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Organic matter in coal seam-VI of the Barapukuria coal field in Bangladesh

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Abstract

Organic matter sources and depositional environments of Permian Gondwana coals in the coal seam-VI, Barapukuria coal field, northwestern Bangladesh have been investigated using CHNS elemental analysis. A total of 8 coal samples were collected from the coal seam-VI within depth range between 118 and 509 m. Total organic carbon (TOC), hydrogen, nitrogen and sulfur contents range from 48 to 75 (average 61 wt.%), 3.89 to 5.80 (average 4.59 wt.%), 2.87 to 3.87 (average 3.28 wt.%), and 0.19 to 0.37 wt.% (average 0.30 wt.%), respectively. C/N ratio range between 14 and 21 (average 19), and a positive correlation between TOC and TN (r = 0.51) suggested that organic matter in the coals was derived largely from terrestrial plants. However, C/S ratio values (up to 376) and scatter distribution between TOC and TS indicates the coal seam-VI was deposited in fresh water dominated inland lake environments.

Keywords: Organic matter; Depositional environment; Gondwana coal; Barapukuria coal field; Bangladesh.

Introduction

Coal is one of the most important and relatively inexpensive energy resources around the globe. Bangladesh has enormous coal reserves, most of these coal deposits located in the northwestern part of the country (Fig. 1). The major coal deposits of Bangladesh as well as adjacent Indian landmass occur in Permian Gondwana sequences. The Gondwana coal in Bangladesh has been identified in the vicinity of Barapukuria, Khalaspir, Dighipara, Phulbari and Jamalganj (Fig. 2a) and it occurs mostly at shallow depths (Fig. 2b). The Barapukuria coal field is the first major coalfields in Bangladesh, which discovered in 1985 by Geological Survey of Bangladesh (GSB). The Gondwana Barapukuria coal basin islocated in the Parbatipur thana of Dinajpur district, which is close to the West Bengal of India (Fig. 1). The basin covers an area of approximately 5.25 km^2 (Wardell-Armstrong, 1991). Coal in this coal field is encountered at a depth range between 195 and 535 m from the surface (Bakr *et al.*, 1996; Hossain *et al.*, 2013a). The estimated coal reserve of the Barapukuria coal field is about 390 million tons (Imam, 2013). Six major coal seams of different thickness were identified in the basin, and coal seam-VI alone represents approximately 36 m (Norman, 1992; Bakr *et al.*, 1996). The coals are mainly high volatile sub-bituminous to bituminous rank (Bakr *et al.,* 1996; Imam, 2013). This coal was deposited in cool climatic conditions during the Permian period (Wardell-Armstrong, 1991).

Several authors have been investigated in this coal-bearing Gondwana sequence, Barapukuria basin atdifferent points of view (Norman, 1992; Hussain and Curtin, 1995; Bakr *et al.*, 1996; Islam *et al.*, 2003; Islam and Hossain, 2006; Islam and Hayashi, 2008; Farhaduzzaman *et al.*, 2012; Hossain *et al.*, 2013a, b). However, investigation of organic matter source and depositional environment in Barapukuria coal using CHNS elemental analyses has not been quantified. The main objective of this study is to provide the results of organic geochemical analyses, in order to compare these coals in terms of organic matter composition, source and environment of deposition in the coal-bearing Barapukuria basin, northwestern Bangladesh. The results obtained will also be compared with similar studies of Gondwana coals from neighboring countries.

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Geological setting

The Bengal Basin is situated in the northeastern part of the Indian sub-continent occupying whole Bangladesh, Assam, Tripura, Myanmar and part of the Bay of Bengal (Fig. 1). The Barapukuria coal field is placed in the north western part of Bangladesh, which was primarily joined with Precambrian Indian landmass, South Africa, Antarctica and Australia building an extensive continental landmass named Gondwanaland (Imam, 2013). This coal is also called Gondwana coal because it was deposited within the Gondwana Supergroup. The coal in the basin was formed by the geologic processes of burial forest debris during the Permian (~250 million years) and coalification of buried organic matter by subsurface heat and pressure (Imam, 2013). The Barapukuria basin comprises an elongated asymmetrically half-graben type (Fig. 2, Norman, 1992), is thus equivalent to the Gondwana basins in India (Ward, 1984; Mishra, 1996), consists mainly of Precambrian Basement Complex, Early Permian GondwanaGroup, Late Miocene to Middle Pliocene Dupi Tila Formation, Late Pliocene to Pleistocene Barind Clay Formation and recent alluvium in an ascending order (Wardell-Armstrong, 1991). Stratigraphy of the Gondwana succession in the Barapukuria basin is summarized in Table I and Fig. 3.

