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Origin of Sulfur Content in Lignite Deposit at Thar Coal Mine, Block II, Pakistan

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ABSTRACT: *Thar geology encompasses of three main stratigraphic sedimentary formations i.e. Bara - coal bearing (oldest), sub-recent and dune sand (youngest). These formations are overlain on granitic igneous basement throughout the Thar region. Pakistan's first largest open pit lignite mining operations are on-going at Thar Coal Mine Block II which is located at the centre of Thar region. This study is focused in exploring the origin of sulfur content in lignite at Thar Coal Mine. Coal seams (part of Bara formation) at Thar Coal Mine are comprised of pollen streaks which are plant micro-organisms (organic) and generally found in lignite indicating the un-maturity and low grade of coal. Pollens can be easily identified in coal seams by its shiny lustre (yellow color). These pollen streaks are dominant in CL 2-3 coal seam which has the highest sulfur content (2.1%). The geochemistry of pollen streaks was studied separately to evaluate its influence in the overall coal quality. The results revealed that it contains very high sulfur content along with high net calorific and ash values. Thus, pollens are considered as one of the main contributors of sulfur in Thar coal seams. Another contributing factor for the presence of sulfur in Thar coal is the occurrence for pyrite mineral which is very common in coal deposits since coal formation is mainly takes place in reducing environment.*

KEYWORDS: sulfur, origin, pollen, thar coal mine, pyrite, lignite

INTRODUCTION

Fossil fuels, coal in particular, are indispensable sources of energy necessary for today's technological and economic progress throughout the world (Medunic et al., 2018). Generally, coal is being classified into 4 ranks/grades; lignite (low rank), sub-bituminous, bituminous and anthracite (high rank) based on its maturity. Coal geochemistry is broadly comprised of moisture, ash, volatile matter, fixed carbon and calorific value and total sulfur content. Sulfur being one of the most important quality parameter in coal especially in terms of coal rank/maturity and power generation is the main focus of the present study. Sulfur occurs in coal as pyritic, organic and sulphate where the most important forms are pyritic and organic sulfur (Ryan and Ledda, 1998). Pearson and Kwong (1979) suggest that, in coals with low pyrite concentrations, organic sulfur can oxidize to form gypsum, which can also be introduced by groundwater.

Pakistan has lignite reserves of around 186 Bt out of which 176 Bt are situated at Thar coal field of Thar Parker district. The study area; Thar Coal Mine present in Thar Block II which

has 2.4 Bt of coal resources is located at the centre of Thar coal field (Fig. 01). The extension of entire lignite bearing area in Thar is about 9100 Km^2. Lignite being considered as the most un-matured coal in general thus it tends to have relatively higher sulfur content. The present study emphases in exploring the origin of sulfur content in lignite at Thar Coal Mine.

General Geology

Thar geology consist of three sedimentary formations with an underneath igneous basement. The coal-bearing layers at Thar Coal Mine are included in Bara formation and are divided into 02 main groups (non-minable and minable). The non-minable coal group includes 1-1, 1-2, 2-1 and 2-2 coal seams while the minable coal group contains 2-3, 2-4, 2-7 and 2-8 coal seams (Fig. 02). These coal seams are separated by non-coal layers (waste material) which are dominantly composed of carbonaceous claystone. The cumulative average coal thickness in Thar Coal Mine is 25 meters. The overall weighted average net calorific value of minable coal seams at Thar Coal Mine is 11.2 MJ/Kg while its ash content is 8.5% with a very low sulfur value of 1.5%. The 2-7 coal seam is the main mineable coal seam and is laterally spread all over the Block II. This seam is the thickest and most persistent among all seams and makes up approximately 73% of the total coal resource. As a result, the coal quality of seam 2-7 has the largest degree of influence on the overall coal resource quality. Coal seam CL 1-1 and 2-1 due to their limited lateral extent and localized structural existence have not been considered in the present study since both seams are also excluded from Thar Block II total coal resources.

