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Geochemical Characterization of Exposed Black Shale Sediments of Matamuhari Anticline, Bangladesh

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Abstract: Matamuhari anticline is one of the largest anticline situated at Bandarban district of Bangladesh which is 60 km long and 20 km wide in the part of Bangladesh. Like other part of Bangladesh the whole sequence of exposed rocks of Matamuhari anticline is composed with Neogene sedimentary rocks of shale, sandstone, siltstone, silty shale, tabular and spheroidal calcareous concretions. Seven outcrop black shale samples were collected during February 2013 from the different parts of the anticline namely Boro Bari Jhiri, Ochir Jhiri and Tak Chara area were mineralogically and geochemically examined in order to better understand the depositional environment in the studied area. The major and trace elements were analyzed using XRF spectrometry. Two distinct lithofacies assemblages, mud and silty-clay are found in the collected sediments. The shales are predominated by quartz, kaolinite, chlorite, mica (muscovite and biotite), feldspar (both K-feldspar and plagioclase), and in addition to a variable ratio of vermiculite. Lithologial characteristics and geochemical data demonstrated that sediments are enriched with TOC contents ranging from 0.39 to 0.67 wt.% and shows distinctive correlation to concentration of TOC and particle size distribution. These shales are characterized by a low Mg and K as well as high Al and Fe contents. Studies also show that the values of MgO/Al_2O_3 and K_2O/Al_2O_3 ratio ranges from 0.15 to 0.17 and 0.18 to 0.20 respectively. The relationship of log MgO/Al_2O_3 and log K_2O/Al_2O_3 values reveals that all of the studied samples fell within the marine environment. Ni/Co ratio of the studied shale samples ranges from 2.84 to 3.88 (average 3.36). These low values of Ni/Co ratio suggest that the sediments were deposited under oxic conditions. However, the above observations suggest that the sediments of Matamuhari anticline area were deposited in a shallow marine environment under oxic conditions.

Keywords: Matamuhari Anticline, Black Shale, Environment, Total Organic Carbon

1. Introduction

Matamuhari anticline is one of the largest anticline situated in the eastern and northern frontier hilly region of folded area of Bandarban, Bangladesh. Matamuhari Structure is about 150 km in length, but in Bangladesh part it is about 60 km in length and 20 km in width having axial trend along NNW-SSE direction. It is elongated asymmetric anticline, southward to the Rakhain area of Myanmar, which is complicated due to the longitudinal and transverse fault. Matamuhari anticline has an average elevation ranges from 245 m to 330 m in the northern part, but in the southern part it increases from 410 m to 570 m and the maximum elevation is 710 m in the south. Matamuhari anticline is uniqueanda little amount of work has been done on it. Geological survey party of Bangladesh Petroleum Exploration and Production Company Ltd. (BAPEX) worked there to find out the

geological feature, collect rock samples for petrophysical study [1]. Before 1999, the dispositional environment of the exposed rock throughout the southern fold belt of Bengal Basin of the Neocene Surma group was thought Deltaic to Shallow marine. However, [4-7] revises the earlier views and proposed that the lower Surma group succession represents an overall basinward propogradation from deep marine to coastal marine depositional settings. Based on sedimentological evidences [3] has suggested that the lower Surma group represents a slope apron the growth of which is thought to have been governed by westward migrating accretionary prism complex within the active margin setting of the Indo Burmese plate convergence. Comparable deep-sea clastics with thicker intervals of sandstone turbidities contained within a submarine canyon are present in the Miranja anticline. Alam, et al. (2003) also gives an overview regarding the sedimentary geology of the Chittagong-Tripura Fold Belt (CTFB) and shows a stratigraphic classification for the CTFB that has been established for four groups [2]. Matamuhari anticline fall into the Miranja Group which is analogous under the traditional upper Surma Group and this group has possibly the thickness ranges from 1200 m to 1600 m [2]. It is interesting to get more information about the shale facies as one of the most important oil and gas source rocks. Thereafter, this study will focus on (i) to draw a general picture of geochemical characteristics of exposed black shale sediments, and (ii) to better understand the depositional environment in the area studied.