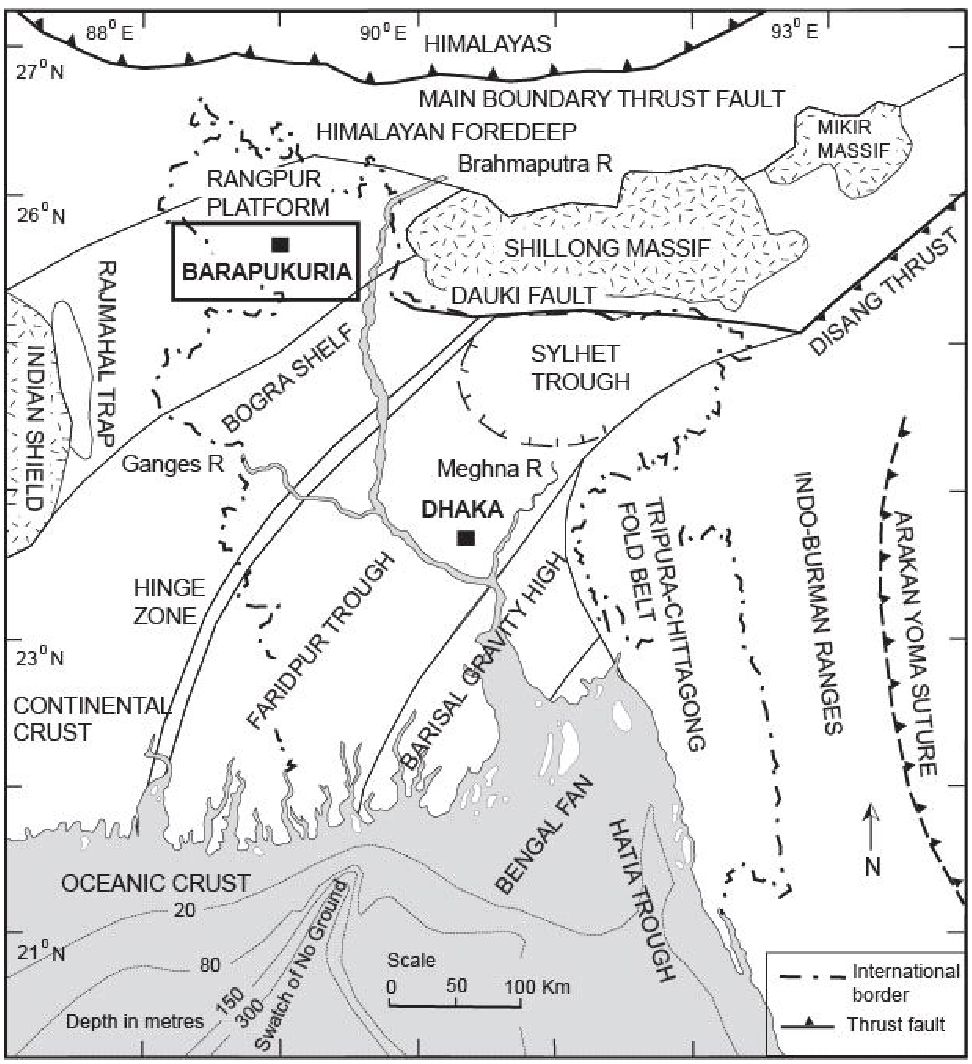

Fig. 1. Generalized map showing location of the Barapukuria coal field and major physiographic and geotectonic features of the Bengal Basin and adjoining areas (modified after Hossain *et al.*, 2013a)