In general, lignite coal seams at Thar Coal Mine varies in sulfur content where the highest amount of sulfur is found in coal seams 1-2 and 2-2 of non-minable seam category and specifically in coal seam 2-3 of minable coal group (Table 01). In general, the variation of sulfur in coals is closely related to the depositional environments of coal seams (Chou, 2012). Marine-influenced peats generally have a higher sulfur content than freshwater-influenced peats (Spackman et al. 1976, Casagrande, 1984).

MATERIAL AND METHODS

Coal samples from different seams were collected from Thar coal mine to analyse the geochemical properties especially total sulfur content. Based on field surveys, pyrite and pollen streaks were evident in different coal seams. Thus, some of the samples were also collected from pyrite and pollen streaks that were found within coal seams in order to investigate its impact on the coal quality. Pollens are basically parent plant material (organic) which indicates the degree of maturity and rank of coal.

3.1. Sample Collection

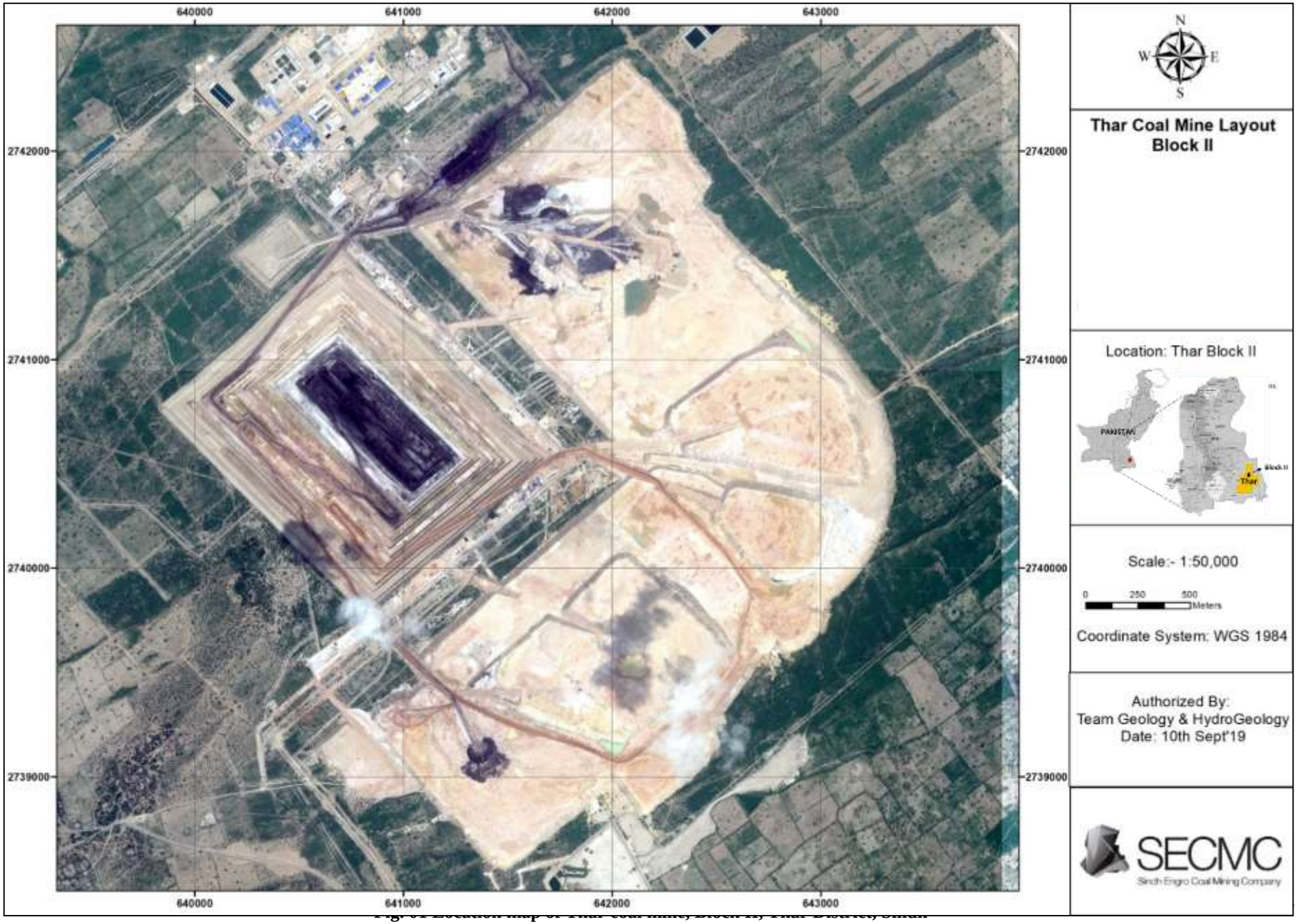
The sample collection methodology adopted was Manual Bench Face Sampling from exposed coal benches during coal production and by digging small trenches from seam top prior to coal production. Some of the studied samples were also extracted from In-Pit Borehole Drilling.

