showing location of the sample sites and major geographic features of the Himalayas (after Hossain et al., 2010).

The Bengal Basin is placed between the Indian Shield Platform and the Indo-Burman Ranges. It is bounded to the north by Shillong Plateau and plunges into the Bay

of Bengal to the south (Fig. 1). The Sylhet basin is located immediately after the Shillong Plateau (Fig. 1). This basin is an elongated trough consists mainly of alternating bed of sandstone and mudstone (Alam et al., 2003). The present study groups in the Sylhet succession are mainly Jaintia and Barail which is located in the lower part of Sylhet succession, northeastern Bengal Basin, Bangladesh (Fig. 2). Biomarker studies of the Bengal Basin mudstones are very few (Alam and Pearson, 1993; Hossain et al., 2009a, b, 2013a). The aim of this study was to recognize distribution and sources of penta-aromatic hydrocarbons in Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh.

Geological setting

The Bengal Basin was formed during the collision between India, Eurasia and Myanmar (former Burma), beginning at about 45 Ma (Gansser, 1964) building the Himalayan orogen and Indo-Burman ranges (Uddin and Lundberg, 1998). Based on the results of geophysical investigations, geological mapping, and on well data, Reimann (1993) classified the principal structural elements of the Bengal Basin and of the adjacent areas into a number of environments. These are the Fold Belt, Bengal Foredeep, Continental Slope, Western Foreland Shelf, Dauki Fault Zone, Rangpur Saddle, and the Himalayan Foredeep. The sediment fill of the Bengal Basin comprises about 22 km of Paleocene to Holocene deposits (Alam et al., 2003). The Basin is bounded to the east and west by Indo-Burman ranges and West Bengal. North is bounded by Shillong Plateau and to the south it plunges into the Bay of Bengal (Fig. 1). The Bengal delta, world's largest delta, has been formed through the seaward prograding deltas of the Ganges, Brahmaputra and Meghna rivers system (Einsele et al., 1996; Alam et al., 2003).

The Sylhet basin, a sub-basin in the northeastern Bengal Basin, consists of a thick Tertiary sedimentary succession, comprising sandstones, shales, fossiliferous limestones, deposited on the passive continental margin. Sediments of the Sylhet basin were derived from Himalayan detritus (Hossain et al., 2010). Alam et al. (2003) reported the Sylhet basin is tectonically placed in the central deep part of the Bengal Basin. The Sylhet basin is situated in the Bengal Foredeep of the Bengal Basin (Reimann, 1993). Aeromagnetic interpretation map suggests gradual deepening of the basement towards the center of the basin, and also shows the presence of subsurface synclinal features and faults within the basin (Mannan, 2002). The syncline was a major depocentre for accumulation of late Neogene and Holocene deposits. Anticlinal structures

within the Sylhet basin include the Atgram, Sylhet, Chattak, Kailas Tila, Beani Bazar, Patharia, Fenchuganj, Rasidpur and Habiganj anticlines (Fig. 2). The Tertiary Sylhet succession comprises into several groups including Jaintia, Barail, Surma, Tipam and Dupitila and their constituent formations in ascending order of Paleocene to Plio-Pleistocene in age Table 1. In this paper we examined only the lower part of the Sylhet succession including Paleocene to late Eocene Jaintia and late Eocene to early Miocene Barail Groups respectively (Hossain et al., 2010).

The Jaintia Group is the lowermost unit of the Tertiary stratigraphic succession in the Sylhet basin. Rocks of the Group are mainly fossiliferous and calcareous in nature, and are divided into three distinct and prominent formations, the Tura Sandstone, Sylhet Limestone and the Kopili Shale Formations, in ascending order. These units were deposited during repeated transgressive and regressive phases during the Paleocene to Eocene period.