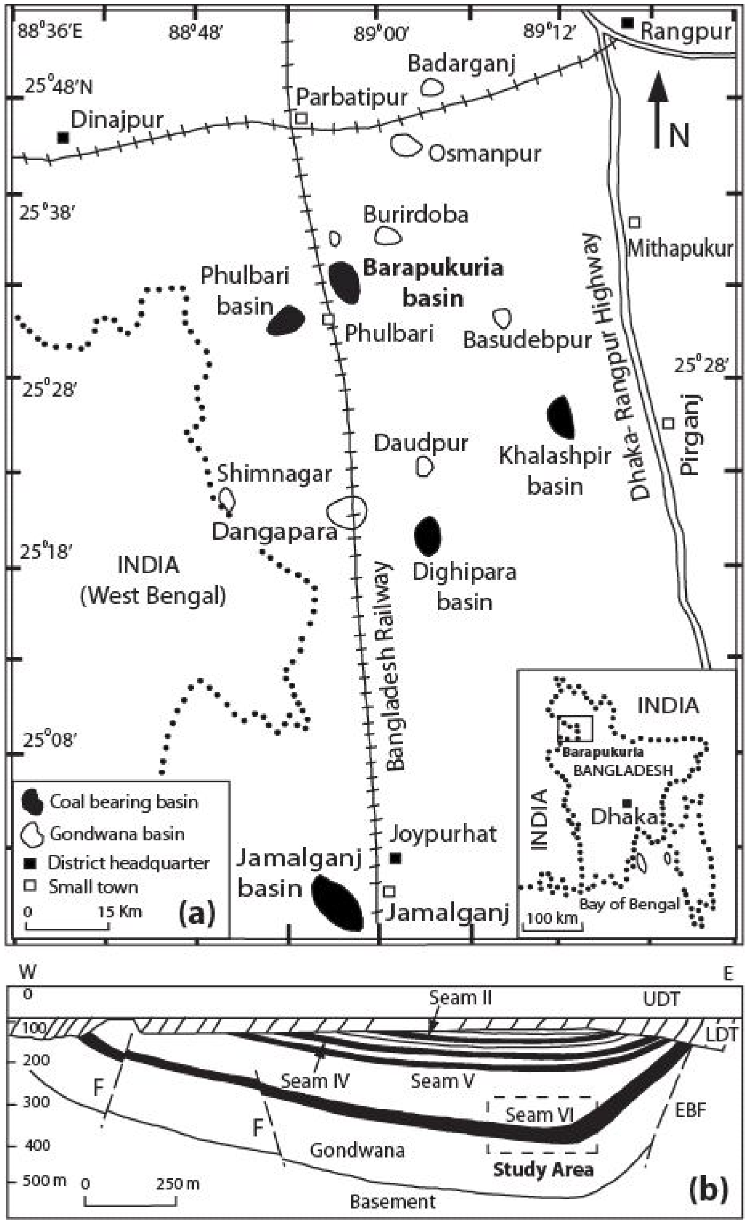

Fig. 2(a). Map showing the sampling site and distribution of major coal fields in northwestern Bangladesh (modified after Islam and Hayashi, 2008, Farhaduzzaman *et al.,* 2012), and (b) East-west crustal cross-section through the Barapukuria basin, northwestern Bangladesh (modified after Norman, 1992)

The Precambrian rocks from the basement of all geological formations referred to as a Basement Complex, consists mainly of granite, granodiorite, diorite and gneiss (Hussain and Curtin, 1995; Bakr *et al.,* 1996). The oldest sedimentary rock unit in Bangladesh is the Lower Gondwana Group of Permian age which unconformably overlies the Basement Complex (Fig. 3 and Table I). It is composed predominantly of feldspathic sandstone, carbonaceous sandstone, siltstone, shale with thick to thin coal seams and occasionally conglomerate (Bakr *et al.,* 1996; Islam and Hossain, 2006). The major coal deposits found in the northwestern Bengal Basin of Bangladesh are all encountered in this Gondwana Group. The quality and characteristic of this coal is very similar to the coal deposits of near by Gondwana basins of India (e.g. Raniganj, Jharia, Jharkhand etc., Norman, 1992). Lithofacies associations in the Gondwana succession of Barapukuria

Table I. Stratigraphy of the Barapukuria basin, northwestern Bangladesh (Modified after Bakr *et al.*, 1996, Hossain *et al.*, 2013a, b).

