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DISTRIBUTION OF SULPHUR IN PALEOGENE COALS OF NORTH-EAST INDIA AND ITS PALEO-ENVIRONMENTAL IMPLICATION

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ABSTRACT

The present paper entails theoccurrence and distribution of different forms of sulphur in Paleogene coals of North-eastern India. All the coal seams present in the North-eastern region of India are characterized by the presence of high sulphur with total sulphur content ranging from 3 to 7%. Among all the forms of sulphur recognised, organic sulphur is dominant. The total sulphur content originated in the Platform area for Eocene coal under stable shelf condition vary from 4.20 % to 6.01 % for Jaintia Hills, from 2.78 % to 4.01 % for Khasi Hills and from 1.90 % to 3.00 % for Garo Hills, whereas Oligocene coal evolved under the foredeep basin have Total sulphur ranging in between 2.90 % and 6.60 %. In Eocene coals, there is a distinct lateral and vertical variation of sulphur i.e. sulphur content increases from the bottom to the top seam and also from western side of Meghalaya to the Eastern side. This lateral variation of sulphur was because of the more marine nature of the Eastern Meghalaya than Western Meghalaya. Both Eocene and Oligocene coals have derived from seawater as evidenced by the presence of Pyritic form of sulphur. Study of forms of sulphur also suggests that the deposition of coals in a different part of the region is influenced by roof strata, peat-forming plant communities, tectonic uplifting, and marine or freshwater incursion.

Key Words : High sulphur, Paleogene coal, North-East India, Foredeep basin, Platform area

INTRODUCTION

In the Paleogene coals of India, the occurrence of sulphur attracted the attention of early workers[1-3]. The large deposits of the Paleogene coals are mainly distributed in the states of Assam, Arunachal Pradesh, Nagaland and Meghalaya. Considering the Gondwana coals of India the Paleogene coals contribute only a meagre portion of the total Indian coal inventory[4]. But the Paleogene coal deposits are of good quality having characteristically low ash content and medium caking property. The only drawback of this coalis the presence of a highamount of sulphur (inorganic constituent) which render it unsuitable for commercial utilization. However, energy demands have increased recently, which focused attention on the utilization of high sulphur coals.

Sulphur is an undesirable but economically important constituent of all coals. The amount of sulphur in coal ranges from traces to as high as 10% or more. The maximum permissible amount of sulfur existing in fuels demonstrates a descending trend on a global scale that raises the importance of accurately measuring the total amount of sulfur in coals and its desulphurization steps, if required[5-8]. Inorganic, as well as organic forms of sulphur, remain present in coal. Combustion of sulfur-containing fossil fuels, especially coal/lignite and oil, by boilers and industry emits over 90% of atmospheric SO_2. At high temperature, the organic sulfur compounds of coal degrade to elemental sulfur or inorganic sulfur species, such as hydrogen sulfide, and further converted to SO_2. This SO_2 get oxidized to sulfur trioxide (SO_3), which combines with water vapourto produce ultrafine (<0.1 μm diameter) sulfuric acid particles[9]. Sulphur in coals of north-eastern region of India occurs as 1) Pyritic sulphur 2) Sulphate sulphur and 3) organic sulphur. Pyritic and sulphate sulphur together commonly referred to as inorganic sulphur. Iron sulphide (FeS_2), the primary inorganic form of sulphur, occurs in two crystalline forms, pyritic (cubic) and marcasite (orthorhombic).The major classes of organic sulphur include thiols, disulfides,organic sulfides, polysulfides, thiophene derivatives and sulfonates[10]

The sulphur content of coal seams is an important factor in resource development and utilization. The studies have shown that the coals with marine roof rocks have higher sulphur contents than those with fresh or brackish water roof rocks[11,12].

The coal deposits of north-east India dealing mainly with sulphur have been studied from time to time, but the earlier studies lack systematic sampling and regional approach. Mention may be made of the work of worth meaningful[13-16].

The present study is mainly focused on the determination and detailed description of the amount and occurrence of the different forms of sulphur(total, pyritic, sulphate and organic)in the coals of north-east India. Lateral as well as the vertical distribution of the sulphur forms has also been discussed with emphasis to the paleoenvironmental aspects of these coals.

Geological setting

In north-eastIndia, coal deposits occur in the states of Assam, Arunachal Pradesh, Nagaland and Meghalaya mainly **(Fig. 1)** confined to the Oligocene and Eocene arenaceousFormations. The coal deposits of these areasare understood to have been formed in two distinct tecto-sedimentary settings. The coal deposits of Assam, Arunachal Pradesh (Namchik-Namphuk coalfield) and Nagaland (Borjan coalfield) have probably originated in a foreland basin[17], whereas the coalfields of Garo, Khasi, and Jaintia Hills of Meghalaya represent the development of coal facies over platform areas[18]. In the Garo, Khasi and Jaintia Hills of Meghalaya the coal seams occur in the LakadontgSandstone Formation of Jaintia Group **(Table 1)** and are sandwiched between the overlying umlatholoh limestone and underlying Lakadong Limestone. The result of intermittent marine transgression and regression during the Eocene period has created the deposition of these formations[19].

Drifting of the Indian plate during the Cretaceous period towards north and north-east and finally, its collision with the Burmese plate (Asian plate), which proceeded with the subduction of Indian plate margin in the foredeepare believed to be the result of such activity. The development of theforedeep (trench/ technogenic) is because of subduction, which provided the site for deposition of Tertiarysequencesofstupendousthickness (~15,600 m). In the Early Eocene period the sedimentation began with the deposition of DisangGroupfollowed by Barail Group of Oligocene age **(Table 2)**. In the Tikak Parbat Formation of Barailgroup, the coal seams of the fore deeps occur **(Fig. 1)**. Alternate bands of Sandstone, shale and carbonaceous shale is the lithology of the Barail Group. The Tikak Parbat Formation of Barail Group is disturbed more tectonically than the underlying BorgagolaiFormation (alternatingsandstone and shale with carbonaceous shale and thin lamination of coal) and Naogaon Formation (splintery, grey or brownishcoloured, iron-stained shales with occasionally interbedded thin bands of fine-grainedsandstone and sandy shale)[15].