The Tura Sandstone Formation consists mostly of quartz arenites with some carbonaceous materials and impure limestones, and was deposited in shallow to deep marine environments (Khan, 1991; Uddin and Lundberg, 1999; Najman et al., 2008). The Sylhet Limestone Formation, which conformably overlies the Tura Sandstone Formation, consists mainly of nummulitic limestone interbedded with minor sandstone (Reimann, 1993). It is interpreted as a shallow-marine carbonate deposit (Shamsuddin and Abdullah, 1997). The overlying Kopili Shale Formation consists mainly of argillaceous and fossiliferous materials, interbedded with sandstones and occasional thin limestones. Presence of fossiliferous sediments in the formation indicates distal deltaic to shelf or slope depositional environments (Alam et al., 2003).

The Barail Group was deposited during the Oligocene period, when basin-wide emergence and a major marine regression occurred (Alam et al., 2003). It is divided into the Jenam and Renji Formations. The Jenam Formation consisting mainly of siltstones, silty shales, and sandstones with minor carbonaceous shales (Khan, 1978). The Renji Formation, consisting mostly of fine grained sandstones, alternating with siltstones, and calcareous and carbonaceous shales with a few coal lenses (Reimann, 1993). The contact between the Renji Formation and the overlying Surma Group is marked by a distinct unconformity. Barail Group sediments have been deposited in shallow-marine and deltaic environments (Banerji, 1981; Johnson and Alam, 1991). Alam (1991) reported that the Barail Group sediments were deposited in tide-dominated shelf environments.