1.1. Area of Investigation

The study was conducted in Lama, Alikadam and Naikhangchari Upazilla of Bandarban at South-Eastern part of Bangladesh. The studied area encompasses mostly Neogene Hills of Matamuhari anticline. The study area lies between 21°34' N to 21°45' N latitude and 92°04' E to 92°19' E longitude (Figure 1a). The area is bounded by Matamuhari River to the north and east, Cox's Bazar to the west and Rakhaing of Myanmar to the south (Figure 2). The accessibility of the area is trouble- free in the northern part of the structure, but very tough in the southern part. The study area is in tropical climatic region and receives heavy rainfalls during monsoon. The extent of rainfalls take place during the monsoon that prevails month of June to September. The maximum average temperature is about 32^0C which prevails in the month between April and May weather the minimum average temperature is 13^0C which prevails in the month of December. The intense rainfall is the main cause of developing abundant growth of vegetation in the study area (Figure 1b). The entire hilly area is covered with dense mixed forest with a variety of trees and wide number of bamboos, shrubs and herbs.
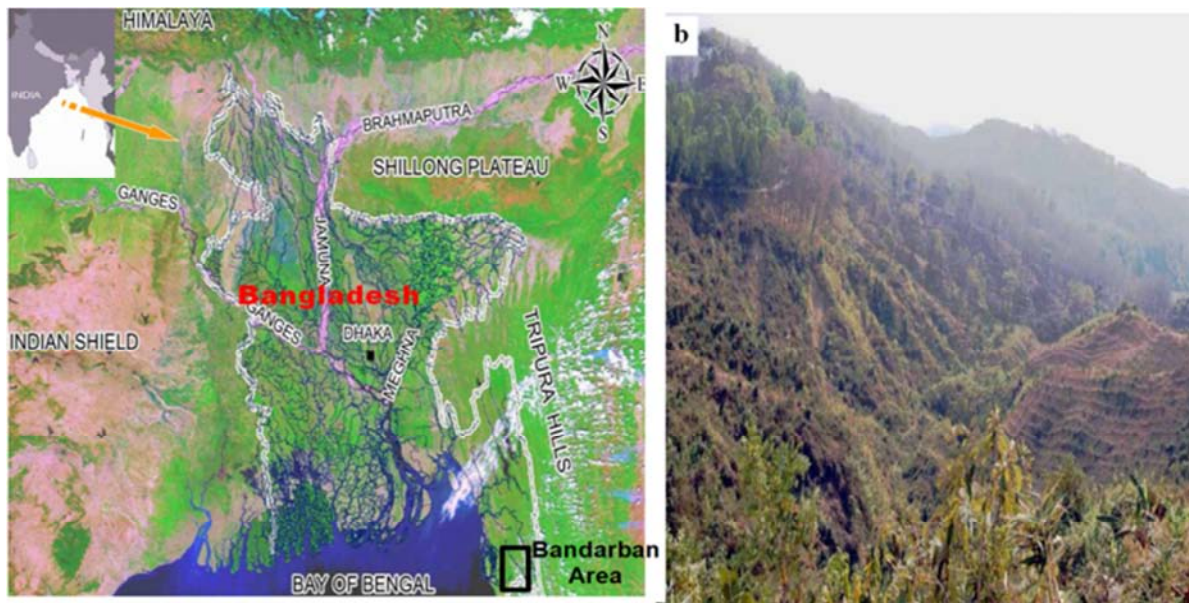
Figure 1. (a) Map showing the location of Bandarban study site in the South-Eastern part of the Folded Belt and location of the study area is indicated by a small rectangle;(b) Photograph showing the sky view of Neogene Hills of Matamuhari anticline.