MATHODOLOGY

Coal samples [pillar/channel/run-of-mine (ROM)] were collected from different collieries of the coalfields of Assam (Dilli-Jeypore), Arunachal Pradesh (Namchik-Namphuk coalfield), Nagaland (Borjan andMoulong-Kimong) and Meghalaya (Garo, Khasi and Jaintia Hills) covering whole Paleogene coals of north-east India. To cover the entire north eastern coals sulphur studies done by different workers have also been taken into account. The data of Sulphur for West Daranggiri coalfield has been collected from Phukan[20], for Makum coalfield the data was collected from[21] and for Tiru coalfield the data was taken from[22].

The lithology of the seam, roof and floor strata, intervening dirt band and partings were also studied while collecting the samples from the collieries.

The samples were prepared to pass a 72 mesh (212 µm) sieve. The total sulphur was determined by digesting the coal with Eschka mixture containing 2 parts of MgO (Magnesium Oxide) and 1 part of Na_2CO_3 (Sodium Carbonate).

The sulphur was extracted by using (barium chloride) to precipitate $BaSO_4$ (Barium Sulphate) from solution. The total sulphur was then determined by the gravimetric method. The sulphate sulphur concentration was determined by treating the coal samples with dilute HCl and the concentration of the combined pyrite and sulphate sulphur fraction was determined by treating the coal with dilute nitric acid. The organic sulphur was calculated by subtracting total sulphur from pyritic and sulphate sulphur.

RESULTS AND DISCUSSION

The results of various forms of sulphur of Paleogene coals of north-eastern regions of India are furnished in **Table 3, Table 4, Table 5, Table 6, Table 7** and **Table 8** (average value). These coals are high sulphur, which varies from 7.03% to 1.62% (with an average of 2.98%). Coal with less than 1% sulphur is placed under low-sulphur coals. Coal[23] with 1% to <3% sulphur is medium sulphur coal and coal with ≥ 3% sulphur is considered high sulphur coal. Forthecoalfields of north-east India, all the coals contain sulphur more than 3% (except, few samples) and therefore placed under high sulphur coal category. Due to the complex nature of organic sulphur, its desulphurization is difficult while chemical desulphurization processes help in reduction of aliphatic sulphur structure but to achieve a high desulphurization rate biological techniques are useful[24]. These coals have high organic sulphur compared to inorganic sulphur.

Coalfield of platform basins

The coalfields of platform areas are shown[19] in **Table 9** however mining activity is confined to the areas listed in **Table 3, Table 4** and **Table 5**.

The total sulphur content of Eocene coals (Meghalaya) varies from 1.62 to 7.03% **(Table 3, 4, 5)**. In the Jaintia Hills **(Table 3)** the total sulphur content ranges from 3.57 to 7.03%, i.e. all the samples showa value above 3% and the coals are classified as high sulphur coals. The sulphate sulphur of the coal is 0.21 to 0.78%. The pyritic sulphur varies from 0.14 to 1.21%. The organic sulphur from 2.69 to 5.04%.

In the Khasi Hills **(Table 4)**, the total sulphur varies from 1.62 to 5.01% (average). Although all the samples show sulphur content above 3% except 3 samples ofMawhehlakhan area, where the value is slightly above 1. The sulphate sulphur varies from 0.11 to 0.94%, pyritic sulphur- 0.09 to 0.94% and organic sulphur- 1.31 to 3.91%.

In the Garo Hills **(Table 5)**, only 2 working coalfields are exposed. The total sulphur content of Garo Hills coals varies from 1.8 to 5.2% having sulphate sulphur fromtrace to 0.4%,Pyritic sulphur trace to 1.9% and organic sulphur from 0.8 to 3.2%.

The sulphur percentage of the bottom seam is less than 3 and is placed in the medium S category. All other coals are classified as high S coal and only a few samples have characteristics of medium sulphur coal.

From the sulphur study of coals of Jaintia, Khasi, and Garo Hills, it is revealed that there is a lateral and vertical variation of total sulphur concentration which increases from the bottom seam to the top seam **(Table 3, Table 4** and **Table 5)** having highest amount in Jaintia Hills followed by Garo Hills and Khasi Hills.The seams at the top have a higher content of sulphur than that of the seam at the bottom. The Bapung and Jaintia coalfields of Jaintia Hills are located in the Eastern part of Meghalaya whereas West Daranggiri and Siju coalfields of Garo Hills are located in the western part of Meghalaya.

Stratigraphically Bapung coalfield belong to the Lakadong Sandstone member of Shella Formation of Jaintia Group of Lower to Middle Eocene age and is the oldest in Meghalaya whereas West Daranggiri and Siju coalfields belong to the Tura Formation of Lower Eocene age **(Table 1)**.

Pyritic sulphur content is found to increase from west to the eastern part of Meghalaya, which is mainly due to the prevalent marine condition at the time of the deposition in the eastern part of the basin[25] Coals of Meghalaya are characteristically higher in sulphate sulphur content. High sulphate sulphur content in coals of Meghalaya generally occur with thin overburden, which suggests that the high sulphate sulphur may be due to the weathered nature of coal[14].

The regional lateral variation of coals of Meghalaya is strictly a palaeo environmental effect. In other words, the increase in sulphur content from western to eastern is due to the more marine nature of the peat-forming swamps of Khasi and Jaintia Hills as compared to that of Garo Hills[14].

The coalfields of foreland basins

The coalfields of foreland basins are placed[19] in **Table 10**. These fields are confined to the states of Assam, Arunachal Pradesh and Nagaland.

The principal coalfield of Oligocene coal is the Makum coalfield of Assam **(Table 6)** having main collieries,Tipong, Ledo, Borgolai, Tirap and Namdong. There are 5 seams known as 60 ft., 20 ft., 8 ft., 5 ft., and New seam present in the area. The sulphur content varies as follows- total sulphur- 2.20 to 6.88% (average 4.43%), sulphate sulphur 0.04 to 0.95% (average 0.64%), pyritic sulphur 0.15 to 1.90% (average 1.01 to 1.1) and organic sulphur 1.60 to 5.76% (average 3.38 to 3.18%). In general, sulphate sulphur is lesser than another form of sulphur. There is no uniformity in the variation of sulphur in the seam both in the lateral and vertical direction.

The other coalfield of Assam in Jeypore **(Table 6)** and Dilli coalfield from there run-of-mine samples were collected and analyzed which contain total sulphur from 2.8 to 6.2%, sulphate sulphur from 0.7 to 4.3%, pyritic sulphur from 0.75 to 1.43% and organic sulphur from 1.9 to 4.02%.