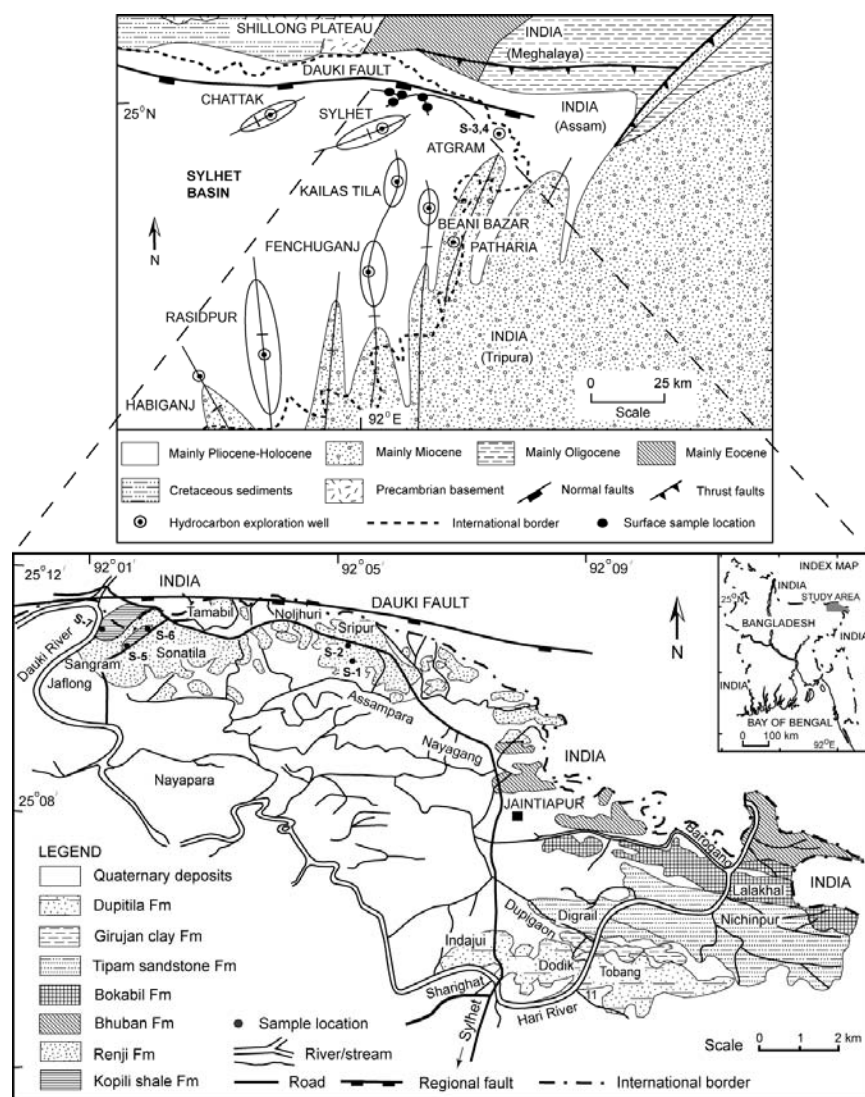


Fig. 2 (a) Map showing location of the study area and hydrocarbon exploration wells in the Sylhet basin, Bangladesh (after Hossain et al., 2009a, 2013a), (b) geological map showing lithotypes and surface sample points in the study area (after Hossain et al., 2009a). Cores were taken from Atgram-1X well.

Materials and Methods

Sample collection and preparation

Indurated mudstone samples were collected from Jaintia and Barail Group, Sylhet basin, Bangladesh. The sections were located between latitude $25^{\circ}04'N$ to $25^{\circ}12'30''N$ and longitude $92^{\circ}01'E$ to $92^{\circ}12'30''E$. Locations of the sample points are indicated in Fig. 2. Rock samples were manually disaggregated or chipped, and then rinsed in deionized distilled water and dried at $110^{\circ}C$ prior to crushing. The oven-dried samples were then crushed in a tungsten-carbide ring mill, with mills times generally between 30 to 45 seconds.

Solvent extraction and separation

Powdered mudstone sample (~40 gm) was used to extract hydrocarbon solvent in a Soxhlet apparatus for 72 hours using a mixture of dichloromethane and methanol (9:1). Copper granules were used to separate elemental sulfur. Hydrocarbon solvent was separated using a rotary evaporator. Aromatic hydrocarbon was then separated using thin layer chromatography on activated silica gel (Kieselgel 60 PF254, Merk) eluting with *n*-hexane.

Gas chromatography-mass spectrometry (GC-MS)

The aromatic hydrocarbon fractions were performed using gas chromatography-mass spectrometry (GC-MS)

with a Shimadzu QP2010 instrument at Shimane University, Japan. The chromatograph was equipped with an automatic temperature programmable injection system and a fused silica capillary column coated with chemical bound (DB-5MS of 30 m × 0.25 mm i.d.). The interface temperature was held at 50 to 300 °C. The oven temperature program was used at heating rate 8 °C min⁻¹. Helium was used as a carrier gas with flow rates

of 1.69 ml min⁻¹. GC-MS analyses were performed initially on a Finnigan MAT TSQ-700 system using electron impact potential ionization of 70 eV with a mass range 50 to 850 *m/z* and a cycle time of 0.5 seconds. The total ion chromatograms (TICs) of the GC-MS were identified based on their mass spectra and retention times (Killips and Massoud, 1992; Jiang et al., 1998, 2000; Grice et al., 2007).

Table 1 Stratigraphy of the Sylhet succession, northeastern Bengal Basin, Bangladesh (after Hossain et al., 2010).

Age	Group	Formation	Lithology	Depositional Environments
Recent	Alluvium	Alluvium	Sand, silt, clay	Fluvial
Late Pleistocene	Dihing	Dihing	Sandstone, shale	Fluvial ^a
Pliocene–Pleistocene	Dupi Tila	Dupi Tila	Sandstone, shale	Fluvial ^{b,f}
Late Miocene–Pliocene	Tipam	Girujan Clay	Clay, sandstone	Fluvial ^c , lacustrine ^c
		Tipam Sandstone	Sandstone, shale	Fluvial ^{b,f}
Middle–Late Miocene	Surma	Boka Bil	Sandstone, shale	Marine ^{c,d} , deltaic ^{b,c,f}
		Bhuban	Sandstone, shale	
Late Eocene–Early Miocene	Barail	Renji	Sandstone, shale	Shallow marine ^c , deltaic ^f
		Jenam	Shale, sandstone	
Late Eocene		Kopili Shale	Shale, minor lst.	Shallow marine ^c , deltaic ^c
Early–Middle Eocene	Jaintia	Sylhet Limestone	Limestone	Shallow marine ^{c,d}
Paleocene–Early Eocene		Tura Sandstone	Quartz arenites	Shallow marine ^{a,b,f}

