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Abundance of Sulphur in Paleogene coals of North-East India and Its Paleo-environmental Implications

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Abstract :
The present paper entails the occurrence 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 from 2.90 % and 6.60 %. In the 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 the Western Meghalaya. Both Eocene and Oligocene coals have been derived from seawater as evidenced by the presence of pyritic form of sulphur. Study of forms of sulphur also suggests that the deposition of the coals in a different part of the region was influenced by roof strata, peat-forming plant communities, tectonic uplifting, and marine or freshwater incursion.

Keywords: 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 (LaTouche, 1882; Ghosh, 1964; Ahmed, 1971). 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 (Indian Bureau of Mines, 2018). But the Paleogene coal deposits are of good quality having characteristically low ash content and medium caking property. The only drawback of this coal is the presence of a high amount of sulphur (inorganic constituent) which render it unsuitable for commercial utilization. However, energy demand has 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 (Borah et al., 2001; Mukherjee and Borthakur, 2001; Hashan, 2016; Singh et al., 2018). 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 SO2. At high temperature, the organic sulfur compounds of coal degrade to elemental sulfur or inorganic sulfur species, such as hydrogen sulfide, and further converts to SO2. This SO2 get oxidized to sulfur trioxide (SO3), which combines with water vapour to produce ultrafine (<0.1 μm diameter) sulfuric acid particles (Schlesinger, 2010). 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 is commonly referred to as inorganic sulphur. Iron sulphide (FeS2), 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 (Tissot and Welte, 1985)

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 (Diessel, 1992; Chou, 1997).

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 (Ahmed and Bora, 1981; Chandra et al., 1983; Ahmed and Rahim, 1996; Nath, 2016).

The present study is mainly focused on the determination and detailed description of the amount and occurrence of 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 on the paleoenvironmental aspects of these coals.

Geological setting

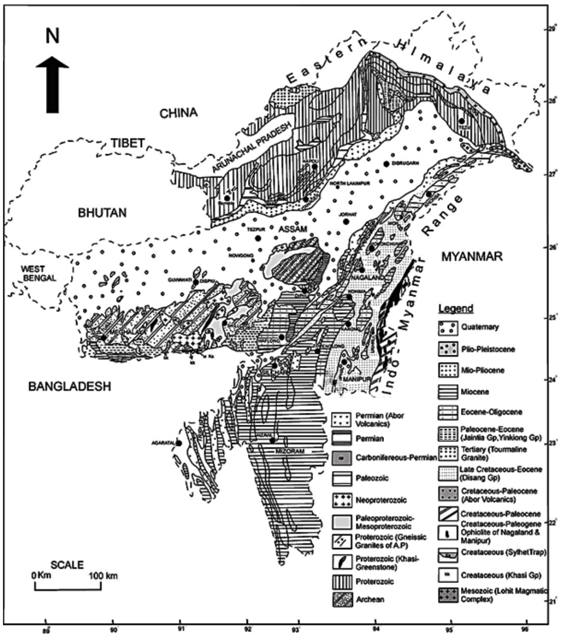

Figure 1 Geological map of North-Eastern India (after Singh et al., 2017)

In north-east India, coal deposits mainly occur in the states of Assam, Arunachal Pradesh, Nagaland and Meghalaya (Figure 1) confined to the Oligocene and Eocene arenaceous formations. The coal deposits of these areas are understood to have been formed in two distinct tecto-sedimentary settings. The coal deposits of Assam (Makum and Dilli- Jeypore), Arunachal Pradesh (Namchik-Namphuk coalfield) and Nagaland (Borjan coalfield) have probably originated in a foreland basin (Singh et al., 2013), whereas the coalfields of Garo, Khasi, and Jaintia Hills of Meghalaya represent the development of coal facies over platform areas (Singh and Singh, 2000). In the Garo, Khasi and Jaintia Hills of Meghalaya the coal seams occur in the Lakadontg Sandstone 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 resultd in the deposition of these formations (Raja Rao, 1981).

Drifting of the Indian plate during the Cretaceous period towards the 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 foredeep are believed to be the result of such activity. The development of the foredeep (trench/ technogenic) is because of subduction, which provided the site for deposition of Tertiary sequences of stupendous thickness (~15,600 m). In the Early Eocene period the sedimentation began with the deposition of Disang Group followed by Barail Group of Oligocene age (Table 2). In the Tikak Parbat Formation of Barail Group, the coal seams of the fore deeps occur (Figure 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 Borgagolai Formation (alternating sandstone and shale with carbonaceous shale and thin lamination of coal) and Naogaon Formation (splintery, grey or brownish coloured, iron-stained shales with occasionally interbedded with thin bands of fine-grained sandstone and sandy shale) (Ahmed, 1996).

