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Vertical distribution and sources of perylene in miocene mudstones from the northeastern Bengal Basin of Bangladesh

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Abstract

The distributions and sources of perylene (Pery) and penta-aromatic hydrocarbons were investigated in 10 Miocene mudstones from the NE Bengal Basin, Bangladesh using gas chromatography-mass spectrometry (GC-MS). Pery concentrations were high in most of the samples, and highest levels were found in samples of strong terrigenous influence of organic matter deposited under reducing environmental conditions. Mudstones in the Bokabil Formation (upper Surma Group) contain relatively high abundances of benzofluoranthenes (Bflas), benzo[e]pyrene (BePy) and benzo[a]pyrene (BaPy) attributed to their combustion origin. Pery to pentacyclic aromatic isomers (PAIs) ratios implies an influence of diagenetic sources, whereas low BaPy/BePy ratios indicate that these compounds were originated from combustion sources. Wood degrading fungi in arid to humid and seasonal climatic conditions was also the possible source of Pery in the studied succession.

Keywords: GC-MS; Perylene; Mudstone; Bengal Basin; Bangladesh

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are widespread in both modern and ancient sediments (Aizenshtat, Z., 1973, Wakeham *et al.*, 1979, 1980, Jiang *et al.*, 1998, 2000, Grimalt *et al.*, 2004, Grice *et al.*, 2007). Generally, PAHs occur in soils (Blumer and Youngblood, 1975, Youngblood and Blumer, 1975), modern lake sediments (Giger and Schaffner, 1978, Laflamme and Hites, 1978, Wakeham *et al.*, 1980, Gschwend and Hites, 1981), modern marine sediments (Youngblood and Blumer, 1975, Hites *et al.*, 1977, Gschwend and Hites, 1981), river and river estuary sediments (Giger and Schaffner, 1978, Hites *et al.*, 1980, Brown and Maher, 1992) and in river particulates (Giger and Schaffner, 1978, Brown and Maher, 1992).

Perylene (Pery) and pentacyclic aromatic isomers (PAIs; benzofluoranthenes (Bflas), benzo[e]pyrene (BePy) and benzo[a]pyrene (BaPy) consist of five condensed benzene rings, and are abundant in modern sediments (Jiang *et al.*, 1998, Baumard *et al.*, 1998, Readman *et al.*, 2002, Bakhtiari *et al.*, 2009). Pery is derived from biological precursors during post-depositional diagenesis in terrigenous sources (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). It has been derived from both aquatic and continental organic matters under the influence of prolong

reducing environments (Aizenshtat, 1973, Wakeham *et al.*, 1979, Silliman *et al.*, 2000, Suzuki *et al.*, 2010). Pery may also originate from diatomaceous sediments (Wakeham *et al.*, 1979, Venkatesan, 1988).

Bangladesh is located in the northeastern part of the Indian subcontinent (Fig. 1), which is bounded to west by the Precambrian Indian Shield Platform, to the east by the Indo-Burman Ranges, to the north by the crystalline Shillong Massif, and to the south it plunges into the Bay of Bengal (Fig. 1). The study area covers Middle to Late Miocene Surma Group (Bhuban and Bokabil formations) within the Surma Basin, NE Bangladesh (Fig. 2), in terms of concentration and potential sources of Pery in mudstones. Biomarker study of the Tertiary mudstones in the eastern Bengal Basin of Bangladesh has been reported by Alam and Pearson (1993), Farhaduzzaman *et al.* (2012) and Hossain *et al.* (2009a, 2009b, 2013). The main objective of this study was to identify distribution and possible sources of Pery and PAIs in the Miocene mudstones, Surma Basin, NE Bangladesh.

Geological setting

The Bengal Basin is placed just south of the crystalline Shillong Massif, occupying entire Bangladesh, West Bengal,

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Assam, Tripura, Myanmar and part of the Bay of Bengal (Fig. 1). This basin has been originated between the collision of three plates- Indian plate, Eurasian plate and Myanmar plate, building the extensive Himalayas and Indo-Burman Ranges, and thereby loading the lithosphere to form flanking sedimentary basins (Uddin and Lundberg, 1998). The Bengal Basin comprises three geo-tectonic provinces- the stable shelf zone, the central deep basin and the Chittagong-Tripura fold belt (Alam et al., 2003). The basin fills comprise ~22 km thick sedimentary record (Alam et al., 2003), and it has been developed by the seaward prograding deltas of the Ganges and Brahmaputra river systems (Einsele et al., 1996). The Ganges and Brahmaputra rivers drain opposite flanks of the Himalaya, and drastically rising mountains carrying sediments by Brahmaputra from the northeastern Himalayas (Fig. 1). The Himalayas originated between collision of Indian and Eurasian plates and significantly uplift its northeastern part. The Ganges-Brahmaputra delta and the Bengal fan contain large volume of sediments that were input from the collision zone, characterizing physical and chemical weathering and drained by many Himalayan streams to join into the Ganges and Brahmaputra rivers.