All the samples were preserved in plastic bags to avoid air ingress and moisture loss, labelled with sample description followed by representative seam id and then dispatched to mine-mouth coal laboratory for sample preparation and testing.

3.2. Geochemical Analysis

The collected insitu samples were crushed into small pieces, reduced thoroughly and pulverized into fine powder to obtain representative sample for geochemical analysis (moisture, ash, volatile matter, fixed carbon, calorific value, total sulfur along with sulfur forms).

Sample preparation was performed using Automatic Crusher Divider, moisture and ash content were analysed by Automatic Proximate Analyzer while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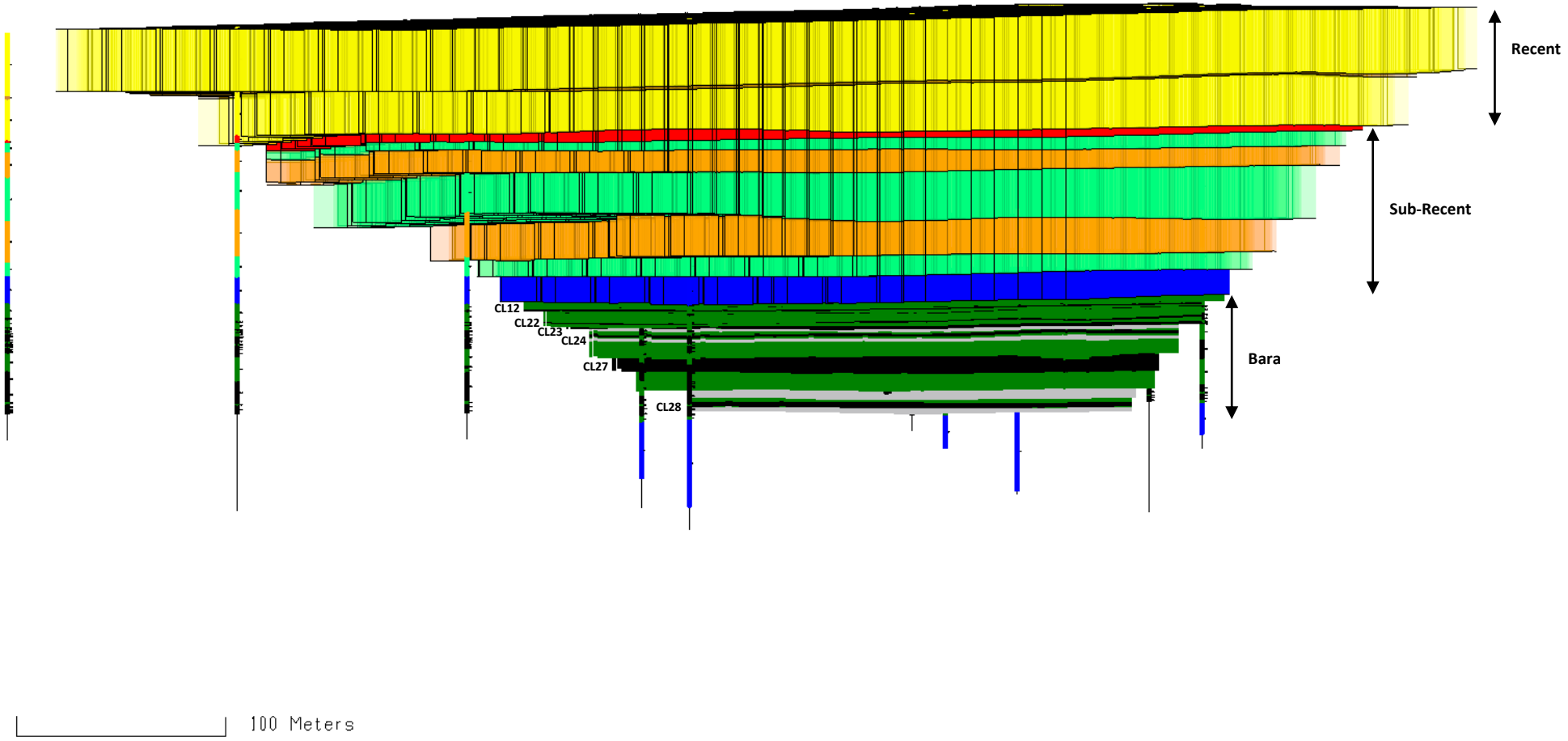

