Central European Geology, Vol. 55/3, 293–306 (2012)
DOI: 10.1556/CEuGeol.55.2012.3.4

Distribution of sulfur in power supply lignite from North Hungary

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The present article discusses the results of measurements carried out to assess the distribution of different sulfur types in lignite samples deriving from two opencast lignite mines near the villages of Bükkábrány and Visonta. These mines ensure the continuous supply of fuel for one of Hungary's largest thermal power plant. According to our findings no significant differences could be identified between the samples of the two mines based on their total sulfur (S_t) content. Both lignite types were classified as coals with medium–sulfur content according to the system of Chou (1990). A majority of total sulfur is accumulated in lignite, while in the intercalated carbonaceous shale total sulfur is present in minor amounts. Usually the sequence of the distribution of sulfur among the different bond forms in lignite collected from opencast mine of Visonta is as follows: pyritic sulfur (S_p) > organic sulfur (S_{org}) > sulfate sulfur (S_{SO_4}).

In the samples collected from Visonta and Bükkábrány quantities of total sulfur were similar. However, some difference in their distribution among various sulfur types were noted. Although half of the samples were weathered and the amount of pyrite sulfur must have been higher in the weathered lignite of Bükkábrány preceding the oxidation process, the sequence of the distribution of sulfur was likely as follows $S_{org} \geq S_p > S_{SO_4}$.

Key words: lignite, total sulfur, pyritic sulfur, sulfate sulfur, organic sulfur, Bükkalja Lignite Formation

Introduction

The Mátra Power Plant Co. Ltd., is Hungary's largest electricity producer with a lignite–fired power station, which produces as much as 13% of the Hungarian economy's electric energy consumption. The continuous supply of fuel is ensured

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Received: March 7, 2013; accepted: March 25, 2013

from two opencast lignite mines near the villages of Visonta and Bükkábrány. These productive areas in the southern foreland of the Mátra and Bükk Mountains belong to the Bükkalja Lignite Formation. One of the most significant pollutants of solid fuels (coal, lignite) burnt in thermal power plants is their sulfur content; however, retrofit systems have been installed and used in the last decade, reducing the emission of SO_X (Ludányi 2001). Knowledge of the abundance and nature of sulfur in solid fuels is important in their utilization because sulfur oxides released during solid fuel combustion can be a major cause of acid rain. The purpose of the present paper is to compare distribution of different sulfur types in power supply lignite from opencast mines of Visonta and Bükkábrány.

Sulfur compounds in coal

It is generally known that sulfur in coal is derived partly from original plant material and partly from ambient fluids in the coal-forming environment.

The total ratio of sulfur in coal varies in the range of 0.2 to 10% (Deurbrouck 1972). In most samples it is in the range of 1.0 to 4.0%. According to Smith et al. (1982), in Australian coal the total sulfur content is $< 1\%$ if deposition and coalification occurred in a freshwater environment, and is $>$ or » 1% if marine conditions prevailed during early diagenesis.

The sulfur in fossil fuels exists in inorganic (e.g. iron mono– and disulfides, sulfates) and organic form (Renton and Bird 1991). In coal the bulk of the inorganic S fraction is present as iron disulfides, mostly as pyrite (FeS_2 cubic) and from time to time marcasite (FeS_2 orthorhombic). Pyrite and marcasite are the most abundant minerals (e.g. Balme 1956; Berner 1984; Casagrande 1987). Other sulfide minerals may be present in minor quantities depending on the geochemical and mineralogical composition of the geologic deposit. Prolonged exposure of coal seams to rainwater, humidity and air causes pyrite oxidation to elemental sulfur (S^0), thiosulfate ($S_2O_3^{2-}$), polythionates ($S_nO_6^{2-}$), and eventually to sulfates (Evangelou 1995). Microorganisms are largely responsible for these oxidative processes.