Namchik-Namphuk coalfield of Arunachal Pradesh also contains sulphur inhigh amount withtotal sulphur varying from 2.8 to 3.8% **(Table 8)**. Sulphate sulphur rangesfrom 0.41 to 0.94%, pyritic sulphur 0.85 to 1.50% and organic sulphur 1.54 to 3.36%.

The sulphur content of Nagaland coalfield **(Table 7)** isalso high. For, Borjan Coalfield the total sulphur content ranges from3.75- 5.52% with sulphate sulphur in the range of 0.24 to 0.73%, pyritic sulphur 0.89 to 1.01%, and organic sulphur- 2.62- 3.78%.

For, MoulongKimang coalfield, the Total sulphur varied in between 2.89 and 3.01% with sulphatesulphur from 0.53 to 0.64%, pyritic sulphur 0.61 to 0.94% and organic sulphur- 1.84 to 1.93%.

In the case of Tiru coalfield,the total sulphur varies[22] from 6-11%(Singh et al. 2012), which is quite high of all the coalfields of north-eastern region, which may be classed as super high organic sulphur (SHOS) coals as a special class of coal that is remarkably enriched in organic sulphur, usually in the range[23] of 4 to 11%.

Sulphur is generally rich in marine influenced coals as observed by Teichmuller[26]. This observation is further supported byPrice and Shieh[27], Sinninghe Damste and De Leeuw[28] and Chou[29] (1990), where they showed that coals usually more than 1% sulphur comes from theseawater. A similar situation prevails in case of Oligocene coals of North-East India, which have a relatively high sulphur content ratio. A relative abundance of different forms of sulphur in coals of the study area is presented in **Fig. 2**.

Sources of sulphur

In coal, sulphur (S) primarilyoriginate from sea water, fresh water, vegetation and extraneous mineral matter. During (syngenetic) or after (epigenetic) coal formation secondary sulphur can be introduced by ground water, which is probably remobilizing sulphur that originated in sea water or as loosely held organic in the vegetation[30]. The study observed that fresh water does not contribute any sulphur in coal as they contain 0 to 10 ppm sulphur. However, their observation conducted that coal about 0.5% sulphur was probably derived from sea water which contains average 0.265% SO_4 or 885 ppm S (SO_4 contain 33.4% S). This is substantiated further by Casagrande (1987) that the marine-influenced peats generally have a higher sulphur content. Similar conditions prevail in North-Eastern coals where sulphur content is more than 3% (excepta few samples) and higher sulphur content is probably derived from seawater.Liang[31] also found that the marine biota release organic sulfur compounds, such as dimethylsulfide (DMS), to the marine boundary layer.Organic sulfur may be a residue of sulfur in proteins of the peat-forming plant communities or may be bounded with organic substances by bacterial activity whereas pyritic sulphur and sulphate sulphur formed due to chemical reactions involving iron, sulphur and other chemicals present in swamp water[32]. A triangular plot showing the distribution of different forms of sulphur in Paleogene coals are presented in **Fig. 3**. The **Fig. 4** clearly shows the dominance of organic form of sulphur in all the coalfields.

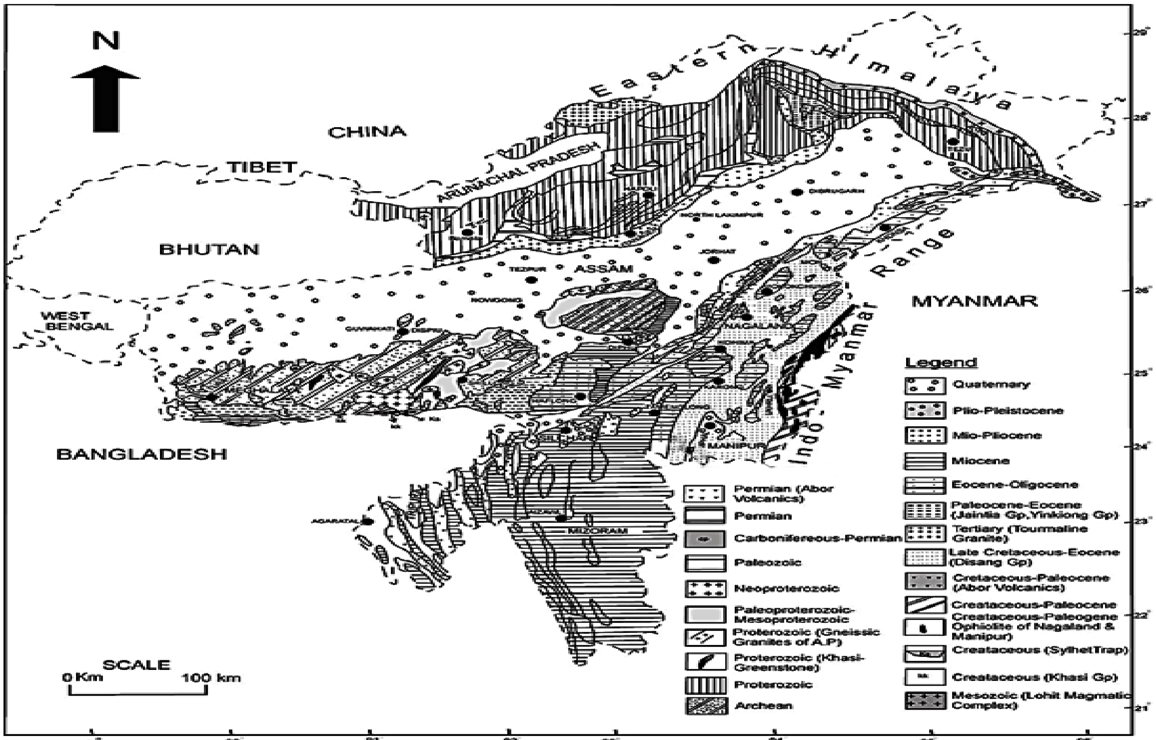

Fig. 1 : Geological map of North-Eastern India[26]