Age	Group	Formation	Member	Lithology	Max. Thickness (m)
Holocene		Alluvium		Sand, silt and clay	1.83
Pleistocene		Barind Clay		Clay and sandy clay	10.36
			Upper	Sandstone, pebbly sandstone and clay/mudstone	126.82
Pliocene		Dupi Tila			
			Lower	Sandstone, mudstone and white clay	
Permian	Gondwana			Feldspathic sandstone, carbonaceous sandstone and shale, ferruginous sandstone, conglomerates, coal beds	457.32
Precambrian		Basement Complex		Diorite, granodiorite, quartz diorite, granite and diorite gneiss	14.32+

basin suggest that the paleoenvironments changed from fluvio-glacial braided stream, peat-forming backswamp to comparatively low sinuous to moderately sinuous stream channel, and floodplain complexes (Islam and Hossain, 2006).

The Dupi Tila Formationis Pliocene in age and lie directly above the coal bearing Permain Gondwana Group. This group is sub-divided into a sandy Upper Dupi Tila Member and a clayey Lower Dupi Tila Member (Imam, 2013). The contact between the Gondwana Group and the Dupi Tila Formation is erosional. The overlying Braind Clay Formation is also known as Madhupur Clay Formation, which consists mainly of mottled clay, yellowish to reddish brown with ferruginous nodules (Bakr *et al.*, 1996). The Barind Clay Formation is unconformably overlying by alluvium of the Holocene in age consists primarily of sand, silt and clay.

Materials and methods

Eight fresh coal samples (ca. 100 g) were collected using a hand cutter from the coal seam-VI, Barapukuria coal field, Bangladesh (Figs. 2b and 3). Depths of the coal sample points range between 454 and 494 m from the surface. The coals are dark black to shiny black and brownish in color and brittle structure containing some laminations and chalcopyrite impressions. The coal samples are dried, and subsequent ly powdered using an iron mortar and pestle.

Total organic carbon (TOC), total hydrogen (TH), total nitrogen (TN), and total sulfur (TS) contents were determined by combustion method about 1800°C in a CHNS elemental analyzer (EA 1108). Approximately 5 mg of powdered coal sample was placed in a thin Ag film cup, and 1M-HCL added several times and dried at 110°C for 30 minutes. The dried coal sample was then wrapped in a thin tin film for combustion. TOC, TH, TN and TS data were calibrated against a BBOT standard (2, 5-Bis-(5-tert-butyl-benzoxazol-2-yl)-thiophen), and standard regression line method was employed for quantitative analysis. All analyses were carried out on a dry weight basis at Bangladesh Council of Scientific and Industrial Research (BCSIR) laboratory.

Results and disscution

Elemental compositions (C, H, N and S) of the studied coal samples are listed in Table II. TOC contents vary from 48 to 75 wt.%, with an average TOC content of 61 wt.%. TH and TN abundances for the coals range from 3.89 to 5.80 wt.% (average 4.59 wt.%,) and 2.87 to 3.87 wt.% (average 3.28 wt.%), respectively. The coals are characterized by low total sulfur contents and range from 0.19 to 0.37 wt.% (average 0.30 wt.%). The TOC to TN ratio (C/N) in the coal samples vary between 14 and 21 (average 19) and TOC to C S ratio (C/S) range between 156 and 376 (average 218) (Table II).

Table II. CHNS elemental analysis (wt.%) of Permian Gondwana coals in the coal seam-VI, Barapukuria coal field, northwestern Bangladesh.

Sample number	TOC (%)	TH (%)	TN (%)	TS (%)	C/N	C/S
BCMCL1	60	4.91	3.03	0.31	20	195
BCMCL2	62	3.89	2.91	0.29	21	215
BCMCL3	56	4.01	2.87	0.35	19	159
BCMCL4	63	4.50	3.05	0.37	20	169
BCMCL5	75	3.91	3.79	0.20	20	376
BCMCL6	50	5.80	3.67	0.19	14	265
BCMCL7	72	4.80	3.87	0.35	19	206
BCMCL8	48	4.92	3.01	0.31	16	156
Average	61	4.59	3.28	0.30	19	218