Sedimentary pyrite is formed under anoxic conditions when sufficient decomposable organic matter, dissolved sulfate and reactive iron are present (Berner 1984). These conditions are fulfilled in several sedimentary environments. Sedimentary pyrite precipitates in a wide range of sedimentary environments from the time of sedimentation, through diagenesis, until metamorphism. Sedimentary pyrite formation usually takes place during early diagenesis, preceding significant burial and compaction, when pore systems can be regarded as prevailingly open. In anoxic, H_2S-bearing environments the distribution of pyrite is homogeneously dispersed (Hámor 1994).

The amount of sulfate is lower in freshwater than in seawater. Marine-influenced peat generally has a higher sulfur content than freshwater-influenced peat (Casagrande et al. 1977). During burial sediment sulfate supply is restricted, so in these circumstances pyrite does not form only via reduction of sulfate alone. According to Altschuler et al. (1983) pyrite formation in organic–rich sediment is commonly linked to H_2S formed within the organic tissues and suggests that the sulfide may derive largely from organic sulfur.

Sulfate sulfur is also generally of only minor importance in fresh coal samples and except in rare instances occurs in significant amounts only as the coals oxidize (weathering). It occurs mainly in the form of iron and calcium sulfates (Gluskoter 1977). In British and US coal the quantity of sulfate sulfur rarely exceeds 0.3% (Wandless 1955; Bhatia 1978; Casagrande 1987). The sulfate content of freshly mined coal is usually less than 0.1%, and it increases with time upon long–term exposure of coal to air (Walker and Hartner 1966; Kargi 1984). The presence of iron sulfates is generally an indication of coal weathering. Because pyrite and marcasite oxidize rapidly when exposed to moist air, a number of different phases of ferrous and ferric sulfates may form (Gluskoter and Simon 1968).

Organically-bound sulfur is primarily derived from two sources: the original organically bound plant sulfur preserved during the coalification process, and biogenic sulfides which reacted with organic compounds during the biochemical alteration of accumulated plant debris (Chambers and Trudinger 1979). The organically bound sulfur compounds in coal are believed to occur as aliphatic or aromatic thiols, sulfides or disulfides, thiophenes, and their numerous derivatives (Given and Wyss 1961; Dai 2000). Thiol and disulfide are likely secondary products because they are thermally rather unstable and would not survive the carbonization process (Tsai 1982). Organic sulfur can represent over 50% of the total sulfur content in some types of coal from the Illinois Basin (Chou et al. 2003).

Geologic setting

In general, the Pannonian Age sequence in the Carpathian Basin consists of the following lithostratigraphic units, reflecting the gradual infilling of the basin (Fig. 1). The basal Békés Conglomerate occurs mainly in the Makó and Békés Grabens. The conglomeratic beds are overlain by the Endrőd Marl and the Tótkomlós Calcareous Marl Member, which was deposited in a deep offshore environment. The oldest series of formations consists of basaltic volcanism in some places (e.g. Kecel Basalt F.). The lower part of the group is represented by the basal marl of the Endrőd Formation which is overlain or interfingered by the sandy turbidite–rich Szolnok Formation. The Algyő Formation (alternating clayey marl and sandstone) represents the sediments of the delta slope and on the margins of nearshore environments. Delta–front and delta-plain environments are indicated by the sandy Újfalu Formation. The sandy-clayey Zagyva Formation, which represents an alluvial–plain environment, interfingers with the paludal Bükkalja Lignite. Lignite and variegated clay with sandy intercalations are frequent. The