The Surma Basin, is a rapidly subsiding sub-basin in the northeastern part of the Bengal Basin, Bangladesh and con tains hydrocarbon reserves, the only natural energy resource discovered so far in Bangladesh. Tectonically, the Surma Basin is located in the central deep basin of the Bengal Basin

(Alam *et al.*, 2003). The stratigraphy of the Surma Basin is listed in Table I. The present paper is concentrated only the Surma Group of the Tertiary succession of Middle to Late Miocene age, NE Bengal Basin, Bangladesh. The Surma Group consists of an arenaceous lower Bhuban and argillaceous upper Bokabil formations (Alam *et al.*, 2003). The Bhuban Formation is comprises mainly of grey sandstones, siltstones and mudstones. The overlying Bokabil Formation consists primarily of light grey mudstones, siltstones and sandstones. Sediments of the Surma Group were deposited under the strong influence of tidal currents in a deltaic to shallow marine depositional systems (Khan, 1991, Reimann, 1993, Shamsuddin and Abdullah, 1997, Alam *et al.*, 2003).

Materials and methods

Sample collection

Ten unweathered mudstone samples were collected from hydrocarbon exploration drill core and surface outcrops in Miocene Surma Group, Sylhet succession, Bangladesh. Outcrop samples (n=3) were taken from section exposed along the Hari River (Hossain and Roser, 2006), and drill core samples (n=7) were taken from three hydrocarbon exploration wells, namely Rashidpur-1, Fenchuganj-2 and Patharia-5 (Fig. 2 and Table II). Location of the sample sites are shown in Fig. 2. Fresh rock samples were manually crushed and subsequently washed with deionized distilled

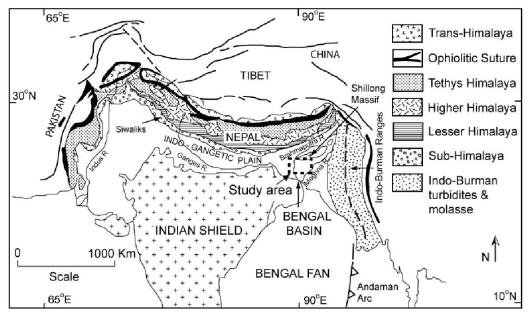


Fig. 1. Geological map of the study area showing location of the sample sites and major geographic features of the Himalaya (Hossain *et al.*, 2010)

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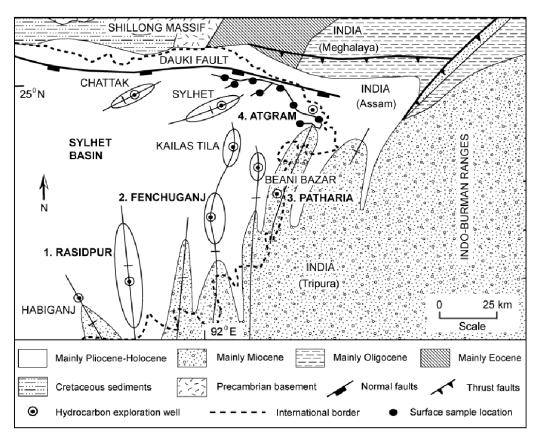


Fig. 2. Map showing location of the study area and hydrocarbon exploration wells in the NE Bengal Basin, Bangladesh (Hossain *et al.*, 2009b). Drill core samples were taken from the Rasidpur-1, Fenchuganj-2 and Patharia-5 wells

water, and dried in an oven at 110°C. The oven-dried samples were then powdered in a tungsten-carbide ring mill for 25 to 45 s.

Column chromatography

Approximately 40 g of each powdered sample were extracted in a Soxhlet apparatus for 72 hours using a mixture (9:1) of dichloromethane (DCM) and methanol (MeOH). Elemental sulfur was removed by using Cu granules. The solvent was separated using a rotary evaporator. Saturated hydrocarbons and aromatic hydrocarbons were then separated using thin layer chromatography on fully activated silica gel (Kiselgel 60 PF254, Merck) eluting with *n*-hexane.