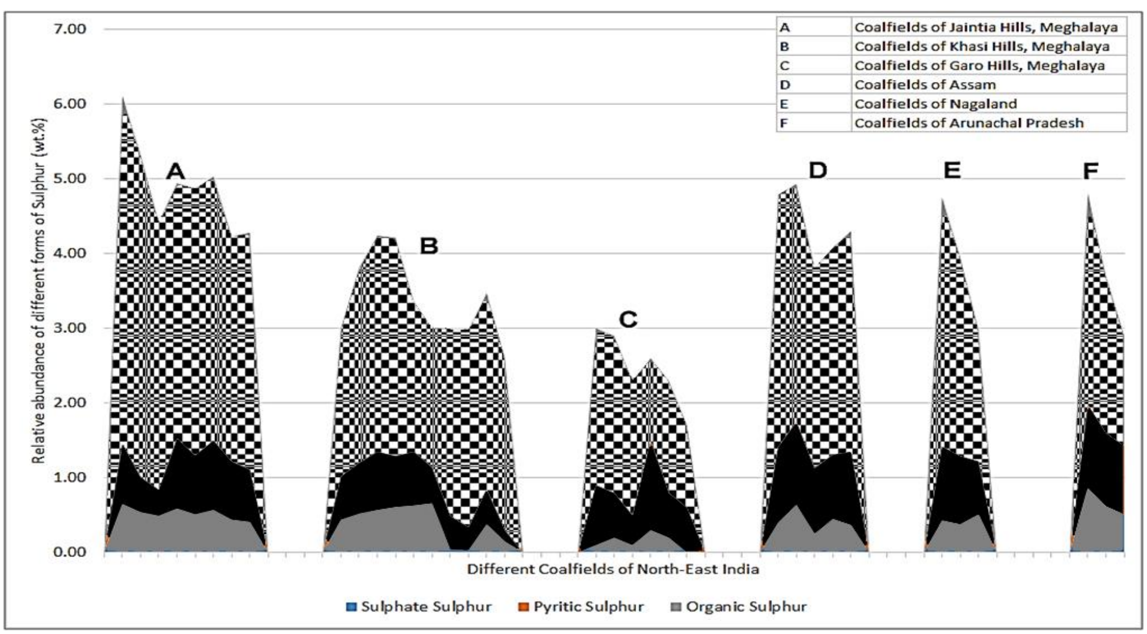

Fig. 2 : The relative abundance of different forms of Sulphur in different coalfields of North-East India

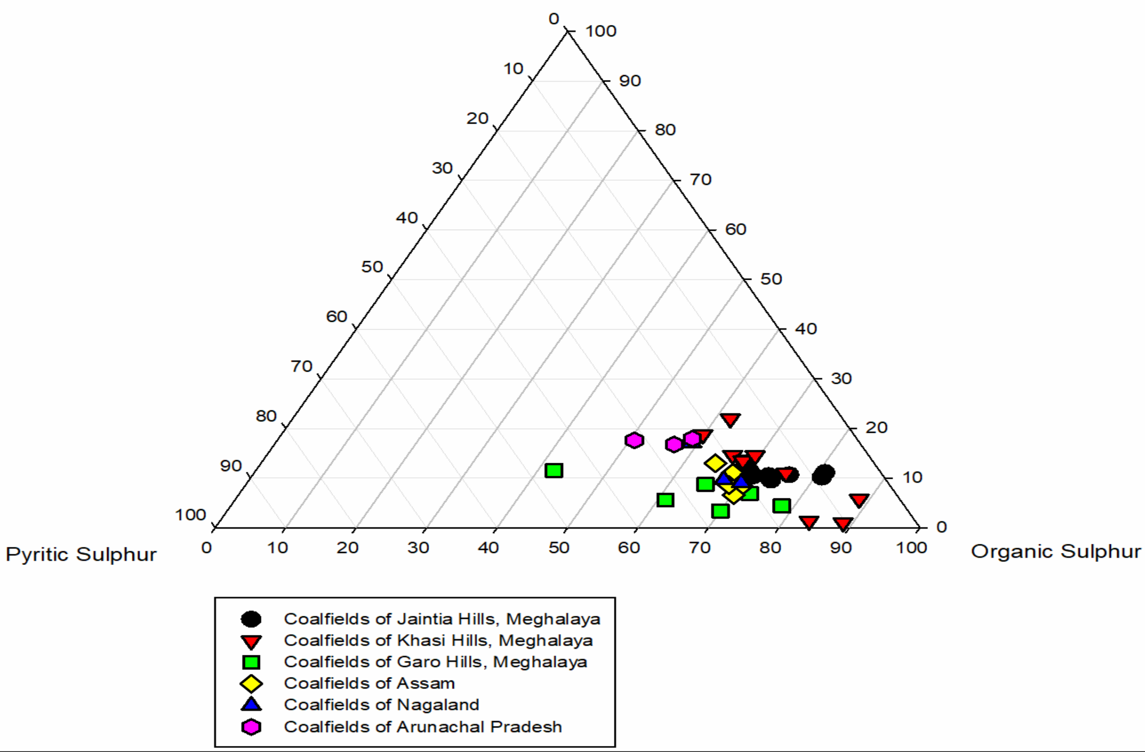

Fig. 3 : Ternary plot showing different forms of Sulphur in different coalfields of North-East India