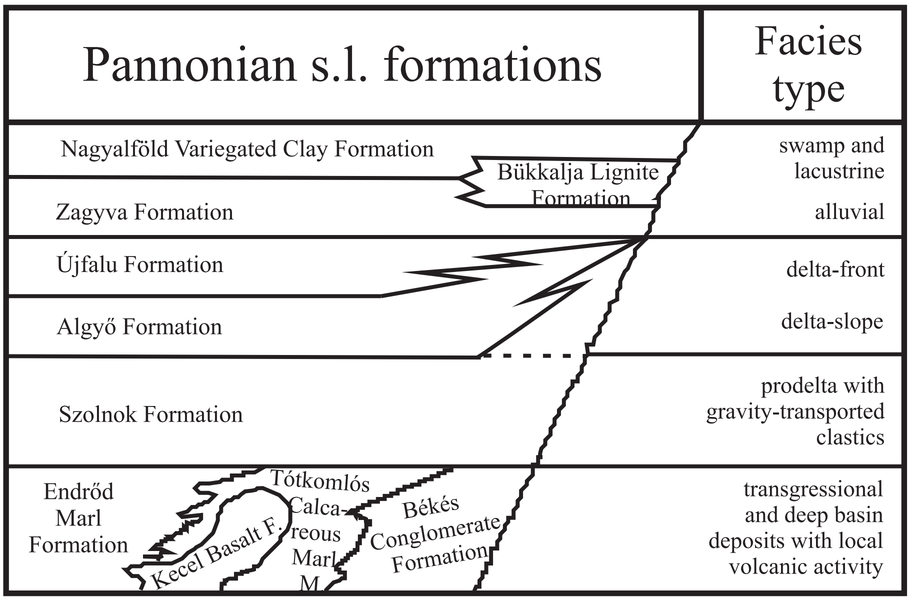

Fig. 1
Lithostratigraphy and facies types of the Pannonian in Hungary (Rasser et al. 2008, modified)

succession ends with the clay of the lacustrine–paludal Nagyalföld Variegated Clay Formation. It represents a typical lacustrine, fluvial and terrestrial sequence. Thickness: several hundreds of meters (Juhász 1991; Császár 1997; Juhász et al. 2006; Rasser et al. 2008).

Lignite formation took place during the Late Miocene (the Pannonian s.l.) between 8.07–7.43 Ma (Lantos et al. 1992), in the central part of the Pannonian Basin, as a result of continuous progradation of delta systems (Haas et al. 1999). Paleoecological studies based on both the macroflora and vertebrate fauna indicate that the overall climate of the Pannonian Basin at this time was favorable for lignite formation. The warm temperate climate was characterized by a mean annual temperature of about 15 ºC and precipitation of about 1000 mm (Bruch et al. 2006; van Dam 2006; Erdei et al. 2007). In the northern embayment of Lake Pannon, relatively thick (>2 m) lignite seams were deposited during the late phase of the aggradational period. These seams, together with the intercalated barren beds, comprise the Bükkalja Lignite Formation (Császár et al. 2009) (Fig. 2).

The productive area of the Bükkalja Lignite Formation is in the southern foreland of the Mátra and Bükk Mountains, of an E–W length of ca. 120 km and of a width varying from 10 to 30 km, where the two opencast mines of Visonta and Bükkábrány exploit raw material for electric power generation. This lignite region is the largest continuous coal area in Hungary.

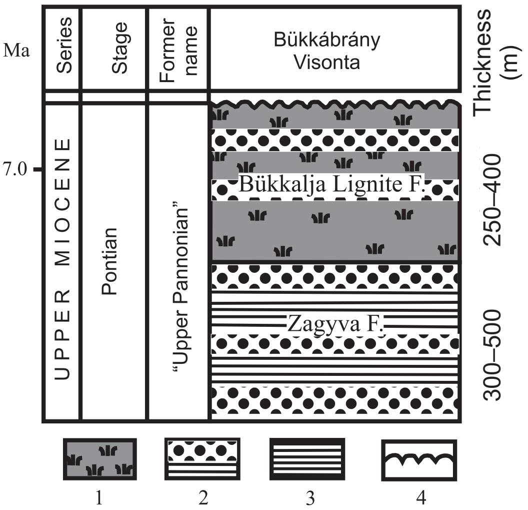

Fig. 2
Stratigraphic column of the Upper Miocene Bükkalja Lignite Formation. Legend: 1. lignite; 2. fluvial; 3. marl; 4. erosional unconformity (Hámor-Vidó and Hámor 2007, modified)

The thickness of the coal sequence is ca. 300–400 m, and the number of seams varies from 15 to 30. The average thickness of a lignite seam is 1.5 m; lignitic shale and clay intercalations are frequent. In the lignite seam the vitrinite reflectance varies from 0.27 to 0.29%. This lignite is of a light fibrous lithotype, where the woody texture is well recognizable, or medium–dark earthy and dull (Hámor–Vidó and Hámor 2007).