Methods of Study

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 and Moulong-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 was collected from Phukan (2002), for Makum coalfield the data was collected from (Gogoi et al., 2010) and for Tiru coalfield the data was taken from (Singh et al., 2012).

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 Na2CO3 (Sodium Carbonate).

The sulphur was extracted by using (barium chloride) to precipitate BaSO4 (Barium Sulphate) from the 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 Tables 3, 4, 5, 6, 7 and 8. These coals have high sulphur content, which varies from 7.03% to 1.62% (with an average of 2.98%). Coal with less than 1% sulphur is placed under low-

Table:1 Geological succession of the coalfields of platform areas (modified after Raja Rao, 1981).

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: Geological succession of the coalfields of Schuppen zone (modified after Raja Rao, 1981).

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		
	No's	Total Tickness (m)	Name	Type	Total No's	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**		
	No's	**Total Tickness (m)**	**Name**	**Type**	**Total No's**	**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**		
	No's	**Total Tickness (m)**	**Name**	**Type**	**Total No's**	**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	2.30	0.10	0.20	0.10	0.90	trace	0.40	2.20	1.50	1.80
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 from Phukan, (2002).*

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**		
	No's	**Total Tickness (m)**	**Name**	**Type**	**Total No's**	**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. (2010).*

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**		
	No's	**Total Tickness (m)**	**Name**	**Type**	**Total No's**	**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. (2012).

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**		
	No's	**Total Tickness (m)**	**Name**	**Type**	**Total No's**	**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 (Raja Rao, 1981).

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 (Raja Rao, 1981; Indian Bureau of Mines 2018).

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

sulphur coals. Coal with 1% to <3% sulphur is medium sulphur coal and coal with ≥ 3% sulphur is considered high sulphur coal (Chou, 1997).For the coalfields of north-east India, all the coals contain sulphur more than 3% (except, few samples) and therefore are placed under high sulphur coal category. Due to the complex nature of organic sulphur, its desulphurization is difficult while chemical desulphurization processes helps in reduction of aliphatic sulphur structure but to achieve a high desulphurization rate biological techniques are useful (Calkins, 1994). These coals have high organic sulphur as compared to inorganic sulphur.

Coalfield of Platform basins

The coalfields of platform areas are shown in Table 9 (Raja Rao, 1981) however mining activity is confined to the areas listed in Table 3, 4 and 5.

The total sulphur content of Eocene coals (Meghalaya) varies from 1.62 to 7.03% (Tables 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 show values 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 varies from 2.69 to 5.04%.

In the Khasi Hills (Table 4), the total sulphur varies from 1.62 to 5.01%. Although all the samples show sulphur content above 3% except 3 samples of Mawhehlakhan 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 from trace 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 sulphur category. All other coals are classified as high sulphur 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 (Tables 3, 4, 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 belongs 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 conditions at the time of the deposition in the eastern part of the basin (Mishra and Ghosh, 1996). 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 (Chandra et al., 1983).

The regional lateral variation of coals of Meghalaya is strictly a palaeoenvironmental effect. In other words, the increase in sulphur content from western to eastern part is due to the more marine nature of the peat-forming swamps of Khasi and Jaintia Hills as compared to that of Garo Hills (Chandra et al., 1983).

The coalfields of foreland basins

The coalfields of foreland basins are placed in Table 10 (Raja Rao, 1981). 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, having main collieries, Tipong, Ledo, Borgolai, Tirap and Namdong. There are 5 seams of 60 ft., 20 ft., 8 ft., 5 ft. thick, and a 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 other forms of sulphur. There is no uniformity in variation of sulphur in the seam both in the lateral and vertical direction.