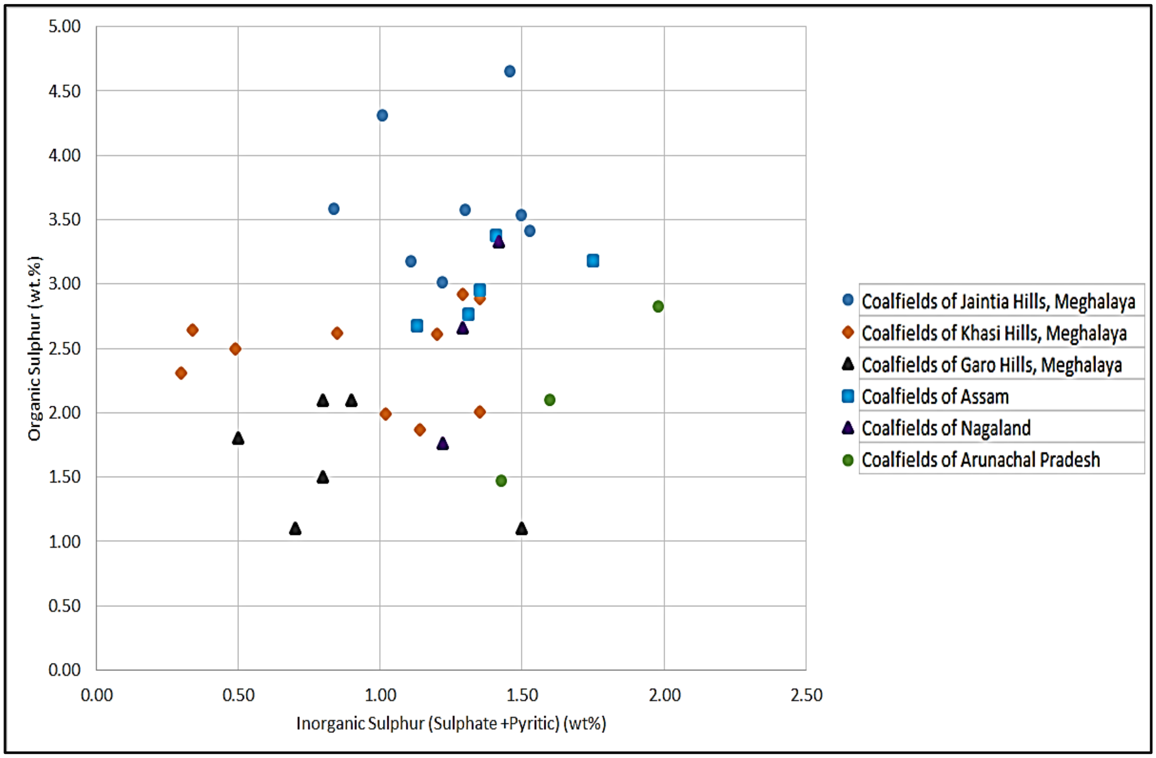

Fig. 4 : Cross plot between Organic and Inorganic forms of Sulphur for different coalfields of North-East India

Table 1 : Geological succession of the coalfields of platform areas[19]

Age	Formation and Member	Thickness	Rock Types
Upper Eocene	Kopili		Ferruginous sandstone, grey siltstone and shale
Middle Eocene	Sylhet Limestone: Prang Limestone/ Siju Limestone	60 to 150 m	Bluish massive to thinly bedded limestone with marlyinterband
Lower Eocene	Nurpuh Sandstone	15 to 26 m	Coarse to medium-grained ferruginous sandstones with bands of sandy limestone
Lower Eocene	Umlatdoh limestone	70 m to 110 m	Grey to pinkish grey limestone, sandy limestone and calcareous sandstone
Lower Eocene to Palaeocene	Lakadong sandstone	35 m to 250 m	Predominantly buff coloured medium grained arkosic sandstone with thin grey and carbonaceous shale and coal seams
Lower Eocene to Palaeocene	Lakadong limestone	25 m to 60 m	Grey to brownish grey limestone,siliceous limestone
Lower Eocene to Palaeocene	Therria sandstone	20 m to 80 m	Buff coloured medium to coarse-grainedarkosic sandstone with thin bands of pyrite-rich silty sandstone
Upper Cretaceous (Danian)	Langpar	10 m to 50 m	Buff coloured calcareous ferruginous sandstones, earthy limestones etc.
Upper Cretaceous (Maestrichtian)	Mahadek	160 m to 335 m	Massive coarse-grained glauconitic sandstones containing dark grey shales and calcareous horizons
Jurassic to lower Cretaceous	Sylhet Trap	250 m to 400 m	Aa and pahoehoe type basalts

Table 2 : A geological succession of the coalfields of Schuppen zone[19]

Age	Group Formation	Thickness	Rock Types
Pliocene	Dihing Group	1800 m.	Mostly pebbly sandstone with thin greyish clay beds
……………………………..Unconformity………………………………………..			
Mio-Pliocene	Namsang Formation	800 m.	Fine to coarse-grained sandstone with bands of clay
……………………………..Unconformity………………………………………..			
Miocene	Tipam Group (i) Girujan Clay (ii)Tipam Sandstone	1800 m 2300 m.	Mottled clay with greyish soft sandstone Ferruginous, fine to coarse-grained micaceous to felspathic sandstone

……………………………Unconformity………………………………………..			
Oligocene	Barail Group (i)Tikak Parbat Formation (ii)Baragolai Formation (iii)Naogaon Formation	600 m. 3500 m. 2200 m.	Greyish to yellowish white sandstone, sandy shale, coal seams Greyish to bluish grey or yellowish red mudstone, shale, sandstone, carbonaceous shale and thin coal seam Compact, fine-grained, dark grey sandstone with bands of splintery shale.
Eocene	Disang Group	3000 m.	Splintery dark grey shales and thin sandstone interband

Table 3 : Distribution of different forms of Sulphur in Eocene Coalfields of Jaintia Hills, Meghalaya

Coalfield	Seam characteristics			Sample characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	Total Tickness(m)	Name	Type	Total Numbers	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.
Bapung Coalfield	3	0.31-1.05	T	Channel	24	7.03	5.23	6.01	0.78	0.60	0.65	1.21	0.52	0.81	5.04	4.11	4.65
			M			6.23	5.01	5.27	0.65	0.52	0.54	0.84	0.28	0.47	4.74	4.21	4.31
			B			5.02	4.09	4.52	0.55	0.48	0.49	0.65	0.14	0.35	3.82	3.47	3.58
Jarain Coalfield	2	0.3-1.0	T	Channel	18	6.06	4.12	5.01	0.75	0.51	0.59	1.01	0.92	0.94	4.30	2.69	3.41
			B			5.95	4.01	4.25	0.66	0.45	0.51	0.90	0.72	0.79	4.39	2.84	3.57
Sutunga Coalfield	2	0.1-1.07	T	Channel	15	5.92	4.52	4.03	0.71	0.49	0.57	1.02	0.89	0.93	4.19	3.14	3.53
			B			4.89	3.92	3.57	0.64	0.31	0.44	0.97	0.65	0.78	3.28	2.96	3.01
Lakadong	1	0.3-2.1		ROM	15	5.03	3.97	4.20	0.69	0.21	0.41	1.05	0.64	0.70	3.29	3.12	3.17

Table 4 : Distribution of different forms of Sulphur in Eocene Coalfields of Khasi Hills, Meghalaya