Fig. 02 Coal seam model representation from Thar coal mine, Block II, Thar District, Sindh

volatile matter was tested via Muffle Furnace. Calorific value was estimated by Bomb Calorimeter whereas total sulfur testing was conducted with Automatic Sulfur Analyzer. Entire sample preparation and testing were executed based on ISO standards at Thar Coal Mine laboratory.

3.3. X-Ray Fluorescence

Pyrite mineral insitu samples collected from mine were analysed using Handheld X-Ray Fluorescence Analyzer to estimate the sulfur and iron contents.

RESULTS AND DISCUSSIONS

4.1. Sulfur Origin

The collected samples were geochemically tested and analysed for sulfur content along with other quality parameters and compared with each other to evaluate the degree of sulfurization in Thar coal lignite deposit. The total sulfur content on as received basis of the studied coal samples are shown in Table 01. The highest amount of total sulfur content is evident in coal seam 1-2, 2-2 and 2-3 which indicates that these seams have undergone greatest degree of sulfurization and are relatively unmatured due to relatively shallower depth of deposition. The high sulfur content particularly in these upper coal seams (1-2, 2-2 and 2-3) also coincides with the generic sulfur trend of Thar coal deposit. Rest of the coal seam samples revealed very low sulfur content.

Table 01 Total sulfur (as received basis) and sulfur forms (dried basis) of coal samples collected from Thar coal mine, Block II, Thar District, Sindh

·am Category	Sample #	Seam ID	Total Sulfur.ar (%)	Pyritic Sulfur.d (%)	Organic Sulfur.d (%)	Sulphate Sulfur.d (%)
Non-Minable	01	CL 1-2	2.34	3.08	1.05	0.09
	02		2.58	2.97	1.39	0.12
	03		4.30	4.91	1.67	0.47
	04	CL 2-2	3.65	4.66	1.19	0.22
	05		2.56	2.78	1.88	0.03
	06		4.92	5.59	1.65	0.82
Minable	07	CL 2-3	1.99	2.08	1.27	0.05
	08		1.89	1.68	1.16	0.17
	09		2.63	2.93	1.12	0.26
	10	CL 2-4	1.58	1.64	1.24	0.18
	11		0.86	0.80	0.66	0.05
	12		0.97	0.64	0.94	0.17
	13	CL 2-7	0.93	0.87	0.66	0.15
	14		0.49	0.11	0.70	0.06
	15		0.45	0.13	0.62	0.09
	16	CL 2-8	0.52	0.29	0.53	0.13
	17		0.99	0.89	0.73	0.20
	18		0.66	0.25	0.93	0.06