1.2. Geological and Tectonic Setting of the Study Area

Matamuhari Structure is situated in the southern part of Eastern Folded Belt in Bengal Foredeep mainly comprises of Neogene Sedimentary rocks. It is located in between Bandarban structure in the east and Olatang structure in the southwest [8]. The development of Matamuhari structure is contemporaneous to the formation of Bengal Foredeep and is affected by faulting like as the other structure of Chittagong-

Tripura Fold Belt (CTFB) [4, 5]. The Matamuhari structure is narrow and elongated with steeper eastern flank than the western flank (Figure 2a & b). The tectonic and structural development of this region is more readily explained by accretionary prism formation [6]. Like other parts of Bangladesh the whole sequence of exposed rocks of Matamuhari structure is composed with Sedimentary rocks. The dominant lithology comprised of shale, sandstone, siltstone, silty shale, intraformational conglomerate and

tabular and spheroidal calcareous concretions of Neogene sediments [4, 5]. Argillaceous rocks occupy the bulk volume of stratigraphic columns with little coverage of arenaceous rocks. Details section wise lithology was studied and lithological columns were produced. However, the columns were correlated and divided into eight units primarily based on lithological features (Table 1). Those lithological units are Dupitila Sandstone, Girujan Clay, Tipam Sandstone, Upper Boka Bil Shale, Middle Boka Bil Sandstone, Lower Boka Bil Shale, Upper Bhuban Sandstone, and Middle Bhuban Shale from younger to older in order.

Table 1. *Correlation of the Local stratigraphy with traditional stratigraphy (personal communication, BAPEX).*