Coalfield	Seam characteristics			Sample characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	Total Tickness(m)	Name	Type	Total Numbers	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.
Langrin Coalfield	6	1.5-2	T	Channel	20	3.82	2.40	3.01	0.58	0.42	0.44	0.78	0.52	0.58	2.46	1.38	1.99
			M			4.51	3.22	3.81	0.65	0.49	0.52	0.82	0.61	0.68	3.04	2.12	2.61
			B			4.61	4.10	4.13	0.69	0.54	0.57	0.89	0.71	0.78	3.03	2.85	2.89
Laitryngew Coalfield	2	0.2-1.9	T	Channel	15	5.01	3.86	4.01	0.89	0.41	0.61	0.94	0.65	0.68	3.18	2.80	2.92
			B			4.60	3.12	3.13	0.94	0.51	0.63	0.84	0.69	0.72	2.82	1.92	2.01
Mawbehlakhar Coalfield	3	0.10-0.50	T	Channel	15	4.60	1.79	3.01	0.10	0.05	0.66	0.75	0.34	0.48	3.75	1.40	1.87
			M			4.39	1.74	2.99	0.08	0.03	0.04	0.67	0.29	0.45	3.64	1.42	2.50
			B			4.40	1.62	2.98	0.09	0.02	0.03	0.40	0.29	0.31	3.91	1.31	2.64
Mawlong-Shella Coalfield	1	0.3-1.5	T	ROM	10	4.01	3.01	3.47	0.52	0.32	0.38	0.66	0.41	0.47	2.83	2.28	2.62
			B			3.00	2.75	2.78	0.24	0.11	0.15	0.25	0.09	0.15	2.51	2.01	2.31

Table 5 : Distribution of different forms of Sulphur in Eocene Coalfields of Garo Hills, Meghalaya

Coalfield	Seam characteristics			Sample characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	Total Tickness(m)	Name	Type	Total Numbers	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.
West Darranggiri*	3	0.3-2.7	T	Pillar	28	5.20	2.50	3.00	0.40	0.10	0.10	1.90	0.60	0.80	3.20	1.70	2.10
			M			3.00	2.30	2.90	0.30	trace	0.20	0.80	0.10	0.60	2.30	1.80	2.10
			B			2.60	2.10	2.3	0.10	0.20	0.10	0.90	trac	0.4	2.20	1.50	1.8

								0					e	0			0
Siju Coalfield	3	0.15-1.1	T	Channel	10	3.00	2.50	2.60	0.40	0.20	0.30	1.80	0.80	1.20	1.50	0.80	1.10
			M			2.80	2.20	2.30	0.30	0.10	0.20	0.90	0.60	0.60	1.60	1.50	1.50
			B			2.10	1.80	1.90	0.20	trace	trace	0.70	0.50	0.60	1.20	1.10	1.10

*Data collected fromPhukan[20].

Table 6 : Distribution of different forms of Sulphur in Oligocene Coals of Assam

Coalfield	Seam Characteristics			Sample Characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	**Total Tickness(m)**	**Name**	**Type**	**Total Numbers**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**
Makum*	5	1.2-33		Channel	49	6.28	2.20	4.43	0.90	0.08	0.40	1.88	0.15	1.01	5.65	1.60	3.38
						6.88	3.09	3.98	0.95	0.04	0.64	1.90	0.45	1.11	5.76	1.80	3.18
						4.82	3.19	4.01	0.40	0.19	0.25	1.20	0.60	0.88	3.78	2.00	2.68
Jajpur Coalfield	7	1.3-2.7		ROM	15	5.83	2.80	4.08	0.87	0.15	0.45	1.18	0.75	0.86	3.78	1.90	2.77
Dilli Coalfield	8	3.0-6		ROM	15	6.20	3.20	4.30	0.75	0.11	0.37	1.43	0.78	0.98	4.02	2.31	2.95

*Data collected from Gogoi et al. [21].

Table 7 : Distribution of different forms of Sulphur in Oligocene Coals of Nagaland

Coalfield	Seam Characteristics			Sample Characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	**Total Tickness( )**	**Name**	**Type**	**Total Numbers**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**	**Max.**	**Min.**	**Av.**
Borjan Coalfield	2	2.0-7.0	T	Channel	15	5.52	4.42	4.75	0.73	0.33	0.43	1.01	0.99	0.99	3.78	3.10	3.33
			B			4.42	3.75	3.95	0.63	0.24	0.38	0.98	0.89	0.91	2.81	2.62	2.66
MoulongKimong	1	<1		ROM	10	3.51	2.89	2.98	0.64	0.53	0.51	0.94	0.61	0.71	1.93	1.84	1.76
Tiru Coalfield*	1	<2		Pillar Sampling	9	11.00	6.00	6.66									

*Data collected from Singh et al. [22].

Table 8 : Distribution of different forms of Sulphur in Oligocene Coals of Arunachal Pradesh

Coalfield	Seam Characteristics			Sample Characteristics		Total Sulphur			Sulphate Sulphur			Pyritic Sulphur			Organic Sulphur		
	Numbers	Total Tickness (m)	Name	Type	Total Numbers	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.	Max.	Min.	Av.
Namchik-Namphuk	8	1-17.4	S-8 (Top)	Channel	10	5.80	4.20	4.80	0.94	0.85	0.86	1.50	0.94	1.12	3.36	2.41	2.82
			S-3			4.30	3.50	3.70	0.74	0.55	0.62	1.11	0.89	0.98	2.45	2.06	2.10
			S-1 (Bottom)			3.10	2.80	2.90	0.75	0.41	0.51	1.04	0.85	0.92	1.31	1.54	1.47

Table 9 : Coalfields of Platform areas[19]

Areas/State	Coalfields
Jaintia Hills	Bapung
	Malwar
	Lumshnong
	Mutang
	Lakadong
	Sutunga
	Jarain
Khasi Hills	Laitryngew
	Mawsynram Area
	LumDidom Hill
	PynurslaPlateau
	Maw-Beh-Larkar Area
	Umrileng Area
	Langrin
	Mawlong-Shella
Garo Hills	West Daranggiri
	East Daranggiri
	Balphakram-Pendengru Area
	Siju
	Baljong, Dogrengg and Hansapal
	Rongrenggiri

Table 10 : Coalfields in the zone of Schuppen[19,4]

State	Coalfields
Assam	Makum
	Dilli-Jeypore
	Mikir Hills
Arunachal Pradesh	Namchik-Namphuk
Nagaland	Borjan
	Jhanzi-Disai Valley
	Tuen Sang
	Tiru Valley
	Monlong-kimong

Paleoenvironment

The abundance of sulphur in coal is pointed towards a sedimentary environment of coal-bearing strata. White et al.[33] from the study of Illinois basin, USA suggests that the high sulphur content of coal was related to the marine and brackish environment of coal deposits. Williams and Keith[34] while studying the sulphur distribution in the lower Kittanning area concluded that sulphate ions from seawater played an important role in the sulphur enrichment of coal. H_2S is formed by reduction of sulphate and pyrite is produced by subsequent reaction with ferrous iron[35-38].