The studies samples were further investigated to determine the contributing factors and source of sulfur in Thar coal deposit. Selected samples from different seams were tested to estimate the pyritic, organic and sulphate sulfur concentrations on dried basis for which the results are given in Table 01. The results show that sulfur content in most of the samples is mainly

contributed from pyritic and organic sulfur. However, the percentage of pyritic sulfur is relatively higher than organic sulfur in nearly all the samples which is due to the prevailing marine condition during peat formation. This is confirmed from the existence of 02 aquifer bodies (named as coal seam roof and bottom aquifers) present at top and bottom of lignite deposit in the study area. Pyritic sulfur plays a major contributing role to sulfur in marine peats as compared to sulfur in freshwater peats (Casagrande, 1987). In addition, availability of ferrous ions for pyrite formation during peatification and diagenesis is also the contributing factor for pyritic sulfur.

4.2. Pyritic Sulfur

In general, low sulfur coals (<1%) contain more organic sulfur than pyritic sulfur while high sulfur coals (>1%) often contain nearly similar proportions of pyritic and organic sulfur (Casagrande, 1987). 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 sulfur and ferrous iron oxide (FeO) to form pyrite and water, which is the only way pyrite can form in peats and low rank coals (Ryan and Ledda, 1998). Organic sulfur and pyritic sulfur in coal can be converted into each other under favourable conditions (Want et al., 2016).