Regional (CTFB) Nomenclature				Nomenclature for the Matamuhari Anticline			
Age	Group	Formation	Thickness (m)	Age	Group	Formation	Thickness (m)
Recent	Holocene	Alluvium		Recent	Holocene	Alluvium	
	Dupitila	Dupitila	500		Dupitila	Dupitila Sandstone	300-520
Plio-Pleistocene		Girujan Clay	350	Pliocene - Pleistocene		Girujan Clay	160-410
	Tipam Group	Tipam Sandstone	900		TipamGroup	Tipam Sandstone	180-470
						Upper Bokabil Shale	260-550
		Bokabil	1200			Middle Bokabil Sandstone	230-700
Miocene	Surma Group			Miocene	Surma Group	Lower Bokabil Shale	370-860
						Upper Bhuban Sandstone	500+
		Bhuban	3000+			Middle Bhuban Shale	300-520
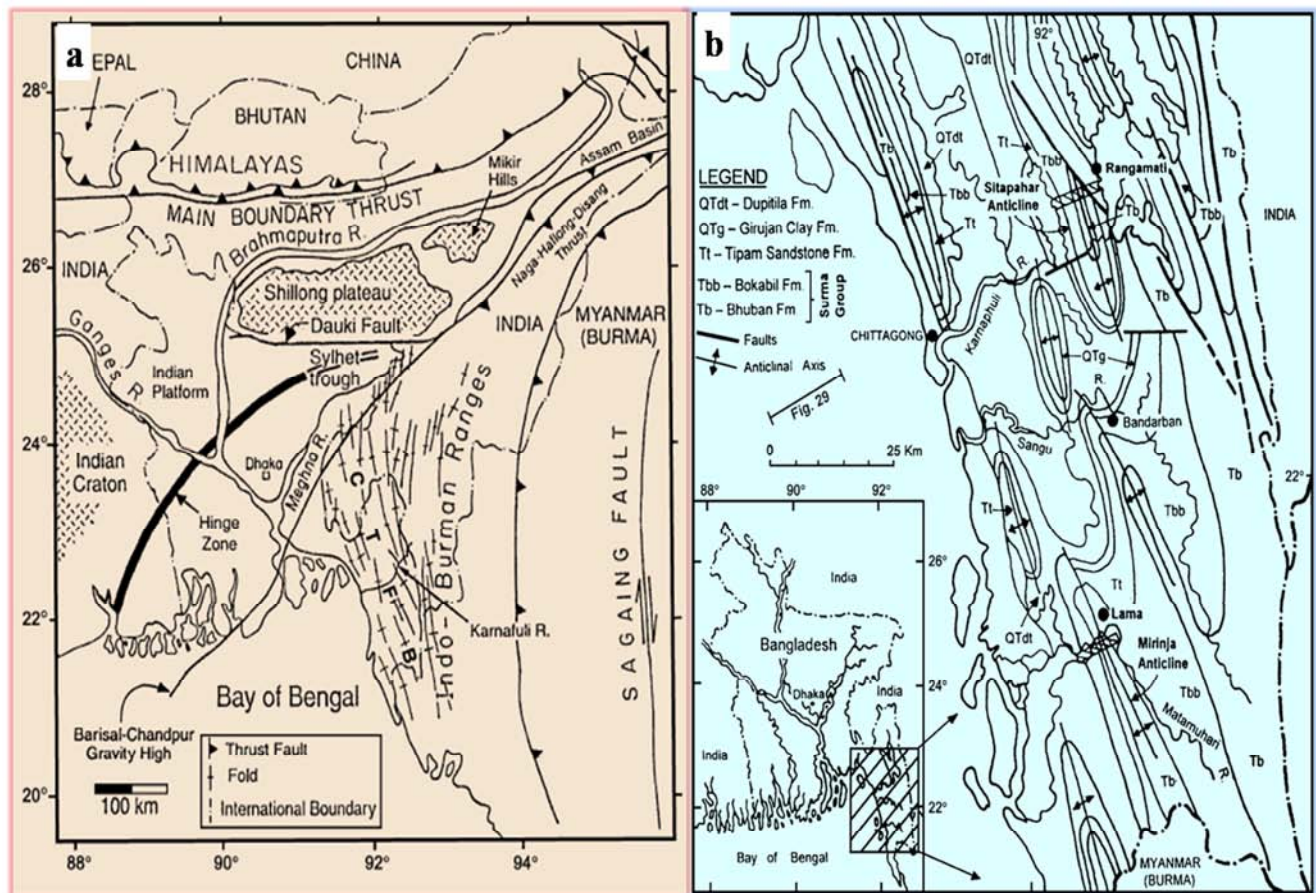
Figure 2. *(a) Regional tectonic setting of the Bengal Basin showing location of the study area within the Chittagong–Tripura Fold Belt (CTFB) (after Johnson and Alam, 1991);(b) Geological sketch map of part of the Matamuhari anticline and adjacent Structure (modified after Alam etal., 2003).*

2. Materials and Methods

2.1. Sample Collection

With the help of satellite image, the sampling location was selected and seven outcrop black shale samples were collected during Feb. 2013 from the different parts of the anticline such as Boro Bari Jhiri (TCB-01, BBJ-01), Ochir Jhiri (OJ-02, OJ-03) and Tak Chara (TC-01, TC-02, TC-03) area to the depth of 26cm below the surface to avoid weathered outcrop (Figure 3). Undisturbed sediment cores were collected from the Bokabil formation shale of Surma group by penetration of a 3.5cm diameter split-barrel sampler (Raymond sampler) by a private drilling company (Eastern