Gas chromatography-mass spectrometry (GC-MS)

The aromatic hydrocarbon fractions of all samples were analyzed by GC-MS with a Shimadzu QP2010 instrument at Shimane University, Japan. The chromatograph was equipped with an automatic temperature programmable

injection system. A fused silica capillary column, DB-5MS of 30 m and 0.25 mm i.d. was used. The column temperature was programmed from 50 to 300° C at 8° C/min. Helium was used the carrier gas. GC-MS analyses were performed initially on a Finnigan MAT TSQ-700 system using electron impact ionization (70 eV). Full scan spectra were recorded over a range m/z 50 to 850 at a scan rate of 0.5 s. Identification of Pery (m/z = 252) and PAIs (m/z = 252) in the biomarkers were monitored by comparing the retention time and mass spectra with published data.

Results and discussion

Distribution of Pery and PAIs were examined from the Miocene Surma Group (Bhuban and Bokabil formations) mudstones, NE Bengal Basin, Bangladesh (Fig. 3). The experimental data (%) is shown in Table II (Hossain *et al.*, 2013). Vertical distribution of Pery and PAIs are illustrated in Fig. 4. Benzo[*b*]fluoranthene, benzo[*j*]fluoranthene and benzo[*k*]fluoranthene could not fully separate with the GC-

Table I. Stratigraphy of the NE Bengal Basin, Bangladesh (Khan, 1991; Reimann, 1993)

Age	Group	Formation	Lithology				
Recent	Alluvium	Alluvium	Sand, silt and clay				
Late Pleistocene	Dihing	Dihing Sandstone with interbedded mudstone					
Pliocene-Pleistocene	Dupitila	Dupitila Pebbly sandstone and sandstone with subordinate siltstone					
Late Miocene-Pliocene	Tipam	Girujan Clay	Mottled clay with subordinate sandy clay and sandstone				
		Tipam Sandstone	Massive sandstone with subordinate mudstone				
Middle-late Miocene	Surma	Bokabil	Mudstone with interbedded, sandstone and siltstone				
		Bhuban	Alternation of sandstone, siltstone and mudstone				
Late Eocene-early	Barail	Renji	Sandstone, mudstone and coal lenses				
Miocene		Jenam	Mudstone, siltstone and sandstone				
Late Eocene		Kopili Shale	Shale with subordinate sandstone and thin limestone				
Early-middle Eocene	Jaintia	Sylhet Limestone	Fossiliferous limestone				
Paleocene-early Eocene		Tura Sandstone	Alternation of sandstone and limestone, with mudstone and				
			coal seams				

MS methods. Bflas are integrated as sum from benzo[b]fluoranthene, benzo[j]fluoranthene and benzo[k]fluoranthene. Among the PAIs, distributions of BePy are relatively higher than Bflas and BaPy (Table II). Abundances of Bflas are high in the upper Bokabil samples, whereas contents decrease with increasing depth of the lower Bokabil Formation (Fig. 4). Bflas in the Bhuban Formation mudstones range from 9 to 22%, whereas in the Bokabil Formation concentrations of Bflas are relatively high range from 9 to 35% (Table II). Vertical distributions of PAHs show almost similar pattern except BaPy (Fig. 4). Relatively high concentrations of Bflas and BePy are present in the upper part of the Bokabil mudstones, subsequently high TOC contents inferring that distribution may controlled by organic carbon. In the Bokabil Formation, abundances of Pery increases with different

depth intervals (e.g., Z-31 and Z-35) and the samples are dominated by higher plant organic matter (Hossain *et al.*, 2009a). The relative abundances of Pery in the underlying Bhuban Formation are ranging from 41 to 73% (Table II; Fig. 4). These highest values of Pery in the studied mudstones together elevated terrestrial organic matter in varying depths moreover imply *in situ* biogenic sources (Agbozu and Opuene, 2009). Grimalt *et al.* (2004) and Grice *et al.* (2009) suggested that Pery concentration increases during depth of burial of organic matter. Vertical distribution of Pery in the Tertiary succession shows variable contents (Fig. 4). T_{max} values in the Surma Group samples range from 435 to 449 °C (Table II; Hossain *et al.*, 2009a), and vitrinite reflectance (Ro) values range from 0.45 to 0.60% (Shamsuddin and Khan, 1991), inferring that the occurrence of Pery in the