Lignite seams at Visonta are numbered from top to bottom. In the K–1 field the lignite seams No. I, II, and III were exploited and in the K-2 field are located seams No. 0, I, and II. Two significant seams (No. 0 and No. I) are located at Bükkábrány; the main lignite seam No. 0 is the one being exploited.

Samples and analytical methods

Altogether 18 samples of lignite and intercalated carbonaceous shale were collected from the section of lignite seam No. 0 of the Visonta eastern field K-2 (Fig. 3). The total thickness of the studied sections in Visonta was ca. 10 m. Samples were placed into double plastic bags and a portable freezer box in the field and were kept at –20 °C in the lab until the beginning of analyses. First the samples were pulverized to ≈ 200 μm size in a ball agate mill, and then their moisture and ash contents were determined immediately. Sulfur content was measured as total (S_t) sulfate (S_{SO_4}) and sulfidic phase (S_p).

The total sulfur (S_t) determination was carried out with the Eschka procedure according to Hungarian standard MSZ 18000/12–72. The Eschka procedure consists of thoroughly mixing coal with Eschka mixture (2 parts calcinated MgO and 1 part anhydrous Na_2CO_3) and ashing it in a muffle furnace at 800° C. The ashed coal is leached with hot water, filtered and the sulfur is converted into $BaSO_4$ and weighed gravimetrically.

The acid-soluble sulfate sulfur (S_{SO_4}) was recovered as $BaSO_4$ using HCl and $BaCl_2$ in the separation method, according to Hungarian standard MSZ 18000/12–72.

Pyritic sulfur (S_p) was determined by the following method. After hydrochloric acid extraction (determination of sulfate sulfur) the residue was oxidized with H_2O_2 and the pyritic sulfur was converted into $BaSO_4$ with $BaCl_2$ and weighed gravimetrically. For a detailed description see Pápay (2001). In every case the organic sulfur (S_{org}) was determined by the difference between the total sulfur and inorganic sulfur.