Geotechnical, Dhaka). Samples were immediately vacuum-packed in O_2-impermeable film bags (Escal film, Mitsubishi Gas Chemical Co., Japan) with deoxidizer. Later these sample were kept in the laboratory of Jessore University of Science and Technology freezer (-18°C) unit analysis.
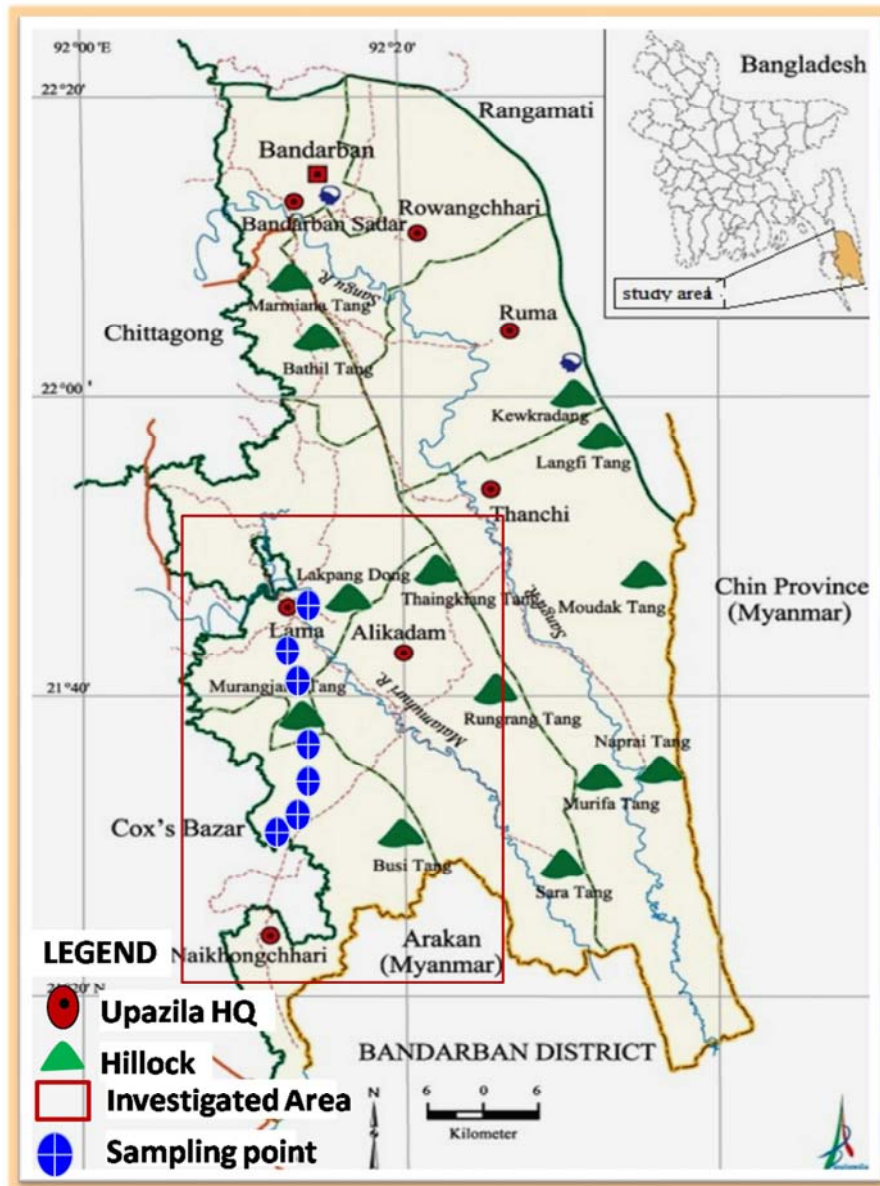
Figure 3. *Enlarged topographic map of the study area showing the location of sediment sampling sites (Figure modified after Banglapedia, http://en.banglapedia.org).*

2.2. Grain Size, Bulk Mineralogy, and Major Chemistry

Prior to analysis, about 50 g of undisturbed sediment core was immediately transferred from the freezer (-18⁰ C) to a vacuum drying oven (Advantec, VO-320) in order to avoid oxidation of the reduced compounds, and freeze dried. The dried samples were ground gently by hand with an agate mortar and pestle, and used for the analyses outlined. Grain size distribution was determined by laser diffraction and scattering spectroscopy (SALD-3000S, Shimadzu) after dispersion of the samples in sodium hexametaphosphate solution. The reproducibility of the results for the duplicate samples was within ± 5 % at maximum. Bulk mineralogy of the sediments was identified by X-ray powder diffraction