^aKhan, 1991; ^bJohnson and Alam, 1991; ^cReimann, 1993; ^dShamsuddin and Abdullah, 1997; ^eAlam et al., 2003; ^fNajman et al., 2008

Results

The measured Penta-aromatic hydrocarbons and their respective concentration (percentage) in individual sample are shown in Table 2. Distribution of Penta-aromatic hydrocarbons is illustrated in Fig. 3. The Jaintia Group mudstone samples contained relatively high concentration of benzo[*b,j,k*]fluoranthene than overlying Barail Group samples. In the Renji Formation, benzo[*b,j,k*]fluoranthene is almost constant (~10%). Similarly, abundances of benzo[*e*]pyrene are similar in the underlying Jenam Formation mudstones (Table 2). Concentration of perylene was abundant in the Jaintia Group samples (up to 37.07 %) than overlying Barail Group samples with high total organic carbon (TOC) content (Hossain et al., 2009a). In the Barail Group, perylene concentration was relatively abundant in the upper part (32.42 to 25.91%) than in the lower part (10.99 to 15.92%). A number of samples contained relatively low perylene content (Table 2). Higher molecular weight PAHs were identified in the top layer of each group than in the bottom layer sample. Abundances of benzo [*b,j,k*] fluoranthene, benzo

[*e*]pyrene and benzo [*a*] pyrene are relatively high in sample of the lower Jaintia Group (Table 2). Samples in the lower Jaintia Group contain relatively low perylene content (12.69%). Abundance of benzo[*e*]pyrene and perylene in the upper Jaintia Group samples are mostly uniform (about 18.21% and 37.07% respectively).

Discussion

The PAHs, benzo[*b,j,k*]fluoranthene, benzo[*e*]pyrene and benzo[*a*]pyrene may be originated from combustion products (Jiang et al., 1998). The distribution of combustion-derived PAHs may be modified by subsequent sedimentary alkylation procedures (Alexander et al., 1995; Smith et al., 1995; Jiang et al., 1998). Benzo[*e*]pyrene is the most stable PAHs among the Bfla, BaPy and perylene (Sullivan et al., 1989). Therefore, benzo[*e*]pyrene may be the least susceptible to post-depositional modifications (Jiang et al., 1998). Combustion derived PAHs take place from combustion of vegetation. Benzo[*e*]pyrene most probably derived from forest fires as well as peat fires (Jiang et al., 1998). High relative abundance of

Table 2 Concentrations (%) of benzo[*b,j,k*]fluoranthene, benzo[*e*]pyrene, benzo[*a*]pyrene and perylene in Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh (Hossain et al., 2013a). Abbreviation: PAI, Pentacyclic aromatic isomers (Bflas, BePy, BaPy) and TOC, total organic carbon.

Sample	Group	Formation	Bflas	BePy	BaPy	Pery	PAI	TOC*	BaPy/BePy	Bflas/(Bflas+BePy)
			<i>m/z</i> 252	<i>m/z</i> 252	<i>m/z</i> 252	<i>m/z</i> 252		(wt%)		
S-1	Barail	Renji	10.16	45.02	18.90	25.91	74.09	1.56	0.42	0.18
S-2	Barail	Renji	10.68	34.48	22.43	32.42	67.58	1.19	0.65	0.24
S-3	Barail	Jenam	14.81	53.58	20.62	10.99	89.01	0.60	0.38	0.22
S-4	Barail	Jenam	22.10	53.00	8.99	15.92	84.08	0.80	0.17	0.29
S-5	Jaintia	Kopili Shale	23.47	18.21	21.25	37.07	62.93	1.20	1.17	0.56
S-6	Jaintia	Kopili Shale	26.54	18.22	18.17	37.07	62.93	0.90	1.00	0.59
S-7	Jaintia	Kopili Shale	31.59	36.73	19.00	12.69	87.31	1.18	0.52	0.46

*: cited from Hossain et al. (2009a)

Benzo[*e*]pyrene in the Jenam and lower Jaintia samples suggested that their derivation from combustion of forest or peat materials.