Origin of organic matter

The sources of organic matter within sediments and sedimentary rocks are important in understanding the roles of terrestrial as well as estuarine originated organic matter as sources of energy and nutrients to the terrestrial and marine systems. The origin of organic matter in sedimentary systems is often diverse, as they receive organic matter from both autochthonous and allochthonous sources such as phytoplankton, algae, bacteria, and terrestrial plants (Dale, 1974; Meyers, 1994, 1997). Abundances of TOC in the studied coal samples exhibit wide variation, ranging between 48 and 75 wt.%, while TN contents are mostly fall between 2.91 and 3.87 wt.% (Table II). Large variation of TOC in the studied coal seam-VI is likely to be derived from diverse origin of organic matter as well as periodic fluctuation of water tables during accumulation of organic matter in the peat-forming mires (Hossain *et al*., 2015). Generally, terrestrial plants are consisting of high carbon with low nitrogen (protein) and sulfur, while planktonic organic matters primarily supply nitrogen (Müller, 1977). (Tissot and Welte 1984) reported that higher plants are key organic producer in the terrestrial environment, and consist largely of cellulose and lignin which contain small amounts of nitrogen and sulfur. High TOC content in the coal samples indicate the influence of terrestrial organic matter to the peat bogs. The TOC in the coals show a positive correlation with TN (r = 0.51, Fig. 4a), and increase TOC with increasing TN contents, reflecting a comparable source material input to the paleomire.

The C/N ratiois widely used to the indicative of sedimentary organic matter, as it is distinguish between algal and terrestrial plant origins of organic matter (Meyers, 1994; Sampei and Matsumoto, 2001; Perdue and Koprivnjak, 2007; Hossain *et al.*, 2009). Terrestrial vascular plants and their derivatives in sediments have C/N ratios of greater than 15 (Ertel and Hedges, 1984; Sampei and Matsumoto, 2001) and those of wood, leaf and macrophytee specially high at 209, 28 and 39, respectively (Hedge *et al.,* 1986). Generally, planktonic organic matters deposited in deep-sea sediments contain C/N values higher than 10 to 15 (Müller, 1977) owing to the preferential loss of nitrogen relative to carbon (Müller, 1977). Perdue and Koprivnjak (2007) documented that terrestrially derived organic matter is rather depleted in nitrogen abundances. Planktonic organic matters predominantly supply nitrogen, therefore, freshly deposited planktonic organic matter is most reactive for sulfate reduction (Berner, 1984) and have the C/N ratio is usually between 6 and 9 (Bordowskiy, 1965). C/N ratio of planktonic organic matter in sedimentary environments increases with decomposition rate of the organic matter (Tissot and Welte, 1984). High cellulose and lignin composition of vascular terrestrial plants, along with low protein content, result in C/N ratio valueslarger than 15 (Meyers, 1994, 1997; Sampei and Matsumoto, 2001). The C/N ratios in the studied coal samples are 20 (BCMCL1), 21 (BCMCL2), 19 (BCMCL3), 21 (BCMCL4), 20 (BCMCL5), 14 (BCMCL6), 19 (BCMCL7), and 16 (BCMCL8), respectively, with mean C/N ratio of 19 (Table II). Most of the coals in the coal seam-VI have C/N ratios higher than 15, suggesting the influx of organic matter is mainly derived from terrestrial higher plants. However, the TOC also show a marked positive correlation with C/N (r = 0.70), suggests the dominance of vascular plants organic matter input to the peat-forming mire. The coaly organic matter of the Barapukuria basin is likely to be controlled by terrestrial higher plant materials (Farhaduzzaman *et al*., 2012; Hossain *et al.*, 2013a, 2015). This result is also consistent with the study of tri-aromatic hydrocarbons in Permian Gondwana coals and coaly shales from the Barapukuria basin, Bangladesh and concluded that the presence of substantial amounts of 1,7-dimethylphenanthrene, anthracene and methylanthracene in coals and coaly shales indicating a significant influx of land derived organic matter like coniferous