(XRD) using a Rigaku Geigerflex instrument (RAD-IA system) with Ni filtered Cu Ka radiation (30 kV, 10 mA), operating in step scan mode, over an angular range of 2⁰ to 65⁰ 2θ with 0.02⁰ 2θ steps and a 2s count time on 200 mg unoriented side-packed powder mounts. Approximate relative abundance ratios of major minerals were estimated from the relative intensities of the most intense and specific peak of each mineral from an XRD chart. The major chemical composition of bulk sediments were determined by X-ray fluorescence (XRF) photometry (VXQ-160S, Shimadzu) using glass bead samples, which were prepared by fusion of sediment samples with lithium borate (1:3 ratio). The analytical errors for major elements were within ± 3 %.

2.3. TOC Analysis

Total Organic Carbon of bulk sediment samples was measured by dry combustion according to a method described by [14] using an elemental analyzer (FLASH EA-1112 series, IRMS) after HCl digestion. Analytical precision for duplicate samples was within ± 5 %.

3. Result and Discussion

3.1. Sediment Characteristics

Two distinct lithofacies assemblages, mud (clayey-silt) and silty-clay are found in the collected sediments. The sediments consist dominantly of dark gray to black thinly parallel laminated clayey-silt with mean grain diameter of < 22 μm, except the sediments collected from Ochir Jhiriarea with mean grain diameter < 40 μm. The collected sediments were pale soft, sticky and soapy feel when wet.

3.2. Mineral Species and Their Relative Abundance Identified by XRD

Based on XRD analysis, abundant quartz was found in every sample, and clay minerals (mainly kaolinite), mica (muscovite and biotite), feldspar (both K-feldspar and plagioclase), chlorite and minor vermiculite were observed in the silty-clay sediments (Figure 4). Although the mineral assemblages in the clayey-silt sediments were similar to those of silty-clay sediments, but small amount of illite was also identified.
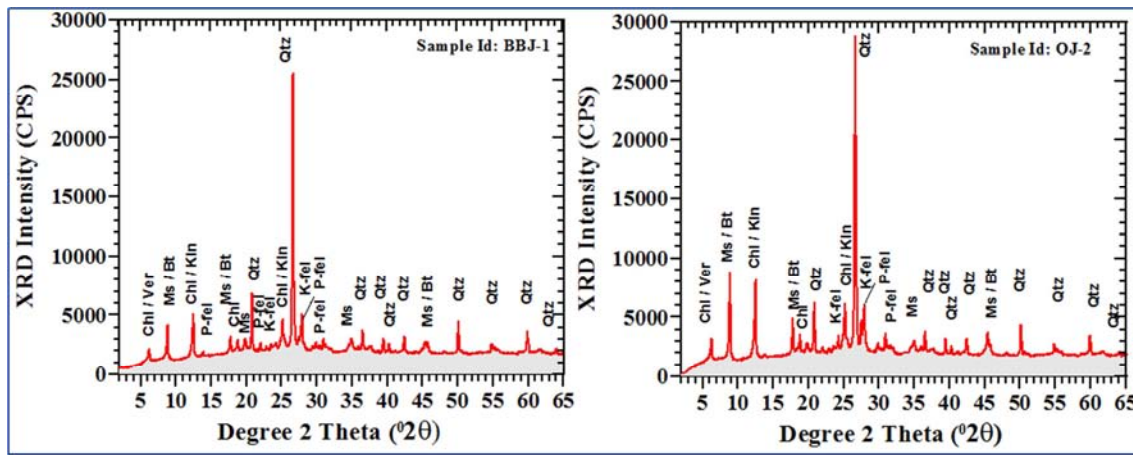