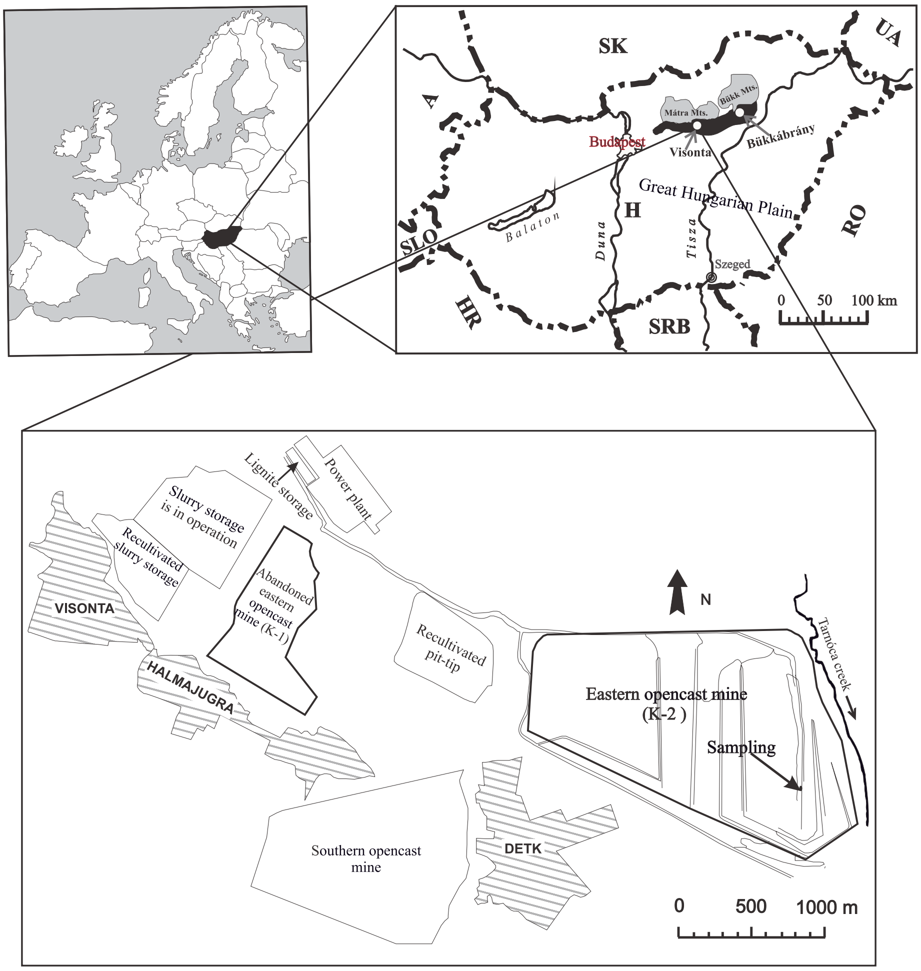

Fig. 3
Location and sampling site of the studied area

The total carbon (C_t) content was measured in samples free of carbonate, at 1000 °C under intense oxygen flow by combustion in Carmhograph–8 (Wösthoff) equipment. CO_2 content of the samples was determined by the gas-volumetric method. These analytical methods are presented in previously published studies (Pápay 1995, 1997). The carbon content (C_{carb}) data of carbonate bond forms were calculated from the CO_2 content. The total organic carbon content (TOC) is determined by the difference between C_t and C_{carb}.

Results and discussion

Measurement results of samples from lignite seam No. 0 of Visonta opencast mine K-2 are summarized in Table 1. Particularly the ash content (range 4.2–73.5%) as well as TOC (range 2.7–50.7%) displayed large fluctuations in the samples, implying different quantities of inorganic and organic component in them. Thus, for adequate evaluation samples were artificially divided into two major groups (see Table 2):

1) Relatively high carbon (mean TOC: 36.7%) and relatively low inorganic content (mean A^d: 14.8%): lignite group.

2) High ash content (mean A^d: 65.3%) and low carbon content (mean TOC: 8.4%): inorganic-rich (carbonaceous shale) group.

This division is naturally theoretical as both types are exploited together and fired in the power plant. There seems to be significant differences in the sulfur content of the two artificially created groups. The total sulfur content was higher in samples of the lignite group (average S_t: 1.85%) than in the inorganic-rich group where mean S_t is 0.4%.; i.e. more sulfur accumulated in the relatively high carbon content lignite then in the less useful carbonaceous shale.

Samples were further subdivided according to the classification of Chou (1990), considering the level of sulfur content in the coal: a) low-sulfur coal that contains less than 1% total sulfur, b) medium-sulfur coal that contains 1–3% total sulfur, and c) high-sulfur coal that contains more than 3% total sulfur. The lignite from seam No. 0 of Visonta belongs to subcategory b, that is, low-rank coal with moderate sulfur content. The moisture in lignite is fairly high (mean value: 49.6%); therefore its average total sulfur content >3% on dry, ash-free basis, indicating that it belongs to the organic facies, which is very rich in sulfur. The sequence of the distribution of sulfur among the different bond forms in lignite from the K-2 exploitation is as follows: pyritic sulfur > organic sulfur > sulfate sulfur. This trend has also been observed in the majority of the inorganic-rich samples (see Table 1).