From the other coalfield of Assam in Jeypore and Dilli coalfield run-of-mine samples were collected and analyzed which contain total sulphur from 2.8 to 6.2%, sulphate

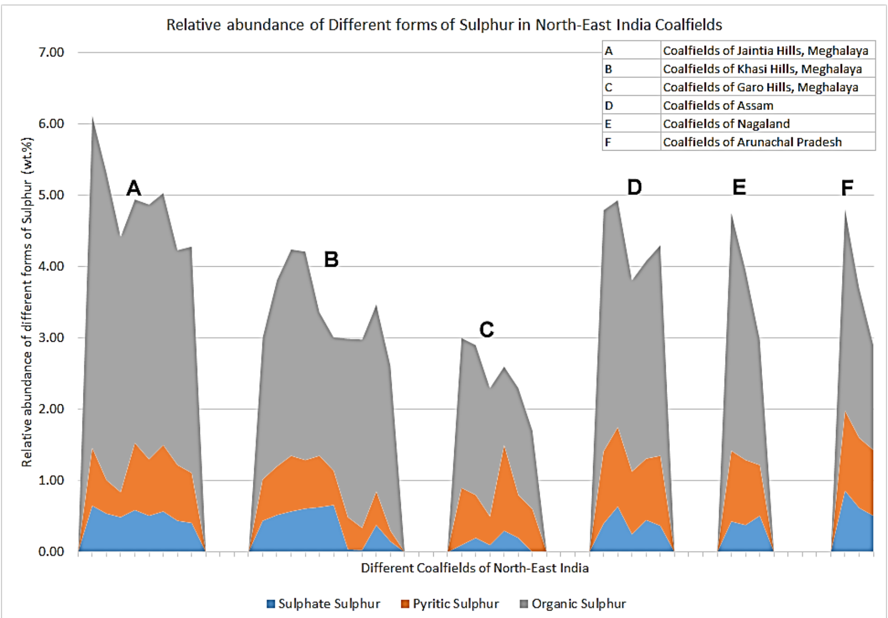

Figure 2: The relative abundance of different forms of sulphur in different coalfields of North-East India.

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 in high amount with total sulphur varying from 2.8 to 3.8% (Table 8). Sulphate sulphur ranges from 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) is also high. For, Borjan Coalfield the total sulphur content ranges from 3.75- 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%.

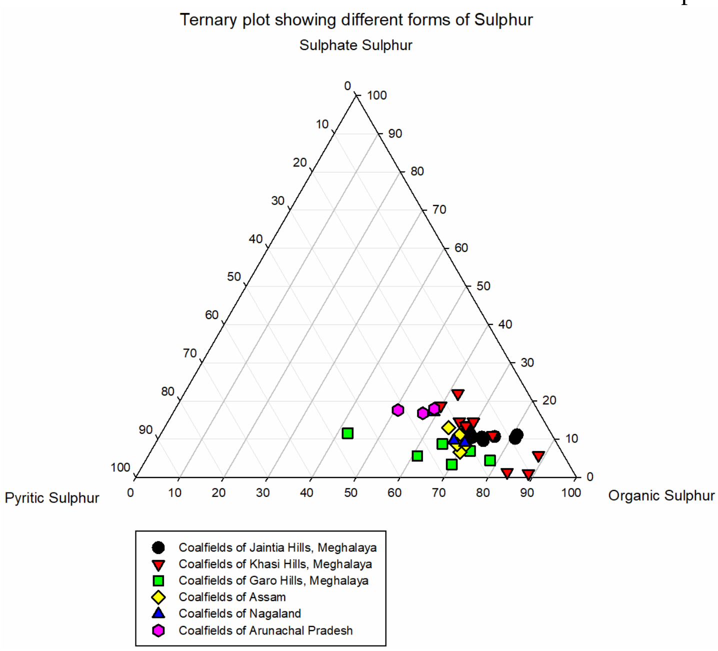

Figure 3: Ternary plot showing different forms of sulphur in different coalfields of North-East India.

For, Moulong Kimang coalfield, the total sulphur varies from 2.89 and 3.01% with sulphate sulphur 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 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 of 4 to 11% (Chou, 2012).

Sulphur is generally rich in marine influenced coals as observed by Teichmuller (1962). This observation is further supported by Price and Shieh (1979), Sinninghe Damste and De Leeuw (1990) and Chou (1990), where they showed that coals usually more than 1% sulphur comes from the seawater. A similar situation prevails in case of Oligocene coals of northeast India, which have a relatively high sulphur content. A relative abundance of different forms of sulphur in coals of the study area is presented in Figure 2.