Figure 4. *Powder X-ray diffraction charts of two typical silty-clay (Sample Id: QJ-2) and clayey-silt (Sample Id: BBJ-1) sediments. Qtz: quartz, Bt: biotite, Ms: muscovite, Kln: kaolinite, Chl: chlorite, Ver: vermiculite, Kfel: Potash feldspar, P-fel; Plagioclase feldspar.*

3.3. Organic Richness

The organic richness represented by the total organic carbon (TOC) ranges from 0.39 to 0.67 wt.%. The studied samples lie in the poor to fair organic richness of the studied samples zone (Figure 5) [8, 11, 12]. It is also noted TOC is generally low in the studied sediment column, in which most of the samples contain < 0.50 wt.% except one sample (TC-01), which is rich in TOC with the maximum concentration of 0.67 wt.% and no intervals rich in carbonaceous matter (e.g., peat) were apparent in the studied samples.
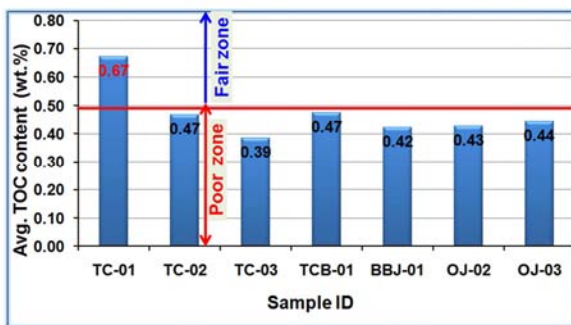
3.4. Major Inorganic Geochemistry

Geochemical signatures of clastic sedimentary rocks provide important sources of information that record different aspects of provenance and environments of deposition. The frequencies of the chemical components of the studied shale samples are shown in (Figure 6). Chemical composition was associated with mineral composition. As shown in Figure 7, the content of major inorganic constituents of all of shale samples is almost similar and uniformly distributed.
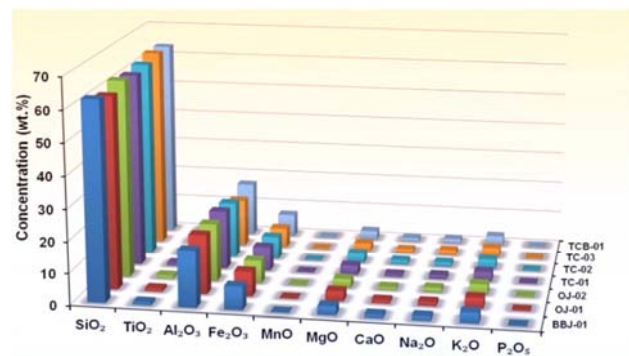

Figure 5. *Organic richness i.e., average TOC wt.% content in the studied sediments.*

Figure 6. *Showing the concentration of some major inorganic components of the studied sediments.*

Comparison of these data with the published average upper continental crust (UCC) [15]. (Table 2) illustrates that the studied shales are within the range of SiO_2, Mn, and Mg; with higher contents of Al, Ti, and Fe while their contents of Ca, Na and K are lower than the UCC data.

The higher Al and Fe content may indicate fineness in grain size and presence of clay minerals, especially kaolinite and chlorite; while the lower content of K may indicate marine condition [12].

Table 2. Major and trace element concentrations of the studied shale samples. Average upper continental crust (UCC) from [15].