The coal deposits of the NE region formed under the marine condition as evidenced by the high content of sulphur (3-7%). Both Eocene and Paleocene coals were developed under the marinecondition of sedimentation. The Eocene coals of Meghalaya were probably deposited during the Eocene time under stable shelf conditions. The Oligocene coal of foredeep basins evolved as a consequence of subduction of Indian plate margins with that of the Burmese plate so there was miogeosyncline of flysh and molasses type of sediments[19]. Dasgupta and Biswas[39] have shown that the prevailed a brackish water condition during Barail Formation. Peat is either connected to Brackish water[40,41] or it is overlain by marine sediments as revealed by the high sulphur amount. Further studies supported that on modern peats under the marine influence which has shown the enrichment of sulphur due to sulphate reducing bacteria which results into precipitation of pyrite in peat[42]. Pyrite in the form of iron sulphides is found in coal as the dominant sulphide.Euhedral and massive pyrite also marcasite generally form during early syngeneticprocesses in uncompressed peat whereas early to late syngenetic processes are responsible for the formation ofcell-filling pyrite in cell cavities of macerals. Pyrite formed in the cleatsof coal indicate its origin

by late syngenetic or epigenetic processes whereas dendritic pyrite forms at thelater stages of coal formation[43].

Pyrite in coal typically forms from H_2S and Fe in solution. The process involves bacterial reduction of SO_4 to H_2S at pH values of 7 to 4.5 followed by the combining of H_2S, elemental sulphur and ferrous iron oxide (FeO) to form pyrite and water. This is the only way Pyrite can form in peat and low-rank coals. Consequently the presence of bacteria and required pH range are very important controls on pyrite formation in coals. The SO_4 may come from sea or vegetation, but either of these sources provides iron, which is usually in plentiful supply and comes from other sources[44]. It is probably derived from the breakdown of clay minerals and is possibly carried in solution as stabilized organic colloids[23]. In coal with a high amount of total sulphur, the more proportion comes from seawater[29]. A cross plot between organic and inorganic forms of sulphur is presented in **Fig. 4**, which clearly shows the separate clusters formed by coalfields of different regions of North-East India. This indicates that all these coals are having high sulphur,whose variation is mainly controlled by tectonic uplifting, peat-forming plant communities, roof strata, and marine or freshwater incursion.

CONCLUSION

On the basis of a detailed study of the distribution of sulphur of NE region of India following conclusion can be drawn-

1. The coals of Meghalaya were deposited in Platform areas under stable condition duringthe Eocene Period,whereas Oligocene coals of Assam, Arunachal Pradesh, and Nagaland developed in foredeepbasins of the Barail Group of Tikak Parbat Formation.
2. The coals are rich in S content which ranges from 3-7%.
3. All the forms of S recognised and out of these organic sulphur is the dominant one.
4. There is a vertical and lateral variation of sulphur in Eocene coals of Meghalaya which is absent in Oligocene coals of Assam, Arunachal Pradesh and Nagaland. The sulphur content of platform basins increases towards the top seam from bottom one. More so sulphur content increases from the western part of Meghalaya to the Eastern part.
5. The main sources of both Paleogene coals are the sea as there was a marine incursion during that period as evidenced by the pyrite content of sulphur (0.20 to 1.42%).
6. Pyrites were typically formed from bacterial reduction of SO_4 to H_2S at pH values of 7 to 4.5.

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REFERENCES

1. La Touche T.H.D., The Daranggiri Coal-field, Garo Hills, Assam, *Rec Geol Surv India*, **15**, 175–178, **(1882).**
2. Ghosh T.K., On Tertiary coal from Daranggiri, Assam, *QJ Geol Soc India.,* **36**, 91–94, **(1964).**
3. Ahmed M., Petrochemical study of coal, Litryngew coalfield, K & J Hills, Assam, *J Geol. Soc.*, **1**, 15–20, **(1971).**
4. Indian Bureau of Mines, *Indian Minerals Yearbook 2017, 56th edn.* Ministry of Mines, Nagpur, **(2018).**
5. Borah D., Baruah M.K. and Haque I, Oxidation of high sulphur coal. Part 1. Desulphurisation and evidence of the formation of oxidised organic sulphur species. *Fuel.*, **80**, 501–507, **(2001).**
6. Mukherjee S., Borthakur P.C., Chemical demineralization / desulphuriza -tion of high sulphur coal using sodium hydroxide and acid solutions, *Fuel.,* **80**., 2037–2040. doi: 10.1016/S0016-2361(01)00094-1, **(2001).**
7. Singh A.K., Kumar A. and Singh P.K., et al Bacterial desulphurization of low-rank coal: A case study of Eocene Lignite of Western Rajasthan, India. *Energy Sources*, Part A Recover Util Environ Eff 40:1199–1208. doi: 10.1080/15567036. 2018.1476608, **(2018).**
8. Hashan M., Sulphur Concentration in Imported Coal Around the Tamabil Area, Sylhet, Bangladesh. *Int J Environ Monit Anal.*, **4**, 27. doi: 10.11648/j.ijema. 20160401.15, **(2016).**
9. Schlesinger R.B., Sulfur Oxides. In: Comprehensive Toxicology. *Elsevier*, 277–290, **(2010).**
10. Tissot B.P. and Welte D.H., Petroleum Formation and Occurrence. Eos, Trans Am Geophys Union 66:643. doi: 10.1029/EO066i037p00643, **(1985).**
11. Diessel C.F.K., Coal-Bearing Depositional Systems. *Springer*, Berlin Heidelberg, Berlin, Heidelberg, **(1992).**
12. Chou C.L., Geologic factors affecting the abundance, distribution, and speciation of sulfur in coals. In: Geology of Fossil Fuels, *Proc 30th Int Geol Congress*, 47–57, **(1997).**
13. Ahmed M. and Bora J.P., Geochemistry of tertiary coal, Bapung coalfield, Jaintia Hills, Meghalaya, *J. Assam Sci. Soc.*, 24, 9–10, **(1981).**