Table 3 presents parameters of samples previously collected from lignite seam No. I of the former Visonta K–1 mine (unpublished to date) and those from the work of Pápay (1997) on samples from open air lignite mine of Bükkábrány, for comparison.

Table 1
Distribution of sulfur varieties in samples from lignite seam No. 0 of Visonta pit K-2, including data for moisture, ash, carbonatic carbon content and organic carbon content

Number of samples	W^a %	A^d %	C^a_{carb} %	TOC %	S^a_t %	S^a_p %	$S^a_{SO_4}$ %	S^a_{org} %	Remark
1	51.6	15.3	0.04	46.9	1.95	1.20	0.12	0.63	
2	53.4	9.7	0.07	42.1	1.80	1.39	0.07	0.34	
3	46.5	60.6	0.04	2.7	0.65	0.38	0.14	0.13	carbonaceous shales
4	47.9	20.6	0.03	36.6	2.59	2.01	0.16	0.42	
5	51.5	11.6	0.04	46.0	2.49	1.93	0.11	0.45	
6	42.2	66.0	0.04	15.4	0.56	0.29	0.00	0.27	carbonaceous shales
7	52.7	15.6	0.11	43.4	2.07	1.26	0.12	0.69	
8	41.9	57.7	0.04	11.3	0.44	0.27	0.00	0.17	carbonaceous shales
9	50.5	15.2	0.03	48.2	1.58	1.05	0.03	0.50	
10	37.7	73.5	0.07	4.1	0.16	0.09	0.00	0.07	carbonaceous shales
11	52.1	12.3	0.07	47.9	3.38	3.14	0.12	0.12	
12	38.2	69.1	0.05	7.4	0.38	0.23	0.00	0.15	carbonaceous shales
13	34.7	64.9	0.03	9.5	0.19	0.04	0.00	0.15	carbonaceous shales
14	48.5	4.2	0.02	50.7	0.51	0.11	0.00	0.40	
15	45.5	19.4	0.02	20.1	1.87	1.26	0.21	0.40	
16	45.8	16.8	0.03	20.7	1.49	0.85	0.13	0.51	
17	48.0	18.9	0.03	19.2	1.28	0.67	0.09	0.52	
18	47.7	18.0	0.02	19.4	1.24	0.65	0.08	0.51	

W^a: analitical moisture wt%; A^d: ash (air-dried sample) wt%; C^a_{carb} carbonatic bond forms carbon content in air-dried sample wt%; TOC: total organic carbon wt%;
S^a_t, S^a_p, $S^a_{SO_4}$, S^a_{org}: total, pyritic (+ sulfide), sulfate, organic (by difference) sulfur content in air-dried sample wt%

Table 2
Range and mean of different sulfur varieties in samples from lignite seam No. 0 of Visonta field K-2 including data for moisture, ash, carbonatic carbon and total organic carbon

Number of samples	W^a %	A^d %	C^a_{carb} %	TOC %	S^a_t %	S^a_p %	$S^a_{SO_4}$ %	S^a_{org} % (diff.)	Remark
18	34.7–53.4 46.5	4.2–73.5 31.6	0.02–0.11 <0.05	2.7–50.7 27.3	0.2–3.4 1.4	<0.1–3.1 0.95	0.0–0.2 0.1	<0.1–0.7 0.35	
12	45.5–53.4 49.6	4.2–20.6 14.8	0.02–0.11 0.04	19.2–50.7 36,7	0.5–3.4 1.85	0.1–3.1 1.3	<0.1–0.2 0.1	0.1–0.7 0.45	lignite
6	34,7–46.5 40.2	57.7–73.5 65.3	0.03–0.07 0.045	2.7–15.4 8.4	0.2–0.7 0.4	<0.1–0.4 0.2	0.0–0.1 < 0.05	<0.1–0.3 0.16	carbonaceous shales

Table 3
Range and mean of different sulfur varieties in lignite from seam No. I of Visonta former field K-1 including data for moisture, ash, carbonatic carbon, total organic carbon and similar data from lignite seam No. I of Bükkábrány (Pápay 1997)