Elements	BBJ-01	OJ-01	OJ-02	TC-01	TC-02	TC-03	TCB-01	UCC(wt.%)
SiO_2(wt.%)	62.83	60.91	62.85	61.8	62.52	63.7	63.41	66
TiO_2(wt.%)	0.9	0.95	0.92	0.95	0.91	0.8	0.93	0.5
Al_2O_3(wt.%)	17.95	18.84	18.44	18.8	17.82	15.14	17.07	15.2
Fe_2O_3(wt.%)	7.42	7.91	7.29	7.74	7.21	6.27	7.21	4.5
MnO(wt.%)	0.08	0.13	0.1	0.11	0.1	0.08	0.11	0.1
MgO(wt.%)	2.77	3.12	2.8	2.96	2.98	2.42	2.87	2.2
CaO(wt.%)	0.98	1.02	0.86	0.8	1.36	1.15	1.09	4.2
Na_2O(wt.%)	1.29	1.25	1.38	1.26	1.37	1.51	1.47	3.9
K_2O(wt.%)	3.21	3.38	3.3	3.09	3.13	2.96	3.12	3.4
P_2O_5(wt.%)	0.14	0.16	0.15	0.15	0.15	0.14	0.17	-
Co(ppm)	22.2	21.1	20.5	22.5	18.5	18.2	19.2	10
Ni(ppm)	76	81.9	75.2	83.2	65.8	51.6	71	20
Ni/Co	3.42	3.88	3.67	3.7	3.56	2.84	3.7	2

4. Depositional Environment

The relationship between MgO/Al_2O_3 and K_2O/Al_2O_3 was used by Roaldest (1978) to differentiate between marine and non-marine clay [13]. The value of MgO/Al_2O_3 and K_2O/Al_2O_3 ranges from 0.15 to 0.17 0.18 to 0.20 respectively. Application of this relation on the studied black shales (Figure 7) revealed that all the studied samples fell within the marine environment. According to Jones and Manning (1994) values of Ni/Co ratio ranges from below 5 indicate oxic environment whereas values above 5 suggest suboxic and anoxic environment [10]. Ni/Co ratio of the studied shale samples ranges from 2.84 to 3.88 (avg. 3.36) (Table 2). These low values of Ni/Co ratio suggest deposition under oxic conditions. Thereafter, it is suggested that the studied shales were deposited in a shallow marine environment under oxic conditions.

5. Conclusion

Two distinct lithofacies assemblages, i.e. clayey-silt and silty-clay are found in the collected Bokabil black shales sediments. Lithological characteristics and geochemical data demonstrated that the sediments are enriched with TOC contents ranging from 0.39 to 0.67 wt.% and show distinctive correlation to concentration of TOC and particle size distribution. The mineralogical composition of the studied sediments is predominated quartz, kaolinite, chlorite, mica (muscovite and biotite), feldspar (both K-feldspar and plagioclase), and in addition to a variable ratio of vermiculite and illite. These shales are characterized by a low Mg and K as well as high Al and Fe contents indicating that they were formed through intensive weathering of intermediate and mafic igneous rocks [12]. Mineralogical and geochemical studies (organic and inorganic) indicate deposition of these shales under a shallow marine environment with some terrestrial input. The studied shales have poor to fair organic richness. They indicate the capability to produce gas and fair oil source.
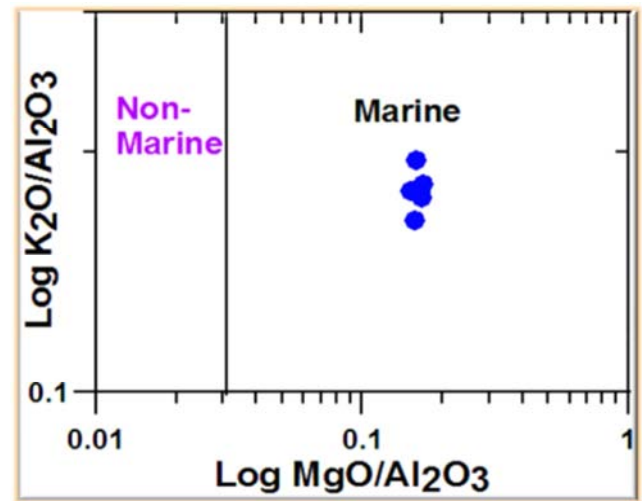
Figure 7. Depositional environment diagram on log(MgO/Al_2O_3) and log(K_2O/Al_2O_3) (after Roaldest,1978).

Acknowledgements

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