Perylene concentration increases during depth of burial of OM (Grimalt et al., 2004; Grice et al., 2009). In the Jaintia Group, abundances of perylene are uniform towards upper part, where as low values in the bottom part samples. Concentration of perylene is relatively lower in the overlying Barail Group mudstones (Table 2). The high values therefore suggest *in-situ* biogenic sources. Perylene is abundant in marine, lake and river sediments either aquatic or terrigenous in anoxic environments (Hites et al., 1980; Silliman et al., 1998; Jiang et al., 2000; Grimalt et al., 2004). Perylene could also originate from fungi (e.g., wood-degrading fungi) within anoxic marine environment (Grice et al., 2009; Nabbefeld et al., 2010). Sediments in the Sylhet succession were accumulated in deltaic to marine environmental conditions (Reimann, 1993). OM of the Jaintia and Barail Groups was deposited mainly in marine dominated oxic conditions, with abundant inputs of terrestrial higher plant material (Hossain et al., 2009a). Therefore, influx of perylene in the Jaintia and Barail Group mudstones probably linked with deltaic OM sources subsequently deposited in oxic-anoxic environmental conditions.

Abundances of perylene relative to pentacyclic aromatic isomers (PAI, summed values of Bflas, BePy, BaPy) are important for derivation of combustion or diagenetic sources (Hites et al., 1980; Venkatesan, 1988; Baumard et al., 1998; Readman et al., 2002; Bakhtiari et al., 2009; Fan et al., 2011). Perylene abundance >10% relative to PAI representing diagenetic sources (Hites et al., 1980; Venkatesan, 1988) and <10% representing combustion sources (Baumard et al., 1998; Readman et al., 2002; Bakhtiari et al., 2009). Abundance of perylene to PAI in the

Jaintia and Barail Group mudstones ranging from ~10% to over 20% suggesting both diagenetic and combustion sources.

Elevated level of perylene concentration in sediments is occasionally favored by the control of TOC (Agbozu and Opuene, 2009; Bakhtiari et al., 2009; Fan et al., 2011). In the Barail Group, perylene versus TOC concentrations show a marked positive correlation ($r = 0.80$) and clear linear trend (Fig. 4), inferring that concentrations of perylene in the Barail Group mudstones may be dependent on the abundances of TOC. Conversely, correlation between perylene versus TOC concentrations in the Jaintia Group shows no correlation, suggesting that TOC does not influence the concentration of perylene in these mudstones. Perylene in the Jaintia Group most likely derived from different sources. Fan et al. (2011) reported that high influx of perylene in mudstones probably originated from erosion of soils/sedimentary rocks under strong influence of steady state environmental condition. This is consistent with the study of Hossain et al. (2010) and concluded that Paleocene to late Eocene Jaintia Group mudstones contained relatively high chemical index of alteration (CIA) values (up to 85) subsequently originated from a stable cratonic source.

The ratios of BaPy/BePy and Bflas/(Bflas + BePy) is used to identify source of PAHs. The values of BaPy/BePy in the Jaintia and Barail Group mudstones ranging from 0.52 to 0.17 and 0.17 to 0.65, respectively (Table 2), inferring that combustion and petrogenic sources throughout the sequence (Khalili et al. 1995). The ratio values of Bflas/(Bflas + BePy) are less than 0.20 indicate petroleum combustion, between 0.20 and 0.50 indicate fossil fuel combustion, and values greater than 0.50 are characteristics of grass, wood and coal combustion (Shengli et al., 2010). In the late Eocene Jaintia Group, Bflas/(Bflas + BePy) ratios