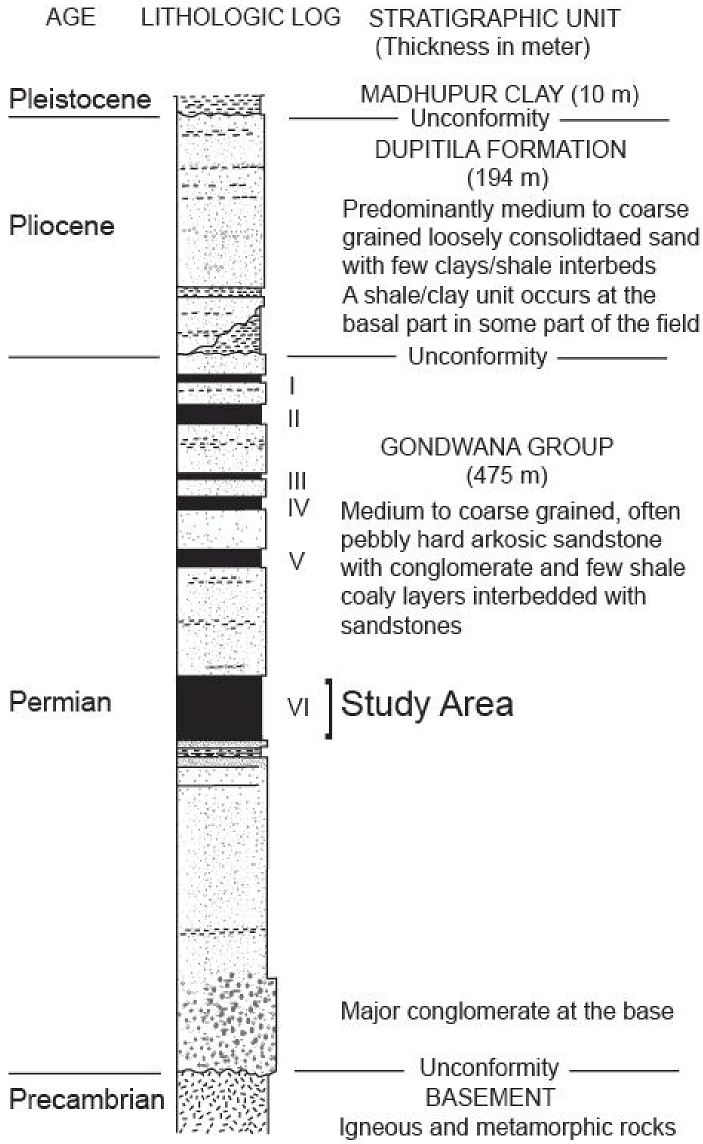

Fig. 3. Summarized lithologic log with major stratigraphic units of the Barapukuria coal field, northwestern Bangladesh (after Imam, 2013)

gymnosperms (Hossain *et al.*, 2013a). On the other hand, high C/N ratios ($\geq$12) are indicative of low biodegradation, and low C/N ratios (<12) are a characteristic of increased bacterial degradation (Hart, 1986). In the present study, C/N ratios fall in the range between 14 and 21 (Table II), reflecting lower degree of biodegradation.

Depositional environments

In this study, a schematic model has been developed based on relative abundances of TOC, TN, TS and C/S ratio sin order to decipher organic matter depositional environments of coal seam-VI of the Barapukuria basin (Fig. 5). The C/S ratios are widely used as an indicator for inferring depositional conditions of organic matter accumulated in marine or non-marine water-bottom environments (e.g. Berner, 1984; Sampei *et al.*, 1997; Hossain *et al.*, 2009). Since the biogeochemical cycle of sulfur in sediments is in separable from that of carbon, and the relationship depends largely on the depositional environment during or after burial of organic matter. Sediments deposited in marine environment have