14. Chandra D., Mazumdar K. and Basumallick S., Distribution of sulphur in the tertiary coals of Meghalaya, India. *Int J Coal Geol.*, **3**, 63–75, doi: 10.1016/0166-**5162**(83), 90013-7, **(1983).**
15. Ahmed M. and Rahim A., Abundance of sulfur in Eocene coal beds from Bapung, northeast India, *Int. J. Coal Geol.*, 30:315–318. doi: 10.1016/0166-5162(95)00050-X, **(1996).**
16. Nath M., Utilisation Prospects of Bapung Coal, Meghalaya, Northeast India. In: Neo-Thinking on Ganges-Brahmaputra Basin Geomorphology, *Springer*, 139–151, **(2016).**
17. Singh A. K., Singh M. P. and Singh P.K. Petrological Investigations of Oligocene Coals from Foreland Basin of Northeast India. Energy Explor Exploit 31:909–936. doi: 10.1260/0144-5987.31.6.909, **(2013).**
18. Singh M. P. and Singh A. K., Petrographic characteristics and depositional conditions of Eocene coals of platform basins, Meghalaya, India, *Int. J. Coal. Geol.*, **42**, 315–356, doi: 10.1016/S0166-5162(99)00045-2, **(2000).**
19. Raja Rao C. S., Coalfields of India : Coalfields of North Eastern India, *Bull Geol Surv India*, Ser A, **45**(1), 75, **(1981).**
20. Phukan S., *Study of the West Daranggiri coal field of Garo Hills*, Meghalaya. Gauhati University, **(2002).**
21. Gogoi K., Dutta M.N. and Das P.K. Distribution and Seamwise Variation of Sulphur in Makum Coal Field of Assam, India, *Appl Geochemistry*, **12**, 2010–2010, **(2010).**
22. Singh P. K., Singh M. P., Singh A. K. and Naik A.S., Petrographic and Geochemical Characterization of Coals from Tiru Valley, Nagaland, NE India, *Energy Explor Exploit.*, **30**, 171–191. doi: 10.1260/0144-5987.30.2.171, **(2012).**
23. Chou C., Sulfur in coals: A review of geochemistry and origins, *Int. J. Coal. Geol.*, 100, 1–13, doi: 10.1016/j.coal.2012.05.009, **(2012).**
24. Calkins W.H., The chemical forms of sulfur in coal : A review, *Fuel.*, **73**, 475–484. doi: 10.1016/0016-2361(94)90028-0, **(1994).**
25. Mishra H.K. and Ghosh R.K., Geology, petrology and utilisation potential of some Tertiary coals of the northeastern region of India, *Int J Coal Geol.*, 30:65–100. doi: 10.1016/0166-5162(95)00038-0, **(1996).**
26. Singh Y.R., Singh B.P. and Devi S.R., Source rock characterization, Diagenesis and Depositional Environment of the Upper Disang Formation from Gelmoul area of Manipur, NE Indiam, *J. Ind. Assoc. Sedimentol*., **34**, 1–7, **(2017).**
27. Price F.T. and Shieh Y.N., Fractionation of sulfur isotopes during laboratory synthesis of pyrite at low temperatures, *Chem. Geol.*, 27, 245–253, **(1979).**
28. Sinninghe Damste J.S. and De Leeuw JW, Analysis, structure and geochemical significance of organically-bound sulphur in the geosphere: State of the art and future research, *Org Geochem.*, 16, 1077–1101. doi: 10.1016/0146-6380(90)90145-P, **(1990).**
29. Chou C. L., Geochemistry of Sulfur in Coal. In: Orr WL, White CM (eds) Geochemistry of Sulfur in Fossil Fuels, *Ame. Chem. Soc.*, 30–52, **(1990).**
30. Ryan B. and Ledda A., A review of sulphur in coal: with specific reference to the Telkwa deposit, north-western British Columbia, *Geol Fieldwork*, 22, **(1997).**
31. Liang J., Acid rain, In: Chemical Modeling for Air Resources, *Elsevier*, 115–142, **(2013).**
32. Dai S., Ren D. and Tang Y et al., Distribution, isotopic variation and origin of sulfur in coals in the Wuda coalfield, Inner Mongolia, China, *Int. J. Coal. Geol.*, 51, 237–250. doi: 10.1016/S0166-**5162**(02), 00098-8, **(2002).**
33. White C.D. and Thiessen R., The origin of coal. US Government Printing Office, **(1913).**
34. Williams E.G. and Keith M.L., Relationship between sulfur in coals and the occurrence of marine roof beds, *Econ. Geol*., **58**, 720–729, **(1963).**

35. Kaplan I.R., Emery K.O. and Rittenbebg S.C. The distribution and isotopic abundance of sulphur in recent marine sediments off southern California, *Geochim Cosmochim Acta*., **27**, 297–331, **(1963).**
36. Goldhaber M.B. and Kaplan I.R. Mechanisms of sulfur incorporation and isotope fractionation during early diagenesis in sediments of the Gulf of California, *Mar. Chem.*, **9**, 95–143, **(1980).**
37. Olson J.S., Garrels R.M. and Berner RA, et al, The natural carbon cycle, *Atmos Carbon Dioxide Glob Carbon Cycle.*, 239, 175–214, **(1985).**
38. Morse JW, Berner RA, What determines sedimentary C/S ratios? *Geochim Cosmochim Acta*, **59**, 1073–1077, **(1995).**
39. Dasgupta A.B. and Biswas A.K., *Geology of Assam : Geological Society of India*, Bangalore, 169, **(2000).**
40. Bustin R. M. and Lowe L.E., Sulphur, low temperature ash and minor elements in humid–temperate peat of the Fraser River Delta, British Columbia, *J. Geol. Soc. London.*, **144**, 435–450, **(1987).**
41. Casagrande D.J., *Sulphur in peat and coal. In: Geological Society*, London, Special Publications, 87–105, **(1987).**
42. Phillips P., Bender J. and Simms R, et al, Manganese and iron removal from coal mine drainage by use of a green algae-microbial mat consortium. *In: Proc. of the III Int. Conf. on The Abatement of Acidic Drainage*, Bureau of Mines SP 06A-94 Publication., Pittsburgh, PA, 99–109, **(1994).**
43. Grady W., Microscopic varieties of pyrite in West Virginia coals. Trans Soc Min Eng AIME;(United States) 262, **(1977).**
44. Nayak B., Mineral matter and the nature of pyrite in some high-sulfur tertiary coals of Meghalaya, northeast India. *J. Geol Soc India*, **81**, 203–214. doi: 10.1007/s12594-013-0023-9, **(2013).**