Table II. Abundances (%) of perylene (Pery), benzofluoranthenes (Bflas), benzo[e] pyrene (BePy) and benzo[a] pyrene (BaPy) in Miocene mudstones, NE Bengal Basin, Bangladesh (Hossain et al, 2013). Abbreviation: pIAs, Pentacylic aromatic isomers (Bflas + BePay + BaPy); TOC, Total organic carbon. TOC* and T_{max} °C data from Hossain et al (2009_a)

Age	Well/local name	Group	Formation	Pery	Bflas	BePy	BaPy	PAIs	TOC*	$T_{max}{}^{\rm o}{\rm C}^*$
Z-31	BDR camp	Surma	Bokabil	74.89	10.59	12.09	2.43	25.11	0.31	438
ZH-122	Rashidpur-1	Surma	Bokabil	18.90	34.28	40.64	6.18	81.10	0.53	449
ZH-126	Rasidpur-1	Surma	Bokabil	23.22	35.01	39.56	2.21	76.78	0.48	448
Z-35	Baragang	Surma	Bokabil	74.52	9.05	14.53	1.90	25.48	0.39	439
ZH-128	Rasidpur-1	Surma	Bokabil	42.56	17.44	38.20	1.80	57.44	0.40	447
ZH-55	Fenchuganj-2	Surma	Bokabil	48.52	19.30	28.66	3.52	51.48	0.32	442
ZH-59	Fenchuganj-2	Surma	Bhuban	41.41	21.87	35.30	1.41	58.59	0.38	442
ZH-88	Fenchuganj-2	Surma	Bhuban	72.57	9.05	14.88	3.50	27.43	0.39	445
ZH-14	Patharia-5	Surma	Bhuban	53.78	13.87	27.32	5.03	46.22	0.46	436
Z-43	Tetultala	Surma	Bhuban	65.54	14.44	18.26	1.75	34.46	0.38	438

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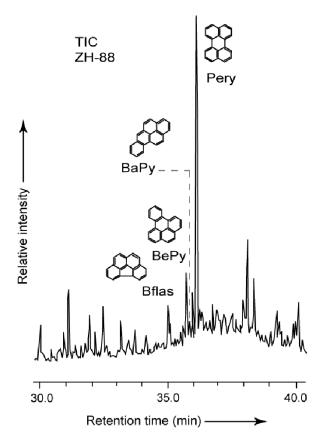


Fig. 3. Total ion chromatogram (TIC) for aromatic fractions of representative Miocene mudstone (sample no. ZH-88), NE Bengal Basin, Bangladesh. Abbreviations as in Table II

mudstones is seem to be organic source dependent and does not show the dependence on maturity. This result is in good agreement with previous study of tri-aromatic hydrocarbons in the Surma Group mudstones (Hossain *et al.*, 2009b), NE Bengal Basin, Bangladesh suggesting that phenanthrene, methylphenanthrenes and dimethylphenanthrene isomers were likely to be primarily controlled by source input rather than by thermal maturity.

Pery 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). Grice *et al.* (2009) and Nabbefeld *et al.* (2010) reported that Pery probably attributed to a source from wood-degrading fungi. Pery may also originate from combustion sources (Baumard *et al.*, 1998, Readman *et al.*, 2002, Bakhtiari *et al.*, 2009, Fan *et al.*, 2011). Hossain *et al.* (2009a) suggested that organic matter in the Surma Group

was deposited in a sea-water influenced anoxic environmental condition, as indicated by lower pristane/phytane ratios (~1). Sediments in the Surma Group were accumulated in deltaic to shallow marine environments (Reimann, 1993, Alam et al., 2003). Organic matter in the Surma Group was originated from mainly phytoplankton and minor influence from terrestrial higher plants (Hossain et al., 2009a). Pery is formed by diagenetic transformation of marine precursors (Readman et al., 2002). The Surma Group sediments were deposited during repeated marine transgression and regression phases, and last marine incursion occurred at the top of the Bokabil Formation (Alam et al., 2003, Hossain et al., 2010). The high relative abundance of Pery (~75%) is present in upper most part of the samples, ascribed to be deposited in reducing environment. Pery has been derived from perylenequinone structures of fungus comb as well as in modern plants, and it is generally developed in anoxic condition (Aizenshtat, 1973, Grice et al., 2009, Suzuki et al., 2010). The fungus combs are widely distributed on woody materials in the tropical environments, e.g., South Asia, North America and Brazil (Bakhtiari et al., 2009). Therefore, Pery in the studied Surma Group may be input from the activity of fungus comb on woody plants. Recently, Hossain et al. (2013) reported that Pery in the Tertiary Sylhet succession, NE Bengal Basin was derived from fungi with different climatic conditions mostly arid to humid and seasonal.