Sources of Sulphur

In coal, sulphur (S) primarily originate 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 matter in the vegetation (Ryan and Ledda, 1997). They observed that fresh water does not contribute any sulphur in coal as they contain 0 to 10 ppm sulphur. However, their observation infer that in coal about 0.5% sulphur was probably derived from sea water which contains average 0.265% SO4 or 885 ppm S (SO4 contain 33.4% S). This is substantiated further by Casagrande (1987) that the marine-influenced peats generally have a higher sulphur content. Similar conditions prevailed in northeastern coals where sulphur content is more than 3% (except a few samples) and higher sulphur content is probably derived from seawater.Liang (2013) also found that the marine biota release organic sulfur compounds, such as dimethyl sulfide (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 (Dai et al., 2002). A triangular plot showing the distribution of different forms of sulphur in Paleogene coals are presented in Figure 3. The figure clearly shows the dominance of organic form of sulphur in all the coalfields.

Paleoenvironment

The abundance of sulphur in coal is pointed towards a sedimentary environment of coal-bearing strata. White et al. (1913) 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 (1963) 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. H2S is formed by reduction of sulphate and pyrite is produced by subsequent reaction with ferrous iron (Kaplan et al., 1963; Rickard, 1975; Goldhaber and Kaplan, 1980; Olson et al., 1985; Raiswell and Berner, 1985; Morse and Berner, 1995).

The coal deposits of the northeast region formed under the marine conditions as evidenced

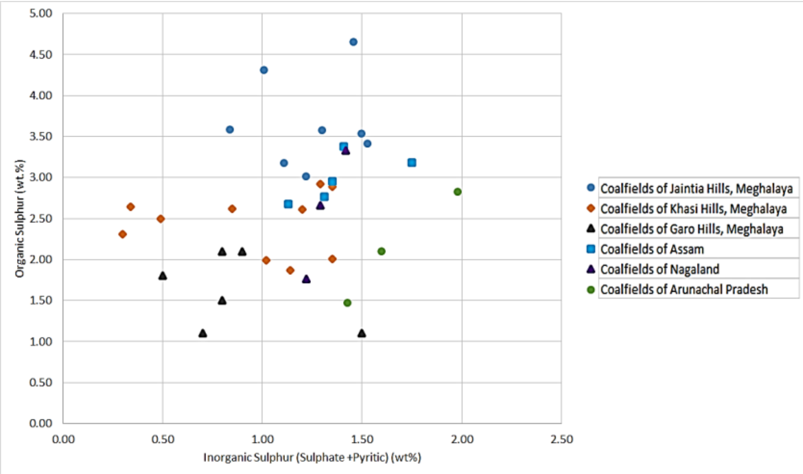

Figure 4A: Cross plot between organic and inorganic forms of sulphur for different coalfields of North-East India.

by the high content of sulphur (3-7%). Both Eocene and Paleocene coals were developed under the marine conditions 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 (Raja Rao, 1981). Dasgupta and Biswas (2000) have shown that brackish water condition prevailed during Barail Formation. Peat is either connected to Brackish water (Bustin and Lowe, 1987; Casagrande 1987) 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 enrichment of sulphur due to sulphate reducing bacteria which results into precipitation of pyrite in peat (Phillips et al., 1994). 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 syngenetic processes in uncompressed peat whereas early to late syngenetic processes are responsible for the formation of cell-filling pyrite in cell cavities of macerals. Pyrite formed in the cleats of coal indicate its origin by late syngenetic or epigenetic processes whereas dendritic pyrite forms at the later stages of coal formation (Grady, 1977).

Pyrite in coal typically forms from H2S and Fe in solution. The process involves bacterial reduction of SO4 to H2S at pH values of 7 to 4.5 followed by the combining of H2S, 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 SO4 may come from sea or vegetation, but either of these sources provides iron, which is usually in plentiful supply and comes from other sources (Nayak, 2013). It is probably derived from the breakdown of clay minerals and is possibly carried in solution as stabilized organic colloids (Chou, 2012). In coal with a high amount of total sulphur, the more proportion comes from seawater (Chou. 1990). A cross plot between organic and inorganic forms of sulphur is presented in Figure 4, which clearly shows the separate clusters formed by coalfields of different regions of northeast 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:

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 foredeep basins of the Barail Group of Tikak Parbat Formation. The coals are rich in S content which ranges from 3-7%. All the forms of S recognised, organic sulphur is the dominant one.

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.

The main sources sulphur of both the Paleogene coals are the sea water as there was a marine incursion during that period as evidenced by the pyrite content of sulphur (0.20 to 1.42%).
Pyrites were typically formed from bacterial reduction of SO4 to H2S at pH values of 7 to 4.5.

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