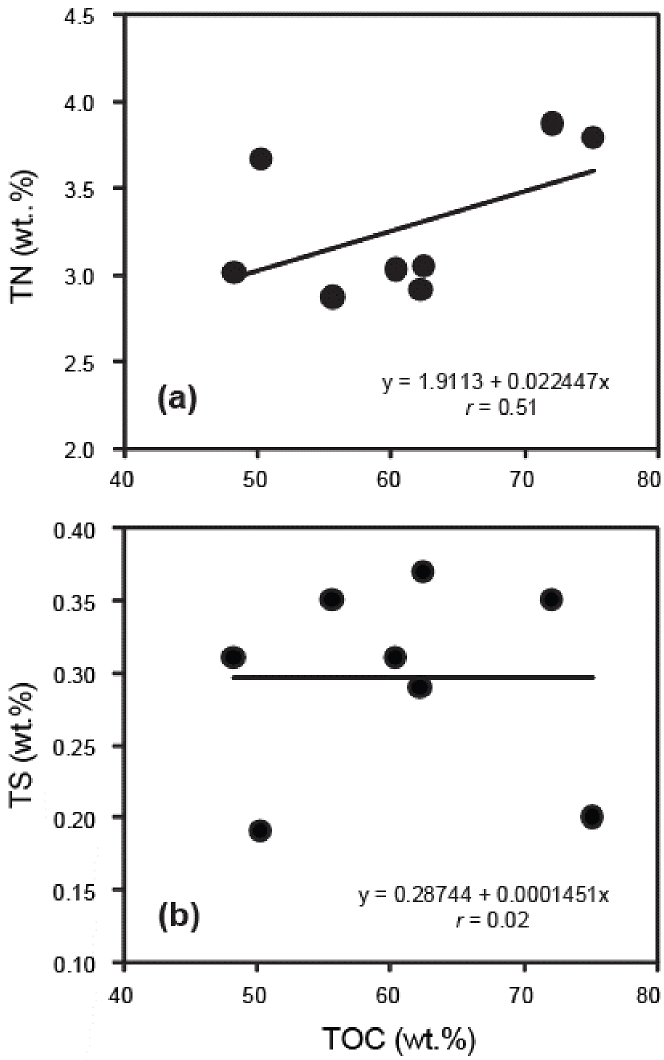

Fig. 4. Variation diagrams of TOC versus TN and TS with correlation coefficients for the Gondwana coals in seam-VI, Barapukuria coal field, north western Bangladesh

low C/S values of 0.5 to 5, whereas freshwater sediments have relatively high C/S values >10 (Berner and Raiswell, 1984). In the studied coals, sulfur content is relatively low (<0.5 wt.%), with average value of 0.3 wt.% (Table II), indicates a freshwater influence and suggests that the coaly organic matters in seam-VI were communication with a non-marine environmental conditions. Kumar *et al.* (2015) reported that the organic carbon and sulfur contents in the Permian bituminous coal from Jharia coalfield, India vary from 70 to 85 wt.% (average 76 wt.%) and 0.10 to 0.17 wt.% (average 0.14 wt.%), respectively with elevated C/S ratios of 403 to 704 (average 577). The Jharia coal basin may be attributed to deposition in fluvio-lacustrine environments (Dasgupta, 2005). In addition, Indian Permian coals were accumulated predominantly in swamps of medium wetness conditions (Mishra, 1996). However, TOC in this study show no correlation and scatter distribution with TS (r = 0.02, Fig. 4b). Some coal samples contained relatively slight elevated TS and low TOC contents (Table II). This sulfur probably considers inland lake environment and/or brackish coastal lakes with swamps, and perhaps the cyclic variation of climate over the region. However, presence of minor terrestrial palynomorp hacritarch (e.g. *Circulisporites, Peltacystia, Haplocystia, Leiosphaeridia*) in coal seam-VI signifi esinland basin with brackish water influence (Islam *et al*., 2003; Hossain *et al.*, 2015). On the other hand, increases in sulfur accumulation in lakes may be due ultimately to changing organic producers and/or decline to the sediment or water column from anoxic to oxic conditions (Peters, 1986). This interference is ascribed to be different sulfur sources input to the paleomires or possibly least bacterial degradation under oxic or anoxic events. Amijaya and Littke (2005) suggested that low sulfur content in coal is well known forrestricted sulfur input and / orabsence of sulfate-reducing bacteria in acidic conditionsduring organic matter deposition in peat-bogs.Accordingly, high sulfur content in organic mattertypically indicate marine environment where high amount of sedimentary pyrite deposit occurred (Tissot and Welte, 1984). The pyrite content in the studied coals is less abundant, implying that sulfate reducing bacterial activity to the paleomire was rather low. Freshwater lakes often describe the characteristics of vascular plant and planktonic organic matter inputs, where the sulfate levels are enormously low (Sampei *et al*., 1997). Furthermore, bacterial activity in marine and lake sediments is dominated by sulfate reducers, and the sulfur content is relatively higher in marine water than in freshwater (Hughes *et al*., 1995). C/S ratios in the coals vary from 156 to 376 (Table II), indicate that the depositional environment of the coal seam-VI is characterized by dominantly freshwater lake/swamp. In addition to the relatively high C/S ratios in Gondwana coaly shales occurs at thick coal seam-VI, Barapukuria basin support the inference of a freshwater lacustrine environment during organic matter preservation (Hossain *et al*., 2015). Hossain *et al.* (2015) also mentioned that lower part of the Gondwana succession was accumulated in dominantly oxic conditions with abun-