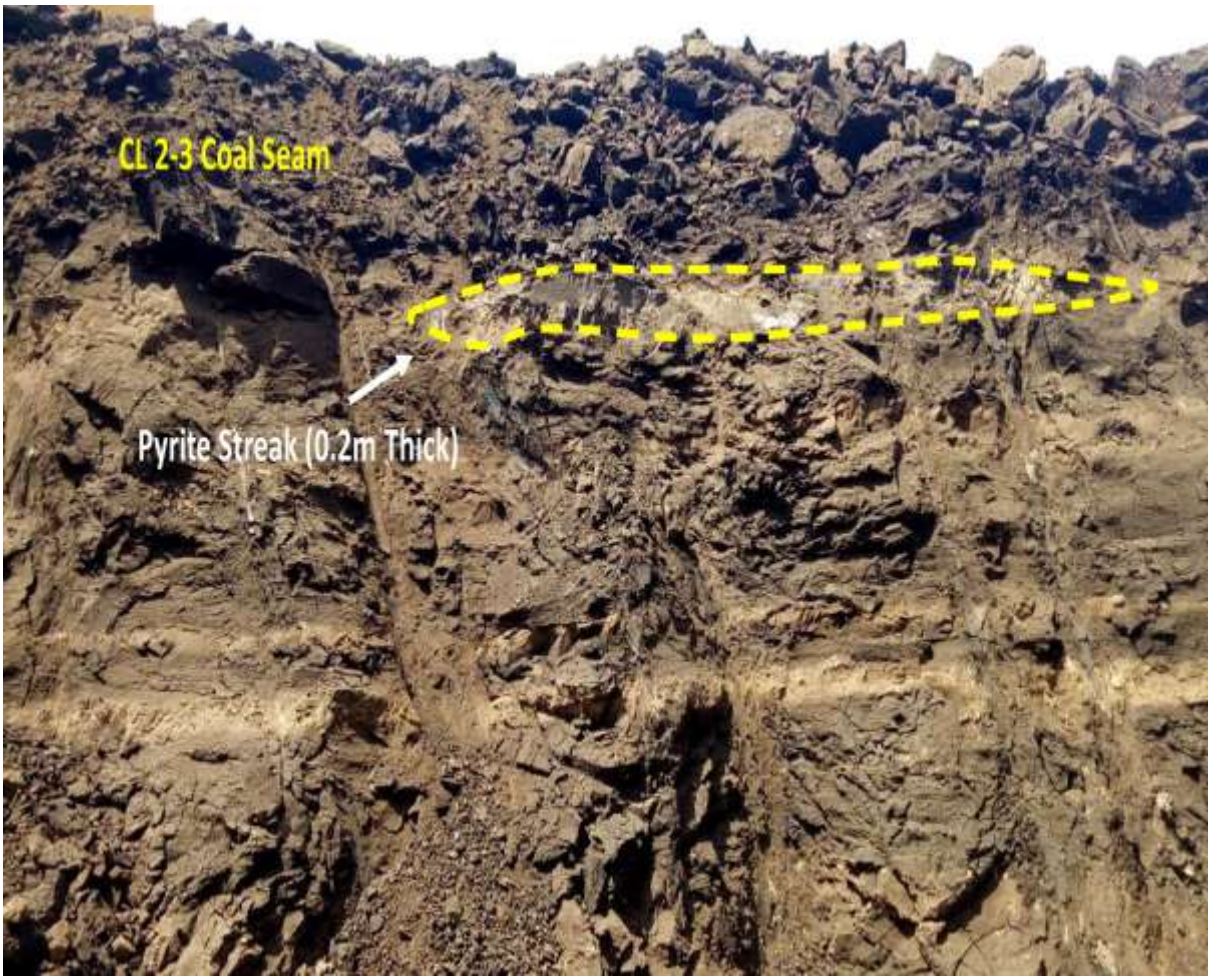

Fig. 03 Localized pyritic streak evident at the top 0.5m portion of CL 2-3 coal seam

The XRF analysis of pyrite mineral which is found in traces with coal at Thar coal mine disclose that it contains around 55% sulfur, 38% iron, 2% silica and 5% light elements. Pyrite mineralization is very common in coal deposits since coal formation mainly takes place in reducing environment. In the study area, pyrite mineralization is commonly found within coal seams where most pyritic streaks are evident at the upper 0.5m portion of CL 2-3 seam (Fig. 03) which is also confirmed from its relatively higher pyritic sulfur content (Table 01) as compared to other minable seams. Pyritic sulfur content is often higher in upper and lower portions of a seam than in the middle, perhaps implying coal sulfur content is influenced by roof or floor strata (Williams & Keith 1963, Gluskoter & Simon 1968, Gluskoter 1977). Pyritic streaks in CL 2-3 seam are very thin (max. 0.2m), non-continuous and have a very limited lateral extent (Fig. 03). In general, pyrite is considered to have a density of 4.9–5.2 g/cm3 (Wang, 1996), which is much higher than coal-derived pyrite, especially than fine-grained pyrite in coal (Wang et al., 2016).

4.3. Organic Sulfur

Field evidence and results from Table 01 indicates that most of the organic material in the form of pollen streaks is dominant in 1-2, 2-2 and 2-3 coal seams (Fig. 04). Since CL 2-3 is the thickest among these three seams, thus the geochemistry of pollen streaks in CL 2-3 was studied separately which is of vital importance in order to evaluate its influence in the overall

coal quality. For this purpose, pollen and pure coal samples were examined and the results were compared to confirm the organic origin of sulfur (Table 02). The results clearly suggest that all the pollen samples have very high ash, volatile and sulfur contents with quite low fixed carbon confirming that these pollens are the sign of unmatured coal. On the other hand, pure coal samples comprised of low ash, volatile and sulfur content along with high fixed carbon since it is clean and free from any impurities. The total moisture content in pollen samples is comparatively very low (~28%) whereas they have much higher calorific value (~18 MJ/Kg) than coal which is due to the fact that calorific value is believed to be directly proportional with sulfur and is usually triggered with increasing sulfur content. Much of the organic sulfur associated with plants and microorganisms are in the form of sulfur containing amino acids such as methionine (Freney, 1967).

Table 02 Geochemical analysis of CL 2-3 pollen and coal samples collected from Thar coal mine, Block II, Thar District, Sindh

ample #	**Sample Type**	**Total Moisture (%)**	**Ash.ar (%)**	**Volatile Matter.ar (%)**	**Fixed Carbon.ar (%)**	**Net Calorific Value.ar (MJ/Kg)**	**Total Sulfur.ar (%)**
01	Pollen	28.79	16.91	43.55	10.75	17.00	10.71
02	Pollen	26.66	14.78	46.22	12.34	18.51	9.78
03	Coal	48.12	4.64	25.82	21.42	11.94	0.79
04	Coal	49.70	6.47	24.40	19.42	11.38	1.22