Locality (Remark)	Number of samples	W^a %	A^d %	C^a_{carb} %	TOC %	S^a_t %	S^a_p %	$S^a_{SO_4}$ %	S^a_{org} % (diff.)
Visonta	7	40.1–52.9 45.6	8.9–17.7 14.5	0.03–0.1 0.06	25.4–32.2 28.2	1.0–2.1 1.4	0.5–1.3 0.7	0.1–0.3 0.2	0.4–0.8 0.5
Bükkábrány	12	30.9–50.3 40.5	5.3–24.8 12.8	<0.1–0.1	23.0–40.5 29.5	0.6–2.3 1.3	<0.1–1.0 0.3	<0.1–1.0 0.45	0.3–1.2 0.55
Bükkábrány (lignites)	6	36.7–50.3 42.8	5.9–24.8 13.0	<0.1–0.1	23.8–36.7 29.3	0.6–1.7 1.0	<0.1–1.0 0.3	<0.1–0.3 0.2	0.4–0.6 0.5
Bükkábrány (lignites oxidized or weathered)	6	30.9–49.8 38.2	5.3–17.4 12.6	<0.1–0.1	23.0–40.5 29.8	1.2–2.3 1.6	0.2–0.6 0.3	0.6–1.0 0.7	0.3–1.2 0.6

W^a: analytical moisture wt%; A^d: ash (air-dried sample) wt%; C^a_{carb} carbonatic bond forms carbon content in air-dried sample wt%; TOC: total organic carbon wt%;
S^a_t, S^a_p, $S^a_{SO_4}$, S^a_{org}: total, pyritic (+ sulfide), sulfate, organic (by difference) sulfur content in air-dried sample wt%

Lignite samples were collected from two distinct seams (No. 0 and No. I) of Visonta, yet only minor differences could be observed in their carbon and sulfur contents. The average TOC in samples from lignite seam No. 0 was 36.7% while in those from seam No. I it was 28.2%. The corresponding average total sulfur values were S_t = 1.85% and S_t = 1.4%, respectively. More sulfur accumulated in lignite, with its higher total organic carbon content, then in samples with lower TOC. In both lignite seams the distribution of sulfur among the different bond forms is the same. The sequence is as follows: pyritic sulfur > organic sulfur > sulfate sulfur. Pyritic and organic sulfur account for the bulk of sulfur in the lignite of Visonta; sulfate sulfur occurs only in small amounts, with average values of 0.1 and 0.2%.

Samples from lignite seam No. I of Bükkábrány tend to display good correlation in their measured average geochemical parameters with the lignite from seam No. I of Visonta K-1 field (see Table 3). The distance between two the opencast lignite mines is ca. 60 kilometers. There is some difference in the distribution of various sulfur types between samples collected from Visonta and Bükkárány. In the Bükkábrány lignite organic sulfur exceeds sulfide sulfur content and $S_{SO_4} > S_p$. The sequence is as follows: $S_{org} > S_{SO_4} > S_p$, in numerical value S_{org} 0.55%, 0.45%, S_p 0.3%. The range of sulfate sulfur content in half of the Bükkábrány lignite samples is 0.6 to 1.0% with an average of 0.7%. In comparison, in the other half of the Bükkábrány samples the quantity of sulfate sulfur is small, with an average value of 0.2%. The relatively high sulfate concentration of lignite indicates that these samples are weathered or oxidized. There is general agreement in the literature (e.g. Gluskoter and Simon 1968; Smith and Batts 1974) that a part of the ferrous and ferric sulfates derive from inorganic disulfides. Sulfate is usually formed as result of chemical and biological oxidation of pyrite in the presence of air. The sulfate present in samples is secondary in origin and resulted from the weathering of iron sulfides (e.g. Smith and Batts 1974; Kargi 1984). It is very likely that despite careful sampling and storage, these lignite samples were exposed to weathering for too long a time in the mine or during oxidation in the laboratory.