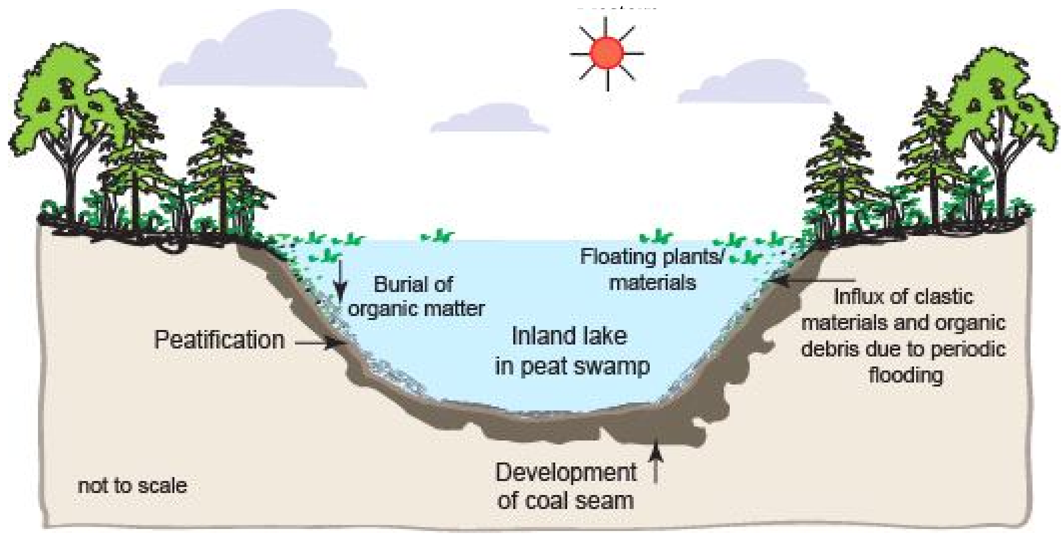

Fig. 5. Schematic diagram for deposition of coal seam-VI, Barapukuria coal field, north-western Bangladesh

dant input of terrestrial derived organic matter. Plant production is relatively high in areas of hot humid climatic conditions and is rather low in dry/cool climatic conditions. Large-scale swamps are normally connected with some sorts of rivers and lakes in arid or seasonally dry regions of the globe. Islam and Hossain (2006) stated that low- to moderately-sinuous stream channel is commonly associated with peat-forming mire complexes in the Gondwana Barapukuria basin. Organic matter in the channel sediments has normally high C/N ratios and substantially low sulfur contents (Hart, 1994). The low- to moderately-sinuous stream channel exhibits the inland lake or brackish coastal lakes environments, where the organic matter normally reflecting freshly deposited least planktonic matter with high contributions of terrestrial plants (Sampei *et al.*, 1997). Additionally, the abnormally thick coal seam-VI in the present study contained thin layers of clastic sediments. The clastic sediments in the coal seam are interpreted as fluvial origin. Therefore, the elemental compositions and ratio values suggest that the organic matter in the thick coal seam-VI, Barapukuria basin might be originated from higher land plants, and preserved in a diverse array of inland lake environments under fluvial regime.

Conclusion

Eight Gondwana coal samples of Permian age were collected from coal seam-VI of the Barapukuria coal field, north-Bangladesh have been investigated using CHNS elemental analysis to identify sources and depositional environments of organic matter. TOC, TN, TH and TS contents of the coal samples were ranging from 48 to 75 wt.% (average 61wt.%), 3 to 4 wt.% (average 3 wt.%), 4 to 6 wt.% (average 5 wt.%) and 0.19 to 0.37 wt.% (average 0.30 wt.%), respectively. The C/N and C/S ratio values range from 14 to 21 (average 19) and 156 to 376 (average 218), respectively suggesting land plant derived organic matter subsequently deposited in entirely terrestrial environments.

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