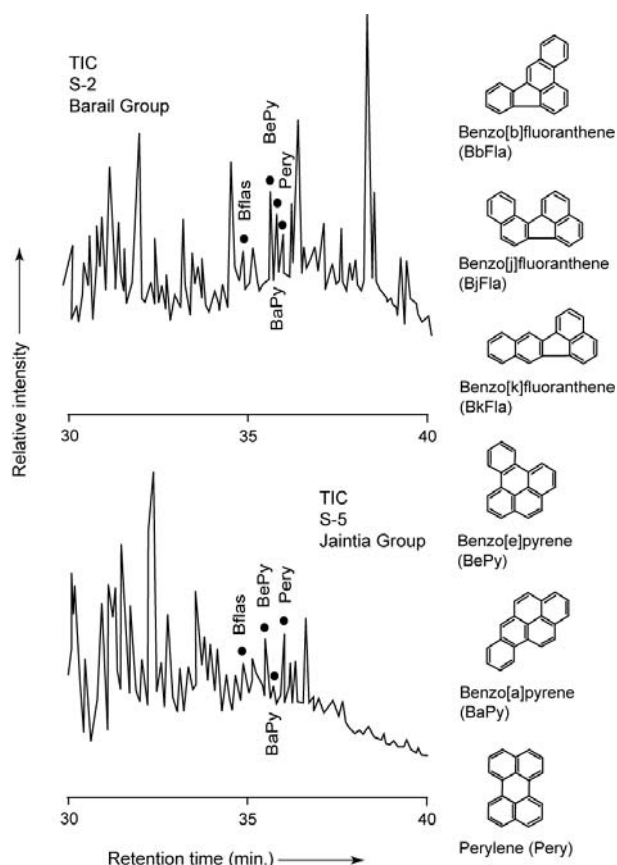


Fig. 3 Representative total ion chromatograms (TICs) showing distributions of benzo[*b,j,k*]fluoranthene (m/z 252), benzo[*e*]pyrene (m/z 252), benzo[*a*]pyrene (m/z 252) and perylene (m/z 252) in the Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh.

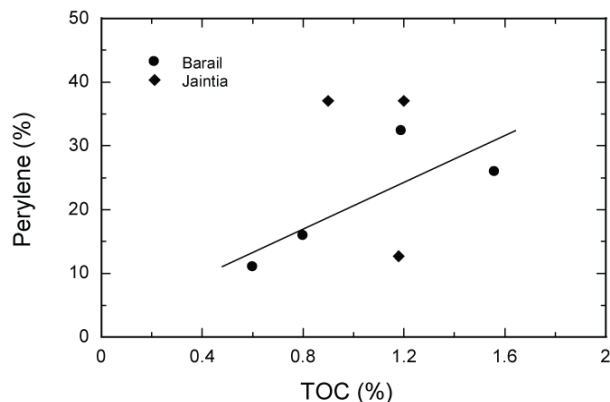


Fig. 4 Variation diagram between TOC (%) and perylene (%) in the Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh. TOC data from Hossain et al. (2009a).

range from 0.46 to 0.59 suggesting combustion of grass, wood and coal. The overlying Jenam and Renji

formations in the late Eocene to middle Miocene Barail Group, ratios of Bfla/(Bfla + BePy) range from 0.22 to 0.29 and 0.18 to 0.24, respectively suggested that PAHs in the group were derived from combustion sources.

Conclusions

Penta-aromatic hydrocarbons in the Jaintia and Barail Group mudstones, northeastern Bengal Basin, Bangladesh have been studied. Distributions of Bfla, BePy and perylene were abundant in Barail Group than underlying Jaintia Group. High concentration of perylene in the Barail Group mudstones suggesting diagenetic influences subsequently derived from terrigenous organic matter sources. Abundances of perylene increase with increasing depth of the succession inferring that influx of perylene may deposited under oxic/anoxic environmental condition. Combustion sources of PAI were also identified as evidences by high Bfla/(Bfla + BePy) ratio values in the studied mudstones.

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