These weathered samples were treated separately in the evaluation from the rest of the samples. Small differences could be observed in the total sulfur content between the two groups. In half of the lignite samples from Bükkábrány the quantity of total sulfur (mean value: 1.6%) was higher than in another group, where the mean value was 1.0%.

The weathered lignite group with higher total sulfur content was also characterized by higher S_{SO_4} values. In these lignite samples sulfate sulfur and organic-bond sulfur content dominate, and pyrite is the lowest in quantity. The sequence is as follows: $S_{SO_4} > S_{org} > S_p$, with mean values of 0.7% > 0.6% > 0.3%.

In the remaining samples of Bükkábrány lignite the sequence of the distribution of sulfur is as follows: $S_{org} > S_p > S_{SO_4}$; mean values are 0.5% > 0.3% > 0.2%. Moreover, a third sequence is observed in the averages of all the samples as well: $S_{org} > S_{SO_4} > S_p$, with mean values of 0.55% > 0.45% > 0.3%. Due to the low number of samples, it was not possible to determine the exact sequence of the distribution of sulfur in the Bükkábrány lignite. The amount of pyrite sulfur must have been higher in the weathered lignite of Bükkábrány preceding the oxidation (weathering) process. It is likely that there is a difference in distribution of sulfur in lignite from Visonta and that of Bükkábrány, but the verification of this issue requires further investigation.

It should be noted that no significant differences between the samples of the two artificially created groups were observed regarding their total organic content, with mean values of 29.8% and 29.3%, respectively. Therefore the relationship between the TOC and total sulfur content is not as obvious here as in case of the lignite of Visonta mentioned earlier.

Conclusions

The bulk of the sulfur content in the studied area of the opencast mine of Visonta is found in the low-rank coal-bearing seams. The sulfur content was higher in the relatively high carbon-bearing lignite than in the intercalated carbonaceous shale. The lignite samples were collected from two distinct lignite seams of Visonta, but only small differences could be observed in their carbon and sulfur content. The higher total organic carbon content lignite contained more sulfur then the samples with lower TOC. The distribution of sulfur among the different bond forms was the same in both lignite seams. Pyritic and organic sulfur account for the bulk of sulfur in Visonta lignite; sulfate sulfur occurs only in a small amount. The sequence is as follows: pyritic sulfur > organic sulfur > sulfate sulfur.

Although the quantities of total sulfur were similar in the samples collected from Visonta and Bükkábrány, there is some difference in the distribution among various sulfur types. Although half of the samples were weathered and the amount of pyrite sulfur must have been higher in the weathered lignite of Bükkábrány prior to the oxidation process, the sequence of the distribution of sulfur was likely as follows $S_{org} \geq Sp > S_{SO_4}$.

In both study areas the process of peat deposition occurred in the lower delta plain of the Pannonian Basin (Hámor-Vidó and Hámor 2007). The above-mentioned slight difference in distribution of sulfur among the different bond forms is less significant than what was observed during the analysis of the samples from the opencast mines of Visonta and Ilnice.

In our previous study lignite formations were studied in two locations in the central and distal part of the delta environment from the Late Miocene (Pannonian s.l.) Bükkalja Lignite Formation (BL), at Visonta the open–pit mine (Hungary) and the Trans-Carpathian Ilnice (IN) open-pit mine (Ukraine). The total sulfur content shows a significant difference between the two sites. In the central part of the Pannonian Basin BL, the average S_t is higher than in IN.

Although pyrite sulfur values at BL and IN indicate different peat–forming environments, higher values are found in the central delta system due to higher salinity, while lower values were recorded in the distal freshwater part of the delta system. Nevertheless the composition and preservation of organic matter is similar (Hámor-Vidó et al. 2003; Ivanova et al. 2004).

Acknowledgement

The author would like to express his appreciation to Dr. Mária Hámor Vidó, geologist and mining engineer, for her generous help in sampling and access to the mine in Visonta.

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