The concentrations of Perv relative to PAIs are significant for combustion as well as diagenetic sources (Hites et al., 1980, Venkatesan, 1988, Baumard et al., 1998, Readman et al., 2002, Agbozu and Opuene, 2009, Bakhtiari et al., 2009, Fan et al., 2011). Pery concentrations >10% relative to the PAIs suggesting diagenetic origin of terrestrial organic matter (Hites et al., 1980, Venkatesan, 1988) and <10% representing pyrolytic/combustion origin (Baumard et al., 1998, Readman et al., 2002, Bakhtiari et al., 2009). Tolosa et al. (2004) reported that Pery over PAIs ratios <20% probably originated from pyrolytic sources. The value of Pery to PAIs in the Surma Group mudstones are all >20% (range from ~25 to 77%) therefore suggested that a greater influence of diagenetic sources. Among the combustion derived PAHs, Bflas and BePy may be the best example of the original combustion products (Jiang et al., 1998). Smith et al. (1995) suggested that combustion derived PAHs have been modified through sedimentary alkylation procedures. Bflas and BePy are relatively less susceptible to alkylation processes and/or post-depositional modifications (Jiang et al., 1998). BePy may also originate from forest fires and peat fires (Jiang et

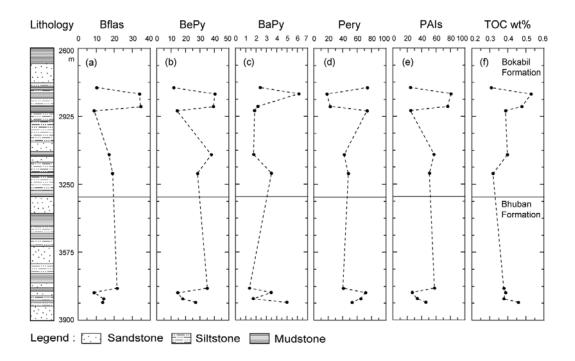


Fig. 4. Vertical distributions of Bflas, BePy, BaPy, Pery, PAIs and TOC in Miocene mudstones, NE Bengal Basin, Bangladesh

al., 1998). The high relative abundances of BePy and Bflas to BaPy in the samples studied suggesting that a significant contribution of these PAHs from combustion of paleovegetation either forest fires or peat fires. The isomer concentration ratio of BaPy/BePy has been used to identify origin of PAHs (Khalili et al., 1995). The ratios of BaPy/BePy in the Surma Group mudstones range from 0.04 to 0.24 indicating combustion sources (Khalili et al., 1995).

High contents of Pery in mudstones are likely to be derived from erosion of soils or ancient sediments during steadystate environmental condition (Fan et al., 2011). High abundances of Pery in sediments are sometimes attributed to preferential retaining of PAHs by high total organic carbon (TOC) in the sediments (Opuene et al., 2007, Bakhtiari et al., 2009, Fan et al., 2011). TOC data sources are from Hossain et al. (2009a). Concentration of TOC varies between 0.31 to 0.53%, with relatively high values in middle of both formations. Low Pery values in mudstones therefore contain high TOC (Fig. 4). Correlation between Pery and TOC contents are poor (figure not illustrated), and even shows negative trend, inferring that organic carbon may not be fully responsible for the influx of Pery in the Surma Group mudstones. Distribution of PAIs and TOC is mostly identical (Fig. 4), suggesting a comparable source trends.

Conclusion

The occurrences of Pery and penta-aromatic hydrocarbons were investigated in 10 Miocene mudstones from the NE Bengal Basin, Bangladesh. The highest concentrations of Pery were identified in outcrop samples, and highest levels were found in samples of strong terrigenous influence of organic matter deposited under anoxic environmental conditions. Bflas, BePy and BaPy in the upper Surma Group are relatively high, suggesting that the PAHs were derived from combustion sources. Pery to PAIs ratios implies an influence of diagenetic sources and/or in situ biogenic origin, whereas low BaPy/BePy ratios indicate that these compounds were originated from combustion sources. Wood degrading fungi in arid to humid and seasonal climatic conditions was also the possible source of Pery in the studied succession.

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