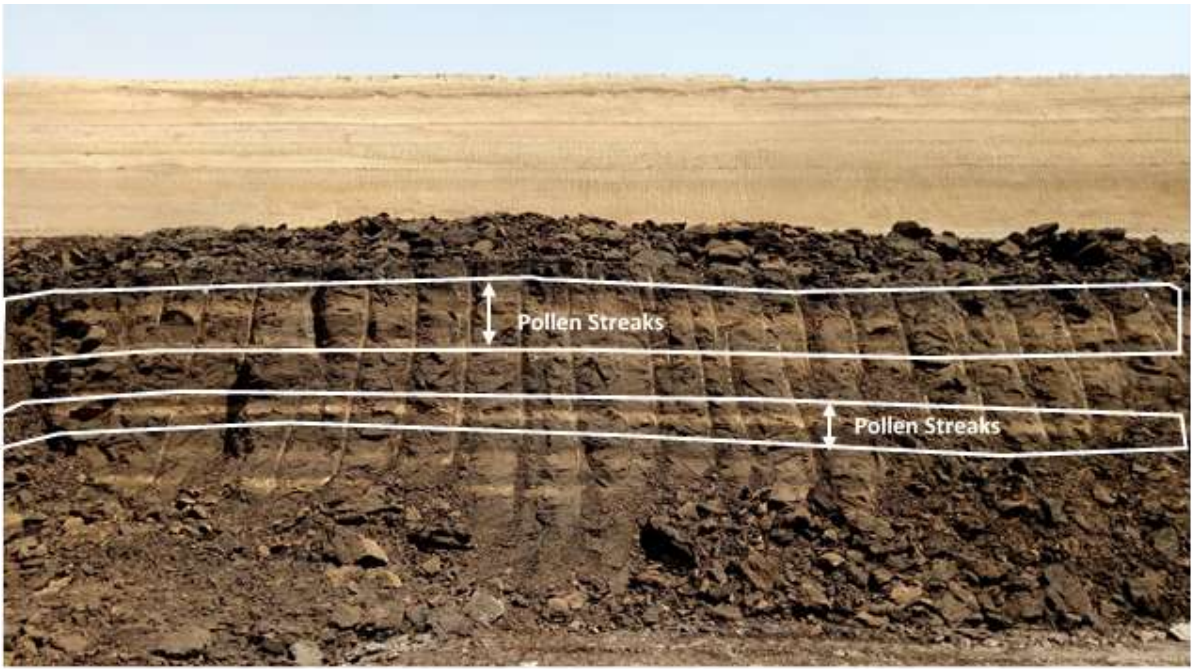

Fig. 04 Laterally consistent pollen streaks within CL 2-3 coal seam at Thar coal mine

SUMMARY AND CONCLUSIONS

Based on the results and analysis being carried out in the present research to evaluate the origin of sulfur in lignite deposit of Thar coal mine, it can be summarized that there are two main contributors of sulfur that basically represents its origin. One of them is inorganic; derived from pyrite while another one is organic; derived from plants.

In total, 06 coal seams were examined for sulfur analysis, 02 seams from non-minable category (1-2 and 2-2) and 04 from minable seams group (2-3, 2-4, 2-7 and 2-8). The results reveal that the total sulfur content decreased from top-most to bottom-most coal seam indicating the sulfur depends upon the depth of deposition i.e. shallower the coal seams, greater will be the sulfur content and vice versa. The highest amount of total sulfur was recorded in 1-2, 2-2 and 2-3 (shallower coal seams) though most of the total sulfur content was dominated by pyritic and organic sulfur in all seams. It was also revealed that nearly all the seams had greater percentage of pyritic sulfur than organic sulfur. This is due to the following reasons: (a) peat formation under marine

depositional environment generally tends to have higher pyritic sulfur; (b) the availability of ferrous ions during coalification had enhanced the formation of pyrite; (c) greater amount of insitu sulfur in pyrite (~55%) as compared to pollens (~10%) at study area resulted in relatively higher pyritic sulfur content in Thar coal seams.

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